FORMATION OF INVERSION IN JET OF $CO_2-H_2O-N_2$ GAS MIXTURE EXPANDING THROUGH A SLOT

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A calculation is made of the kinetics of vibrational relaxation of CO_2 molecules in a $CO_2-H_2O-N_2$ mixture escaping into a vacuum from a slot. The examination of vibrational relaxation led to a solution of the kinetic equations corresponding to the most important channels of energy exchange in vibration–vibration and vibration–translation processes. It proved possible to consider the dynamics of a nonequilibrium gas in an approximation of the adiabatic motion of a medium with an effective adiabatic index γ corresponding to a certain degree of freezing in of the vibrational component of the heat capacity of the gas. The calculated values of the gain index α agree well with experimental data. The gain index was calculated with allowance for Doppler and Lorentz mechanisms of line broadening. The results of the calculation were analyzed.

1. Experiments on $CO_2-H_2O-N_2$ gasdynamic lasers (GDL) were reported on in [1, 2]. In [3] experimental studies were conducted on GDL with a high water content in the mixture, up to a ratio of concentrations of (1 CO_2 : 1 H_2O). In contrast to the experiments conducted with ordinary supersonic nozzles which had an opening angle of 10-15° [1, 2], the rapid cooling of the gas in [3] was accomplished by expansion through a nozzle-slot with an opening angle of 120°, which we will henceforth call a slot. The use of a slot provided higher initial cooling rates compared with those reached in [1, 2].

The vibrational relaxation of the CO_2 molecule in CO_2-N_2-He and CO_2-N_2 mixtures during free gas-dynamical dispersion simulating the discharge from a slot is examined in [4]. A comparison of these calculations with the data of experiments [5] indicated only qualitative agreement. In the present work a calculation was made of the kinetics of relaxation for the $CO_2-H_2O-N_2$ mixture with more correct allowance for the gas dynamics of discharge from a slot [6].

2. Following the classification given in the review [7] and on the basis of the results of [8-10] one can isolate the main channels of energy exchange in the vibration–vibration (V–V) and vibration–translation (V–T) processes which determine the kinetics of relaxation of the model of the CO_2 molecules in the $CO_2-H_2O-N_2$ mixture:

$$N_2^* + CO_2 \underset{}{\overset{k_{43}}{\rightleftarrows}} N_2 + CO_2^*(\nu_3) \tag{2.1}$$

$$k_{43} = 6.55\cdot 10^{10} T^{-3/2} \quad [8]$$

$$N_2^* + H_2O \overset{k_{45}}{\rightleftarrows} N_2 + H_2O^*(\nu_5) \tag{2.2}$$

$$k_{45} = 10.5\cdot 10^{6} \exp(30.6 T^{-1/3}) \quad [7]$$

$$CO_2^*(\nu_3) + M_j \overset{k_{32}^{(j)}}{\rightleftarrows} CO_2^*(\nu_1, \nu_2) + M_j \tag{2.3}$$

$$k_{32}^{(1)} = 2.24\cdot 10^{-2} T^{2.75}, \quad k_{32}^{(2)} = {}^1/_2 k_{32}^{(1)} \quad [8]$$

$$k_{32}^{(3)} = 2.32\cdot 10^{7} \quad [9]$$

(Here and later j = 1 corresponds to the CO_2 molecule, j = 2 to the N_2 molecule, and j = 3 to the H_2O molecule.)

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$$CO_2^*(\nu_1, \nu_2) + M_j \overset{k_{20}^{(j)}}{\rightleftharpoons} CO_2 + M_j \tag{2.4}$$

$$k_{20}^{(1)} = 8 \cdot 10^7 \exp(-41.6T^{-1/3}), \quad k_{20}^{(2)} = 3k_{20}^{(1)} \quad [7]$$

$$k_{20}^{(3)} = 9.2 \cdot 10^6 \exp(-34.5T^{-1/3}) \quad [10]$$

where $k_{em}^{(j)}$ are the constants of the reaction rates of (2.1)-(2.4) obtained by approximation of the experimental data of [7-10] in the range of gas temperatures $T \approx 300\text{-}1000°K$, which have the dimensionality $[\sec^{-1} \cdot atm^{-1}]$. The symmetric, deformation, and asymmetric types of vibrations of the CO_2 molecule are denoted by ν_1, ν_2, and ν_3, the vibration of the N_2 molecule by ν_4, and deformation-type vibrations of the H_2O molecule by ν_5. An excited state of the molecule is marked by an asterisk in the reactions, and which vibrations are excited is shown in parentheses. With allowance for these processes the system of relaxation equations takes the form

$$\begin{aligned} d\varepsilon_4/dt &= -k_{43}p^{(1)}[\varepsilon_4 - \varepsilon_3] - k_{45}p^{(3)}[\varepsilon_4 - \varepsilon_4°] \\ d\varepsilon_3/dt &= k_{43}p^{(2)}[\varepsilon_4 - \varepsilon_3] - \sum_j k_{32}^{(j)}p^{(j)}[\varepsilon_3 - \varepsilon_3°] \\ d\varepsilon_2/dt &= Z^{-1}\left\{\sum_j k_{32}^{(j)}p^{(j)}[\varepsilon_3 - \varepsilon_3°] - \sum_j k_{20}^{(j)}p^{(j)}[\varepsilon_2 - \varepsilon_2°]\right\} \end{aligned} \tag{2.5}$$

Here $\varepsilon_i = g_i\theta_i x_i(1 - x_i)^{-1}$ is the energy of the harmonic oscillator of the i-th type of vibration under conditions of an equilibrium distribution with a vibrational temperature T_i, which differs in general from the gas temperature T; $\varepsilon_i°$ is its energy when $T_i = T$; $\theta_i = h\nu_i/k$ is the characteristic temperature (θ_1 = 1990°, θ_2 = 960°, θ_3 = 3380°, θ_4 = 3350°), g_i is the multiplicity of degeneracy of the i-th vibration ($g_1 = g_3 = g_4 = 1$, $g_2 = 2$); $x_i = \exp(-\theta_i/T_i)$; $p^{(j)}$ is the partial pressure of the j-th component of the mixture. The multiplier Z in the last equation of the system (2.5) is connected with the presence of a resonance interaction of the vibrations ν_1 and ν_2 ($\theta_1 \approx 2\theta_2$) which produces joint relaxation of the vibrational energies ε_1 and ε_2. Since $T_1 \approx T_2$,

$$\frac{d}{dt}(\varepsilon_1 + \varepsilon_2) = Z\frac{d\varepsilon_2}{dt}, \quad Z = (1 + 4x_2 + x_2^2)/(1 + x_2)^2$$

In writing the kinetic equations (2.5) it is kept in mind that the vibrational exchange reaction (2.1) satisfies the resonance condition ($\theta_3 \approx \theta_4$), while the reactions (2.2)-(2.3) can be considered in the approximation of V−T relaxation since the vibrational temperatures of the modes ν_1 and ν_2 of the CO_2 molecule and mode ν_5 of the H_2O molecule are close to the gas temperature because of the smallness of the corresponding relaxation times [7].

3. For a complete description of the kinetics of relaxation in a jet of expanding gas the system (2.5) must be supplemented by the corresponding gasdynamical equations. In the present case it proves possible to consider the dynamics of the nonequilibrium gas in the approximation of the adiabatic motion of a medium with an effective adiabatic index γ which corresponds to a certain degree of freezing in of the vibrational component of the heat cpacity of the gas. This enables one to use in the kinetic equations (2.5) the solutions of gasdynamical equations corresponding to the isentropic flow of the gas with a constant γ. In isentropic processes the pressure p and temperature T of the gas are determined by the equations

$$p = p_0(1 + {}^1/_2(\gamma - 1)M^2)^{-\gamma/(\gamma-1)}, \quad T = T_0(1 + {}^1/_2(\gamma - 1)M^2)^{-1} \tag{3.1}$$

Here $M = u/c$ is the local Mach number, u and $c = \sqrt{\gamma p/\rho}$ are the local flow velocity and speed of sound, γ is the effective adiabatic index, ρ is the gas density, T_0 is the stagnation temperature, and p_0 is the total gas pressure.

In the case of discharge from a slot into a vacuum the values of M(X, Y) are found by numerical integration of the gasdynamical equations for steady plane isentropic flow. The calculations were conducted by É. A. Ashratov and G. K. Bunina by the method of characteristics [11]. The results of the calculations are presented below, in particular, the values of $M(\xi)$ along the central flow line (jet axis) at γ = 1.3, 1.4, and 1.5 (second, third, and fourth lines, respectively). The distance ξ from the slot is expressed in units of h_0, where h_0 is the halfwidth of the slot: $\xi = X/h_0$.

ξ	1	2	5	10	20	50	100
M	1.48	2.0	2.87	3.58	4.2	5.15	5.9
M	1.5	2.06	3.0	3.82	4.68	5.95	7.0
M	1.51	2.1	3.2	4.16	5.23	6.9	8.37

4. In Eqs. (2.5) it is convenient to change to the dimensionless variable ξ in accordance with $dt/dt = uh_0^{-1}\, d/d\xi$.

The system (2.5) together with (3.1) was solved by the Runge–Kutta method on an M-220 electronic computer. The dependence $M(\xi)$ for the central flow line was provided by interpolation of the corresponding data presented above. In the temperature region where experimental data are absent the rate constants were taken in accordance with their approximate expressions for the reactions (2.1)-(2.4). The initial state of the gas at $\xi = 0$ was assumed to be an equilibrium state with a single intial temperature $T_i = T_* = T_0 2/\gamma + 1$, initial pressure $p_* = p_0(2/\gamma + 1)^{\gamma/(\gamma-1)}$, and flow velocity $u = c_* = \sqrt{\gamma p_*/\rho_*}$ for the vibrational and translational degrees of freedom. Before the slot the gas was at rest ($u = 0$) and its temperature and pressure were T_0 and p_0, respectively. The calculations were conducted for a $CO_2-H_2O-N_2$ mixture with a large water vapor content. The vibration–rotation population inversion $\Delta N = N' - N$ was calculated for the transition ($\lambda_0 = 10.6\ \mu$) of the CO_2 molecule (001, J' = 21) → (100, J = 22):

$$\begin{gathered}\Delta N = g' \frac{2hc}{kT}\left\{n_v' B' \exp\left[-\frac{F(J')}{kT}\right] - n_v B \exp\left[-\frac{F(J)}{kT}\right]\right\} \\ n_v' = x_3(1-x_3)(1-x_2^2)(1-x_2)^2 \rho_{CO_2}, \\ n_v = x_1(1-x_3)(1-x_2^2)(1-x_2)^2 \rho_{CO_2}\end{gathered} \tag{4.1}$$

where n_v' and n_v are the populations of the vibrational levels (001) and (100); ρ_{CO_2} is the concentration of CO_2 molecules in the mixture; F(J') and F(J) are the energies of the rotational terms of the upper and lower states; B' and B are the corresponding rotational constants; g' and g are the statistical weights of the upper and lower laser levels (001, J') and (100 J). The rotational temperature was taken as equal to the gas temperature. For the gain index $\alpha(\lambda_0)$ at the center of the line an expression [12] was used which takes into account the combined effect of Doppler and Lorentz mechanisms of broadening:

$$\begin{gathered}\alpha(\lambda_0) = (\lambda_0^2/8\pi)\,\Delta N \tau_{21}^{-1} S(\lambda_0) \\ S(\lambda_0) = 2\pi^{-1/2}(\lambda/v)\, a \int_0^\infty (a^2 + y^2)^{-1} \exp(-y^2)\, dy \\ a = (\Delta\nu_L/\Delta\nu_D)\sqrt{\ln 2}, \quad v = \sqrt{2kT/m}, \quad \Delta\nu_D = v\sqrt{\ln 2}/c\lambda_0\end{gathered} \tag{4.2}$$

$\Delta\nu_D$ and $\Delta\nu_L$ are the Doppler and Lorentz halfwidths of the lines. Because of the absence of experimental data on the collisional broadening of CO_2 in water vapor it was assumed to be the same as in the case of pure carbon dioxide. In this case $\Delta\nu_L = 0.1\, p\sqrt{300/T}\ \mathrm{atm^{-1}\cdot cm^{-1}}$ and the transition probability $\tau_{21}^{-1} = 0.21$ $\mathrm{sec^{-1}}$ [13].

The effective index γ was determined in accordance with the equation (ignoring dissociation of the gas)

$$\begin{gathered}\gamma = 1 + 2(5 + 6/\varkappa + 3\varkappa_{H_2O}/\varkappa)^{-1} \\ \varkappa_{H_2O} = \rho_{H_2O}/\rho_{CO_2}, \quad \varkappa_{N_2} = \rho_{N_2}/\rho_{CO_2}, \quad \varkappa = 1 + \varkappa_{H_2O} + \varkappa_{N_2}\end{gathered}$$

where ρ_{CO_2}, ρ_{H_2O}, and ρ_{N_2} are the concentrations of the corresponding molecules. This equation takes account of the fact that the vibrations ν_4 of the N_2 molecule and ν_3 of the CO_2 molecule can be considered as frozen in when estimating γ, while the vibrations ν_1 and ν_2 of the CO_2 molecule and ν_5 of the H_2O molecule, being in equilibrium with the gas temperature, can be assumed to be fully excited in the most important region of the jet for the relaxation and gasdynamics of the discharge at stagnation temperatures $T \approx 2000°$ which are of interest.

Typical distributions of the gas temperature T and the vibrational temperatures T_i along the central flow line of the jet for a $CO_2-H_2O-N_2$ mixture are presented in Fig. 1. The composition and parameters of the mixture are as follows: $\varkappa_{N_2} = 4$, $\varkappa_{H_2O} = 0.5$, $T_0 = 2000°K$, $p_0 = 20$ atm, $h_0 = 0.04$ cm. Curves 1, 2, and 3 show the distributions of T_4, T_3, and T. It is seen from Fig. 1 that separation of the vibrational temperature T_4 of the N_2 molecule occurs immediately behind the slot, followed by separation of the vibrational temperature T_3 of the CO_2 molecule from the gas temperature at distances of two to three units. The vibrational temperatures of modes ν_1 and ν_2 of the CO_2 molecule coincide with the gas temperature ($T_1 = T_2 = T$) because of the high rate of V–T relaxation caused mainly by the presence of water vapor. The behavior of the vibrational degrees of freedom, in which some of them are rapidly frozen in while others remain in equilibrium with the translational degrees of freedom in the process of expansion, is responsible for the possibility of an adiabatic description of the motion of a gas with vibrational nonequilibrium.

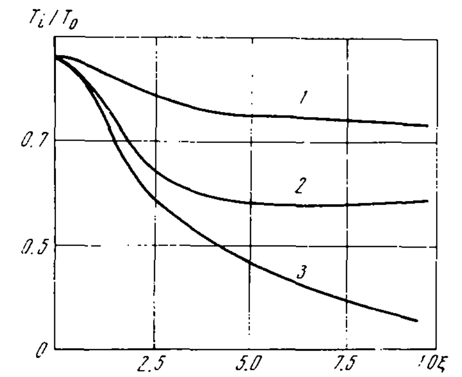

Fig. 1

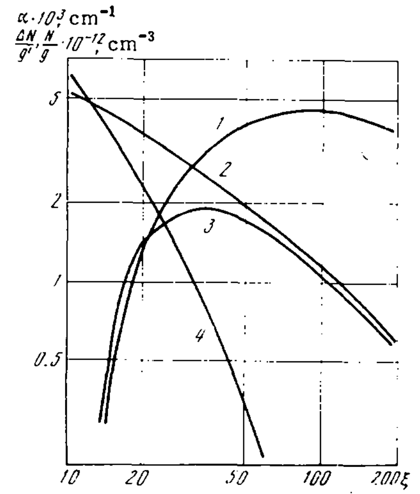

Fig. 2

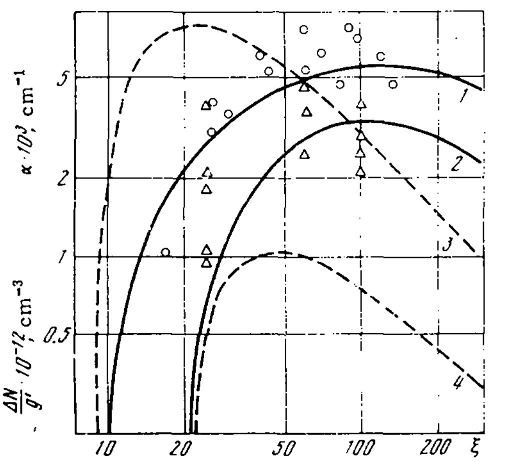

Fig. 3

Typical dependences of α, N'/g', $\Delta N/g'$, and N/g on the distance ξ along the jet axis from the slot for the $CO_2-H_2O-N_2$ mixture are presented in Fig. 2. The composition and parameters of the mixture are the same as for Fig. 1. Curves 1, 2, 3, and 4 correspond to the distributions of α, N'/g', $\Delta N/g'$, and N/g. It is seen from Fig. 2 that the gain index $\alpha(\xi)$ and the population inversion ΔN increase rapidly at first (the moment of formation of the inversion in Fig. 2 corresponds to the point of intersection of the curves N'/g' and N/g) and then, passing through maxima, slowly decrease with increasing distance. The maximum $\alpha(\xi) = \alpha_m$ in general does not coincide with the position of the maximum ΔN_m and is located at distances where the gas density is low ($\rho \approx 2\text{-}3 \cdot 10^{17}$ cm^{-3}). It is convenient to analyze the dependence $\alpha(\xi)$ presented in Fig. 2 by simplifying Eq. (4.2):

$$\alpha \sim \Delta N S(\lambda_0) \sim \begin{cases} T^{-1/2} \exp(-\theta_3/T_3^*) & \text{when } \Delta\nu_L \gg \Delta\nu_D \\ \rho T^{-1} \exp(-\theta_3/T_3^*) & \text{when } \Delta\nu_L \ll \Delta\nu_D \end{cases} \quad (4.3)$$

Here it is considered that when $n_V' \gg n_V$ for levels with J_m corresponding to the maximum in the vibration–rotation inversion ($J_m \sim T^{1/2}$)

$$\Delta N \sim \rho T^{-1/2} \exp(-\theta_3/T_3^*)$$

where T_3^* is the frozen-in value of the vibrational temperature T_3. In the region of ξ where the gas density is high and the shape of the line is Lorentzian ($\Delta\nu_L \gg \Delta\nu_D$) the value of α does not depend on ρ. Since $T_3^* \approx$ const while the gas temperature T decreases with an increase in ξ, according to Eq. (4.3) the gain index α must increase. At large distances ξ where there is a Doppler line shape ($\Delta\nu_L \ll \Delta\nu_D$) the variation in the density and not in the gas temperature ($T \sim \rho^{\gamma-1}$; when $\gamma = 1.3$, $\rho T^{-1} \sim \rho^{0.7}$) is the determining factor for $\alpha(\xi)$, and a decrease in α is observed with an increase in ξ. The maximum in $\alpha(\xi)$ is reached at the distance ξ_m where the collisional and Doppler line widths are approximately comparable in value: $\Delta\nu_L \approx \Delta\nu_D$. According to the calculations this corresponds to pressure of 5-10 mm Hg and distances of $\xi \sim 100$ (see curve 1 in Fig. 2). With an increase in T_0 and ρ_0 the position of the maximum of $\alpha(\xi)$ is shifted in the direction of larger ξ_m. According to the calculations conducted the greatest values of the gain index and population inversion in a jet of the $CO_2-H_2O-N_2$ gas mixture ($\varkappa_N = 4$, $\varkappa_{H_2O} = 0.5\text{-}1$) expanding through a slot with $h_0 = 0.04$ cm are reached at $T \approx 2200°K$ and $p \approx 20$ atm and are $\alpha_m = 0.5 \cdot 10^{-2}$ cm^{-1} and $\Delta N_m = 1 \cdot 10^{14}$ cm^{-3} at $\varkappa_{H_2O} = 0.5$ and $\alpha_m = 0.3 \cdot 10^{-2}$ cm^{-1} and $\Delta N_m = 0.5 \cdot 10^{14}$ cm^{-3} at $\varkappa_{H_2O} = 1$. The presence of the optima α_m and ΔN_m with respect to the pressure and stagnation temperature is explained by the fact that with an increase in T_0 and p_0 the relaxation rate of CO_2 (001) increases along with the increase in the initial stock of excited CO_2 molecules. While the first circumstance contributes to the increase in α_m and ΔN_m the second circumstance opposes it.

The water vapor in the mixture considerably increases the rates of the relaxation processes, including the process of deactivation of the upper laser level CO_2 (001). In connection with this high cooling rates are required in the gas jets to obtain the necessary population inversion and gain per unit length. One possible means of increasing the cooling rate dT/dt is a decrease in the size of the slot. As calculations show (see [6]), a marked increase in α_m and ΔN_m can be obtained with a constant value of the so-called similarity parameter p_0h_0 = const through a decrease in h_0. The optimum value of the similarity parameter for the $CO_2-H_2O-N_2$ mixture ($\varkappa_{N_2} = 4$, $\varkappa_{H_2O} = 0.5$-1) proves to be $p_0h_0 \approx 0.8$ atm · cm. The cooling rate is also determined by the composition of the mixture or more accurately by its effective adiabatic index γ. High cooling rates dT/dt correspond to large values of γ. Therefore it is expedient to increase γ by adding to the mixture components which have a large γ and are inefficient in deactivating the upper laser level CO_2 (001), such as He, Ar, and N_2. In the present case nitrogen fulfills a double function: it increases the effective relaxation time of CO_2 (001) and raises the effective adiabatic index γ of the mixture.

5. Experimental values of α [3] and those calculated in the present work as a function of the distance ξ from the slot along the jet axis are presented in Fig. 3. The calculated dependences of the population inversion $\Delta N/g'$ are shown by dashed lines in Fig. 3. The experiments of [3] were conducted on a shock tube. The gas, heated by a reflected shock wave to the temperature T_5 (pressure p_5), escaped through a narrow slot of halfwidth $h_0 = 0.04$ cm into a chamber with a low inflation pressure $p_\infty < 1$ mm Hg.

In the experiments the gain index for the weak signal of CO_2 laser emission at $\lambda_0 = 10.6\,\mu$ was measured at the jet axis of a $CO_2-H_2O-N_2$ gas mixture; the optical axis was parallel to the slot. Curves 1 and 3 (Fig. 3) correspond to the gain index α and population inversion $\Delta N/g'$ calculated for a $CO_2-N_2-H_2O$ mixture with $\varkappa_{N_2} = 4$, $\varkappa_{H_2O} = 0.1$, $T_0 = 1700$°K, and $p_0 = 47$ atm. The experimental points of α corresponding to these conditions are denoted by circles. The other data presented in Fig. 3 pertain to a $CO_2-N_2-H_2O$ mixture with the parameters: $\varkappa_{N_2} = 4$, $\varkappa_{H_2O} = 1$, $T_0 = 2250$°K, $p_0 = 22$ atm. In comparing the theory and experiment it was assumed that the flow was stabilized and the parameters T_0 and p_0 coincide with the gas parameters T_5 and p_5 behind the reflected shock wave.

The time of establishment of stationary discharge τ_s can be estimated as in [14], where it is shown that for one-dimensional flow of an ideal gas in nozzles $\tau_s \approx 10\,l/u$, where l is the nozzle length and u is the flow velocity. In the present case the nozzle length can be taken as $l \approx 5$ cm; u $\approx 10^5$ cm/sec and $\tau_s \approx$ 500 μsec. In the same time ($\sim 300\,\mu$sec) following the moment of arrival of the discharge front in the experiments of [3] stable gain was observed which lasted ~ 2 msec. There is satisfactory agreement between the calculations and the results of the experiment [3].

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