EXPERIMENTAL STUDY OF THE AMPLIFICATION (INTENSIFICATION) OF LIGHT IN $CO_2 + N_2 + He$ (H_2O) MIXTURES DURING EXPANSION IN A SUPERSONIC NOZZLE

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The results of an experimental investigation on the amplification of light by carbon dioxide molecules in various mixtures during the expansion of the gas through a supersonic nozzle at the end of a shock tube are presented. The experimental results are compared with theory, and satisfactory agreement is obtained.

The idea of creating an inverted population in gases by sharp changes of pressures was considered in [1, 2]. The use of adiabatic expansion in a $CO_2 + N_2$ gas mixture for this purpose was first proposed in [3, 4]. Subsequently numerical calculations of the parameters of a gas-dynamic laser were published for a variety of gas mixtures. Experimental data at present existing in the literature [5-10] relate to a wide range of CO_2, N_2, He, and H_2O concentrations, but this cannot be said of the range of variation of the retardation temperature ($T < 1800$-2000°K). A range of variation of the retardation pressure amounting to $P = 2$-16 atm, with a fixed retardation temperature of $T = 1250$°K, was studied in [8, 10].

In the present investigation we measured the amplification factor of a 10% CO_2 + 40% N_2 + 50% He mixture subjected to supersonic expansion over a wide range of experimental conditions (retardation temperature $T = 800$-3600°K, gas pressure 2.5-40 atm), the relative cross section of the nozzle taking several fixed values $S/S_* = 18.2$, 36.2, 49, 59, where S_* is the critical cross section of the nozzle and S is the cross section in the measuring region. A mixture of 10% CO_2 + (90 − X) % N_2 + X% H_2O (where X = 0-5%) was studied for retardation temperatures of $T = 800$-2200°K and pressures of $P = 1.5$-40 atm, with relative nozzle cross sections of $S/S_* = 18.2$, 36.2, 59.

The resultant amplification factors were compared with the values calculated by the method proposed in [11, 12].

The amplification factors of the foregoing mixtures were studied experimentally in an apparatus comprising an aerodynamic shock tube and an optical system. The aerodynamic shock tube consisted of the shock tube proper (length 5 m, diameter 80 mm), a receiver (length 4 m, diameter 60 cm), and a supersonic nozzle situated at the end of the shock tube. The nozzle was axisymmetrical, the area of the critical cross section $S_* = 0.785\ \text{cm}^2$, the confusor section was rounded to a radius of $R = 0.5$ cm, the diffusor section was given by the equation

$$S/S_* = \begin{cases} 1 + 0.29l^2, & l < 5\ \text{cm} \\ 4 + 0.7l + 0.0305l^2, & 5 \leqslant l \leqslant 32.5\ \text{cm} \\ 59, & l > 32.5\ \text{cm} \end{cases}$$

where l (cm) is the distance from the critical cross section along the axis of the nozzle.

Before the experiment, the entrance into the nozzle was covered with a membrane so as to be able to evacuate the shock tube and receiver individually to pressures of under $2 \cdot 10^{-2}$ mm Hg. After evacuation, the low-pressure chamber of the shock tube was filled with the test mixture to a pressure of $P_1 = 15$-400

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mm Hg, which was measured with a vacuum gage. A mixture of the desired composition was prepared in a stainless steel vessel; a small inbuilt fan was used for the mixing. The rate of air inflow into the vessel was less than 1.5 mm Hg/h.

The mixture containing water vapor was made up in the following way: a vessel containing water and a thermometer was placed in the evacuated volume; the thermometer readings gave the pressure of the saturated water vapor. The CO_2, N_2, and H_2O were agitated for more than 2 h. After careful mixing, the gases passed into the shock tube and were heated to the conditions specified by being subjected to the incident and reflected shock waves. After this the mixture was expanded through the supersonic nozzle into the receiver. A synchronization system enabled the whole apparatus to be set into action at the precise instant required.

The amplification factor was measured in the following way. Continuous radiation from an OKG-15 laser with a specially stabilized supply passed at right angles to the flow of gas through the KCl inspection windows of the receiver and the inspection windows in the nozzle (built-in flush with the inner surface of the nozzle). Then the radiation was converged on to the photoresistive receiver (Ge–Au, $T = 55°K$) by means of a lens.

In front of the lens was a diaphragm limiting the intensity of spontaneous emission from the test mixture. The signal from the receiver was applied to an oscillograph, which only recorded its varying component, i.e. the increment of the light intensity over the constant laser intensity, due to amplification in the mixture. The constant laser intensity was determined at the exact instant of determining the amplification factor, so as to allow for fluctuations in laser power. In order to take account of this instability of generation, some of the laser energy was carried out of the beam by means of a plane-parallel plate, interrupted with an obturator, and recorded by means of a Ge-Au receiver on the oscillograph at the instant at which the mixture flowed through the nozzle. This arrangement enabled relative amplifications of the order of 10^{-3} to be recorded ($\Delta K \sim 2 \cdot 10^{-4}$ cm^{-1}).

Spontaneous emission was not recorded in these experiments (as it was in [13]), since a receiver with a high sensitivity in the region of $\lambda = 10.6\ \mu$, due to the more intensive cooling of the active element, was used in the present case.

The retardation temperature and pressure were calculated from the measured initial pressure and the velocity of the incident shock wave, allowing for the excitation and dissociation of the components. The velocity of the incident shock wave was measured to an accuracy of 1.5%, which corresponded to an accuracy of ~3% in determining the retardation temperature. The indeterminacy in measuring the initial pressure of the mixture was no greater than 1 mm Hg.

The results of the experiments relating to the amplification factor of the gas mixture K are presented in Figs. 1 and 2 and the table below for a mixture of 10% CO_2 + 40% N_2 + 50% He in relation to the retardation temperature T, pressure P, and relative nozzle cross section S/S_*.

$T \cdot 10^{-3}$, °K	1.3	1.8	2.2	2.9	3.2	3.6
$K \cdot 10^3$, cm^{-1}	1.65 1.15	2.7 2.9 3.0	2.5 2.9 3.1	1.9 1.65	1.15	0.65 0.8
P, atm	2.9	4.8	6.9	12	15	19

The horizontal axis of Fig. 1 gives the retardation temperature T, and the vertical axis gives the amplification factor K. The experimental points and the calculated curves indicated by the numerals 1, 2, 3, and 4 relate to nozzle cross sections $S/S_* = 18.2, 36.2, 49, 59$, respectively. The retardation pressure is determined by the shock adiabat for an initial pressure of the mixture equal to $P_1 = 167$ mm Hg.

Figure 2 shows the experimental points and calculated data (continuous curve) for the amplification factor K (measured along the vertical axis on the left-hand side) as a function of the retardation pressure P (along the horizontal axis). The retardation temperature is fixed at $T = 1900 \pm 100°K$ and the nozzle cross section at $S/S_* = 59$.

In addition to this, for convenience in using the data of Fig. 1, the broken line (in Fig. 2) represents the retardation temperature T as a function of the retardation pressure P for $P_1 = 167$ mm Hg. The retardation temperature is shown on the right-hand vertical axis.

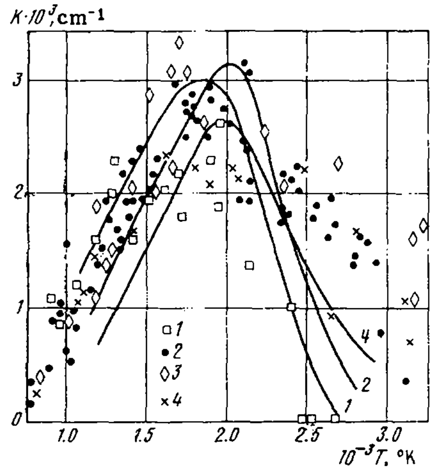

Fig. 1

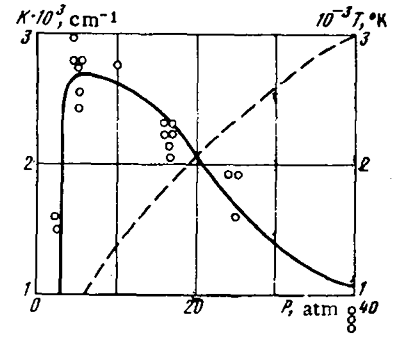

Fig. 2

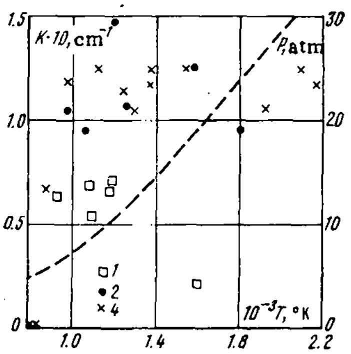

Fig. 3

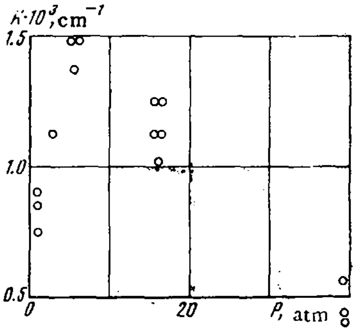

Fig. 4

The table presents some experimental data relating the amplification factor K to the retardation temperature T for $S/S_* = 59$, with an initial pressure of $P_1 = 50$ mm Hg. The corresponding retardation pressure P is also shown in the table.

The amplification factor K passes through a maximum of $K = 3 \cdot 10^{-3}$ cm^{-1} at T = 1800-2000°K. The reason for this behavior of the amplification factor lies in the following. On raising T from 800 to 1800°K, the populations of the asymmetric valence and deformation (shear) vibrations increase, and so does their difference. On further increasing the temperature, the rate of deactivation of the asymmetric valence vibration increases very considerably, as a result of which the population of the levels relating to this form of vibration does not increase so rapidly as that of the shear-vibration levels. The value of K therefore falls in the range $T > 1800$°K.

The curve relating the amplification factor K to the retardation pressure P for a fixed retardation temperature $T \simeq 1900$°K also passes through a maximum ($K = 3 \cdot 10^{-3}$ cm^{-1}) at P = 5 atm. This behavior is explained by the fact that with falling pressure the rate of vibrational relaxation diminishes and hence the populations of all the CO_2 modes increase. The inverted population and the amplification factor also first increase. However, the population of any given mode is unable to exceed the specific limiting value which it assumes at the retardation temperature. Hence, as soon as the population of the asymmetric vibration reaches this limiting value, any further increase in pressure leads to a fall in the inverted population and the amplification factor (by virtue of an increase in the population of the shear or deformation type of vibration).

It should be noted that in [8, 10], K was found to be independent of P in the range P ~ 2-16 atm at T = 1250°K; however, the configuration of the nozzle differed from that employed in the present investigation.

The curve relating the amplification factor K to the relative cross section of the nozzle S/S_* exhibits a maximum at $S/S_* \sim 35$ for $T > 1800$°K. For $T < 1800$°K the amplification factor does not depend on S/S_* in the range $S/S_* = 18.2$-59, within the limits of experimental error.

The results of our study of the 10% CO_2 + 90% N_2 mixture are presented in Figs. 3 and 4. The horizontal axis in Fig. 3 gives the retardation temperature T, the vertical axis on the left gives the amplification factor K. The points 1, 2, 4 were obtained for nozzle cross sections of $S/S_* = 18.2, 36.2, 59$ respectively. The retardation pressure P corresponds to an initial pressure of $P_1 = 167$ mm Hg. For convenience the value of P is shown in the figure by a broken line, the pressure being measured along the vertical axis to the right.

In Fig. 4 the horizontal axis gives the retardation pressure P, and the vertical axis gives the amplification factor K. All the points relate to a retardation temperature of $T = 1500 \pm 100$°K and to a nozzle cross section $S/S_* = 59$. A comparison between Figs. 1-4 shows that the dependence of the amplification factor K on the retardation pressure P and the relative cross section of the nozzle S/S_* qualitatively reproduces the $K = K(P, S/S_*)$ relationship for the $CO_2 + N_2 + He$ mixture, with the sole difference that the coefficient K is much lower in this case. The experimental data relating K to the retardation temperature T differ from those corresponding to the $K = K(T)$ relationship for the $CO_2 + N_2 + He$ mixture not only in respect of the scale of K, but also in respect of the fact that K only diminishes at high T for small S/S_* values. However, it is essential to remember that the maximum temperature in these experiments was $T = 2200$°K.

In addition to this, several experiments were carried out with atmospheric air instead of commercial nitrogen. Within the limits of experimental error the results of the experiments coincided.

We should emphasize that the results obtained for the mixture of 10% CO_2 + 90% N_2 disagree with published experimental data [7, 8, 10]. We ourselves found intensification of the light in the absence of He or H_2O, while in the earlier investigations absorption was recorded. We therefore made a series of control experiments with specially purified gas.

The commercial gas (N_2, CO_2) was dried in a heat exchanger containing liquid nitrogen. The carbon dioxide was then deposited on the walls of the heat exchanger, and the sublimation products were used for composing the mixture.

The results of the control experiments are presented in Fig. 5, in which the amplification factor is set out along the vertical axis and the retardation temperature along the horizontal. The retardation pressure corresponds to an initial pressure of $P_1 = 167$ mm Hg and a nozzle cross section of $S/S_* = 59$. The numeral 1 denotes points obtained after drying the mixture. For comparison, the same figure shows the results of experiments with an undried mixture, as indicated by the numeral 2.

The amplification factor K for the dried mixture is 1.5 times lower than that for the undried mixture at $T = 1000$-1200°K and approximately equal to it for $T > 1300$°K. This result leads to the conclusion that, under the conditions of the present experiment relating to the 10% CO_2 + 90% N_2 mixture, the effects of an uncontrolled quantity of water vapor were insignificant.

The results of our study of the 10% CO_2 + $(90 - X)$% N_2 + X% H_2O mixture with water vapor contents of $X = 2$-5%, $S/S_* = 59$, and $P = 167$ mm Hg are presented in Fig. 5. The points are denoted by the figures 3 ($X = 2$%) and 4 ($X = 5$%). The amplification factor K fell by about 3 ($X = 2$%) and 8 ($X = 5$%) times by comparison with that of the undried mixture. We were unable to study lower concentrations of water vapor owing to difficulties in the method of preparing the working mixture.

Here it should be noted that the results of the preliminary experiments revealed that K was entirely independent of the water vapor content X in the range $X = 0$-5%. This may be explained if we assume that water vapor was adsorbed by the surface of the shock tube while the experiment was being prepared. If the evacuated shock tube is filled with water vapor before the experiment, and after 5-10 min again evacuated to a pressure of 0.5 mm Hg, the experiments only then being initiated, the results show a marked dependence on the concentration of water vapor in the mixture. We note that the experimental points relating to the mixture with a water concentration of 2 and 5%, and partly with an undried mixture of 10% CO_2 + 90% N_2 (Fig. 5), were obtained after the preliminary filling of the shock tube with water vapor in the manner indicated.

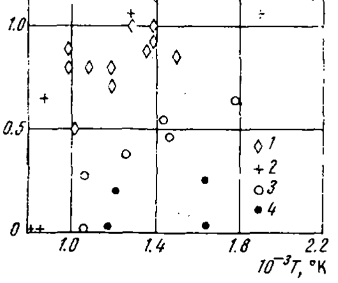

Fig. 5

It is a matter of fundamental interest to compare the experimental values of the amplification factor K with calculation. In calculating K for the conditions of our experiment with a mixture of 10% CO_2 + $(90 - X)$% N_2 + X% H_2O we used the method proposed in [11, 12]. For a mixture of 10% CO_2 + 40% N_2 + 50% He, in conformity with [13], we obtained excellent quantitative agreement between the experimental and theoretical data relating to the amplification factor for a calculation based on the foregoing method. We accordingly attempted to perform an approximate analytical calculation of K for the mixture containing helium [14]. Here we assumed that the true gas-dynamic parameters in the nozzle might be approximated by those relating to the flow of gas with a constant adiabatic index. In this case the coefficients of the kinetic equations become known functions, and an approximate solution may be found for the equations. The probabilities of the processes were taken in accordance with [11, 12].

The amplification factor was calculated [15] from

$$K(\nu) = \frac{c^2 A_{vJ}^{v'J'}}{8\pi^2 (\nu_{vJ}^{v'J'})^2} \left(\frac{n_{v'J'}}{g_{J'}} - \frac{n_{vJ}}{g_J} \right)_2 S(\nu - \nu_0)$$

where $S(\nu - \nu_0)$ is the form factor of the emission line, $A_{vJ}^{v'J'}$ is the Einstein coefficient for spontaneous emission, $n_{v'J'}$, n_{vJ} are the populations of the vibrational–rotational levels, $g_{J'}$, g_J are the statistical weights, $\nu_{vJ}^{v'J'}$ is the transition frequency, and c is the velocity of light.

The calculation was carried out for the center of the line $\nu = \nu_0$, taking $A = 0.2 \text{ sec}^{-1}$. The line contour was of the mixed type under the experimental conditions employed; the collision-broadening probabilities were taken from [15].

The theoretical data in Figs. 1 and 2, indicated by continuous lines, relate to the P20 line. On changing the rotational quantum number J by two units $\Delta J = \pm 2$ (P 18, 22) the theoretical data relating to the amplification factor K change by about 20%, which corresponds to the scatter of the experimental data.

A comparison between the theoretical and experimental data for the $CO_2 + N_2 +$ He mixture (Figs. 1 and 2) reveals not only qualitative but also excellent quantitative agreement.

In the case of the 10% CO_2 + 90% N_2 mixture, the experiments reveal considerable amplification (intensification), whereas calculations in accordance with [11, 12] predict absorption $K \simeq -5 \cdot 10^{-1}$ cm^{-1}. Absorption remains operative up to a water vapor concentration of $X = 0.12$–0.3% (for $S/S_* = 18$–59). This difference may be explained either by assuming that we underestimated the value of the deactivation probability of the deformation (shear) type of vibrations of the CO_2 molecule used in the calculation (inversion was obtained in [17] or higher values of the probability), or else by assuming the presence of impurities. The most likely impurity under the conditions of the present experiment would be oil vapor.

For the 10% CO_2 + (90 – X)% N_2 + X % H_2O with X = 2–5% we find quantitative agreement between the theoretical and experimental data.

The foregoing results lead us to conclude that the principle underlying the numerical calculation [11, 12] and the proposed analytical procedure remains valid.

The set of reaction velocity constants proposed in [11, 12] for the $CO_2 + N_2 + He(H_2O)$ mixture describes the system quite accurately for large helium concentrations (~50%) and for water vapor contents $X \geq 2\%$.

In order to describe the $CO_2 + N_2 + H_2O$ system for low water concentrations, it may be that we shall have to correct the deactivation velocity constant for the deformation type of vibrations of the CO_2 molecule.

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