VIBRATIONAL TEMPERATURES OF CARBON DIOXIDE IN THE REACTION PRODUCTS OF CARBON MONOXIDE AND NITROUS OXIDE UNDER GASDYNAMIC LASER CONDITIONS

I. EXPERIMENTAL METHOD

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A method is developed for the experimental determination of the vibrational temperatures of asymmetric and combined symmetric modes of carbon dioxide, based on the simultaneous recording of the spontaneous and stimulated radiation characteristics of the CO + N_2O reaction products under gasdynamic laser (GDL) conditions.

The detailed investigations [1-4] of the kinetics of the vibrational relaxation of mixtures containing CO_2-N_2 performed in recent years in connection with the development of gasdynamic lasers (GDL) permitted the study of the fundamental dependences of the relaxation processes on the thermodynamic parameters, the mixture composition, the flow gasdynamics for chemical equilibrium of flows being cooled. At the same time, from both the viewpoint of investigating the fundamental singularities of the influence of vibrationally excited particles on the progress of chemical reactions [5], and the viewpoint of investigating the possibility of producing highly efficient chemical gasdynamic lasers [6], the investigation of the chemically reacting, laser-active gas media is of interest.

The results of a systematic investigation of the amplifying and radiation characteristics of vibrationally nonequilibrium expanding fluxes of products of the reaction between nitrous oxide and carbon monoxide diluted by nitrogen and helium are described in a series of papers exposed by this methodological paper. Such mixtures have been successfully used to produce the working body of CO_2 gasdynamic lasers [7, 8].

An investigation of the amplification of the population inversion and the vibrational temperatures of the CO_2 molecule lasing levels as a function of the gas temperature in the space ahead of the nozzle and of the composition of the chemically reacting mixtures and the inert comparison mixtures has been performed at the wavelength $\lambda = 10.6\ \mu m$. Besides the general investigation of the influence of the reacting mixture composition and its initial temperature on the mechanism of population inversion formation in the reaction products, the main purpose of the research was to find the conditions under which superequilibrium chemical pumping of the vibrational degrees of freedom of the CO_2 molecules, the products of chemical reactions, would be observed.

A single experimental and theoretical method was used in this set of papers. The scheme of carbon dioxide gas relaxation with two vibrational temperatures T_3 and T_2 [3] was used to describe the kinetics of the vibrational energy exchange. These temperatures characterize the Boltzmann distribution over the vibrational levels within the asymmetric and symmetric CO_2 modes, respectively. The principles for the measurement of the vibrational temperatures T_3 and T_2 of CO_2 molecules in mixtures with N_2 and CO are elucidated in [9].

Let us briefly examine results on the kinetics of chemical transformations in the mixtures investigated. According to [10, 11], progress of the reaction in the mixture CO + N_2O can be characterized by a period of induction over whose extent abrupt ignition of the mixture occurs. The duration of the induction period is described by the empirical relationship [10]

$$\Delta\tau_f^{-1}\,[N_2O]^{-1} = 3.47\cdot 10^{13}\exp\left[-\frac{25600}{RT}\right], \qquad (1)$$

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TABLE 1. Rate Constants of Reactions (2)-(6) (k_1, k_2, k_4, k_5, $cm^3 \cdot mole^{-1} \cdot sec^{-1}$; k_3, $cm^6 \cdot mole^{-2} \cdot sec^{-1}$)

Rate constants	Approximate dependence	Temperature range, °K	Reference
k_1	$2{,}1 \cdot 10^{11} \exp\left(-\frac{17300}{RT}\right)$	1216—1655	[12]
	$1{,}1 \cdot 10^{11} \exp\left(-\frac{23000}{RT}\right)$	1317—1908	[13]
k_2	$5 \cdot 10^{14} \exp\left(-\frac{58000}{RT}\right)$	1500—2500	[14]
	$5 \cdot 10^{13} \exp\left(-\frac{49500}{RT}\right)$	1700—2200	[15]
	$5 \cdot 10^{14} \exp\left(-\frac{59200}{RT}\right)$	840—1050	[16]
k_3	$3{,}4 \cdot 10^{11} \exp\left(\frac{23800}{RT}\right)$	1500—3000	[13]
	$\frac{1{,}8 \cdot 10^{19}}{T} \exp\left(-\frac{4000}{T}\right)$	1000—3500	[17]
	$5{,}05 \cdot 10^{15} \exp\left(-\frac{1800}{RT}\right)$	1000—3500	[17]
k_4	$10^{14} \exp\left(-\frac{28000}{RT}\right)$	900—2300	[17]
	$4{,}5 \cdot 10^{13} \exp\left(-\frac{24100}{RT}\right)$	1600—3000	[19]
	$3{,}63 \cdot 10^{13} \exp\left(-\frac{27200}{RT}\right)$	900—2300	[18]
k_5	$k_4/k_5 = 1{,}0$		[17]
	$0{,}5$		[20]
	$0{,}317$	1216—1655	[16, 12]

where $[N_2O]$ is the nitrous oxide (N_2O) concentration in moles/cm³. Let us use the analysis of chemical transformations in a $CO + N_2O$ mixture performed in [12], according to which the following five elementary stages are essential

$$N_2O + CO \xrightarrow{k_1} CO_2 + N_2 + 87.3 \text{ kcal/mole} \tag{2}$$

$$N_2O + M \xrightarrow{k_2} N_2 + O + M - 38 \text{ kcal/mole} \tag{3}$$

$$CO + O + M \xrightarrow{k_3} CO_2 + M + 125 \text{ kcal/mole} \tag{4}$$

$$N_2O + O \xrightarrow{k_4} 2NO + 36 \text{ kcal/mole} \tag{5}$$

$$N_2O + O \xrightarrow{k_5} N_2 + O_2 + 79.3 \text{ kcal/mole}. \tag{6}$$

The rate constants of the processes (2)-(6), recommended in [12-20], are presented in Table 1. Values of the constants k_2, k_4, k_5 obtained by different authors are in satisfactory agreement. The mean of the quantity k_1 from [12], [13] was used to compute the chemical transformations in reaction (2), and the estimate of k_3, equal to 10^{15} $cm^6 \cdot mole^{-2} \cdot sec^{-1}$, averaged from the data in [13, 17, 18], was used for reaction (4). According to [21], if the method of quasistationary concentrations of active particles is used, then we obtain for the relative content of atomic oxygen

$$\xi_0 = \frac{[O]}{[M]} = k_2 \left[k_3 \frac{\xi_{CO}}{\xi_{N_2O}} [M] + (k_4 + k_5) \right]^{-1}, \tag{7}$$

where [M] is the total number of molecules per unit volume.

TABLE 2. Ratios between the Rates of Consumption of the Nitrous Oxide Molecules in Reactions (3) and (2) and Atomic Oxygen in (5), (6), and (4) for Different Gas Temperatures T. Pressure P Was Assumed to Equal 5.0 atm, while the Ratio of the Concentrations Was $\xi_{CO}/\xi_{N_2O} = 5$

T, °K	1000	1300	1600	1700	1800	1900	2200	2500	2800
$\frac{d[N_2O]_3}{d[N_2O]_2}$	$1{,}6 \cdot 10^{-5}$	$0{,}3 \cdot 10^{-3}$	0,13	0,23	0,45	0,79			
$\frac{d[O]_5+d[O]_6}{d[O]_4}$	$2{,}5 \cdot 10^{-4}$	0,021	0,14	0,19	0,41	0,61	1,77	3,19	5,66

The comparative effiency of the progress of elementary stages (2)-(6) for the typical experimental conditions of this paper is presented in Table 2. For moderate temperatures $T_0 \leq 1700$-1800°K the rate of consumption of the N_2O molecules because of direct interaction with CO (2) essentially exceeds the rate of thermal decomposition of the nitrous oxide (3) $d[N_2O]_3/d[N_2O]_2 \ll 1$. Hence, for $T_0 \leq 1700$-1800°K the chemical transformation scheme in the mixture of carbon monoxide and nitrous oxide can be reduced approximately to the reaction of direct exchange interaction between these molecules (2). Since the quantity hence is $(d[O]_5 + d[O]_6)/d[O_4] \ll 1$, the atomic oxygen being formed because of thermal dissociation of the nitrous oxide (3) is consumed mainly in the recombination reaction (4). Therefore, the estimates made permit the consideration that the reaction products form a mixture of carbon dioxide and nitrogen molecules, the basis of the working mixture of a CO_2 gasdynamic laser, during the oxidation of carbon monoxide by nitrous oxide at the temperature $T_0 < 1700$-1800°K. At higher temperatures $T_0 = 1800$-2500°K, the efficiencies of the progress of the thermal dissociation reaction for N_2O (3) compared to exchange reaction (2), and the oxidation reactions of the N_2O molecules by atomic oxygen (5) and (6) compared to the recombination reaction (4) are sharply elevated (Table 2). This results in an increase in the content of nitrous oxide and molecular oxygen in the CO + N_2O reaction products. Thus, if estimates are carried out by means of (7) for the atomic oxygen concentration, then we obtain the value of 0.1-0.3 for the magnitude of the initial nitrous oxide concentration $\xi^0_{N_2O}$ or $\xi_{NO} \approx \xi_{O_2} \approx 1.0$-3.0% for the nitrous oxide and molecular oxygen concentrations at $T_0 = 1800$-2500°K for typical conditions of this research. The carbon dioxide content in the reaction products hence diminishes to 0.7-0.8 of that computed by means of (2)-(4). According to [22], admixtures of molecular oxygen in such quantities do not influence the magnitude of the population inversion in $CO_2 + N_2 + He$ (H_2O) mixtures. Analysis of the rate constants of vibrational energy exchange with the participation of nitrous oxide [3] shows that 1-5% of the NO molecules do not alter the mechanism of population inversion formation in the $CO_2 + N_2 + He$ mixture in practice. The use of mixtures with a certain excess of carbon monoxide in this paper permits a sharp reduction in the molecular oxygen content in the reaction products because of its consumption in the oxidation of the CO molecules.

Comparison mixtures from inert components were formed for reacting mixtures on the basis of the relationships

$$\alpha N_2O + \beta CO + \gamma N_2 + \delta He \rightarrow \alpha CO_2 + (\alpha + \gamma) N_2 + (\beta - \alpha) CO + \delta He \tag{8}$$

Comparing the vibrational nonequilibrium parameters, the gain coefficient, the population inversion, and the vibrational temperatures of the carbon dioxide lasing levels in an expanding reacting mixture and in a chemically inert comparison mixture, permits clarification of the role of the chemical transformations in vibrational energy exchange.

The use of a chemical nonequilibrium gas mixture with an elevated content of active particles to 0.1-0.4%, oxygen atoms, in gasdynamic lasers can result in a substantial change in the mechanism for population inversion formation. In particular, such a change is due to the efficient progress of exothermal reactions with a significant yield of vibrationally excited particles of the reaction products in the expanding flow. An example of such a reaction is the recombination of CO and O (4) examined in [6].

The vibrational nonequilibrium flux of chemical reaction products in a mixture of carbon monoxide and nitrous oxide was experimentally investigated in this research on an apparatus of the type of a pulsed gasdynamic laser in a shock tube. Chemical reactions were realized in the investigated mixtures in the shock reflected from the tube endface. The parameters of the tube used were: inner diameter, 94 mm; length of the

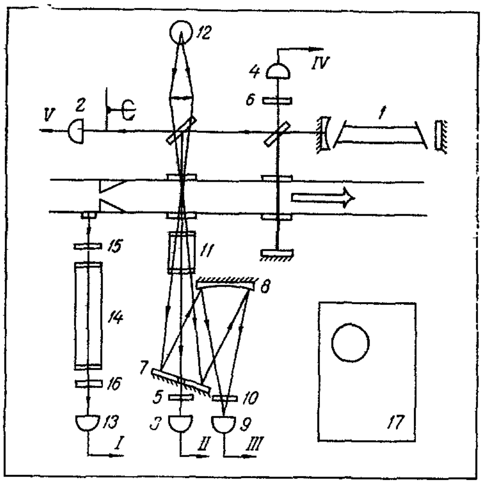

Fig. 1. Optical diagram of the experimental setup.

low pressure section, 5.0 m. A flat wedgelike nozzle with a critical section height of $h_* = 1.5$ mm, 80 mm length, and $\varphi = 22.5°$ halfangle was placed on the tube endface. The divergent part of the nozzle made the transition into a rectangular channel of constant section (expansion section). The degree of flow expansion is $h/h_* = 33$. The degree of purity of the gases used was the following: nitrogen of special purity with not less than a 99.5% content of the main component, medicinal nitrous oxide 99.6%, technical carbon monoxide subjected to careful drying, nutritive carbon dioxide purified by triple distillation from the solid state with recovery of the middle fraction [23].

The series of experiments described in this paper were conducted in such a way that the pressure behind the reflected shock P_5 remained constant, while the temperature T_5 varied within broad limits. The values of P_5 and T_5 were determined on the basis of measurements of the incident shock velocity. These measurements were accomplished by two piezoelectric sensors of 500-mm length and an electronic chronometer F-599. The errors in determining the gas temperature behind the reflected shock, due to inaccuracy in measuring the time of passage of the shock over the base segment, do not exceed 20-30°K. Let us note that according to [24] the inhomogeneity in the temperature along the length in the "hot plug" can reach 100°K. Additional heating of the gas in the region ahead of the nozzle behind the reflected shock can be computed in the case of investigating chemically reacting mixtures from the results on the thermal effects of reactions (2)-(4) under assumptions about the constancy of the pressure, $P_5 = P_0 = \text{const}$ [11] and the completeness of execution of the chemical transformation

$$\Delta T = \frac{\xi^0_{N_2O}}{\bar{c}_p}; \quad T_0 = T_5 + \Delta T, \tag{9}$$

where $\bar{c}_p$ is the mean value of the specific heat of the gas mixture in the temperature band (T_5, $T_5 + \Delta T$), and Q is the thermal effect of the reaction (2) or (3), (4). For the value $\xi^0_{N_2O} = 0.05$ characteristic for this research, the temperature rise ΔT was 450-650°K.

Taking account of the progress of reactions (5) and (6) at high temperatures $T_0 \geq 2200°K$ results in a 30-80°K diminution in heating of the gas in the region ahead of the nozzle. For $T_0 \geq 2500°K$ significant dissociation of the carbon dioxide accompanied by a temperature drop can occur in the "hot plug." Computations performed permit making the deduction that for $T_0 \approx 3000°K$ the degree of carbon dioxide dissociation in the time interval $t = (0-0.3) \cdot 10^{-3}$ sec which corresponds to and measures the amplifying and emission properties of the stream, does not exceed 20% with a corresponding $\alpha_d = 1 - \xi_{CO_2}/\xi^0_{CO_2}$ reduction in the temperature; hence, the influence of carbon dioxide dissociation on the change in the chemical composition of the mixture and the reduction in the temperature T_0 is neglected. For $T_0 \leq 2000°K$ underreaction of the mixture $CO + N_2O$ in the space before the nozzle and the reduction in the temperature T_0 compared to the rated values, which is due to this, are possible. This reduction was determined by means of the diminution in the IR radiation intensity from the space before the nozzle for the reacting mixture as compared with the chemically inert comparison mixture and did not exceed 100-120°K.

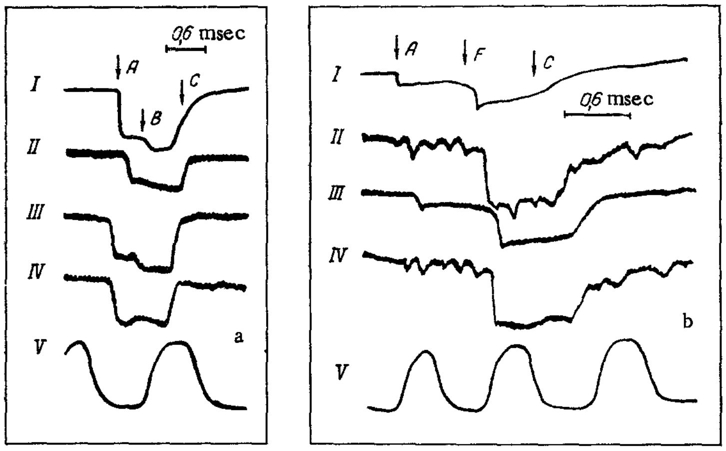

Fig. 2. Typical oscillograms: a) inert mixture 0.05 CO_2 + 0.05 N_2 + 0.2 CO + 0.7 He; P_0 = 5.3 atm; T_0 = 2000°K; b) reacting mixture of the initial composition 0.05 N_2O + 0.25 CO + 0.7 He; P_0 = 5.1 atm; T_0 = 1790°K (T_5 = 1160°K). A, F, B, C, respectively, are the times of reflected shock arrival, reacting mixture ignition, perturbation arrival on reflection from the contact surface, and rarefaction wave arrival.

Values of the pressure and gas temperature in the working section of the supersonic stream were computed from relationships for one-dimensional, isentropic ideal gas flow. The influence of vibrational relaxation processes on the supersonic flow parameters was taken into when using the effective adiabatic index γ_{ef} in the form

$$\gamma_{ef}=\frac{\Sigma c_{p_i}\xi_i}{\Sigma c_{v_i}\xi_i}=\frac{5/2R\xi_{He}+7/2R(\xi_{CO_2}+\xi_{N_2}+\xi_{CO})+3R\xi_{CO_2}}{\Sigma(c_{p_i}-R)\xi_i}. \tag{10}$$

Total freezing of the vibrations of the N_2 and CO molecules as well as of the asymmetric mode of the CO_2 (ν_3) vibrations and thermalization of the symmetric CO_2 modes were assumed. The gas temperature T in the supersonic stream under investigation (under the conditions of this research) was (0.1-0.2)T_0. The total error in determining T, due to inaccuracy in the obtained values of T_0 and the simplified taking account of the influence of the vibrational energy exchange, did not exceed 20-40°K and did not influence the results of measuring the vibrational temperatures T_3 and T_2.

The optical diagram of the apparatus is presented in Fig. 1. The gain coefficient was measured in two sections of the stream at 140 and 350 mm from the plane of the critical nozzle section. An LG-23 power-stabilized electrical discharge CO_2 laser 1 was used as source of probing radiation. The constancy of the radiation intensity and its measurement during the experiment were checked by using a reference photoresistor 2. The photoresistors 3 and 4 recorded changes in the laser radiation intensity during passage of the active section into the gas stream in the first and second working sections. Extraction of the needed spectrum range in the gain measurement channels was performed by the dispersion filters 5 and 6 with the passband λ = 10.6 ± 0.6 μm.

The IR radiation intensity in the CO_2 4.3 μm and CO 4.7 μm bands was measured in the first stream section. The flat 7 and spherical 8 mirrors focused the gas radiation on the sensor of the photoresistor 9. Spectrum selection was by the dispersion filter 10 with the passband λ = 4.75 ± 0.6 μm. The absorptive cuvette 11 was filled with a mixture of nitrous oxide and carbon dioxide in a 1 : 1 ratio for a total pressure, equal to the atmospheric value. Use of a cuvette permits the establishment of the possible influence of absorption in the cold, near-wall layers of the stream on the results of measuring the spontaneous radiation intensity in the vibrational–rotational 4.3 μm CO_2 and 4.7 μm CO bands.

Calibration of the IR radiation recording diagram was carried out by measuring the carbon monoxide radiation intensity (the 4.7 μm band) behind the incident shock being propagated in pure CO and the mixture CO + N_2 ($0.1 \leq \xi_{CO} \leq 1.0$). To do this, a connecting section assuring the smooth passage of the cylindrical shock tube section into the 50 × 80 mm rectangular channel of the expansion section was mounted on the shock tube endface in place of the nozzle. The optical diagram for IR radiation intensity measurement during calibration was kept the same as in the tests with the supersonic flows under investigation.

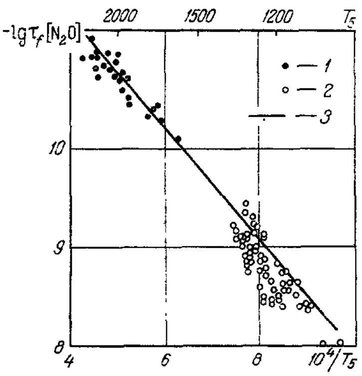

Fig. 3. Results of measuring the duration of the induction period in the mixture $CO + N_2O$: 1) data from [10]; 2) this paper; 3) formula (1) $\log(\tau_f[N_2O])$, mole • sec/cm^3.

The carbon monoxide IR radiation intensity behind the incident shock was computed by means of semiempirical formulas [25, 26]. The error in calibrating the response of the optical system in this research, determined by means of the spread in the experimental results, did not exceed ±15%. The constancy of the recording system response was checked during execution of the experiments by means of the radiation of a standard blackbody 12 at the temperature $T = 1800 \pm 10$°K.

Determination of the duration of the existence of unperturbed conditions in the "hot plug" and of the delay in ignition in the reacting mixtures was carried out by recording the IR radiation intensity in the $\lambda = 4.6$-5.2 μm spectrum range by the photoresistor 13. The spectrum selection in this channel was accomplished by the absorptive cuvette 14 filled with a $N_2O{:}CO_2 = 1{:}1$ mixture at atmospheric pressure, and by the filters 15 and 16.

Photoresistors with a Ge–Au based sensor cooled by liquid nitrogen were used in this research. After preamplification the signals from the photoresistors were delivered to a five-beam oscillograph Cl-33 17. Time resolution of the optical channels was not more than 10 μsec.

A typical oscillogram obtained for the unreacting mixture $CO_2 + N_2 + CO + He$ is represented in Fig. 2a. Curves I-V are records of the IR radiation intensity from the space ahead of the nozzle (I), the amplification of the test laser radiation (I), and the IR radiation intensity (III) in the first section of the stream, the radiation amplification of the test laser in the second stream section (IV) and the intensity of the reference beam of laser radiation during the experiment (V). The constant level of IR radiation intensity during 0.5 msec from the time of shock reflection (the section A–B, Fig. 2a) corresponds to the unperturbed state of the gas in the "hot plug" at which the amplifying and radiation properties of the supersonic stream were measured in this research. Ignition of the reacting $CO + N_2O$ mixture (point F) in Fig. 2b results in an abrupt increase in the radiation intensity from the space ahead of the nozzle because of the rise in the temperature T_0 and from the stream at the same time as the appearance of a gain in the probing radiation. Results of measuring the delay in ignition in the mixture $CO + N_2O + He + N_2$ at the temperature $T_5 = 1000$-1350°K ($P_5 = 5.0$-20.0 atm, $\xi^0_{CO} = 0.05$-0.65) are in good agreement with the results of extrapolation, according to the dependence (1), of the data [10] obtained at higher temperatures T_5 (Fig. 3).

The vibrational temperatures of the asymmetric T_3 and symmetric (combined) T_2 modes of carbon dioxide, and therefore, the upper (001) and lower (100) working levels of the lasing transition in this molecule, were determined by the gain coefficient K_0 and the spontaneous radiation intensity of the molecules CO_2 (4.3 μm) and CO (4.7 μm) measured in test. The relative population inversion of the carbon dioxide lasing levels $\Delta N = (N_{001} - N_{100})/N_{CO_2}$ was calculated on the basis of measured values of the gain coefficient K_0 according to [26, 27], where N_{CO_2} is the density of CO_2 molecules.

The error in measuring the gain coefficient and therefore the relative population inversion is determined under the experimental conditions of this research by mainly the shift in the generation frequency within the lines of active medium amplification of the probing CO_2 laser, and is +0, −20-30% [28]. This uncertainty can result in a rise in the measured value of T_2 by 40-50°K as compared to its actual value for $T_2 = 400$-600°K.

Generation in the probing laser was assumed at the line P20. The appearance of the lines P18 and P22 in the generation spectrum does not affect the results of measuring the vibrational temperature T_3. The assumption of generation at the line P18 results in a rise in the computed value $T_2 \approx 7$-8% and to a reduction of 20-30% in ΔN as compared with the assumption of generation at the line P20. The maximum relative errors in measuring the quantities T_2 and ΔN for the appearance of the line P22 in the generation spectrum have the same magnitude but opposite sign. The calibration accuracy ±15% achieved in this research for the optical system response corresponds to the error in determining the temperature T_3, which does not exceed ±50°K for T_3 = 1000-1500°K and $T_2 \pm$ 20-30°K for T_2 = 400-700°K. Possible uncertainties in the concentrations of the radiating components of the CO_2 and CO molecule components do not exert noticeable influence on the results of measuring the vibrational temperature T_3, and at moderate temperatures $T_0 \leqslant$ 1800-2000°K on the results of measuring the quantities ΔN and T_2. A diminution by 20-30% in the carbon dioxide content in the reaction products at T_0 = 2000-3000°K as compared with the computed value from (8) introduces a systematic error in the measurement of the quantities T_2 and ΔN in this range of temperatures T_0. The values of T_2 obtained from tests under the assumption about the composition of the reaction products determined from (8) turn out to be exaggerated by 5-7%, and of ΔN are reduced 20-30% as compared to the actual values of the vibrational temperature of the lower lasing level of CO_2 and the population inversion in the stream under investigation at T_0 = 2000-3000°K.

The method described for the measurement of the vibrational energy exchange characteristics in gas mixtures containing CO_2 was tested in experiments with the well-known mixtures $CO_2 + N_2 + He$ (H_2O). Good agreement between the results obtained and theoretical and experimental data of other authors was established.

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