AMPLIFICATION OF THE EMISSION OF A CO_2 LASER IN THE PRODUCTS OF THE REACTION OF CARBON MONOXIDE AND NITROUS OXIDE

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UDC 621.375

To produce laser-active gaseous media, and to investigate their characteristics, it is convenient to use expanding gasdynamic flows [1-4], and high-speed chemical reactions [5, 6]. The idea of a chemical-gasdynamic laser [7, 8], based on a combination of the chemical and gasdynamic methods of producing population inversion is of interest. In this paper we investigate the conditions under which population inversion of the vibrational levels of the CO_2 molecule can be produced in expanding flows of different gaseous mixtures containing CO_2, including the products of the reaction between NO and CO with the addition of a small amount of hydrogen, and when the initial mixture is diluted with nitrogen or argon. The effect of the composition of the initial mixture and the temperature in front of the nozzle on the gain of the coherent emission at a wavelength of 10.6 μm in the working cross section of the flow is investigated.

To produce a vibrationally nonequilibrium flow of the mixture we used an arrangement employing a pulsed gasdynamic laser in a shock tube with a nozzle [9-11]. The gain was measured by the direct method [12, 13] by illuminating the working cross section of the flow with the beam from a CO_2 laser.

The experimental arrangement is shown in Fig. 1. The shock tube 1 of internal diameter 80 mm was connected to the expansion section 2, in which there was a plane wedge-shaped nozzle. The expansion section was connected to a receiver 3. The overall length of the shock tube is 4.2 m, and the length of the high-pressure section is 0.9 m. A mixture of hydrogen and nitrogen was used as the propellant gas. The initial pressure of the mixture in all the experiments was greater than 50 torr. Before filling with the mixture, the low-pressure section was pumped down to a pressure of 0.1 torr. Leakage of air into the shock tube did not exceed 0.1 torr in 10 min. The mixtures were made in a mixing system which carefully dried the components.

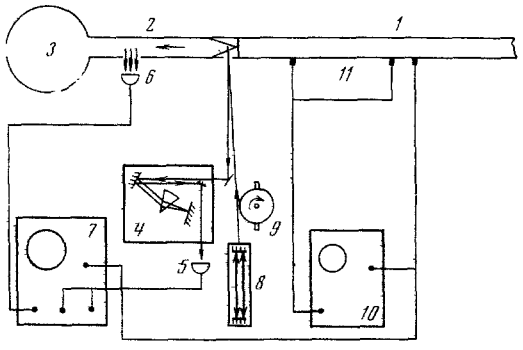

Fig. 1

The pressure p_0 and the temperature T_0 in the reflected shock wave in front of the nozzle was calculated from the experimentally measured shock-wave velocity. The velocity of the shock wave was measured from the signals of two piezoelectric pressure pickups 11 a distance of 0.5 m apart. The signal from a third pickup a distance of 0.2 m higher up the flow was used to trigger and synchronize all the recording apparatuses.

In this investigation the pressure p_0 was maintained constant from experiment to experiment, and the operating conditions of the discharge tube were calculated assuming this condition. For given values of p_0 and T_0 we determined the required initial pressure of the mixture p_1, the

Moscow. Translated from Zhurnal Prikladnoi Mekhaniki i Tekhnicheskoi Fiziki, No. 5, pp. 9-15, September-October, 1974. Original article submitted April 18, 1974.

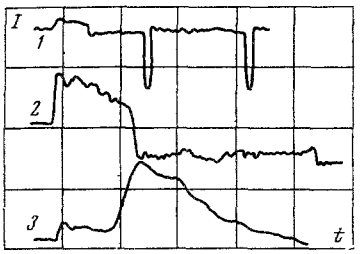

Fig. 2

pressure p_4 of the propelling mixture of gases, and the ratio of hydrogen to nitrogen in the propellant mixture. The final calculation using the measured Mach number M of the shock wave gave values of p_0 that did not differ very much from the assigned values.

To produce the required initial flow conditions, a diaphragm of annealed copper 50 μm thick was placed between the end of the shock tube and the nozzle. The expansion section and the receiver were evacuated down to a pressure of less than 1 torr before the experiment.

The plane wedge-shaped nozzle had a semiangular aperture of 15°, and a controlled critical cross section the height of which could be varied from 0 to 3 mm. The diffuser of the nozzle changed into a rectangular channel 60×80 mm^2 in cross section. To allow the emission to emerge there were two pairs of windows made of zinc sulfide in the expansion section which transmitted infrared radiation in the wavelength range from 0.5 to 12.0 μm.

An LG-23 CO_2 laser with a power of 1 W operating under single-mode conditions was used as the source of probing coherent radiation 8 at a wavelength of 10.6 μm. The laser supply circuit ensured that the emission power remained constant during the experiment.

To measure the gain the probing beam twice crossed the active part of the expanding gas flow, and was then received through the input slit of an IKM-1 monochromator, 4, and was recorded by a photoresistor 5 consisting of germanium doped with gold. To obtain the zero line, the beam of the probing laser was cut off by a shutter 9 for 0.5 msec every 10 msec.

We also recorded the natural infrared emission of the gases investigated. To do this we used a second window of the zinc sulfide situated lower down the flow, and a photoresistor 6. The signals from the photoreceivers 5, 6 were recorded on the screen of an SI-33 5-beam oscilloscope 7. The time resolution of the infrared radiation recording system was 0.05 msec.

Before each experiment the optical system for measuring the gain was adjusted so that the center of the beam of the probing laser after passing through the active part and the monochromator coincided with the sensitive element of the photoreceiver. To eliminate the effect of vibrations all the optical components were acoustically insulated from the shock tube. A series of control experiments with an inert mixture not containing CO_2 showed that these measures were quite effective, and also showed that the schlieren effect had no appreciable influence on the results. The signal fluctuations of the photoreceiver due to vibrations and due to the optical nonuniformity of the flow were less than 10% of the useful amplification signal normally observed. Control experiments showed that the spontaneous infrared emission of the mixtures had a negligibly small effect on the gain measurements made at a wavelength of 10.6 μm.

A typical oscillogram of a record of the amplified radiation of the probing laser (beams 1 and 2), and of the natural infrared radiation (beam 3) is shown in Fig. 2. We measured the ratio $\Delta I/I_0$ where $\Delta I = I_1 - I_0$, I_0 is the intensity of the emission of the probing laser, and I_1 is the intensity of the beam that passes through the active part of the flow. To increase the accuracy with which the quantity ΔI could be read on the oscilloscope screen the recording was doubled on the second channel of the SI-33 oscilloscope with a high sweep rate and a high sensitivity. The gain of the laser emission is given by

$$G = \frac{1}{L} \ln\left(1 + \frac{\Delta I}{I_0}\right) \approx \frac{1}{L} \frac{\Delta I}{I_0} \quad \left(\frac{\Delta I}{I_0} \ll 1\right) \tag{1}$$

Here L is the length of the path of the laser beam in the active medium, which in the experiments was 16 cm. It can be seen from the oscillogram shown in Fig. 2 that the intensity of the natural infrared radiation (beam 3) increases sharply at a time $\tau \simeq 2$ msec after the beginning of the flow. This is obviously due to the increase in the gas density in the expansion part of the apparatus.

From the measured value of G we calculated the relative population of the laser levels of the CO_2 molecule and the vibrational temperature of CO_2 in the flow of gas.

Calculations of the pressure p_6 and the temperature T_6 in the working cross section of the expanding flow under typical experimental conditions gave $p_6 = 10$-25 torr and $T_6 = 150$-350°K. A comparative estimate of the Doppler and collision broadening of the CO_2 emission line at a wavelength of 10.6 μm for these values of p_6 and T_6 showed that the broadening of the CO_2 emission line was mainly due to collisions.

TABLE 1

Mixture No.	T_0, K	G, m^{-1}	δn_{21}	T_{v_2}	Mixture No.	T_0, K	G, m^{-1}	δn_{21}	T_{v_2}
	1100	0.5	1.9	850	2	1590	0.74	1.6	800
	1240	0.73	2.9	940		1720	0.57	1.1	760
	1300	0.74	2.85	940		1780	0.45	0.9	730
	1480	0.62	2.5	920		1820	0.81	1.8	830
		0.37	1.4	800		1980	0.78	1.7	830
1	1740	0.24	1.0	740		2140	0.46	1.1	760
		0.5	2.1	860		2420	0.39	0.9	750
		0.32	1.4	800		740	0.5	0.45	610
	1960	0.23	1.4	770		1110	0.6	0.6	680
		0.37	1.6	830		1200	0.74	0.7	690
		0.5	2.3	910	3	1400	0.39	0.45	640
	2000	0.27	1.25	790		1530	0.28	0.3	620
	980	0.44	0.82	700		1600	0.35	0.45	650
	1230	0.41	0.78	690		1740	0.64	0.8	740
	1300	0.53	1.0	740		1970	0.35	0.5	770
		0.75	1.4	790		2320	0.27	0.36	760
	1400	0.62	1.2	760					
		0.64	1.25	780					
		0.72	1.5	790					

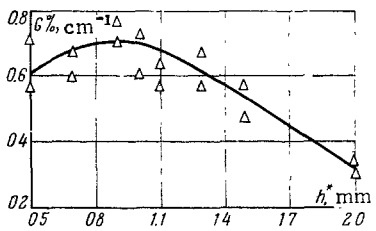

Fig. 3

Using the conclusions and assumptions made in [14], we obtained for the gain at the center of the CO_2 emission line, broadened due to collisions,

$$G = \frac{\lambda_{12}A_{12}}{8\pi Z_c}(N_{00°1} - N_{10°0})\frac{45.6}{T_6}\exp\left(-\frac{239}{T_6}\right) \quad (2)$$

Here λ_{12} is the wavelength of the emission corresponding to the (00°1-10°0) transition of the CO_2 molecule, A_{12} is the probability of a spontaneous transition between the (00°1) and (10°0) levels, $N_{00°1}$ and $N_{10°0}$ are the populations of the upper and lower laser levels of CO_2, respectively, and Z_c is the number of collisions of the CO_2 molecule in the mixture [15]. It is assumed that the transitions occur from the rotational level J = 20 of the 00°1 vibrational state [16, 17].

Equation (2) was used to calculate the population inversion of the CO_2 laser levels from the measured value of G. The vibrational temperature of the upper laser level was estimated from the equation

$$T_{v_2} = -\theta_2\left\{\ln\left[\frac{N_{00°1} - N_{10°0}}{N_{CO_2}} + \exp\left(-\frac{\theta_1}{T_6}\right)\right]\right\}^{-1} \quad (3)$$

assuming the population of the lower levels to be small, and assuming that the vibrational temperature of the lower laser level is approximately equal to the translational temperature T_6 [18]. Here θ_2 and θ_1 are the characteristic temperatures of the upper and lower laser levels, and N_{CO_2} is the number of CO_2 molecules per unit volume of the working cross section.

To use the method we carried out a series of experiments to measure the gain of the laser emission at a wavelength of 10.6 μm in an expanding mixture of $mCO_2 + nN_2 + kHe$, flowing from the nozzle of the shock tube. The temperature and pressure of the mixture in front of the nozzle corresponded to the condition behind the reflected shock wave. We investigated the gain G as a function of the ratio of the mixture components, the height of the critical cross section of the nozzle h*, and the braking temperature T_0 at a constant pressure $p_0 \approx 6$ atm. The values for G obtained for a population of the CO_2 levels $\delta n_{21} = (N_{00°1} - N_{10°0})/N_{CO_2}$ in % and the vibrational temperature of the upper laser level T_{V2} in degrees K, shown in Table 1, agree well with the results obtained in [3, 16-18]. In all the experiments h* = 0.9 mm, and the degree of expansion of the flow in the working cross section of the nozzle A/A* = 24. The number 1 denotes the mixture of 0.05 CO_2 + 0.45 N_2 + 0.5 He, the number 2 denotes the mixture of 0.1 CO_2 + 0.4 N_2 + 0.5 He, and the number 3 denotes the mixture of 0.2 CO_2 + 0.3 N_2 + 0.5 He.

The gain as a function of the height of the critical cross section of the nozzle h* for a mixture of 0.1 CO_2 + 0.4 N_2 + 0.4 He for $p_0 = 6$ atm and $T_0 = 1500$°K is shown in Fig. 3. A maximum in the gain occurs at a value of h* = 0.8-1.0 mm. The optimum value of h* obtained in these experiments was then used in experiments with a reacting mixture of $N_2O + CO$.

The use of the reaction between nitrous oxide and carbon monoxide to produce a laser-active medium in a gasdynamic laser is of considerable interest, since, firstly, the final products of the reaction are CO_2

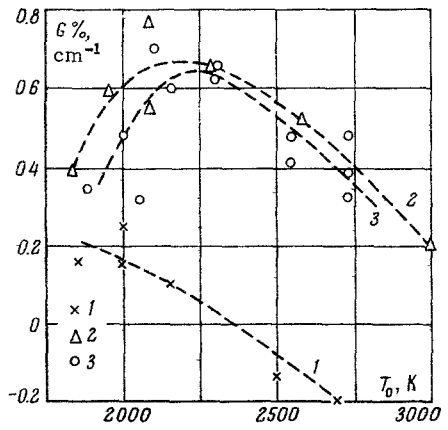

Fig. 4

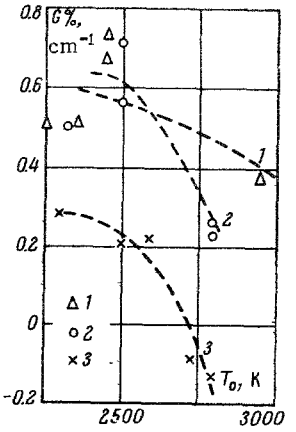

Fig. 5

and nitrogen – the main components of the operating mixture of a CO_2 laser, and, secondly, the energy and kinetic characteristics of the reaction suggest the possibility of obtaining nonequilibrium chemical pumping of the products in supersonic expansion in the nozzle [7]. In this investigation we studied the conditions for population inversion of the vibrational levels of CO_2 to occur in an expanding flow of the products of the N_2O+CO reaction assuming that the reaction is fully completed in the reflected shock wave in front of the nozzle.

The pressure in front of the nozzle p_0 was maintained constant in all the experiments and was approximately 10 atm. The temperature T_0 was varied from 1800 to 3500°K. The thermal effect of the reaction,

$$N_2O + CO \rightarrow CO_2 + N_2 \qquad (4)$$

according to the data in the literature [19] is 87.3 kcal/mole. To eliminate excessive heating of the reaction products a stoichiometric mixture of CO and N_2O was mixed with a chemically inert diluent (nitrogen or argon) as a ballast in amounts up to 80%. In some of the experiments we introduced molecular hydrogen (up to 10%) into the initial mixture to accelerate the reaction (4) [20], and to introduce water molecules into the products of the reaction, to deactivate the lower laser level of CO_2.

Hence, the general form of the reaction is

$$\alpha CO + \beta H_2 + (\alpha+\beta)\, N_2O + \gamma\, N_2 \rightarrow \alpha\, CO_2 + \beta\, H_2O + (\alpha+\beta+\gamma)N_2 \qquad (5)$$

The content of CO in all the experiments was 10%. We investigated mixtures with an initial content of hydrogen of 0, 1, 2, 5, and 10%. In [21] an initial mixture with a ratio of the components $CO : N_2O : H_2 : He = 35 : 35 : 2 : 28$ was used in a gasdynamic laser employing the products of gaseous detonation.

The temperature of the gas in front of the nozzle T_0 was calculated assuming that the chemical transformation of the initial mixture was completed in the region behind the reflected shock wave. Data on the delays in the combustion of the initial mixture in reaction (4) [20] and a calculation of the time for which a hot bottleneck existed confirmed this assumption. At high temperatures T_0 we also took into account the dissociation of the CO_2 and H_2O molecules. In these cases it was possible for a high content of carbon dioxide to exist in the working cross section. The height of the critical cross section of the nozzle in all the experiments was 0.9 mm.

The results of the experiments are shown in Figs. 4 and 5 in the form of curves of the gain G against the temperature in front of the nozzle T_0 for different ratios of the components of the initial reacting mixture. The amount of hydrogen was varied in the various experiments. The graphs in Fig. 4 show the experimental results for initial mixtures with a hydrogen content of 0, 1, and 2 mole %. The corresponding experimental curves are denoted in Fig. 4 by the numbers 1, 2 and 3. In Fig. 5 curves 1 and 3 show the results of experiments for initial mixtures with a hydrogen content of 5 and 10%, respectively. Curve 2 in Fig. 5 shows G as a function of the temperature T_0 for a mixture with an addition of 0.5% of H_2, in which 79 mole % of argon was used instead of nitrogen as a diluent.

In all the initial reaction mixtures investigated we observed amplification of the emission at a wavelength of 10.6 μm due to population inversion in the expanding flow of CO_2 molecules when the products of the reaction (5) emerged from the nozzle.

For the initial mixtures with an addition of hydrogen the highest values of the gain were obtained for a hydrogen content of 1%, and the least value of gain was obtained for a mixture with an addition of 10% of hydrogen.

Comparison of the results obtained for a nonreacting mixture (see Table 1) shows that in the case of a flow of the products of the reaction (5) amplification is observed at higher values of the temperature T_0.

It is seen from Fig. 4 that in the case of an initial mixture without added hydrogen amplification is observed in the temperature range $T_0 = 1850$-$2150°K$. It is possible that despite the measures taken to dry the shock tube and the initial mixture the flow contained traces of water vapor. It was also not possible to eliminate the chance of nonequilibrium vibrational excitation of CO_2 or N_2 during the reaction.

For a mixture diluted with 79% of argon (Fig. 5) values of the gain were obtained which were not less than that obtained in the case when the initial mixture was diluted with nitrogen, despite the absence of an additional reservoir of vibrational energy, like heated nitrogen. It is shown in [22, 23] that argon has a considerable effect on the relaxation characteristics of CO_2 in shock waves. Hence, an investigation of the nonequilibrium characteristics of the triple mixture $CO_2 + N_2 + Ar$ should be of considerable interest.

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