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The effect of temperature on the creation of population inversion in the active media of electric-discharge CO_2 lasers

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***Abstract*.** **It is shown that population inversion can be created on the $00^01 - 10^00$ transition in the active medium of an electric-discharge CO_2 laser even if its temperature increases to ~ 1200 K. By retaining the discharge stability and increasing the selectivity of the upper level pumping, it is possible to build electric-discharge CO_2 lasers without water cooling.**

Keywords: *electric-discharge CO_2 laser, active medium temperature, population inversion, pumping selectivity.*

It is known that the output power of an electric-discharge CO_2 laser can be increased by increasing the power supplied to the discharge. However, the energy spent on the heating of its active medium also increases inevitably in this case. According to the generally accepted point of view (see, for example, review [1], the frequently cited monograph by Witteman [2], or the later book [3]), it is the temperature T of the active medium of an electric-discharge CO_2 laser that limits its lasing power. This is due to the fact that an increase in the active medium temperature increases the thermal population of the lower laser level and accelerates the relaxation of the upper laser level so that the population inversion in the active medium disappears after the attainment of a certain critical temperature. This idea is formulated most clearly in Ref. [1], where it is assumed that the population N_{100} of the lower laser level 'corresponds to the Boltzmann law, i. e., it rises exponentially with increasing T. In view of this, the active-mixture population inversion vanishes when a certain critical temperature of $T_{\text{cr}} \sim 500-600°\text{C}$ is reached'.

Thus, the effective cooling of the active medium of a gas-discharge CO_2 laser is a necessary condition of its operation. The CO_2 lasers are even classified according to the methods of active medium cooling. It should be noted, however, that the authors of the above-mentioned publications give different estimates for T_{cr}. For example, in contrast to the above estimates from Ref. [1], Witteman believes (see p. 16 in his monograph [2]) that the laser power starts decreasing when the gas temperature becomes higher than $\sim 150°\text{C}$, while the authors of Ref. [3] state on p. 249 that the critical temperature lies in the range 250 – 300°C, 'which basically rules out the CO_2 laser action (irrespective of the method of active medium pumping).'

At the same time, some experimental facts contradict this point of view. For example, it was found [4, 5] that the temperature $T = 600 - 700$ K corresponds to the maximum output power of electric-discharge CO_2 lasers. An increase in the output power of a waveguide CO_2 laser with increasing temperature of the waveguide surface was observed in Refs [6, 7], which is obviously a consequence of an in-crease in T.

The aim of this work is to study the effect of T on the creation of population inversion on the $00^01 - 10^00$ transition in the active media of electric-discharge CO_2 lasers. In our opinion, the incorrectness of the generally accepted point of view is due to the inapplicability of the concept of the exponential increase in the population of the lower laser level with T to the 10^00 level of the CO_2 molecule. This can be proved most convincingly using the temperature model of the CO_2 laser [8], according to which quasistationary distributions of populations with corresponding vibrational temperature $T_i > T$ are established for each of the vibrational modes ν_1, ν_2, ν_3 of the CO_2 molecule. The populations of laser levels of the $00^01 - 10^00$ transition are determined by the expressions

$$N_{100} = NQ_{\text{v}}^{-1}X_1, \qquad N_{001} = NQ_{\text{v}}^{-1}X_3, \tag{1}$$

where N is the concentration of CO_2 molecules; $Q_{\text{v}} = (1-X_1)^{-1}(1-X_2)^{-2}(1-X_3)^{-1}$ is the vibrational partition function for CO_2; $X_i = \exp(-h\nu_i/kT_i)$; and $h\nu_i$ is the vibrational quantum of the ith mode of the CO_2 molecule.

The initial state of any vibrational nonequilibrium system is its equilibrium state. For this reason, we will first consider CO_2 molecules in a state when all vibrational temperatures are identical and equal to T. In this case, relations (1) imply that the value of N_{100} first increases with temperature indeed, but more slowly than according to an exponential law (cf. the curves for 10^00 in Fig. 1), achieves its maximum value at $T \sim 1200$ K, and then starts decreasing. Already at a temperature of 700 K, the population N_{100} calculated by formula (1) is about half the value calculated according to the exponential law.

In addition, since we are speaking of population inversion, we must consider the behaviour of the population both of the lower and of the upper energy level. It can easily be verified that an increase in T results in a simultaneous increase in the population N_{001} of the upper laser level (see the curve for 00^01 in Fig. 1), the ratio N_{100}/N_{001} decreasing

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rapidly. For example, the population N_{001} at $T = 300$ K is smaller than N_{100} by two orders of magnitude, while at $T = 600$ K these populations differ only by an order of magnitude. A further increase in T leads to a further decrease in this ratio which tends to unity.

As a result, the difference in the populations of the levels under study increases more slowly with temperature than the population of the lower level (from $\sim 0.94\,\%$ at $T = 400$ K to $3.2\,\%$ at $T = 1000$ K). For $T > 1000$ K, the difference in populations even starts decreasing. In other words, an increase in temperature at $T > 1000$ K does not deteriorate, but, on the contrary, improves the conditions for creating a population inversion. For example, at $T = 1200$ K, when the thermal population of the lower level is maximum and amounts to $\sim 4.38\,\%$, the difference in the populations of the levels under study is the same as at $T \sim 800$ K. In this case, almost a quarter of all CO_2 molecules are in the unexcited state, which makes it possible to create the population inversion at this temperature as well.

Consider now CO_2 molecules in a vibrational nonequilibrium state created in the active media of CO_2 lasers. The expressions (1) for the laser level populations imply that the population inversion in such a medium can be produced when the condition $T_3 > (h\nu_3/h\nu_1)T_1$, or $T_3 > 1.7T_1$, which is valid for any T_1, is satisfied. Therefore, a state with a high vibrational temperature T_3 differing considerably from the vibrational temperatures T_1 and T_2 is typical of CO_2 molecules under the conditions of the active medium. This means that the inversion is determined not by the thermal population of the lower level, but mainly by the population of the upper laser level or the vibrational temperature T_3.

Fig. 2 shows the temperature dependences of calculated populations of the laser levels for several typical vibrational temperatures T_3. For the sake of simplicity, we assume that $T_1 = T_2 = T$. In a rigorous quantitative analysis, their difference (i.e., the fact that actually $T < T_2 < T_1$ should be taken into account. One can see from Fig. 2 that an increase in the difference between T_3 and $T_{1,2}$ not only increases the population of the upper level, but simultaneously decreases the population of the lower level.

The difference between temperatures T_3 and $T_{1,2}$ depends on the selectivity of the upper level pumping. Large values of T_3 can be obtained if the N_{001} level is pumped, for example, mainly due to energy transfer from excited nitrogen molecules. Accordingly, the statement that an increase in the temperature of CO_2 deteriorates the conditions for creating the population inversion on the $00^01 \rightarrow 10^00$ transition is incorrect without an analysis of the mechanism of this inversion because the population inversion may be created at higher values of T if the pumping conditions provide the corresponding vibrational temperatures. For example, the same population inversion as for the vibrational temperatures $T_{1,2} = 400$ K and $T_3 = 100$ K can also be realised in the active medium at higher temperatures $T_{1,2} \sim 1100$ K and $T_3 = 2500$ K.

Note that the dependences presented in Fig. 2 illustrate the worst case for electric-discharge CO_2 lasers, when the increase in temperatures $T, T_{1,2}$ occurs at a constant temperature T_3. In actual practice, an increase in the temperatures $T, T_{1,2}$ takes place upon an increase in the energy deposition in the discharge. However, the energy of all modes of the CO_2 molecule, including the antisymmetric mode, increases in this case. In other words, the vibrational temperature T_3 also increases with the energy deposition in the discharge. Therefore, the actual behaviour of the laser level populations upon a change in the energy deposition to the discharge is determined by the relation between all varying vibrational temperatures.

To substantiate the possibility of creating a population inversion on the $00^01 \rightarrow 10^00$ transition at high temperatures of the active media of cw electric-discharge CO_2 lasers, we should analyse the effect of an increase in T on the processes creating a population inversion. For this purpose, we consider a simple three-level model including the ground level and vibrational laser levels. The main processes in such a system are the excitation and relaxation of the laser levels as well as stimulated emission. The presence of the lower energy levels of the deformation mode through which the relaxation of the 10^00 laser level takes place does not change qualitatively the pattern.

In the case under study, CO_2 molecules with population inversion on the $00^01 \rightarrow 10^00$ transition are in the laser cavity. If radiation of intensity I and frequency ν is generated in the cavity, the behaviour of the populations N_{001}, N_{100} and N_{000} is described by a simple system of kinetic equations

$$\begin{aligned}
\frac{dN_{001}}{dt} &= M_{\rm up} + \frac{I}{h\nu}\sigma f(J)N_{100} - \frac{N_{001}}{\tau_{\rm up}} - \frac{I}{h\nu}\sigma f(J)N_{001},\\
\frac{dN_{100}}{dt} &= M_{\rm low} + \frac{I}{h\nu}\sigma f(J)N_{001} - \frac{N_{100}}{\tau_{\rm up}} - \frac{I}{h\nu}\sigma f(J)N_{100}, \qquad (2)\\
\frac{dN_{000}}{dt} &= \frac{N_{001}}{\tau_{\rm up}} + \frac{N_{100}}{\tau_{\rm low}} - M_{\rm up} - M_{\rm low}.
\end{aligned}$$

Here, $M_{\rm up}$, $M_{\rm low}, \tau_{\rm up}, \tau_{\rm low}$ are the pumping rates and the lifetimes of the upper and lower vibrational levels, respectively; σ is the stimulated emission cross section; $f(J)$ is the distribution function for the populations of a vibrational level over rotational levels, which is given by (see, for example, Ref. [2])

$$f(J) = \frac{2hcB_1}{kT}(2J+1)\exp\left[-\frac{hcB_1J(J+1)}{kT}\right], \qquad (3)$$

where B_1 is the rotational constant of the vibrational level. We assumed in Eqns (2) for simplicity that functions $f(J)$ are identical for both vibrational levels. Using Eqns (2), we can easily prove that the expression for the saturated gain on the line under study,

$$g_{\rm s}(J) = \sigma f(J)(N_{001} - N_{100}) \qquad (4)$$

under the steady-state lasing can be written in the form

$$g_{\rm s}(J) = \frac{(M_{\rm up}\tau_{\rm up} - M_{\rm low}\tau_{\rm low})\sigma f(J)}{1 + I\sigma f(J)(\tau_{\rm up} + \tau_{\rm low})/h\nu}. \qquad (5)$$

This relation immediately leads to the following expressions for the unsaturated gain

$$g_0(J) = \sigma f(J)(M_{\rm up}\tau_{\rm up} - M_{\rm low}\tau_{\rm low}) \qquad (6)$$

and the saturation parameter

$$I_{\rm s} = \frac{h\nu}{\sigma f(J)(\tau_{\rm up} + \tau_{\rm low})}. \qquad (7)$$

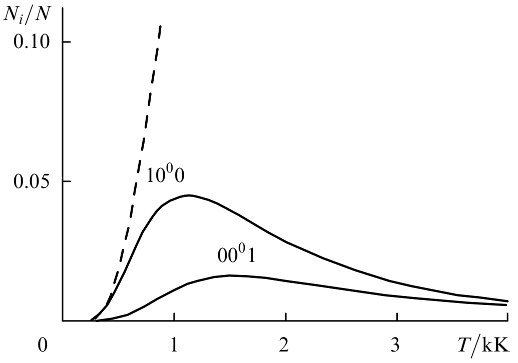

Figure 1. Temperature dependences of the relative populations of the lower (10^00) and upper (00^01) laser levels of the CO_2 molecule in the equilibrium case calculated on the basis of an exponential law (dashed curve) and by formulas (1) (solid curves).

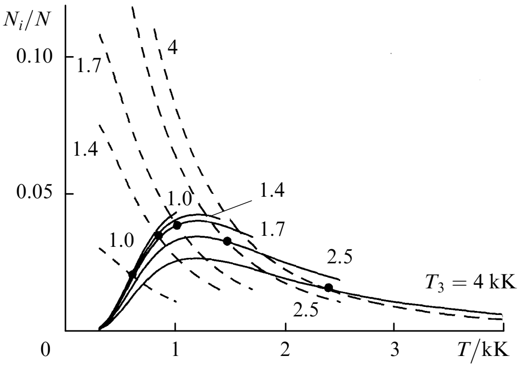

Figure 2. Dependences of the relative populations of the lower (10^00, solid curves) and upper (00^01, dashed curves) laser levels of the CO_2 molecule on temperature $T = T_{1,2}$ for various vibrational temperatures T_3.

The output power of a cw laser can be written in the form [9, 10]

$$P_{\mathrm{out}} = I_{\mathrm{s}} A \frac{t_2\sqrt{R_1}\left[g_0 L + \ln(R_1 R_2)^{1/2}\right]}{\left(\sqrt{R_1} + \sqrt{R_1}\right)\left[1 - (R_1 R_2)^{1/2}\right]}, \qquad (8)$$

where A is the laser beam cross section; R_1 and R_2 are the reflection coefficients of the cavity mirrors; t_2 is the transmission of the output mirror; L is the length of the active medium. It follows from Eqn (8) that the active medium temperature affects the output power of the laser through the gain and the saturation parameter, which are described by expressions (6) and (7), respectively. Therefore, we should analyse the effect of the gas temperature on the quantities appearing in these expressions, namely, the stimulated emission cross section, the distribution function for the populations of the vibrational levels over rotational levels, the pumping rates, and the lifetimes of the vibrational levels under study.

Since the stimulated emission cross section is proportional to the form factor $F(J)$ of the gain line and the line profiles for most CO_2 lasers are collisionally or homogeneously broadened and obey the relation $F(J) \sim \sqrt{T}$ (see, for example, Ref. [2]), we have $\sigma \sim \sqrt{T}$. Thus, the stimulated emission cross section increases with the temperature of the active medium of a CO_2 laser. For example, the cross section σ increases approximately by a factor of ~ 1.83 as the temperature T increases to 1000 K.

The effect of the gas temperature on the distribution of populations of a vibrational level over rotational levels (3) is well known: the population of vibrational levels becomes more uniform upon an increase in T. In this case, the populations of levels with lower rotational quantum numbers J decrease, whereas the population of levels with larger values of J increase. For example, an increase in T from room temperature to 1000 K leads to a decrease in the population of levels with $J < 30$ (the population of the level with $J = 20$ decreases almost by half), while the populations of levels with $J > 30$ increase.

Thus, the product $\sigma f(J)$ for the lines with $J \sim 20$ (which are usually considered in CO_2 lasers) is virtually independent of T, and expressions (6) and (7) imply that an increase in the gas temperature affects the gain and the saturation parameter through a change in the pumping rates and lifetimes of the upper and lower levels. The lifetimes of the levels in CO_2 lasers are determined by the collision relaxation (see, for example, Ref. [8]). An increase in the gas temperature accelerates these processes, reducing the lifetimes of both levels. One can see from Eqn (6) that this leads to a decrease in the gain. At the same time, it follows from Eqn (7) that the saturation parameter increases with decreasing times τ_{up} and τ_{low}.

Finally, we must analyse the effect of the gas temperature on the pumping rates M_{up} and M_{low} of laser levels. Consider the conventional $CO_2 - N_2 -$ He mixture in which an electric discharge with an electron number density n_{e} takes place. Let $\sigma_{\mathrm{ec}}^{\mathrm{low}}$ and $\sigma_{\mathrm{ec}}^{\mathrm{up}}$ be the electron impact cross sections for excitation of the vibrational levels 10^00 and 00^01 of the CO_2 molecule, respectively. Then, the pumping rate for the lower level is given by

$$M_{\mathrm{low}} = \sigma_{\mathrm{ec}}^{\mathrm{low}} n_{\mathrm{e}} N_{000}. \qquad (9)$$

In addition to the electron impact, the upper 00^01 level is populated due to the quasi-resonance energy transfer from excited nitrogen molecules with the rate K_{43}. The total pumping rate for this level is determined by the expression

$$M_{\mathrm{up}} = \sigma_{\mathrm{ec}}^{\mathrm{up}} n_{\mathrm{e}} N_{000} + K_{43} N_n^{v=1} N_{000}, \qquad (10)$$

where $N_n^{v=1}$ is the density of excited N_2 molecules.

It was mentioned above that the active medium temperature in an electric-discharge CO_2 laser increases with the energy deposition in the discharge. This leads to a variation in such quantities as the electron density, the electron velocity distribution and, hence, the excitation cross sections. In each specific case, these changes are determined by the geometry of the discharge gap and by the pressure and composition of the gas mixture. For this reason, their quantitative effects on the pumping rates of the levels are different. For the sake of generality of our analysis, we will assume that these quantities at least do not decrease upon an increase in the energy deposition in the discharge (in its stability region), i.e., the efficiency of the discharge required for the excitation of both levels is preserved.

Note also that the pumping rates of the levels and, hence, the gain decrease in ordinary sealed-off lasers due to a decrease in N_{000} caused by dissociation of CO_2 molecules in the discharge. For large energy depositions, this decrease is significant because the degree of dissociation may be as high as $\sim 90\,\%$ [11]. At the same time, it is known that the composition of the active medium can be maintained virtually unchanged by using an appropriate catalyst minimising the degree of CO_2 dissociation (for example, gold was used in Refs [12, 13] for this purpose). Thus, we will assume below that the composition of the active medium is constant and independent of the energy deposition in the discharge and, hence, of temperature.

In this case, expressions (9) and (10) imply that an increase in the gas temperature affects the pumping rates of the levels via a change in the quantities N_{000} and K_{43}. From the above discussion it follows that the density N_{000} of CO_2 molecules in the ground state decreases with increasing T due to an increase in the thermal population of the excited vibrational levels. For example, an increase in T from 500 to 1000 K reduces the population N_{000} of the ground state from 72 % to $\sim$23 %, i.e., more than three times.

It is known (see, for example, Ref. [8]) that an increase in the gas temperature also reduces the rate of vibrational energy transfer between $N_2(\upsilon = 1)$ and $CO_2(00^01)$ (approximately by half upon an increase in T from 500 to 1000 K). This results in a decrease in the second term in Eqn (10) and, hence, in the overall rate $M_{\rm up}$, which leads to a decrease in the gain according to Eqn (6).

Thus, under the above assumptions concerning the efficiency of the discharge, the gain decreases with increasing temperature of the active medium. It is assumed that this necessitates the cooling of active media of the existing electric-discharge CO_2 lasers. At the same time, as mentioned above, the saturation parameter increases with the temperature of the active medium. As a result, according to Eqn (8), the effect of an increase in the temperature of the active medium of a CO_2 laser on its output power is determined by the relation between the changes in the gain and in the saturation parameter: if the increase in the saturation parameter is more significant than the decrease in the gain, the value of $P_{\rm out}$ increases, and conversely, if the gain decreases at a higher rate than the rate of the increase in the saturation parameter, the value of $P_{\rm out}$ decreases.

It was found in Ref. [14] that the maximum output power in electric-discharge CO_2 lasers with a minimised degree of dissociation of CO_2 molecules can be achieved without cooling the active medium to low temperatures. On the contrary, the optimal characteristics of such CO_2 lasers are obtained at higher gas temperatures corresponding to large energy depositions, when the decrease in the gain is compensated by the increase in the saturation parameter. It should be emphasised that this refers to the case when the composition of the active medium does not change upon an increase in the energy deposition (if we disregard the dissociation of CO_2 molecules).

At the same time, expression (10) implies that the same pumping rate $M_{\rm up}$ of the upper level and, hence, the same gain as for ordinary active medium temperatures can be also obtained at higher temperatures. For this purpose, the decrease in N_{000} and K_{43} should be compensated by an increase in the density $N_n^{\upsilon=1}$ of excited nitrogen molecules by increasing their fraction in the mixture composition, i.e., by increasing the selectivity of the upper level pumping. In other words, by changing the active medium composition by increasing in the fraction of nitrogen molecules relative to the fraction of CO_2 molecules, we can attain the same gain in the active medium at a high temperature as in the existing CO_2 lasers. According to Eqn (8), the power of such a laser will be higher because the saturation parameter is larger at higher temperatures.

A higher temperature of the active medium may be obtained, for example, by removing water cooling and by controlling its thermal conductivity by varying the helium content. Naturally, such a high-temperature operation of a CO_2 laser presumes the possibility of creating a stable discharge in a mixture with a high nitrogen content. The realisation and features of such a discharge were reported in Refs [15, 16]. Moreover, the authors of Ref. [16] built a cw CO_2 laser without water cooling and with an elevated nitrogen concentration in the active medium. In contrast to the conventionally used mixtures with the ratio CO_2:$N_2 = 1$:1, mixtures with the composition CO_2:$N_2 = 1$:$7 - 33$ were used. The optimal ratio for such a laser was found to be CO_2:$N_2 = 1$:20. However, the authors of Ref. [16] failed to explain the mechanism of the population inversion in the laser.

Thus, the population inversion on the $00^01-\ 10^00$ transition in electric-discharge CO_2 lasers is mainly determined by the vibrational temperature T_3 rather than by the active medium temperature. The increase in the latter to $\sim$1200 K does not inevitably prevents the population inversion on this transition. By retaining the discharge stability and increasing the selectivity of the upper level pumping, for example, by increasing the molar fraction of nitrogen in the gas mixture, the CO_2 laser can operate at an elevated temperature of the active medium. In this case, the requirements to the cooling system of the laser are changed, and such lasers can operate at moderate powers without water cooling at all.

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