

On the temperature model of CO₂ lasers

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Abstract. A refined temperature model of CO₂ lasers is presented, which takes into account the fact that vibrational modes of the CO₂ molecule have the common ground vibrational level. New formulas for the occupation numbers and the vibrational energy storage in individual modes are obtained as well as expressions relating the vibrational temperatures of the CO₂ molecules with the excitation and relaxation rates of lower vibrational levels of modes upon excitation of the CO₂–N₂–He mixture in an electric discharge. The character of dependences of the vibrational temperatures on the discharge current is discussed.

Keywords: temperature model, vibrational temperature, cw CO₂ laser, active medium, electric discharge, relaxation.

1. Introduction

The energy parameters of CO₂ lasers of different types are calculated by using both level and temperature models (see, for example, [1–5]). It is well known that the most correct approach to the solution of this problem consists in writing and solving a system of kinetic equations for the populations of the vibrational levels, which play a substantial role in producing inversion at laser transition levels under concrete conditions of the active medium, i.e. to use the level model. The more levels are considered, the more accurate are these calculations. However, it is also known that the application of this approach is limited by the lack of information on the relaxation channels, the excitation and relaxation rates, and their temperature dependences for the majority of vibrational levels of the CO₂ molecule, especially for those levels, which are located above the 00⁰1 level. For this reason, the level models are only rarely applied to calculate the parameters of high-power technological CO₂ lasers operating on the 00⁰1–10⁰0 laser transition.

Such systems are usually calculated with the help of the temperature model [1–4]. The model is based on relations between the characteristic relaxation times of the CO₂ molecule in the active medium: $\tau_{R-T} < \tau_{V-V} < \tau_{V-T} < \tau_{V-V'}$, where τ_{R-T} , τ_{V-V} , τ_{V-T} , $\tau_{V-V'}$ are the times of the rotational–translational, intramode vibrational–vibrational, vibrational–translational and intermode vibrational–vibrational relaxations, respectively [1]. The vibrational modes ν_1 , ν_2 and ν_3 of the CO₂ molecule and the mode ν_4 of the N₂ molecule are considered as harmonic oscillators. The population distribution over vibrational levels of each of these modes is described by the Boltzmann distribution with its own vibrational temperature T_i ($i = 1, 2, 3, 4$). Therefore, knowing the vibrational temperatures of modes, we can determine the population of any vibrational level of CO₂ and N₂ molecules and the vibrational energy E_i stored in modes. The kinetic equations in the temperature model of CO₂ lasers are usually written for the energy E_i or the occupation numbers e_i of modes equal to the number of vibrational quanta in each mode per one molecule [1–4].

The disadvantage of the existing temperature model is the fact that the CO₂ molecule here is considered as a set of three independent harmonic oscillators, whose energies are determined only by their own vibrational temperatures, these vibrational temperatures being treated as independent parameters [2–4]. The temperature model in this form can be used only for the qualitative description of the active media of CO₂ lasers upon weak excitation of vibrational modes ν_1 and ν_2 , i.e. when the vibrational temperatures T_1 and T_2 are small and do not differ considerably from the gas-kinetic temperature T , which is also small. However, in the active media of real CO₂ lasers, especially in the active media of high-power technological systems characterised by the large level of the energy input, this condition is not fulfilled. It was found experimentally that the temperature of the active medium from 600 to 700 K corresponds the maximum output power of CO₂ lasers (see, for example, [6–8]). Therefore, the vibrational temperatures T_1 and T_2 in this case are even higher and the use of the existing temperature model can lead to great errors in calculations of parameters of real CO₂ lasers and incorrect treatment of physical processes proceeding in such active media.

In this paper, we present the refined temperature model of CO₂ lasers, which takes into account the fact that vibrational modes of the CO₂ molecule are related with each other not only by the processes of intermode relaxation but also by their common ground vibrational level.

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Received 13 February 2006; revision received 3 October 2006
Kvantovaya Elektronika 37 (3) 243–247 (2007)
Translated by I.A. Ulitkin

2. Occupation numbers and storage of the vibrational energy in the vibrational modes of N₂ and CO₂ molecules

In the temperature model, the N₂ molecule is simulated by a harmonic oscillator in which the Boltzmann distribution is realised over the vibrational levels with the vibrational temperature T_4 . The population density N_{v_n} of any vibrational level characterised by the vibrational quantum number v_n is described by the expression:

$$N_{v_n} = \frac{N_N}{Q_N} X_4^{v_n}, \quad (1)$$

where N_N is the concentration of N₂ molecules; $X_4 = \exp(-hv_4/kT_4)$; hv_4 is the vibrational quantum of the N₂ molecule;

$$Q_N = \sum_{v_n=0}^{\infty} X_4^{v_n} = (1 - X_4)^{-1} \quad (2)$$

is the vibrational statistical sum. It follows from (1) that the vibrational temperature T_4 is determined by the ratio of populations of any two adjacent vibrational levels of the N₂ molecule.

The vibrational energy stored in the N₂ molecule is

$$E_4 = \sum_{v_n=0}^{\infty} v_n hv_4 N_{v_n}. \quad (3)$$

By substituting (1) in (3) and taking into account (2), we have

$$E_4 = N_N hv_4 \frac{X_4}{1 - X_4}. \quad (4)$$

The occupation number of the v_4 mode is

$$e_4 = \frac{E_4}{N_N hv_4} = \frac{X_4}{1 - X_4}. \quad (5)$$

One can see from (1), (4) and (5) that both the relative and absolute values of the population of the vibrational levels of the N₂ molecule and the vibrational energy storage in this molecule or the occupation number of the mode v_4 are determined by one parameter – the vibrational temperature T_4 .

The vibrational modes of the CO₂ molecule, as was mentioned above, are simulated by harmonic oscillators, in each of which the Boltzmann distribution is realised over the vibrational levels with their own vibrational temperatures T_1 , T_2 , and T_3 . The population of any vibrational level characterised by the vibrational quantum numbers v_1 , v_2 and v_3 , is described by the expression [1–4]

$$N_{v_1, v_2, v_3} = \frac{N_C}{Q_C} X_1^{v_1} (v_2 + 1) X_2^{v_2} X_3^{v_3}, \quad (6)$$

where N_C is the concentration of CO₂ molecules; $X_i = \exp(-hv_i/kT_i)$; hv_i is the vibrational quantum of the i th mode; and

$$Q_C = \sum_{v_1} X_1^{v_1} \sum_{v_2} (v_2 + 1) X_2^{v_2} \sum_{v_3} X_3^{v_3} =$$

$$= (1 - X_1)^{-1} (1 - X_2)^{-2} (1 - X_3)^{-1} \quad (7)$$

is the vibrational statistical sum for CO₂ molecules.

It follows from (6) that the vibrational temperature of any mode of the CO₂ molecule, as in the case of the N₂ molecule, is determined by the ratio of the populations of any two adjacent levels of this mode. For the occupation numbers of the modes v_1 , v_2 and v_3 , expressions, similar to expression (5) (see [1–4]) are used

$$e_1 = \frac{X_1}{1 - X_1}, \quad e_2 = \frac{2X_2}{1 - X_2}, \quad e_3 = \frac{X_3}{1 - X_3}. \quad (8)$$

(Additional factor in the expression for e_2 is related to the double degeneracy of the deformation mode.)

The energy of the CO₂ laser is determined by the storage of the vibrational energy in the antisymmetric mode v_3 of the CO₂ molecule and in the mode v_4 of the N₂ molecule, which in the approximation $hv_4 \approx hv_3$ and $X_4 \approx X_3$, is usually written in the form:

$$E_3 + E_4 \approx (N_C + N_N) hv_3 \frac{X_3}{1 - X_3}. \quad (9)$$

Therefore, in the existing temperature model, both the relative population distribution of any two adjacent levels of each vibrational mode of the CO₂ molecule and the storage of the vibrational energy in each mode or the occupation number of this mode, as in the case of the N₂ molecule, are determined by only one parameter – the corresponding vibrational temperature. However, to find the storage of the vibrational energy in the mode, one should know not relative but absolute values of the populations of the vibrational levels of this mode, which, as follows from (6), are determined by all the three vibrational temperatures. Therefore, the storage of the vibrational energy in any mode of the CO₂ molecule should be also determined by all the three vibrational temperatures. This is explained by the fact that all the three modes have the same ground vibrational 00⁰0 level of the CO₂ molecule. Indeed, it follows from (6) that populations of the first excited vibrational levels of each mode of the CO₂ molecule can be written in the form:

$$\begin{aligned} N_{10^0 0} &= N_{00^0 0} \exp(-hv_1/kT_1) = N_{00^0 0} X_1, \\ N_{01^1 0} &= 2N_{00^0 0} \exp(-hv_2/kT_2) = 2N_{00^0 0} X_2, \\ N_{00^0 1} &= N_{00^0 0} \exp(-hv_3/kT_3) = N_{00^0 0} X_3. \end{aligned} \quad (10)$$

One can see from (10) that any change in the population $N_{00^0 0}$ of the ground vibrational state 00⁰0 leads to a change in all the three vibrational temperatures $T_1 - T_3$, and, hence, the vibrational temperatures of the CO₂ molecule are interdependent parameters. By using (6) and taking (7) into account, we obtain

$$\begin{aligned} E_3 &= \sum_{v_3=0}^{\infty} (v_3 + 1) hv_3 N_{00^0 v_3} \\ &= N_C hv_3 (1 - X_1)(1 - X_2)^2 \frac{X_3}{1 - X_3} \end{aligned} \quad (11)$$

and

$$e_3 = (1 - X_1)(1 - X_2)^2 \frac{X_3}{1 - X_3}. \quad (12)$$

Similarly

$$e_1 = (1 - X_2)^2(1 - X_3) \frac{X_1}{1 - X_1} \quad (13)$$

and

$$e_2 = (1 - X_1)(1 - X_3) \frac{2X_2}{1 - X_2}. \quad (14)$$

The total vibrational energy stored in the antisymmetric mode ν_3 of the CO₂ molecule and in the ν_4 mode of the N₂ molecule, in the approximation $h\nu_4 \approx h\nu_3$ and $X_4 \approx X_3$, is determined by the expression

$$E_3 + E_4 \approx [N_C(1 - X_1)(1 - X_2)^2 + N_N]h\nu_3 \frac{X_3}{1 - X_3}. \quad (15)$$

The important difference of expressions (12) – (14) from expression (8) used earlier is that the occupation numbers of each mode of CO₂ are determined not by one vibrational temperature of this mode but by the vibrational temperatures of all the three modes.

Consider the CO₂:N₂ ≈ 1:1 mixture used in cw electric-discharge CO₂ lasers. It is easy to verify that according to (12) and (8) the same storage of the vibrational energy corresponds to different vibrational temperatures T_3 , expression (8) yielding lower value of T_3 than (12). If these data are used together with the experimentally measured gain, the understated values of T_3 lead directly to understated values of the vibrational temperatures T_1 and T_2 . The improperly determined vibrational temperatures result, in turn, in misunderstanding of physical processes proceeding in the active medium and leading to the establishment of these temperatures [9]. This is especially important while considering the dependences of the CO₂-laser parameters on the pump energy when all the vibrational temperatures increase with increasing this energy. Let us illustrate it by examples.

Let the occupation number of the antisymmetric mode be $e_3 = 0.2262$ for a certain energy input in the discharge, which according to (8) corresponds to the vibrational temperature $T_3 \approx 2000$ K. If we take into account that for this T_3 the vibrational temperatures of modes ν_1 and ν_2 close to $T_1 \approx 600$ K and $T_2 \approx 500$ K can be realised (see, for example, [9, 10]), expression (12) gives the temperature $T_3 \approx 2396$ K for this occupation number of the mode ν_3 . Therefore, the difference in T_3 from (8) and (12) is $\Delta T_3 \approx 400$ K. ΔT_3 also increases with increasing the energy input, when the occupation numbers of all the modes increase. Thus, for example, for $e_3 = 0.4795$ expression (8) gives $T_3 = 3000$ K. But, if we take into account that T_1 and T_2 also increase in this case (for example, $T_1 = 700$ K and $T_2 = 600$ K), we obtain from (12) $T_3 \approx 4280$ K and $\Delta T_3 \approx 1280$ K. What vibrational temperatures T_3 can be achieved in the active media of e-beam sustained discharge CO₂ lasers with increasing the energy input in the discharge?

According to [10–16] the temperature T_3 measured by different methods is saturated both in the cw and pulsed

discharges. Referring to the experimental results, authors of [16] made a conclusion about the existence of some fundamental restriction of the value T_3 in the e-beam sustained discharge CO₂ lasers, which does not allow obtaining $T_3 > 2000 - 3000$ K (depending on the mixture composition). They consider this fundamental restriction to be the mechanism of de-excitation of the vibrational 00^0_1 level of the CO₂ molecule by the electron impact and even found the cross section of this process. However, the fact that different values of maximum T_{ν_3} were obtained in these papers under different excitations conditions and for different compositions of the active medium, and it was reported in [17] that $T_3 = 3400$ K was achieved, indicates the absence of this fundamental restriction.

3. Vibrational temperatures of the CO₂ molecule upon excitation of the CO₂ – N₂ – He mixture in the electric discharge

CO₂ molecules are excited in the electric discharge in collisions with the discharge electrons. It is known that in this case not one degree of freedom of the CO₂ molecules is excited but many – translational, rotational, vibrational degrees of freedom and molecules undergo transitions to the excited electronic states, and can be ionised and dissociated. The fraction of the electron energy transferred to different degrees of freedom of the molecule during their collisions depends on the value E/N , where E and N are the electric field strength and the total concentration of particles in the discharge. According to [18, 19], even when the energy distribution of electrons is optimal for excitation of the ν_3 mode, the ν_1 and ν_2 modes are also excited, and the gas temperature increases. Figure 1 presents the diagram of lower vibrational levels and processes of their excitation and relaxation considered in

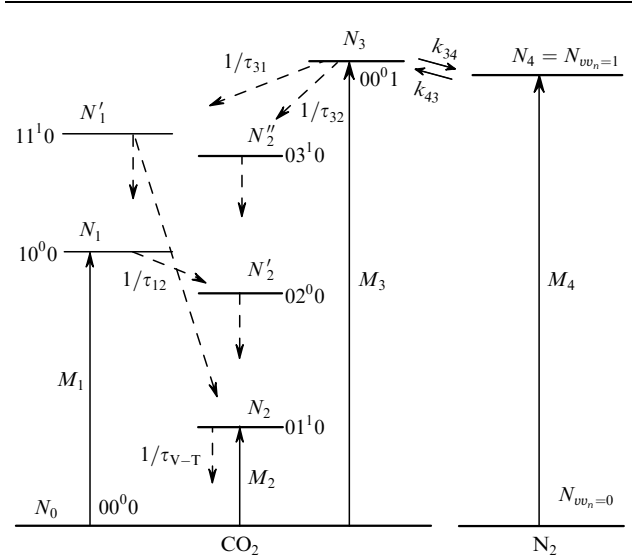


Figure 1. Diagram of the low vibrational levels of the CO₂ and N₂ molecules. Processes of excitation (solid arrows) and relaxation (dashed arrows) used in the kinetic model of the electric-discharge CO₂ laser are shown; N_i and M_i are the populations and the excitation rates by the electron impact of the corresponding vibrational levels; τ_{ij} is the time of the population relaxation of the i th level of CO₂ over different channels; k_{34} and k_{43} are the rate constants of the direct and inverse processes of the vibrational energy exchange between the molecules CO₂ and N₂.

the model. We believe that the $10^0 0$ level is mainly excited from the ground state by the electron impact, while the $00^0 1$ level is excited both by the direct electron impact and quasi-resonance transfer of the vibrational energy from excited N_2 molecules. The model takes into account that the population of the excited vibrational $00^0 1$ level relaxes over two channels: first, through the combination $11^1 0$ level and then $10^0 0$ and $01^1 0$ levels, and, second, through the $03^1 0$ level of the deformation mode. Therefore, $1/\tau_{3\Sigma} = 1/\tau_{31} + 1/\tau_{32}$. Here $\tau_{3\Sigma}$ is the lifetime of the $00^0 1$ level. In turn, the relaxation of the $10^0 0$ level also occurs via the deformation mode. This leads to the additional depopulation of the ground vibrational state due to the collisional relaxation almost by the same value as in the case of electron-impact excitation. In other words, the collisional relaxation nearly doubles the number of CO_2 molecules excited from the ground state to the higher vibrational levels. The corresponding system of kinetic expressions has the form [20]

$$\begin{aligned} \frac{dN_3}{dt} &= M_3 - \frac{N_3}{\tau_{3\Sigma}} + k_{43}N_4N_0 - k_{34}N_3N_{v_n=0}, \\ \frac{dN_4}{dt} &= M_4 - k_{43}N_4N_0 + k_{34}N_3N_{v_n=0}, \\ \frac{dN_1}{dt} &= M_1 + \frac{N_3}{\tau_{31}} - \frac{N_1}{\tau_{12}}, \\ \frac{dN_2}{dt} &= M_2 + \frac{N_1}{\tau_{12}} + N_3 \left(\frac{1}{\tau_{31}} + \frac{1}{\tau_{32}} \right) - \frac{N_2}{\tau_{v-T}}, \\ \frac{dN_0}{dt} &= -M_1 - M_2 - M_3 - \frac{N_3}{\tau_{31}} + \frac{N_2}{\tau_{v-T}}, \end{aligned} \quad (16)$$

where N_1, N_2, N_3 and M_1, M_2, M_3 are the populations and rates of electron-impact excitation of vibrational levels $10^0 0, 01^1 0$ and $00^0 1$ levels, respectively; M_4 and $N_4 = N_{v_n=1}$ are the excitation rate and the population of the first vibrational level of the N_2 molecule, respectively (see Fig. 1). Usually, in the electric discharge $T > 300$ K and we can assume that $k_{34} \approx k_{43}$. In the stationary case, the solution of system (16) has the form

$$\begin{aligned} N_3 &= (M_3 + M_4)\tau_{3\Sigma}, \quad N_1 = \left[M_1 + (M_3 + M_4) \frac{\tau_{3\Sigma}}{\tau_{31}} \right] \tau_{12}, \\ N_2 &= \left[M_1 + M_2 + (M_3 + M_4)\tau_{3\Sigma} \left(\frac{1}{\tau_{3\Sigma}} + \frac{1}{\tau_{31}} \right) \right] \tau_{v-T}. \end{aligned} \quad (17)$$

One can see from (17) that the addition of N_2 molecules into the mixture increases the population of the upper laser level N_3 due to the increase in its excitation rate and lifetime. At the same time, relaxation processes in the CO_2 molecule lead to the increase in the population of other levels, including N_1 and N_2 . Therefore, the achievement of inversion is determined by the relations between the excitation and relaxation rates of the corresponding levels.

Let us represent expressions for $M_1 - M_3$ in the form $M_i = k_i^e n_e N_0$, and for M_4 in the form $M_4 = k_4^e n_e N_{v_n=0}$, where n_e is the electron density in the discharge and k_i^e is the rate constant of the electron-impact excitation of the i th level. If the ratio of partial pressures in the CO_2 and N_2

mixture is $1/Y$, then $N_{v_n=0} = YN_0 Q_C / Q_N$. By substituting expressions for $M_1 - M_4$ in (17) and taking into account that $X_3 \approx X_4$, we arrive at the system of three equations for X_1, X_2 and X_3 . However, it is rather difficult to obtain the solution of this system convenient for the analysis. On the other hand, one can easily see that for temperatures usually realised in active media of electric-discharge CO_2 lasers, the ratio is $Q_C / Q_N \approx Q_C(T)$ and, hence, $N_{v_n=0} \approx YN_0 Q_C(T)$ does not lead to large errors for the value $N_{v_n=0}$. In this case, rather simple expressions for the vibrational temperatures

$$\begin{aligned} T_1 &= \Theta_1 \left(\ln \left\{ \left[k_1^e + (k_3^e + k_4^e Y Q_C(T)) \frac{\tau_{3\Sigma}}{\tau_{31}} \right] n_e \tau_{12} \right\}^{-1} \right)^{-1}, \\ T_3 &= \Theta_3 \left(\ln \{ [k_3^e + k_4^e Y Q_C(T)] n_e \tau_{3\Sigma} \}^{-1} \right)^{-1}, \\ T_2 &= \Theta_2 \left(\ln 2 \left\{ \left[k_1^e + k_2^e + (k_3^e + k_4^e Y Q_C(T)) \tau_{3\Sigma} \right. \right. \right. \\ &\quad \left. \left. \left. \times \left(\frac{1}{\tau_{3\Sigma}} + \frac{1}{\tau_{31}} \right) \right] n_e \tau_{v-T} \right\}^{-1} \right)^{-1}, \end{aligned} \quad (18)$$

necessary for the qualitative analysis are obtained from (17) and (10). Here, $\Theta_1 = 1997$ K, $\Theta_2 = 960$ K, and $\Theta_3 = 3380$ K are the vibrational mode quanta of the CO_2 molecule.

It follows from (18) that the character of dependences of vibrational temperatures $T_1 - T_3$ on the excitation conditions, for example the discharge current (electron density in the discharge) depends on the relation between the rate of change in these conditions and the rate of change in the lifetimes of the corresponding levels (these variations proceed due to the changes in the gas temperature). If the gas is heated enough, a decrease in the lifetimes of levels $\tau_{3\Sigma}, \tau_{12}$ and τ_{v-T} can lead to saturation of the corresponding vibrational temperatures even when the excitation rates of these levels increase.

Figure 2 presents the calculated vibrational temperatures T_3 [curves (1)–(5)] as functions of the electron concentration in the discharge for some mixtures used in cw electric-discharge CO_2 lasers of different types. The

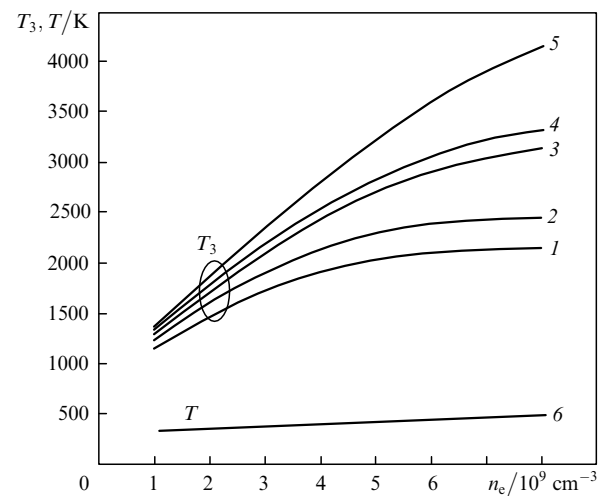


Figure 2. Vibrational temperatures T_3 calculated by (18) for the mixtures $CO_2 : N_2 : He = 1 : 1 : 3$ (1), $1 : 1 : 8$ (2), $1 : 6 : 3$ (3), $1 : 8 : 12$ (4) and $1 : 22 : 5$ (5) for the gas temperature T (6) as functions of the concentration of electrons in the discharge.

excitation rate constants k_3^s and k_4^s were borrowed from [21] and considered equal for all mixtures. We also assumed that the gas temperature T linearly increases from 325 to 500 K [curve (6)] with increasing the discharge current. The relaxation times were calculated by using the data from [22–24]. One can see that the increase in the gas temperature differently affects the character of the dependences: while for the CO₂ : N₂ : He = 1 : 22 : 5 mixture the vibrational temperature increases nearly linearly [curve(5)], for the CO₂ : N₂ : He = 1 : 1 : 8 mixture it exhibits saturation [curve (2)], whereas for the CO₂ : N₂ : He = 1 : 1 : 3 mixture T_3 first increases and then decreases [curve (1)]. Figure 2 shows clearly that different limiting values of T_3 obtained for mixtures of different compositions are not explained by a fundamental restriction to the value of T_3 in e-beam sustained discharge CO₂ lasers, as stated in [16], but are determined by collisional relaxation processes, whose rates depend on the mixture composition and its temperature.

It also follows from (18) that for the same electron concentration in the discharge in the CO₂ – N₂ – He mixture a higher vibrational temperature T_3 will be obtained than in the CO₂ – He mixture due to the appearance of an additional pump channel and the increase in the lifetime of the upper laser level. In this case the increase in the fraction of the nitrogen molecules in the mixture affect both these factors, which results in the increase in T_3 [see curves (1), (3) and (5) in Fig. 2]. This conclusion is consistent with the experimental results (see, for example [10]), which show that the higher vibrational temperature T_3 is achieved in the gas mixture with a lower content of CO₂.

Note that we considered examples when the initial mixture composition did not change. In reality, this can be realised in e-beam sustained discharge fast-flow CO₂ lasers. However, in the case of sealed-off lasers, it is necessary to take into account the dissociation of CO₂ molecules and consider the CO₂ – N₂ – CO – He mixture. In general, the kinetics of processes in the CO₂–CO mixture is similar to that in the CO₂ – N₂ mixture. The use of CO molecules in the mixture leads to an increase in the pump rate and the lifetime of the upper laser level as well as to a decrease in the lifetime of the 01¹0 level, which favours the relaxation of the lower laser level (see, for example, [21]), and, from this point of view, is a positive factor because it favours in the obtaining of inversion. The quantitative influence of CO molecules on the parameters of CO₂ lasers depends on the ratio of CO₂–CO in the active medium.

4. Conclusions

We have shown that for the proper use of the temperature model of CO₂ lasers in which vibrational modes of the CO₂ molecule are considered as harmonic oscillators with the Boltzmann population distribution over the levels with its own vibrational temperature, one should take into account that all the three vibrational modes of the CO₂ molecule have the common ground vibrational level. New expressions have been derived for the occupation numbers and the storage of the vibrational energy in the modes of the CO₂ molecule. It follows from these expressions that for each mode these values depend on the vibrational temperatures of all the three modes. We have also obtained expressions relating the vibrational temperatures of the CO₂ molecule with the relaxation rates and lifetimes of the lower levels of the vibrational modes upon continuous excitation of the

CO₂ – N₂ – He mixture in the electric discharge. It has been shown that the character of the dependences of the vibrational temperatures on the pump energy depends on the ratio of the excitation rate of the 00⁰1 level and the relaxation rate of the corresponding levels. In particular, a decrease in the lifetime of the upper laser level due to the gas heating in the discharge can lead not only to saturation but also to a decrease in the vibrational temperature T_3 with increasing the discharge current.

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