# Hydrocarbon-air mixture combustion gasdynamic laser

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Results are presented of an experimental and theoretical investigation of the unsaturated gain and power of laser radiation from a quasicontinuous gasdynamic laser operating on the products of combustion of hydrocarbon-air mixtures. Acetylene and propane air mixtures and a mixture of carbon monoxide and hydrogen were used. At optimal amplification values of the parameters ( $p_0 = 5-10$  atm,  $T_0 = 1400-1700^{\circ}$ K) the gain was  $8 \times 10^{-3}$  cm<sup>-1</sup>, in good agreement with the calculations. The maximum power generated was 8.5 kW.

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# 1. PRINCIPAL RESULTS OF RESEARCH IN THE DEVELOPMENT OF GASDYNAMIC $CO_2$ LASERS

During the last decade, substantial results were obtained in the development of high-power laser devices. Various types of lasers were produced, differing from one another by the methods of obtaining the laseractive media. At the present time three types of highpower cw lasers are being developed: gasdynamic, gas-discharge, and chemical, in which, in essence, the thermal, electrical, and chemical energy is directly converted into coherent electromagnetic-radiation energy. Of particular interest are gasdynamic lasers (GDL), the development of which, judging from the published data, has made it possible to obtain cw lasers of unprecedented high power.

Population inversion and the laser effect are attained in a gasdynamic laser by rapid gasdynamic expansion of the gas in a supersonic nozzle, as a result of the difference between the relaxation rates of the different energy states of the quantum system.

The ideas of producing inverted population in gases by rapid variation of the temperature were developed by Basov and Oraevskiĭ<sup>[1]</sup> and by Hurle and Hertzberg<sup>[2]</sup>. The invention by Patel of the  $CO_2-N_2$  gas laser in 1964<sup>[3]</sup> has uncovered a new laser system. In 1966, Konyukhov and Prokhorov<sup>[4]</sup> first proposed to use adiabatic expansion of a  $CO_2-N_2$  mixture in a supersonic nozzle to obtain population inversion of the vibrational levels of the  $CO_2$  molecule, and this stimulated the development of high-power GDL. Subsequently, many investigations<sup>[5-9]</sup> were devoted to the theoretical analysis of the process that determine the operation of gas dynamic lasers.

Lasing was observed in experiment by expanding a gas through a supersonic nozzle and a  $slit^{[10-12]}$ . Dzhidzhoev et al.<sup>[13]</sup> described a GDL in which the working medium was obtained by detonating solid substances. Important results were recently obtained in the development of a pulsed GDL using a high-pressure shock tube<sup>[14]</sup>, in which at a stagnation pressure 700 atm and a temperature 2000°K the average radiation power in the multimode regime was 450 kW at a pulse duration 4 msec, thus pointing to great potential of developing pulsed shock-tube lasers with large peak power. Greatest interest, however, was aroused by a paper of Gerry,<sup>[15]</sup> who reported that the Avco Corporation has developed a 60-kW gas laser using combustion of carbon monoxide. It is clear, however, that to produce highpower cw lasers with large operating capacity, the use of the combustion of carbon monoxide in GDL is not

promising. This is precisely why research in this field has recently followed the path of ascertaining the possibility of using the combustion products of hydrocarbon fuels as laser-active media.

Until very recently, this idea was subjected to considerable doubt, since combusion of hydrocarbons produces large concentration of water vapor, the molecules of which have large probability of deactivation of the asymmetric mode of the  $CO_2$ -molecule vibrations. The first experiments in this region seemed to confirm this danger, but further investigations<sup>[16,17]</sup> have shown that the development of high-power GDL using the combustion of hydrocarbon fuels is possible.

In this article we present the results of a systematic experimental and theoretical investigation of the gain and power of laser radiation in a quasicontinuous GDL employing the combusion products of mixtures of hydrocarbons and air. The purpose of the investigation was not only to determine the optimal values of the stagnation parameters, the composition of the working mixture, and the geometry of the nozzle needed to obtain maximum gain and generated power, but also to solve a number of fundamental problems connected with the calculations of the values of the gain and the radiation power for this class of gasdynamic lasers.

## 2. QUASICONTINUOUS GASDYNAMIC COMBUSTION LASER (DESCRIPTION OF THE SETUP AND MEASUREMENT PROCEDURE)

A quasicontinuous gasdynamic combustion laser<sup>[18,19]</sup>, which is the pulsed analog of the continuous GDL, is among the most promising laser systems with high peak powers. At equal peak power values, a combustion GDL is much simpler to operate and much smaller in dimensions than, for example, a shock-tube laser.

The schematic diagram of the setup used in the present study is shown in Fig. 1. The laser consisted of combustion chamber 1, nozzle block 2, multiple-pass resonator cavity 3, and vacuum chamber 4. Prior to the experiment, the entrance to the nozzle block, on the combustion-chamber side, was covered with membrane 5, making it possible to evacuate the resonator and the vacuum chamber to a specified pressure. The combustion chamber was then filled with a hydrocarbon—air mixture of definite composition. The system was ignited with spark plus 6, and the combustion process as well as the parameters of the combustible mixture were monitored with a previously-calibrated piezoelectric pressure pickup 16 mounted in the combustion chamber. From the registered pressure we were able to determine the deceleration parameters and to trace their time variation. The stagnation temperature prior to the breaking of the membrane was calculated from the maximum value of the pressure. The combustionproduct temperatures calculated in this manner, as shown earlier<sup>[18]</sup>, were in good agreement with the results of a direct measurement of the flame temperature.

After the combusion process was completed, the membrane was broken and the combustion products were expanded in the nozzle block. The nozzle block constituted a system of grid-type nozzles consisting of 50 vertically-mounted specially-shaped flat nozzles with a critical-section height 0.4 mm. The nozzle had a corner point with a maximum aperture half-angle  $37^{\circ}$ , to obtain uniform supersonic stream at a degree of expansion equal to 25. The profile of the nozzle was calculated by the method of characteristics. We note that we did not make a special investigation of the possible influences exerted on the results by the discontinuities produced on the edges of the individual nozzles, however, according to data available to us, for a system of nozzles with profiles having sharp edges the discontinuities produced on the edges are weak and exert no noticeable influence on the measurement results.

The supersonic stream of combustion products proceeded from the nozzle block to a resonator cavity 50 mm high and 50 cm wide. The lateral walls of the resonator cavity had each three unified adjustment heads. When the gain was investigated, NaCl plates were installed in the adjustment heads. In the experiments on lasing, mirrors were placed in the adjustment heads and could be used to form one-, two-, three-, four- and five-pass resonators. To prevent rapid contamination of the mirror surface by the gas stream, the mirrors were installed in the adjustment heads at a certain depth relative to the resonator walls. From the resonator cavity, the combustion products proceeded to a vacuum chamber of approximate volume 1 m<sup>3</sup>, which was used to maintain the required pressure in the resonator cavity during the course of the experiment.

The unsaturated gain was measured by probing the stream of the combustion products simultaneously in three cross sections with a low-power laser beam 8 perpendicular to the direction of motion of the gas. The beam of a 7-kW probing single-mode  $CO_2$  laser was split with a system of NaCl plates (9) into three beams. Each of these beams passed through the glass windows of the resonator cavity and through the stream, after which the radiation was collected by lens 10 and registered with a Ge-Au photoreceiver 11. To eliminate the influence of the spontaneous radiation on the registered signal, diaphragms 12 were installed in front of the lens. To monitor the stability of the radiation power of the probing laser, some of the probing-beam energy



FIG. 1. Diagram of experimental setup. The explanation is in the text.

was diverted by a plane-parallel plate 14, interrupted by choppers 13 and 15 and registered with a fourth Ge-Au photoreceiver 11.

An advantage of the procedure described above is the possibility of simultaneously registering the stagnation pressure and the gains in three cross sections of the stream, the first gain-registration channel being located 4 cm away from the critical section of the nozzle, and the succeeding ones at a distance 10 cm from each other. This made it possible to obtain in a single experiment abundant information on how the gain depends on the parameters of the stagnation and on how it varies along the stream. It should be noted that according to the data of our measurement<sup>[20]</sup>, the characteristic time required to establish the stationary flow does not exceed approximately 0.3 msec, which is shorter by almost two orders of magnitude than the time of the experiment and indicates that nonstationary effects cannot influence the results.

Figure 2 shows typical oscillograms of the stagnation pressure  $p_0$  (lower oscillogram) and of the gain in three cross sections along the stream for a 6% acetylene-air mixture. In this experiment, the initial pressure of the working mixture was  $p_{in} = 3$  atm, and the temperature and stagnation pressure products at the instant preceding the breaking of the membrane were respectively 2100°K and 21 atm. From the signal oscillograms shown in Fig. 2 it follows that amplification in the first two channels is observed immediately after the breaking of the membrane. It reaches a maximum at a pressure on the order of 8-10 atm and then decreases during the course of further expansion of the combustion products. It is also important to note that at large stagnation pressures (15-20 atm) the largest gain is observed in the first channel and then it decreases quite slowly downstream, so that it is practically absent from the third channel. This is attributed to the large rates of the relaxation processes with participation of water molecules, which lead to a decrease of the populations of the upper laser level.

With decreasing stagnation pressure, the rates of the relaxation processes decrease strongly and as a result the amplified signals increase noticeably in the second and in particularly in the third channel, although they do remain much smaller compared with the gain in the first channel. In addition, it is seen from the oscillo-grams that at a certain instant of time the gain decreases abruptly in all three channels, and gives way to absorption. This effect, which was analyzed by us in detail earlier<sup>[18,21]</sup>, is due to the fact that during the

FIG. 2. Oscillograms of the gain (curves 1, 2, 3) at distances 4, 14, and 24 cm respectively from the critical cross section of the nozzle, and oscillogram showing the stagnation pressure (curve 4). (The sweep direction of the process is from left to right, one division is equal to 10 msec.)



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course of the expansion of the production products the vacuum chamber becomes pumped by the combustion products, through the supersonic nozzle, and at a certain instant of time the counterpressure gives rise to a shock wave propagating towards the nozzle. The passage of the shock wave to the cross sections in which the probing takes place causes an abrupt decrease of the amplified signals and a change from amplification to absorption. The decrease of the gain behind the shockwave front is due to the increase of the translational temperature of the gas and of the density, which leads to underpopulation of the lower laser level, to a rapid decrease of the inversion, and even to a change from amplification to absorption.

The experiments have shown that the vibrational relaxation downstream is very rapid, so that we shall henceforth focus our attention on the investigation of the maximum values of the gains which are realized at the location of the first channel.

# 3. RESULTS OF THE INVESTIGATION OF THE GAIN IN A GASDYNAMIC COMBUSTION LASER

The result of the measurements of the maximum values of the unsaturated gain  $K_{\nu}$  of the combustion products as functions of the concentration of the acetylene in an acetylene-air mixture, for an initial pressure 3 atm, are shown in Fig. 3 (triangles). The reproducibility results was good, and each point on the diagram is an average of three or four experiments. The concentration of the acetylene in the initial mixture ranged from 4 to 10%. At concentrations lower than 4%, the mixture was not ignited by our ignition source, while at concentrations higher than 10% there was noticeable soot production.

As follows from the diagram, for the speciallyshaped nozzle used in these experiments, with a degree of expansion 25, the gain decreased rapidly with increasing concentration of the acetylene in the mixture, this being due to the increase of the concentration of the water vapor in the combustion products and to the corresponding increase of the rate of deactivation of the upper laser level, which led to a decrease of the inversion (we note that in the acetylene combustion reaction the volume change is small, so that the concentration of the water vapor in the combustion products corresponds to approximately the concentration of the acetylene in the initial mixture). With decreasing acetylene, concentration below 4%, the gain should also decrease, since the combustion temperature decreases abruptly, and this decreases the population of the levels corresponding to the asymmetric valence and deformation oscillations and the difference of these populations.

It is of interest to compare the present data with the results of an earlier study<sup>[8]</sup> where we used a flat wedge-shaped nozzle with half-angle 15°, with the same degree of expansion 25, but with a critical section of 1 mm height. In Fig. 3 the results of<sup>[18]</sup> are represented by circles. From an analysis of the data it follows that the specially-shaped nozzle used by us in the present study, with a critical section 0.4 mm high, is more effective from the point of view of the quenching the vibrational energy accumulated in the CO<sub>2</sub> and N<sub>2</sub> molecules, if the acetylene concentration in the initial mixture is less than 8%. At higher acetylene concentrations, the difference between the gains of the nozzles of these two configurations becomes negligible.

The different character of our procedure has made it possible to obtain in one experiment abundant information concerning the dependence of the gain of the mixture on the stagnation parameters, inasmuch as expansion of the gas through the nozzle changed the stagnation pressure and accordingly the temperature of the combustion products. The running values of the stagnation temperature were calculated from the change of the pressure in the combustion chamber, assuming an adiabatic process with an adiabatic exponent  $\gamma = 1.28$ . The value of the adiabatic exponent depended significantly on the composition of the mixture and on the stagnation parameters, and ranged from 1.27 to 1.29 under the variation of the conditions of our experiments, but we used the indicated constant value in the calculations.

Figure 4 shows the dependence of the gain on the running values of the deceleration parameters for different initial pressures of an initial acetylene-air mixture containing 5.5% acetylene (the initial pressures of the mixtures are shown in the upper parts of the diagrams). Along each curve, the stagnation parameters vary and with them the gain values (in reciprocal centimeters multiplied by  $10^3$ ) represented in Fig. 4 by equal-gain curves. This representation of the results is very instructive and useful, since it makes it possible to determine the region of stagnation parameters corresponding to the optimal values of the gain for the investigated mixture. It is seen from an analysis of Fig. 4 that this region is for the investigated mixture quite



FIG. 3. Dependence of the maximum gain  $K_{\nu}$  ( $\triangle$ -present work,  $\circ$ -[<sup>18</sup>]) and of the laser-radiation power P on the  $C_2H_2$  concentration in the initial mixture.



FIG. 4. Dependence of the beam  $K_{\nu}$  on the running values of the stagnation parameters (pressure  $p_0$ , temperature  $T_0$ ) for different initial values  $p_{in}$  of the initial 5.5% acetylene-air mixture. The change of the stagnation parameters below the dashed line was hindered by the appearance of a shock wave in the resonator cavity.

broad both with respect to pressure, 4-10 atm, and with respect to the stagnation temperature,  $1300-1700^{\circ}$ K.

We note that at pressures lower than 4-5 atm, the gain should decrease, inasmuch at these stagnation pressures the contour of the spectral line in the resonator region assumes predominantly a Doppler shape, and in addition, an appreciable decrease of the relaxation rate (the quenching of the population) of the lower laser level of the CO<sub>2</sub> molecule occurs in a section located close to the throat of the nozzle.

At stagnation pressures higher than 4-5 atm, the contour of the spectral line in the resonator region is determined by the impact broadening and becomes predominantly of the Lorentz type, and in this case the gain, as is well known, depends on the relative value of the inverted population, which depends little on the stagnation pressure. At pressures above 10 atm, the refractive index is affected by the relaxation processes that occur near the nozzle throat and decrease the population of the asymmetric valence oscillations of the  $CO_2$  molecule.

The dependence of the gain on the stagnation temperature is also understandable. When the temperature increases to  $1300-1500^{\circ}$ K, the population of the asymmetric valence oscillation of the CO<sub>2</sub> molecule increases, and this leads to an increase of the gain. However, with a temperature rise above  $1700-1800^{\circ}$ K, the rate of deactivation of this oscillation mode decreases, so that the population of these levels in a certain section of the nozzle increases with increasing deceleration temperature not as rapidly as the level population of the oscillations of the deformation type. This leads to a decrease in the gain at high temperatures.

Similar gain "surfaces" were obtained for acetyleneair mixtures with various compositions. From these plots it is possible to obtain various dependences of the gains on the stagnation parameters, and of particular interest to combustion GDL is the dependence of the gain on the pressure.

Figure 5 shows the dependence of the maximum values of the gains on the concentration of the acetylene in the initial mixture for four values of stagnation pressures: 5, 10, 15, and 20 atm. These diagrams lead to two important conclusions: 1) at a relatively low acetylene concentration and accordingly low water-vapor concentration in the combustion products (on the order of 5%), the gain depends little on the stagnation pressure in accordance with the statements made above; 2) when the acetylene content increases, on the other hand, the dependence of the gain on the stagnation pressure becomes significant. Thus, for example, for an acetylene concentration 7-8%, when the deceleration pressure changes from 5 to 20 atm, the gain decreases by approximately 2.5 times. This behavior of the gain is due to the fact that with increasing acetylene concentration and stagnation pressure, an increase takes place in the rate of deactivation of the upper laser level as a result of the increase in the stagnation temperature and of the concentrations of the  $CO_2$  and  $H_2O$  in the combustion products.

Thus, the higher the acetylene concentration in the initial mixture, the stronger the dependence of the maximum gain on the stagnation pressure. This is of great importance and must be taken into account in the design of gasdynamic lasers based on the combustion of hydrocarbon-air mixtures. We note that these results differ from the data obtained from  $CO_2-N_2$ -He mixtures by Lee et al.<sup>[22]</sup>, where it is found, for a somewhat different nozzle configuration, that the gain is independent of the stagnation pressure in the range from 2 to 16 atm.

Since the concentration of the water molecules in the combustion products is one of the main parameters that determine the behavior of the gain, it would be of interest to establish the extent to which the data on the optimal values of the gain, obtained for acetylene-air mixtures, correlate with data obtained for some other hydrocabron, say propane.

The results of an investigation of a propane-airmixture are shown in Fig. 6, which shows the dependence of the maximum values of the gain on the propane concentration in the initial propane-air mixture at an initial pressure 3 atm. The propane combustion reaction proceeds these with a small increase of volume, so that the concentration of the water molecules in the combustion products is approximately four times larger than the concentration of the propane in the initial mixture.

The experimental data on the dependence of the maximum gain  $K_{\nu}$  on the propane concentration indicate that an appreciable gain,  $(4.5-4.8) \times 10^{-3}$  cm<sup>-1</sup>, can be attained for mixtures of near-stoichiometric composition, when the concentration of the water molecules in the combustion product reaches 14-16%. For lean mixtures, a decrease of  $K_{\nu}$  was noted, and a possible natural explanation is that the combustion temperature is decreased and accordingly the population of the asymmetrical valence oscillations is decreased. In fact, estimates show that when the propane concentration in the initial mixture is decreased from 4 to 3% the temperature of the combustion product is decreased by approximately 600°K. With increasing propane concentration above 4%, the gain decreases, probably owing to the decrease of the  $CO_2$ -molecules concentration, and also owing to the increase of the concentration of the water vapor in the combustion product and the corresponding increase of the rate of deactivation of the asymmetrical valence oscillations, leading to a decrease of the inversion. Thus, relatively high gains, sufficient for the pro-



FIG. 5. Dependence of the maximum values of the gain  $K_{\nu}$  on the acetylene concentration in the initial mixture for different stagnation pressures  $p_0$ .

FIG. 6. Dependence of the maximum values of the gain  $K_{\nu}$  on the propane concentration in the initial mixture for an initial pressure 3 atm.

duction of gasdynamic lasers, are reached also for combustion products of propane-air mixtures. A direct comparison of the data on the gains for acetylene-air and propane-air mixtures is difficult because of the difference in the compositions of the reaction products, but a common correlation does exist.

One of the important problems in the investigation of the gains in GDL with combustion of hydrocarbon-air mixtures is to determine the specific influence of the combustion of the hydrocarbon fuels on the gain. In principle, there is no doubt that the kinetic laws governing the combustion of hydrocarbon fuels can exert a significant influence on the population of the laser levels of the  $CO_2$  molecule. The question is only whether such an influence did take place in our experiments, at our configuration of the nozzle, and at our experimental procedure.

To this end we have performed several series of experiments with model mixtures of carbon monoxide and hydrogen with air, which made it possible, by varying the ratio of the carbon monoxide to the hydrogen, to simulate the composition of the combustion products of various hydrocarbon-air mixtures. Thus, experiments were formed with pseudoacetylene  $(2CO + H_2)$ , pseudopropane  $(3CO + 4H_2)$ , and pseudomethane  $(CO + 2H_2)$ , which span over practically the entire range of compositions of the combustion products of the hydrocarbon fuels that are most promising for GDL.

The results of the investigation of the gains of the model mixtures are shown in Fig. 7. A comparison of the maximum gains  $K_{\nu}$  for hydrocarbon-air mixtures and for their equivalent model mixtures indicate that for both acetylene and propane there is a good correlation between the results. It can therefore be concluded that  $K_{\nu}$  is determined, other conditions being equal, by the composition of the combustion products, and the peculiarities of the kinetics of the combustion of various hydrocarbons do not affect significantly the results for the gasdynamic laser design used in this study.

### **4. CALCULATION OF THE GAIN**

It is of interest to compare the experimental data on the gains with the calculated ones. By now, a number of studies were made, in which a detailed theoretical analysis was made of the relaxation of the vibrational degrees of freedom in mixtures CO<sub>2</sub>-N<sub>2</sub>-He, H<sub>2</sub>O follow-



FIG. 7. Dependence of the maximum values of the gain  $K_{\nu}$  for model mixtures: curve 1-for 2CO + H<sub>2</sub>, 2-3CO + 4H<sub>2</sub>, 3-CO + 2H<sub>2</sub>.

ing rapid expansion in nozzles or free gasdynamic expansion<sup>[5-9,23-26]</sup>. We present below an approach that, in our opinion, is at present the most advantageous for the analysis of kinetic processes in a gasdynamic laser and for the calculation of the gains.

The equations of vibrational relaxation in a system of polyatomic molecules were obtained for the general case, with allowance for all possible relaxation channels, by Biryukov and Gordiets<sup>[23]</sup>. Recent investigations have shown that in the analysis of concrete systems  $CO_2$ -N<sub>2</sub>-He, H<sub>2</sub>O it is necessary to use a simplified system of kinetic equations, taking into consideration only the principal channels of vibrational-energy relaxation, since the presently available information on the transition probabilities make too detailed an analysis of the kinetics unproductive.

In accordance with modern concepts, the main channels of vibrational relaxation in the system CO<sub>2</sub>-N<sub>2</sub>-H<sub>2</sub>O are the following:

$$\operatorname{CO}_{2}^{\bullet}(v_{2}) + M \xrightarrow{W_{20}^{(8)}} \operatorname{CO}_{2} + M,$$
 (1)

$$CO_2 + N_2^{\bullet}(\mathbf{v}_4) \xrightarrow{W_{43}} CO_2^{\bullet}(\mathbf{v}_3) + N_2, \qquad (2)$$

$$CO_{2}^{*}(v_{3}) + M \xrightarrow{W_{4}} CO_{2}^{*}(v_{1}, v_{2}) + M,$$

$$N_{2}^{*}(v_{4}) + H_{2}O \xrightarrow{W_{4}} N_{2} + H_{2}O^{*}(v_{5}).$$
(3)

$$N_2^*(v_4) + H_2O - N_2 + H_2O^*(v_5).$$
 (4)

Here M is any one of the particles  $CO_2$ ,  $N_2$ , or  $H_2O$ ;  $W_{m}^{(S)}$  is the rate constant of the m-th reaction; the superscript s = 1, 2, and 3 pertain respectively to the molecules  $CO_2$ ,  $N_2$ , and  $H_2O_1$ .

We shall henceforth assume the following:

1) The deformation and symmetrical modes of the molecule CO2 relax jointly, inasmuch as, in the harmonic-oscillator approximation used to describe relaxation in a gas, the equilibrium between these oscillation modes is established within the same time as the establishment of the Boltzmann distribution within each vibrational mode.

2) The reaction (3) combines at the present time the two indistinguishable and parallel relaxation channels  $\nu_3 \rightarrow \nu_2 + \nu_1$  and  $\nu_3 \rightarrow 3\nu_2$ .

3) The deformation vibrations of the water molecules are in equilibrium with the translational degress of freedom,

A simplified system of the unique equations for the vibrational energy E<sub>i</sub> of the various modes, obtained from the general system of equations  $[^{23}]$ , can then be written in the form

$$\frac{d(E_{2}+2E_{1})}{dx} = \frac{N}{u} \left\{ -\sum_{s=1}^{3} W_{20}^{(s)} \alpha_{s} (E_{2}-\overline{E}_{2}) + \frac{3}{8} \sum_{s=1}^{3} W_{312}^{(s)} \alpha_{s} \left[ E_{3} (2+E_{2})^{3} - \exp\left\{ -\frac{\theta_{3}-3\theta_{2}}{T} \right\} E_{2}^{3} (1+E_{3}) \right] \right\}, \quad (5)$$

$$\frac{dE_{3}}{dx} = \frac{N}{u} \left\{ W_{43} \alpha_{2} (E_{4}-E_{3}) - \frac{1}{8} \sum_{s=1}^{5} W_{312}^{(s)} \alpha_{s} \left[ E_{3} (2+E_{2})^{3} - \exp\left\{ -\frac{\theta_{3}-3\theta_{2}}{T} \right\} E_{2}^{3} (1+E_{3}) \right] \right\} \quad (6)$$

$$\frac{dE_4}{dx} = \frac{N}{u} [W_{43}\alpha_1 (E_3 - E_4) - W_{45}\alpha_3 (E_4 - \overline{E}_4)]. \tag{7}$$

Here  $E_i$  is the vibrational energy of the i-th mode, expressed in terms of the number of quanta per molecule,

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#### $E_i = g_i [\exp(\theta_i/T_i) - 1]^{-1},$

 $g_i$  is the degree of degeneracy of the i-th mode;  $\theta_i$  is the characteristic vibrational temperature;  $T_i$  is the vibrational temperature of the i-th mode; the values i = 1, 2, 3 pertain to three vibrational modes of the  $CO_2$  molecule, while i = 4 corresponds to the vibrational motion of the molecule  $N_2$ ; N is the molecule density; u is the stream velocity;  $\alpha_S$  is the molar fraction of the s-th component.

The kinetic equations (5)-(7) are solved simultaneously with the equations of gasdynamics of one-dimensional stationary flow of gas through a nozzle of given configuration:

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$$A(x)\rho u = A \cdot \rho \cdot u \cdot , \qquad (8)$$

$$u\frac{du}{dx} + \frac{dp}{dx} = 0,$$
 (9)

$$\frac{3}{2}kT + \left(\alpha_1 + \alpha_2 + \frac{3}{2}\alpha_3\right)kT + \alpha_1\sum_{i=1}^3 hv_i E_i + \alpha_2 hv_i E_i + \alpha_3 hv_5 E_5 + \frac{1}{2}mu^2 + \frac{pm}{\rho} = mH_0, \quad (10)$$

$$p = \rho m^{-1} kT, \quad m = \sum_{s=1}^{3} \alpha_s m_s.$$
 (11)

Here p and  $\rho$  are the pressure and the density, H<sub>0</sub> is the specific stagnation enthalpy, and A(x) is the area of the channel cross section. The asterisks mark the values of the corresponding quantities in the critical cross section of the nozzle.

It should be noted that the simplified vibrationalrelaxation equations proposed by Anderson<sup>[8]</sup>, which are widely used in calculations, can result in noticeable deviations from those given above in cases when the temperature  $T_2$  of the deformation vibrations cannot keep up with the gas temperature T. In mixtures with more water molecules we have  $T_2 \approx T$  and the Anderson procedure practically coincides with that considered here.

The weak-signal gain at the center of a line broadened by collisions and by the Doppler effect can be expressed in the form [27]

$$K_{v} = \frac{\lambda^{2} A (\ln 2)^{\frac{v_{i}}{2}}}{8\pi^{\frac{v_{i}}{2}} \Delta v_{p}} \left( N_{2} - \frac{g_{2}}{g_{1}} N_{1} \right) H\left( \frac{\Delta v_{c}}{\Delta v_{p}} (\ln 2)^{\frac{v_{i}}{2}}, 0 \right), \qquad (12)$$

where  $N_2$  and  $N_1$  are the populations of the upper and lower laser levels;  $g_2$  and  $g_1$  are the corresponding statistical weights; A is the Einstein coefficient for spontaneous emission; H is the Voight function:

$$H(a,0) = \frac{a}{\pi} \int_{-\infty}^{+\infty} \frac{e^{-\nu^{2}} dy}{y^{2} + a^{2}};$$

 $\Delta \nu_D$  and  $\Delta \nu_c$  are the Doppler and collision line half widths and are respectively equal to

$$\Delta v_{\nu} = \frac{1}{\lambda} \left( \frac{2kT \ln 2}{m} \right)^{\frac{1}{2}}, \quad \Delta v_{e} = \frac{1}{2\pi} \left( \frac{8N_{A}}{\pi kT} \right)^{\frac{1}{2}} p \sum_{e} \frac{\alpha_{e} \sigma_{e}}{\bar{\mu}_{e}^{\frac{1}{2}}}, \quad (13)$$

where  $\sigma_{\rm S}$  is the cross section of the optical line broadening in collisions of CO<sub>2</sub> molecules with particles of type s;  $\mu_{\rm S}$  is the reduced molecular weight of the colliding particles; NA is Avogadro's number. The values of  $\sigma_{\rm S}$  were determined earlier<sup>[28-31]</sup> and are given below.

The Einstein coefficient for the spontaneous emission was measured in  $[^{32}, ^{33}]$ . The value obtained in  $[^{133}]$ ,

A = 0.169 sec<sup>-1</sup>, differs significantly from the value A = 0.21 sec<sup>-1</sup> determined by the authors of  $[^{32}]$  and assumed in the present paper.

The density  $N_V(J)$  of the molecules on the vibrational-rotational level, assuming equilibrium of the rotational and the translational degrees of freedom, is

$$N_{v}(J) = N_{v} \frac{2hcB_{v}}{kT} (2J+1) \exp\left[-\frac{hcB_{v}J(J+1)}{kT}\right].$$
 (14)

Here h is Planck's constant, J is the rotational quantum number, and  $B_V$  is the rotational constant.

The rate constants of reactions (1)-(4) were determined by approximating the experimental data<sup>[34-38]</sup> and are given by

$$W_{20}^{(1)} = 1.1 \cdot 10^{-14} T \exp\left(-41.6/T^{'h}\right), \quad W_{20}^{(2)} = 3W_{20}^{(1)}, \quad W_{20}^{(3)} = 3.9 \cdot 10^{-10}/T, \\ W_{43} = 4.2 \cdot 10^{-14} T^{'h} \exp\left[10^{-3}T\left(8.84 \cdot 10^{-4}T - 2.07\right)\right], \\ W_{412}^{(1)} = \frac{0.138 \cdot 10^{-15}T\left[1 - \exp\left(-\theta_2/T\right)\right]^3 \left[1 - \exp\left(-\theta_3/T\right)\right]^{-1}}{\exp\left(24.47 - 592.72/T^{'h} + 4764.1/T^{''4} - 12226/T\right)} \\ W_{312}^{(2)} = \frac{0.138 \cdot 10^{-15}T\left[1 - \exp\left(-\theta_2/T\right)\right]^3 \left[1 - \exp\left(-\theta_3/T\right)\right]^{-1}}{\exp\left(18.666 - 507.83/T^{'h} + 4574.9/T^{''4} - 1274/T\right)}, \\ W_{312}^{(3)} = 3.2 \cdot 10^{-15}T, \quad W_{45} = 0.145 \cdot 10^{-17}T \exp\left(-30.6/T^{'h}\right), \end{cases}$$
(15)

The calculations were performed for a flat speciallyshaped nozzle, which was used by us for the experiment. The result of the calculations for the mixture 11.2%  $CO_2 + 83.2\%$  N<sub>2</sub> + 5.6% H<sub>2</sub>O, corresponding to the combustion products of a mixture 5.5% C<sub>2</sub>N<sub>2</sub> + air, are shown in Fig. 8 in the form of plots of the gains along the nozzle lines for the most interesting variants. From an analysis of the plots it follows that the gain reaches a maximum at deceleration parameters C<sub>0</sub> = 1500°K and p<sub>0</sub> = 10 atm (curve 2 in Fig. 8). Under these conditions, the absolute value of the gain is  $8 \times 10^{-3}$  cm<sup>-1</sup>, which is good agreement with the experimental data Figs. 3 and 4).

In addition, the plots account for all the qualitative dependences of the gain on the deceleration parameters, which were mentioned above in the discussion of the experimental relations. Thus, when the stagnation pressure increases to 15-20 atm, the gain gradually decreases because of the relaxation processes that lead to a decrease of the population of the upper laser level as the gas expands in the nozzle. At relatively low pressures (see curve 5 in Fig. 8), the population inversion of the laser levels takes place farther downstream, owing to the slowing down of the relaxation of the lower laser level. Thus, we can state that there is a sufficiently good qualitative and quantitative correlation between the calculation results and the experimental data.

### 5. MEASUREMENT OF THE POWER GENERATED BY A GASDYNAMIC LASER

To obtain lasing, mirrors were inserted in the adjustment heads of the resonator cavity. The resonator was made up of a flat mirror with reflection coefficient

FIG. 8. Calculated dependences of the gain  $K_{\nu}$  on the stagnation parameters. Curves 1 to 4 are for  $p_0 = 10 \text{ atm}: 1$ )  $T_0 = 1250^\circ \text{ K}, 2$ )  $T_0 = 1500^\circ \text{ K}, 3$ )  $T_0 = 1750^\circ \text{ K}, 4$ )  $T_0 = 2000^\circ \text{ K}$ ; curves 5 to 8 are for  $T_0 = 1750^\circ \text{ K}: 5$ )  $p_0 = 5 \text{ atm}, 6$ )  $p_0 = 10 \text{ atm}, 7$ )  $p_0 = 15 \text{ atm}, 8$ )  $p_0 = 20 \text{ atm}. x$  is the distance from the critical section of the nozzle.



37% and of tiltable flat and spherical mirrors with reflection coefficients 98%. The spherical mirror had a focal length 2.5 m. The apparatus made it possible to investigate multiple-pass resonators with up to five passes. It turned out here that when the number of passes increases beyond three the generation energy and the power decrease, owing to the decrease in the gain downstream. The dependence of the lasing power on the mixture composition and on the stagnation parameters was therefore investigated with a three-pass resonator.

The energy generated by the laser was registered with an energy meter of the IKT-1 type. To measure the waveform of the laser pulse, some of the energy of the laser beam was diverted with a plane-parallel plate and registered with a Ge-Au photoreceiver.

In each experiment we determine simultaneously the stagnation pressure, the waveform of the lasing pulse as a function of the time, and the energy of the laser radiation integrated over the time of the pulse. These data made it possible to determine the radiation power and its time variation as functions of the stagnation parameters, Typical oscillograms of the radiation generation (lower curve) and of the stagnation pressure (upper curve) are shown in Fig. 9 (the sweep direction of the process is from left to right; one division is equal to 10 msec). In this experiment, the maximum stagnation pressure was 24 atm, and the maximum power was 4.6 kW, Figure 3 shows the measured generation power (squares) as a function of the concentration of the acetylene in the initial mixture for an initial pressure 3.7 atm. We note that the reproducibility of the results was very good and that each point on the diagram is an average of three or four experiments. The character of the obtained dependence is analogous to that established in experiments on the measurement of the gain. This was to be expected, since the generation power is a function of the gain and is determined, although not fully, by the same kinetic processes as the gain. The maximum generation power was reached in this series of experiments at an acetylene concentration 5% and amounted to 5.2 kW, with the stagnation pressure equal to approximately 6.3 atm, and the deceleration temperature 1450°K,

When the acetylene concentration was increased above 5%, a gradual decrease of the power took place; on the other hand, when the gain decreased to such an extent that it corresponded to the energy loss level in the resonator, the generation vanished completely.

Of particular interest for GDL of the considered type is the dependence of the generation power on the stagna-



FIG. 9. Oscillograms of the deceleration pressure (curve 1) and of the generated radiation (curve 2).

tion pressure for an optimal mixture containing 5% acetylene. The first four columns of the table list more detailed information on the dependence of the maximum radiation power on the stagnation parameters. From an analysis of these data it follows that with increasing stagnation pressure an increase takes place in the laser radiation power, in almost linear fashion. The character of the variation of the radiation power with changing pressure is determined by two opposite effects: first, the mass flow of the gas increases with increasing pressure, and this increases the radiation power; to the contrary, an increase of the pressure leads to an increase of the rate of de-activation of the upper laser level, causing a decrease in the population inversion of the laser levels and of the gain, and consequently also of the power. In our case, as follows from the table, the radiation power increases continuously with increasing stagnation pressure, thus indicating that the radiation power is predominantly influenced by the mass flow of the gas. At a stagnation pressure 14.5 atm, the radiation power reached 8.5 kW (see the third column in the table). Under these conditions, the laser-pulse energy amounted to 150 J at an approximate duration 20 msec.

It should be noted that in our experiments the output mirror of the resonator has an absorption coefficient 0.13, so that if we exclude the loss due to the radiation absorption in the output mirror, then the laser-generated power increases from 8.5 to 10.7 kW.

To estimate the efficiency of our GDL, it is of interest to compare the obtained experimental data on the generation power with the maximum obtainable GDL radiation density, which is determined by the reserve of the laser energy accumulated in nitrogen molecules and  $CO_2$  at the exit from the nozzle. The maximum attainable radiation power is proportional to the flux of laser energy stored in the asymmetrical vibrations of the  $CO_2$  molecule and in the nitrogen, and is determined by the expression

$$P_{max} = \eta_{q} [\mathscr{E}(T_{s}) - \mathscr{E}(T_{e})]G, \qquad (16)$$

where  $\eta_q = (h\nu_3 - h\nu_1)/h\nu_3$  is the quantum efficiency;  $\mathscr{E}(T_3)$  is the reserve of vibrational energy of the upper laser level when the temperature of this level is equal to T<sub>3</sub>, per unit mass of the gas;  $\mathscr{E}(T_e)$  is the vibrational energy of the upper laser level at a temperature T<sub>e</sub> at which the gain becomes equal to zero; G is the mass flow of the gas.

It follows from gasdynamics that

$$G = S^{\star} \left(\frac{2}{\gamma+1}\right)^{1/(\gamma-1)} \sqrt{\frac{2\gamma}{\gamma+1}} p_0 \rho_0.$$
 (17)

Here and below  $S^*$  is the summary area of the critical section of the nozzle block;  $p_0$ ,  $\rho_0$ , and  $T_0$  are respectively the stagnation pressure, density, and temperature;  $\gamma$  is the adiabatic constant, which for our working medium was 1.28 when the gas when expanded to the critical section of the nozzle and 1.35 with further expansion of the gas, when quenching of the vibrational

p <sub>0</sub> , atm	Τ., К	P, kW	K √·10³, cm <sup>-1</sup>	т, к	G, g/sec	<i>T</i> 3, K	P <sub>max</sub> , kW	$f_i := \frac{P}{P_{max}}$
4.8	1600	2,4	6.0	350	470	1090	8.3	0.29
6.3	1520	4.9	8.5	320	630	1190	14.7	0.33
6.3	1420	5,2	8.0	310	660	1150	14.1	0.37
10.2	1490	6.3	7.1	330	1040	1120	20.0	0.31
14.5	1600	8.5	6.3	350	1430	1100	26.6	0,32

energy of the nitrogen molecule and of the asymmetric  $\mathbf{CO}_2$  vibrations took place.

Estimates show that the quantity  $\mathscr{E}(T_e)$  in Eq. (16) can be neglected in comparison with  $\mathscr{E}(T_3)$ . The vibrational energy of the upper laser level per unit mass and assuming equality of the energy of the vibrational quanta of the asymmetrical mode of the CO<sub>2</sub> molecule and of the nitrogen is equal to

$$\mathscr{E}(T_{s}) = hv_{s}m^{-1}(\alpha_{co_{s}} + \alpha_{N_{s}}) [\exp(\theta_{s}/T_{s}) - 1]^{-1}.$$
(18)

The quenched vibrational temperature of the  $CO_2-N_2$ mixture can be determined from measurements of the gain "surfaces" for the 5% acetylene-air mixture (in analogy with Fig. 4 for the 5.5% acetylene-air mixture), provided that the translational temperature of the gas is known. The translational temperature of the gas can be determined from the given stagnation temperature  $T_0$  and from the number M of the flux from the expression for the isentropic flow of an ideal gas in the nozzle:

$$T/T_0 = [1 + \frac{1}{2}(\gamma - 1)M^2]^{-1}.$$
 (19)

The number M of the flux will be obtained from the ratio of the area of the output cross section of the nozzle to the critical area:

$$\frac{S}{S^{*}} = \frac{1}{M} \left[ \frac{2}{\gamma+1} \left( 1 + \frac{\gamma-1}{2} M^{2} \right) \right]^{(\gamma+1)/2(\gamma-1)} .$$
 (20)

For the area ratio  $S/S^* = 25$  and for  $\gamma = 1.35$  we find that M = 4.6 and  $T = 0.21T_0$ .

We assume further that the temperatures of the symmetrical and deformation modes are in equilibrium with the translational temperature of the gas. Then from the expression for the gain at the center of the line (12) we can determine the vibrational temperature T<sub>3</sub>, which in turn makes it possible to calculate the maximum attainable radiation power  $P_{max}$ . In the calculation we used the following constants<sup>[28-31]</sup>.

$$\theta_1 = 1920 \text{ K}, \quad \theta_2 = 960 \text{ K}, \quad \theta_3 = 3380 \text{ K}, \\ \sigma_1 = 1.3 \cdot 10^{-14} \text{ cm}^2, \quad \sigma_2 = 0.87 \cdot 10^{-14} \text{ cm}^2, \quad \sigma_3 = 0.38 \cdot 10^{-14} \text{ cm}^2.$$

The results of the calculation are shown also in the table in the fifth to ninth columns. From an analysis of these results we can draw a number of interesting conclusions.

First, the quenching of the vibrational energy of the upper laser level occurs downstream of the critical section of the nozzle, where the gas temperature is approximately 0.8-0.9 of the gas temperature at the critical section. As a result, the efficiency of the nozzle in our experiments was approximately 0.5, i.e., half of the vibrational energy of the upper laser level was lost to vibrational de-activation during the gas expansion in the nozzle.

Second, the experimental values of the lasing power amount to approximately 0.32–0.37 of the maximum attainable power  $P_{max}$  for the employed nozzle geometry. The quantity  $\eta = P/P_{max}$  can be regarded as the efficiency of the resonator operation. Thus, approximately one third of the laser energy contained in the stream was converted into laser radiation. The remaining  $\frac{7}{3}$  of the energy comprised the losses, including the losses due to the resonator mirrors, due to collision de-activation in the resonator, and due to gasdynamic effects. In addition, part of the energy remains in the gas as it leaves the resonator, as is evidenced by the results of experiments on the determination of the gain in sections downstream from the resonator in the case of simultaneous generation of the GDL.

Third, in our experiments the specific radiation power was maximal at stagnation pressures on the order of 6.3 atm and amounted to 7.8 kW/kg, and decreased to 5.9 kW/kg with increasing stagnation pressure to 14.5 atm. On the other hand, when account is taken of the laser radiation absorbed by the mirrors, these values of the specific radiation power are equal to 9.7 and 7.3 kW/kg, respectively. Fourth, it is known that the efficiency of gasdynamic lasers with thermal excitation is relatively low, because the vibrational energy of the gas constitutes a small fraction of the total enthalpy under stagnation conditions. Therefore it becomes possible to convert into laser radiation only one or two percent of the enthalpy of the deceleration of the gas. In our experiments, the maximum efficiency was 0.6%.

Further improvement of the characteristics of the gasdynamic laser can be obtained by increasing the operating efficiency of the individual units of the setup, especially the resonator. It is quite realistic to raise the per unit power of the laser radiation in combustion GDL to approximately 15 kW/kg.

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