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

III. SUPEREQUILIBRIUM CHEMICAL PUMPING OF VIBRATIONAL LEVELS OF THE CO_2 MOLECULE

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The conditions under which the superequilibrium chemical pumping of the laser levels of CO_2 molecules occurs in a gasdynamic laser based on products of the reaction $CO + N_2O$ are established experimentally.

The use of exothermic reactions to create the active medium for a gasdynamic laser (GDL) opens up the possibility in principle of obtaining additional superequilibrium pumping of the GDL through the energy of chemical transformations. The theoretical prerequisites for the creation of such a chemical—gasdynamic laser (CGDL) are laid out in [1]. In a CGDL one can use certain exothermic reactions having a considerable release of energy into vibrational degrees of freedom of the type of recombination reactions, which occur efficiently under the conditions of low gas temperatures which are typical of GDL [2], or exchange reactions, for which vibrational excitation of the reagents is essential to overcome the activation barrier [3].

An increase in the generation power of a CO_2 GDL through chemical reactions during the oxidation of carbon monoxide by oxygen was observed in [4]. Generation was obtained in [5] during the supersonic mixing of carbon bisulfide with partially dissociated oxygen. The preliminary results of an investigation of the supersonic chemical pumping of a CO_2, GDL on products of the reaction $CO + N_2O$ are presented in [6].

A systematic comparative investigation of the amplification factor and the vibrational temperatures of CO_2 in an expanding stream of $CO + N_2O$ reaction products and of an inert comparison mixture is carried out in the present report in order to study the influence of chemical reactions on the characteristics of vibrational energy exchange in reacting mixtures based on carbon monoxide and nitrous oxide. The choice of the specific composition of the reacting mixture for this investigation was based on the results presented in [9], where we found an increase in the amplification factor and the vibrational temperature of the asymmetric mode of CO_2 upon a considerable (up to 70%) dilution of the reacting mixture with helium. Reacting mixture 1 had the following initial composition: $0.05\ N_2O + 0.25\ CO + 0.7\ He$. The composition of the comparison mixture 2, $0.05\ CO_2 + 0.05\ N_2 + 0.2\ CO + 0.7\ He$, corresponded to the final products of the chemical reactions in mixture 1.

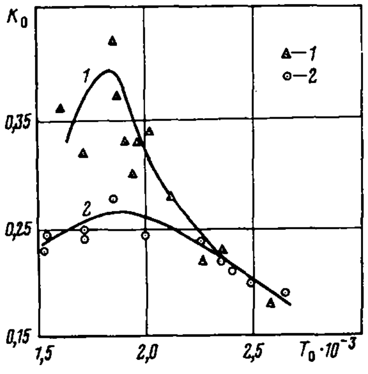

Fig. 1. Dependence of amplification factor K_0 on the gas temperature T_0 ahead of the nozzle: 1) mixture 1; 2) mixture 2.

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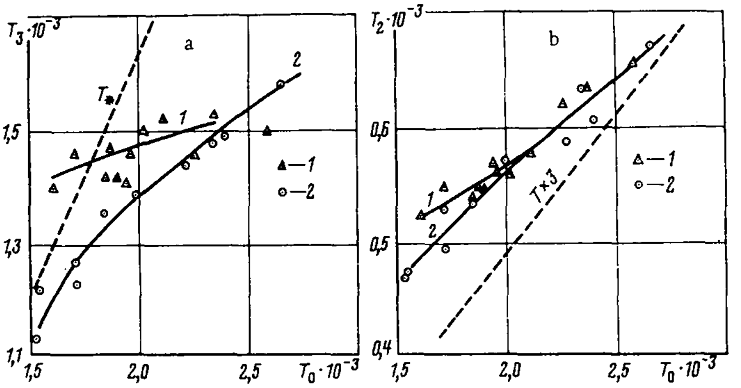

Fig. 2. Vibrational temperatures T_3 (a) and T_2 (b) of carbon dioxide as a function of temperature T_0 ahead of the nozzle. Notation same as in Fig. 1.

The range of variation of the gas temperature T_0 in the prenozzle region was 1500-2800°K. The pressure P_0 was kept within the limits of 5.2 ±0.5 atm.

The temperature dependences $K_0(T_0)$ of the amplification factor in the first stream cross section at a distance of 140 mm from the critical cross section of the nozzle for mixtures 1 and 2 are presented in Fig. 1. The principal feature of the dependences $K_0(T_0)$ shown in Fig. 1 is the considerably higher amplification factors in the reacting mixture 1 than in the inert mixture 2 in the region of temperatures T_0 = 1600-2200°K. The maximum value of K_0 for the reacting mixture is 0.43 m^{-1} at T_0 = 1900°K. The maximum value of K_0 for the comparison mixture is reached at the same temperature T_0 and is 0.28 m^{-1}. At higher temperatures ahead of the nozzle ($T_0 \geqslant 2200$°K) the amplification factors for the reacting mixture and the comparison mixture coincide. An analysis of the kinetics of the chemical transformations in the reacting mixture showed that the reaction CO + N_2O at $T_0 \leqslant 2200$°K cannot proceed fully beyond the reflected shock wave before the moment of rupture of the prenozzle diaphragm (~50 μsec) and occurs partially within the nozzle. At a temperature $T_5 \leqslant 1000$°K beyond the reflected shock wave (which corresponds to $T_0 \leqslant 1500$°K with allowance for heat release) the carbon dioxide content in the stream of reaction products of mixture 1 decreases markedly owing to the strongly incomplete reaction. This leads to a sharp decrease in the amplification factor in mixture 1 at $T_0 \leqslant 1600$°K (curve 1, Fig. 1).

The dependences of the relative population inversion $\Delta N = (N_{001}-N_{100})/N_{CO_2}$ on T_0 for mixtures 1 and 2 are analogous to the corresponding functions $K_0(T_0)$. The maximum value of ΔN in the reacting mixture 1 is reached at T_0 = 1800-1900°K and is 4.0-4.5%. The maximum value of the relative population inversion in the comparison mixture 2 does not exceed 3.3% (T_0 = 2000°K).

The measurements of the CO_2 vibrational temperatures showed that the increased values of the amplification factor and the population inversion observed in the reacting mixture in comparison with the chemically inert mixture at T_0 = 1600-2200°K are connected with the considerable increase in the population of the upper laser level of the CO_2 molecules (the temperature T_3 of the asymmetric vibration mode) in mixture 1 in comparison with that in mixture 2 (Fig. 2a). The vibrational temperature T_3 of the carbon dioxide in the test mixtures as a function of the temperature ahead of the nozzle is presented in Fig. 2a. The gas temperature T_* at the critical cross section is shown by a dashed line. The general character of the dependence of T_3 on T_0 is the same for mixtures 1 and 2: The vibrational temperature of the asymmetric mode of CO_2 increases with an increase in T_0, with the difference T_*-T_3 also increasing owing to the degradation of the conditions for the "freezing in" of the vibrational energy (ν_3) of the CO_2 molecules and those of N_2 and CO at high gas temperatures. The values of T_3 in mixtures 1 and 2 almost coincide at $T_0 \geqslant 2200$°K. In the region of temperatures T_0 = 1600-2200°K the vibrational temperature T_3 in the reacting mixture 1 considerably exceeds the value of T_3 for the inert comparison mixture 1, while at T_0 = 1600-1800°K the value of T_3 in the reacting mixture exceeds the temperature T_* in the critical cross section of the nozzle. For the reacting mixture 1 the vibrational temperature T_3 increases from 1420 to 1500°K as the temperature in the prenozzle space is raised within the limits of 1600-2700°K. For the comparison mixture 2 the value of T_3 varies from 1200 to 1550°K in the same range of values of T_0.

The vibrational temperature of the lower laser level (T_2, combined mode — symmetric vibrations of the CO_2 molecule) for the reacting mixture in the region of temperatures T_0 = 1600-2100°K is somewhat

TABLE 1. Values of Induction Period, τ_{ind} [7], Characteristic Time of Consumption of Nitrous Oxide, $\tau_{N_2O} = \{[K_1\xi_{CO} + K_2 + (K_4 + K_5)\xi_0]\,[M]\}^{-1}$, and Concentration ξ_0 of Atomic Oxygen in Mixture 1 at "Hot Plug." Characteristic Times of Occurrence of CO + O Recombination, $\tau_{rec}{}^* = (K_3\xi_0\,[M]^2)^{-1}$, of Reduction in Translational Temperature, $\tau_T^* = h_*\,(c_*\tan\varphi)^{-1}$, and of V—T Relaxation of Asymmetric Mode of CO_2, $\tau_{\nu_3} = (P_*Q_{23}{}^*)^{-1}\,(\xi_{N_2} + \xi_{CO})/\xi_{CO_2}$ at Critical Cross Section of Nozzle. c_*, P_*, Velocity of Sound and Pressure at Critical Cross Section; h_*, Height of Critical Cross Section; φ, Aperture Half-Angle of Nozzle; Q_{23}, Probability of Vibrational Relaxation in Processes of the Type $CO_2(\nu_3) + M \rightarrow CO_2(\nu_2) + M + \Delta E$ at $T = T_*$ (times τ_i expressed in μsec)

T_0, K	T_*, K	τ_{ind}	$\tau_{N_2O}\left(\frac{T_0 \div T_*}{2}\right)$	ξ_0, %	τ^*_{rec}	τ^*_T	$\tau^*_{\nu_3}$
1500	870	—	2369	$7{,}32\cdot10^{-4}$	5,67	3,24	2,56
1750	1120	1050	625	$9{,}4\cdot10^{-3}$	7,69	3,02	2,14
2000	1380	148	280	0,1	9,1	2,83	1,68
2250	1630	41,4	106	0,26	12,8	2,77	1,32
2500	1890	16,4	24,3	0,76	15,8	2,64	1,01

higher than the corresponding values for the inert comparison mixture (Fig. 2b). The values of T_2 for the reacting and inert mixtures coincide at $T_0 \geqslant 2200$°K. Increasing with an increase in the temperature ahead of the nozzle, the vibrational temperature T_2 of the combined mode exceeds the translational temperature T of the gas (dashed line of Fig. 2b) by more than 3 times for both mixtures.

In the entire range of temperatures T_0 investigated the amplification factors for mixtures 1 and 2 at the second cross section of the stream at a distance of 350 mm from the critical cross section of the nozzle are about half as large as at the first cross section.

The increase in the values of the amplification factor K_0, the population inversion ΔN, and the temperature T_3 of the asymmetric mode of CO_2 vibrations in the reacting mixture 1 in comparison with the values of the same parameters in the inert mixture 2, observed in the tests, indicates that superequilibrium chemical pumping of the vibrational levels of the CO_2 molecules — products of the chemical transformations occurring in the nozzle — takes place under the conditions of the present work. A prolonged induction period, comprising 0.05-1.0 msec (τ_{ind}, Table 1), is traced when the reacting mixture 1 is investigated in the region of temperatures $T_0 = 1500$-2200°K; when it ends there is a sharp increase in the intensity of the IR emission from the prenozzle space and from the gas stream simultaneously with the appearance of amplification of the radiation of the test laser.

The characteristic time of occurrence of the chemical transformations in the reaction CO + N_2O, determined from the time τ_{NO_2} of decrease in the nitrous oxide concentration, is 0.1-0.6 msec under these conditions (see Table 1). The measurements of the amplification and radiation parameters of the supersonic stream in the present work were made over the time interval of 0.0-0.3 msec from the moment of ignition of the mixture. The incompleteness of the chemical transformations in the prenozzle space and the high content of active particles (O atoms) in the reacting mixture (Table 1) create the conditions for the occurrence of the individual stages of the chemical reaction in the expanding stream.

The incompleteness of the chemical reaction at a low temperature, and the appearance of NO and O_2 molecules in the composition of the final products at high temperatures [8] as well, lead to the fact that the concentration of CO_2 molecules in the supersonic stream of the reacting mixture 1 proves to be lower than that in the comparison mixture 2. A calculation of the temperature T_0 in the reacting mixture under the assumption that the chemical transformations are complete and without allowance for the appearance of NO and O_2 as final reaction products gives values of T_0 which are overstated by 50-150° in comparison with the actual gas temperature at the "hot plug." Allowance for the enumerated factors can only lead to an increase in the observed difference in the amplification factor K_0, the relative population inversion ΔN, and the vibrational temperature T_3 in the reacting and inert mixtures 1 and 2.

The increased values of K_0, ΔN, and T_3 obtained for the reacting mixture 1 at a temperature $T_0 \leqslant 2200$°K are evidently due to the formation of vibrationally excited molecules during the occurrence of the individual stages of the chemical reactions in the vibrationally nonequilibrium stream as the gas moves through the nozzle. The formation of vibrationally excited CO_2, N_2, and CO molecules can occur in the following reactions:

$$CO+N_2O\xrightarrow{k_1}CO_2+N_2+87.3 \text{ kcal/mole}, \tag{1}$$

$$O+N_2O\xrightarrow{k_5}N_2+O_2+79{,}3 \text{ kcal/mole}, \tag{2}$$

$$CO+O+M\xrightarrow{k_3}CO_2+M+125 \text{ kcal/mole}. \tag{3}$$

A considerable part of the chemical energy of reactions (1)-(3) can be released into vibrational degrees of freedom [2, 3].

Under the experimental conditions, which correspond to increased values of the amplification factor, in the reacting mixture 1 the concentrations of atomic oxygen and carbon monoxide in the prenozzle space are 0.1-0.3 and 20%, respectively, while the concentration of nitrous oxide molecules does not exceed 0.5-1.0%. A comparison of the rates of occurrence of the reactions (1), (2), and (3), made using the equilibrium constants K_1, K_5, and K_3 (Table 1 of [8]) under the assumption that $T = T_*$, shows that under these conditions the recombination reaction (3) is dominant. The characteristic recombination time under the conditions of the critical cross section of the nozzle, τ^*_{rec}, is presented in Table 1 for mixture 1. In the region of temperatures $T_0 = 1700$-2200°K the value of τ^*_{rec} is 6.0-9.0 μsec. This value has the same order of magnitude as the characteristic times of gas cooling, τ^*_T, and the relaxation time of the asymmetric mode of vibrations of the CO_2 molecules, $\tau^*_{\nu3}$, in the critical cross section of the nozzle (Table 1). Rapid freezing in of the reaction (3) occurs in the critical region of the nozzle owing to the drop in gas density.

It is possible that the increased values of the amplification factor K_0 and the vibrational temperature T_3 in the reacting mixture 1 recorded in the present work are due to the formation of vibrationally excited molecules during the occurrence of the recombination reaction (3) in the region of the nozzle near the critical cross section. If the entire energy effect of the recombination reaction (3) of 125 kcal/mole is distributed into the vibrational degrees of freedom of the products of the chemical transformations then this may explain the observed superequilibrium vibrational excitation of the CO_2, N_2, and CO molecules. In fact, one recombination act (3) can lead to the excitation of $n \approx 18$ quanta of the asymmetric mode of carbon dioxide. Then, with allowance for the relationship of the characteristic times τ^*_{rec}, τ^*_T, and $\tau^*_{\nu_3}$, the increase in the relative population inversion in the stream at $T_0 = 2000$°K due to chemical excitation is

$$\Delta N \simeq \frac{n\xi_0}{\xi_{CO_2}+\xi_{N_2}+\xi_{CO}}\left(\frac{\tau^*_{\nu_3}}{\tau^*_T}\right)\left(\frac{\tau^*_T}{\tau^*_{\text{rec}}}\right)=\frac{18\cdot 0.1}{0.05+0.05+0.2}\cdot\frac{1.68}{9.1}\approx 1{,}2\%.$$

The excess of the relative population inversion in the reacting mixture 1 in comparison with that for the inert mixture 2 recorded in the present work is 0.3-1.0%.

In the high-temperature region ($T_0 \geqslant 2250$°K) the ignition delay τ_{ind}, the characteristic time τ_{N_2O} of occurrence of the chemical transformations in the CO + N_2O interaction (Table 1), and hence the time of existence of high concentrations of atomic oxygen do not exceed the rupture time of the prenozzle diaphragm (~50 μsec). This results in the equilibrium of the concentrations of the molecules in the prenozzle region and, in this connection, the absence of superequilibrium chemical pumping of the vibrational degrees of freedom of the carbon dioxide molecules.

The agreement of the values of the amplification factor in the reacting mixture 1 and the comparison mixture 2 in the second stream cross section indicates the absence of superequilibrium pumping of CO_2 vibrations in the stream between the first and second working cross sections.

Thus, measuring the CO_2 vibrational temperatures and using comparison mixtures made it possible to establish the fact of the existence and the main mechanisms of the superequilibrium chemical pumping of the vibrational levels of CO_2 molecules in a gasdynamic laser based on the products of the reaction of carbon monoxide with nitrous oxide.

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AN APPROXIMATE MODEL OF AN ELECTRIC ARC IN TRANSVERSE MUTUALLY PERPENDICULAR AERODYNAMIC AND MAGNETIC FIELDS

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Expressions are obtained for the characteristics of an electric arc in transverse, mutually perpendicular, aerodynamic, and magnetic fields.

There are quite a few reports devoted to the investigation of an electric arc balanced by aerodynamic and magnetic forces or moving under the action of an external magnetic field. Among them, however, there are few theoretical solutions which admit of practical use. The concept of the impermeability of the core of an arc [1], strengthened in such a way that the arc column itself is treated as a solid cylindrical body, can therefore turn out to be admissible. The experimental investigations of the wake, the frequency of coming off of vortices, the heat exchange, and the resistance presented in [2] indicate that the flow over the core of an arc is similar to the flow over a heated cylindrical body. Indirect considerations about the high viscosity and low density of the arc column in comparison with the envelope also point to the impermeability of the arc. The gas density in the central core comprises several percent of the density of the free stream, so that when the velocities in the core and in the stream do not differ too much about 10% of the gas can pass through the arc.

The model of an arc as a solid cylindrical body naturally requires a cautious approach, since the real physical picture will differ from this idealized scheme.

We note that for such a model as a whole it doesn't matter whether the arc is under steady-state conditions with equality of the resultant aerodynamic and magnetic forces or moves under the action of the magnetic field. In the latter case the velocity, except for the initial period, must be set in such a way that it corresponds to a balance of these forces.

We assume that the properties of the arc do not vary in the axial direction, although it is well known that its characteristics also depend on the interelectrode distance. Such a dependence is primarily due to the influence of the arc sections near the electrodes. In the middle part of the arc column the variations of its properties are slight [2].

Let us write the equation for the energy balance in the electrically conducting zone of the discharge, allowing only for conductive heat transfer in the radial direction.

$$\frac{1}{r}\frac{d}{dr}\left(r\frac{dS}{dr}\right)+\sigma E^2=0, \tag{1}$$

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