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In the present paper a numerical calculation is made of the vibrational relaxation of a binary mixture of molecular nitrogen and carbon dioxide gas. The calculation is performed for the entire range of variation of the concentrations of the components and over a wide range of mixture temperatures and pressures for various geometries of the supersonic part of the nozzle (throat dimensions, degree of expansion). It is shown that population inversion of the  $CO_2$  molecules exists within a certain range of variation of the parameters of the mixture and the nozzle. The population inversion of the vibrational levels and the gain of the gaseous mixture are calculated as functions of these parameters and of distance measured from the critical cross section of the nozzle. The energy characteristics of the two-component gasdynamic laser are optimized.

The gasdynamic CO<sub>2</sub> laser was first suggested by Konyukhov and Prokhorov [1].

In the calculations of Basov et al. [2] it was shown that a population inversion could occur under certain conditions in the supersonic flow of a binary gas mixture in a Laval nozzle. The values of the relaxation constants used in these calculations differ greatly (by one or two orders of magnitude) from the values currently accepted in the literature. When the equations are solved using the improved values of the relaxation constants it is found [3-8] that under the conditions considered in [2] no population inversion occurs in a binary mixture of nitrogen and carbon dioxide gas.

The experiments reported in [3, 5] also showed that under the conditions used in these papers the gain of a binary mixture of nitrogen and carbon dioxide gas is negative.

Population inversion (positive gain) of the medium in a gasdynamic laser was obtained experimentally using three-component mixtures [3-7, 9-12], the experimental results obtained in papers [6, 11, 12] being in satisfactory agreement with theoretical calculations.

The possibility of obtaining population inversion in binary mixtures has been studied experimentally and theoretically in a narrow range of variation of mixture concentrations and of the other parameters of gasdynamic lasers (pressure, dimensions of critical part of nozzle, degree of expansion).

A necessary condition for population inversion in an expanding supersonic flow is that the characteristic time of expansion of the flow  $\tau_0$  must be less than or of the same order as the relaxation time of the upper laser level  $\tau_3$ , and that  $\tau_3$  must be much greater than the relaxation time of the lower laser level  $\tau_2$ , i.e.,

$$\tau_3 / \tau_0 \ge 1, \qquad \tau_3 / \tau_2 \gg 1 \tag{1}$$

Utilizing the expressions for the time constants  $\tau_2 = [k_2p]^{-1}$ ,  $\tau_3 = [\psi_0k_3p]^{-1}$  (this formula holds when the concentration of CO<sub>2</sub> gas in the mixture  $\psi_0 > 10^{-3}$ ),  $\tau_0 = h*/u* \tan \varphi$ , we find that the following conditions are necessary for population inversion of CO<sub>2</sub> molecules in an expanding binary mixture:

$$\omega^{-1} = u_* \operatorname{tg} \varphi / \psi_0 k_3 p_* h_* \ge 1$$
(2)
$$k_2 / k_3 \psi_0 \gg 1$$
(3)

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TABLE 1

Variant No.	T <sub>*</sub> , °K	p*, atm	Ψο	h <sub>*</sub> , cm	$p_*h_*,$ atm · cm	z, atm• cm	S/S*	$ _{\times 10^{-16} \text{cm}^3}^{\Delta N_m \times}$	∝,m <sup>-1</sup>	x, cm
1 2 3 4 5 6 7 8 9	1000 1000 1000 1000 1000 1000 1000 2000 2000	10 20 100 40 200 40 40 40 40	0.1 0.1 0.025 0.005 0.05 0.1 0.025 0.1	0.1 0.05 0.01 0.1 0.1 0.1 0.1 0.1 0.1	1 1 4 20 4 4 4 4 4	0.37 0.37 0.37 0.37 0.37 0.74 1.5 0.37 1.5	9 9 7 6.5 8.5 12.9 11.75 50	0.4 0.9 4 0.6 0.7 0.6 0.4 0.4 0.4	0.4 0.5 0.6 0.15 0.03 0.19 0.1	



Here  $k_2$  and  $k_3$  are the rate constants associated with the relaxation of the lower and upper laser levels, respectively,  $\psi_0$  is the molar concentration of CO<sub>2</sub> gas,  $\varphi$  is the semiangle of the diverging part of the nozzle near the critical cross section,  $h_*$  is the width of the critical cross section of the nozzle, and  $p_*$  and  $u_*$  denote the pressure and the velocity of sound in the critical part of the nozzle.

When inequality (2) is satisfied, the upper laser level of the twocomponent mixture is frozen [7]; inequality (3) means that the lower laser level relaxes more rapidly than the upper. Both conditions (2) and (3) for achieving population inversion are satisfied at sufficiently

small  $CO_2$  concentrations, i.e., a range of parameters exists for which population inversion of the molecules occurs. In accordance with (2) the concentration of  $CO_2$  gas must be reduced with increasing pressure and width of the critical cross section of the nozzle.

A quantitative description of the process of expansion and vibrational relaxation of a binary mixture of molecular nitrogen and  $CO_2$  gas was obtained by solving the appropriate set of differential equations. The probabilities of the relaxation processes were taken from [13-20]. The set of equations is described in detail in [17, 18].

The calculations were carried out for a planar wedge-shaped nozzle with 15° semiangle of the diverging portion, the diverging portion being fitted to a channel of constant cross section. The degree of expansion of the nozzle,  $S/S_*$  was varied over a wide range. From the solution we determined the translational temperature, density, and pressure of the gas along the flow, and also the distribution of the vibrational temperatures of the molecules of nitrogen and  $CO_2$  gas in the bending, symmetric, and antisymmetric modes.

The CO<sub>2</sub> populations of the laser levels,  $N_{001}$  and  $N_{100}$ , were determined by assuming the molecules to be Boltzmann-distributed in energy within each mode of vibration.

The gain of the medium  $\alpha$  for the P branch (J=20) of the collisionally broadened spectral line and the populations of the vibrational levels of the CO<sub>2</sub> molecules were calculated for various temperatures  $T_*$  and pressures  $p_*$  in the critical cross section of the nozzle, throat widths  $h_*$ , and CO<sub>2</sub> concentrations in the mixture  $\psi_0$ . We denote by x the distance from the critical cross section along the flow. The results of the calculations are presented in Table 1, from which it can be seen that for small  $p^*h^*(p_*h < 5atm \cdot cm$ for  $T_* = 1000^{\circ}$ K) maximum population inversion  $\Delta N_m$  and maximum gain occur on the flat part of the nozzle (see Fig. 1). The reason for this is that the lower level remains partially frozen in the diverging part of the nozzle, and its population is depleted more rapidly (compared with the upper level) in the constantsection channel for a constant gas density. The population inversion in the constant-section channel is the greater the smaller the value of  $p_*h_*$ .

In Fig. 1 we have plotted the calculated maximum population inversion (curves 2, 4) and gain (curves 1, 3) in the diverging (curves 3, 4) and constant-section (curves 1, 2) part of the nozzle as functions of S/S\* (variant 2 in Table 1). It can be seen that maximum population inversion in the diverging and constant-section parts of the nozzle occurs at different values of S/S\*. Thus, to achieve maximum population inversion in the constant-section part of the nozzle, this part must be joined to the diverging part at  $(S/S*)_1$  differing from the value  $(S/S*)_2$  corresponding to maximum population inversion in the nozzle. The gain in the diverging and constant-section parts of the nozzle is likewise maximum for different values of S/S\*. We note that population inversion and a positive gain can be achieved at small degrees of expansion  $(S/S* \approx 5)$ , when the pressure in the resonator region can exceed atmospheric.

TABLE 2



Calculation of the energy characteristics of the gasdynamic  $CO_2$  laser in the general case requires the solution of a complex set of equations with a large number of parameters. Optimization of the parameters of the system is accordingly a complicated problem.

With a view to reducing the number of parameters by establishing the most important and discarding the less significant, we utilize in the present paper an approximate model of the relaxation processes in an expanding supersonic flow. In this model the temperature of the upper laser level  $T_3$  is assumed equal to the translational temperature T up to a certain section of the nozzle S', where it becomes "frozen," while the temperature of the lower laser level is assumed everywhere to equal the translational;

$$T_{\mathfrak{z}} = T \quad \text{when} \quad S_{\mathfrak{z}} \leqslant S \leqslant S' \tag{4}$$

$$T_3 = T_3'$$
 when  $S \ge S'$  (5)

$$T_2 = T$$
 (6)

The "maximum gain" model [11, 19] is a particular case of the present model (S' = S<sub>\*</sub>). We shall `` show that within the framework of the present model the relative frozen temperature of the upper laser level,  $T'_3/T_*$ , is approximately expressed as a function of a single parameter  $\omega$ .

The vibrational temperature  $T'_3$  becomes frozen in the cross section S' in which the relaxation time  $\tau_3$  becomes comparable to the time of expansion, i.e., when

$$\tau_0/\tau_3 = \text{const} \sim 1 \tag{7}$$

The ratio  $\tau_0/\tau_3$  is a function of the following dimensionless quantities:

$$\tau_0/\tau_3 = \omega f\left(\xi, \gamma, A_i^3/T_*\right) \tag{8}$$

where  $\gamma = C_p/C_v$  is the adiabatic exponent (assumed constant in the present approximation),  $\zeta = x/h_* \tan \varphi$  is a dimensionless coordinate directed along the flow, and  $A_i$  are factors in the exponent of the exponential in the Landau-Teller expression for the rate constants of the relaxations.

Calculation shows that for expansion of the gas in a Laval nozzle the ratio  $\tau_0/\tau_3$  decreases mainly because of the drop in pressure and not temperature, i.e.,

$$\left|\frac{\partial}{\partial p}\left(\frac{\tau_{0}}{\tau_{3}}\right)\frac{dp}{dx}\right| > \left|\frac{\partial}{\partial T}\left(\frac{\tau_{0}}{\tau_{3}}\right)\frac{dT}{dx}\right|$$
(9)

If the variation of  $\tau_0/\tau_3$  is ascribed solely to the fall in pressure, then

$$\tau_0/\tau_3 = \omega f(\xi, \gamma) \tag{10}$$

On inserting (10) into the condition for freezing (7) and remembering that  $T_3/T_*$  within the framework of the present model is a unique function of  $\zeta$  for a given  $\gamma$ , we see that the relative frozen temperature  $T'_3/T_*$  is a function of the two dimensionless parameters  $(T'_3/T_* = F(\omega, \gamma))$ .

For mixtures consisting mainly of nitrogen ( $\psi_0 \ll 1$ ), the adiabatic exponent  $\gamma$  is only weakly dependent on the composition of the mixture and its temperature. Accordingly, in the first approximation,  $T'_3/T_*$  can be regarded as depending only on the single parameter  $\omega$ :

$$T_{3}'/T_{*} = F(\omega) \tag{11}$$

In the present paper we cite the results of an analysis of a gasdynamic laser using the binary mixture N<sub>2</sub>+CO<sub>2</sub>. A numerical solution of the problem using the complete set of gaskinetic equations (variants 1-8 in Table 2) and also an approximate calculation (variants 9-29 in Table 2) showed that over a wide range of variation of the parameters (P<sub>\*</sub>=10-250 atm, T<sub>\*</sub>=1000-2500°K,  $\psi_0$ =0.002-0.1, h<sub>\*</sub>=0.1-0.01 cm) the relative frozen temperature T'<sub>3</sub>/T<sub>\*</sub> can be regarded approximately as a function of the single parameter  $\omega$  (Fig. 2). This function can be approximated by the following formula:

$$T_{3}'/T_{*} = (0.027\omega^{0.6} + 1)^{-1}$$
(12)

In Fig. 2 the points 1, 2, 3, 4, 5 correspond to  $T_* = 1000$ , 1000, 1800, 2000, 2500°K; the points 2 and 4 were obtained by solving the complete set of equations.

An approximation calculation of the frozen temperature of the upper laser level (variants 9-29 in Table 2) was made using the assumption of equilibrium between the v=1 level of the nitrogen molecule and the 00°1 level of carbon-dioxide gas. The relaxation equations for the energy were solved in the linearized Landau-Teller form [8, 10].

The calculation showed that for values of the product  $p_*h_* \ge 10 \text{ atm} \cdot \text{cm}$  for a binary mixture with  $\psi_0 \ll 1$  in the temperature interval  $T_* = 600-2600$  % for the investigated degrees of expansion, the assumption that the temperature of the lower laser level equals the translational temperature of the gas at the exit from the Laval nozzle  $T_1$  is quite well obeyed.

The specific energy of the stimulated emission per unit mass of the mixture (on the assumption that the vibrational energy stored in the upper laser level is converted into the energy of stimulated emission in an ideal resonator without losses on relaxation until the gain of the medium  $\alpha$  falls to zero) was calculated using the formula

$$E = \eta_0 \left[ E_3 + E_3 \left( \alpha = 0 \right) \right] N / \mu$$
(13)

where  $E_3N/\mu$  is the specific energy of the vibrational mode of the upper laser level, frozen in the Laval nozzle;  $E_3(\alpha=0)$  is the residual energy of the upper laser level at  $\alpha=0$ , which corresponds to the condition  $T_3=1.78T_1$ ;  $\eta_0=0.41$  is the quantum efficiency of the laser; N is Avogadro's number; and  $\mu$  is the molecular weight of the mixture. The energy  $E_3$  was calculated from

$$E_3 = hv \left[ \exp\left(\theta_3 / T_3'\right) - 1 \right]^{-1} \tag{14}$$

where  $\nu$  and  $\theta_3$  denote the frequency and the characteristic temperature of the upper laser level. The quantity  $T_3^i$  was determined from (12).

The radiation energy from unit volume of medium in the resonator region is given by  $W = E\rho$ , where  $\rho$  is the density of the gas. The laser efficiency  $\eta$  was determined through the formula

$$\eta = E \left[ \varepsilon \left( T_0 \right) - \varepsilon \left( T_g \right) \right]^{-1}$$



where  $\epsilon$  is the free energy,  $T_0$  is the temperature in the combustion chamber, and  $T_g$  is the initial temperature (taken as 300°K in the calculations).



The above energy characteristics of the gasdynamic laser will be calculated using the following initial parameters:  $T_*$ ;  $p_*$ ; the parameter  $z = p_*h_*\psi_0/\tan\varphi$ , proportional to  $\omega$  for a given temperature; and the degree of expansion of the nozzle S/S\*, equal to the area of the outlet cross section of the nozzle where it goes over into the plane-parallel channel divided by the area of the critical cross section.

For a binary mixture with  $\psi_0 \ll 1$  and  $\gamma = \text{const}$ , the specific energy of the coherent radiation from unit mass E [see (13)] and the efficiency  $\eta$  can be expressed through the initial parameters

$$E = E(T_*, z, S/S_*), \quad \eta = \eta(T_*, z, S/S_*)$$
(15)

The radiation energy from unit volume of the medium is expressed by the formula

J

$$W = P_* \Phi (T_*, z, S/S_*)$$
(16)

An analysis of (15) shows that E and  $\eta$  increase with increasing S/S<sub>\*</sub> and decreasing z and tend towards their maximal values when z-0 and S/S<sub>\*</sub> -  $\infty_*$  With increasing S/S<sub>\*</sub> the functions E and  $\eta$  rapidly enter a region of saturation, so that we shall restrict S/S<sub>\*</sub> to values for which E(T<sub>\*</sub>, z, S/S<sub>\*</sub>) =0.9 E(T<sub>\*</sub>, z,  $\infty$ ). This value of S/S<sub>\*</sub> corresponding to 0.9E we denote by S<sub>0</sub>/S<sub>\*</sub>. Since S<sub>0</sub>/S<sub>\*</sub> = S<sub>0</sub>/S<sub>\*</sub> (T<sub>\*</sub>, Z), it follows that E can be represented as a function of any two of the variables from S<sub>\*</sub>, T<sub>\*</sub>, z. The results of the calculation of E(S<sub>0</sub>/S<sub>\*</sub>, z, T<sub>\*</sub>) are presented in Fig. 3.

This figure shows two families of intersecting curves corresponding to  $E = E(S_0/S_*)$  for  $T^* = \text{const}$ (solid curves) and for z = const, (dot-dashed curves). The numbers appended to the curves denote the values of  $T_*$  (deg K) and z (atm  $\cdot$  cm), respectively. For given values of  $T_*$  and z these curves can be used to find the optimum degree of expansion  $S_0/S_*$  and the corresponding value of the energy. Figure 4 shows analogous plots of  $\eta(S_0/S_*, T_*, z)$ . Taking  $S/S^* < S_0/S_*$  for a given  $T_*$  and z reduces the energy (efficiency); by increasing the degree of expansion, which is usually undesirable, the energy (efficiency) can be increased by not more than 1/9-th of the value at the point  $S_0/S_*$ . If a single quantity is prescribed ( $T_* = \text{const}$  or z = const), the point whose coordinates determine the optimum values of the two other parameters lies either on the boundary of the investigated region of variation of the parameters (for example, for  $T_* = \text{const}$ we find that z = 0) or within this region. For instance, for z = const ( $z \ge 0.1$ ) maximum efficiency (see Fig. 4) is achieved within the region of calculation for intermediate values of  $S_0/S_*$  and  $T_*$ .

The specific radiation energy from unit volume of the medium W when situated in the resonator is directly proportional to the pressure in the critical cross section of the nozzle (16). All four parameters in (16) are then regarded as independent. The number of independent parameters can be reduced to three by considering the function  $W/P_*$  (specific energy from unit volume at 1 atm pressure in the critical cross section). For each z optimum values of the parameters  $T_*$  and  $S/S_*$  exist for which  $W/P_*$  is a maximum (Fig. 5). Figure 5 shows plots of  $W/P_*$  versus z for various values of  $T_*$  (the numbers appended to the various curves denote  $T_*$  in deg K) and  $S/S_{*}=5$ . This family of curves has an envelope on which is achieved, for each value of z and  $S/S_*=5$ , the maximum  $W/P_*$  and the optimum  $T_*$ . Similar envelopes can be constructed for other values of  $S/S_*$ . In this manner, the optimum value of  $T_*$  is a function of z and  $S/S_*$ .



Figure 6 shows a parametric net of z (dot-dashed curves) and  $S/S_*$  (solid curves). The point on the graph corresponding to a pair of values of z and  $S/S_*$  determines the optimum value of  $T_*$  and the associated maximum (for the given z and  $S/S_*$ ) value of  $W/P_*$ . Choosing a temperature  $T_*$  different from optimal reduces the value of  $W/P^*$ .

Figure 7 corresponds to Fig. 6 plotted in other coordinates. The parametric net of  $S/S_*$  (dotted curve and solid curves) and  $T_*$  (dot-dashed curves) can be used to determine, for given S S\* and z, the optimum temperature  $T_*$  and the associated value of  $W/P_*$ .

It can be seen from Fig. 7 that the radiation energy from unit volume W can be increased by increasing the temperature of the mixture in the combustion chamber of the laser and by letting z tend to zero at  $S/S_*$  equal to around five. This value of  $S/S_*$  can be regarded as the optimum for all  $T_*$  and z, i.e.,  $W/P_*$  is a maximum for  $S/S_*\approx5$ .

The simplified method described in the present paper can be used to determine the approximate optimum values of the parameters of a gasdynamic laser with a two-component mixture and an ideal resonator. If necessary, the position of the optimum can be refined by a more exact calculation within narrow limits of variation of the parameters in the vicinity of the optimum found in the first approximation.

The approach used by us (representation of the relative frozen temperature  $T'_{3}/T_{*}$  as a function of the single parameter  $\omega$ ) may be useful in optimizing the performance of three-component gasdynamic lasers.

The paper by Gembarzhevskii et al. [21] appeared after the present paper had been submitted to the press. These authors obtained amplification using a binary mixture of  $10\% \text{CO}_2$  and  $90\% \text{N}_2$ , which confirms the conclusions of the present work.

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