Investigation of light amplification in a pulsed acetylene-air combustion gasdynamic laser

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The amplification coefficients of the combustion products of acetylene-air mixtures expanding in a hypersonic jet are measured as functions of the suppression parameters and acetylene concentration in the initial mixture. The maximal value of the amplification factor is 6×10^{-3} cm^{-1}, which is about 3 times greater than the theoretical values for the jet geometry employed. It is shown that when a shock wave propagates in a gas with population inversion the amplification first increases and then gives way rapidly to absorption.

The idea of producing inverted population in gases by rapidly changing the temperature was advanced in[1,2] The possibility of obtaining inverted population by adiabatic expansion of a $CO_2 + N_2$ gas mixture was indicated in[3,4]. Lasing was observed experimentally by expanding gas through a Laval nozzle and a slit in[5-7]. Somewhat later (see[8]), data were reported on a continuous gas-dynamic laser (GDL) of high power, amounting to approximately 60 kW with multimode laser operation. The working mixture of this laser was pumped by burning carbon monoxide. Such pumping in high-power continuous GDL is, however, expensive and unpromising. This is why recent research was aimed at ascertaining the possibility of using the combustion products of hydrocarbon fuels as active laser media.

It was only a few years ago that the use of combustion of hydrocarbons to obtain GDL working media raised considerable doubts because of high concentration of water vapor in the combustion products of hydrocarbon fuels, since water is highly effective in the deactivation of the vibrational levels of the CO_2 molecule. Recent investigations[9,10] have shown, however, that high-power GDL based on the combustion of hydrocarbon fuels are quite feasible.

Tulip and Seguin[10] recently measured the emission gain obtained by expanding the combustion products of $CO + H_2$ mixtures in a hypersonic nozzle of a pulsed GDL, and also demonstrated the feasibility of lasing via escape of the combustion products of a number of hydrocarbon fuels. An analysis of this study, however, has shown that it has a number of significant shortcomings: First, the gains for the combustion products of the hydrocarbon fuels were not measured; second, no optimization was carried out with respect to the stagnation parameters; third, the experiments revealed excessively large fluctuations of the gain and power signals, thus indicating the presence of methodological errors; finally, judging from the oscillograms, the maximum gain and power are obtained at the instant when the pressure in the explosion chamber decreases practically to zero, something which is at best not understandable. In the present paper we determine the gain produced when acetylene-air-mixtures with different compositions expand in a hypersonic nozzle, and compare it with the theoretical values obtained by a previously proposed method[11]. Acetylene was chosen as the fuel because of two circumstances: first, its combustion products contain the minimum amount of water vapor per produced CO_2 molecule, and second, its combustion rate greatly exceeds that of other hydrocarbons.

The experimental setup is illustrated in Fig. 1. It consisted of a combustion chamber 1, a flat-throat supersonic nozzle 2, a vacuum chamber 3, and an optical system for registering the gain. Prior to the experiment, the entry into the nozzle, on the combustion chamber size, was covered with membrane 4, making it possible to evacuate the chamber to a specified pressure. The combustion chamber was then filled with an acetylene-air mixture of definite composition, after which the mixture was ignited by spark plug 5 and the combustion front propagated through the chamber. As a result, the pressure in the combustion chamber increased to a certain maximum value at which the membrane broke and the combustion products expanded through a flat-throat supersonic nozzle into the vacuum chamber. The height of the critical nozzle cross section was 1 mm, the width of the nozzle was 14 cm, and the half-angle was 15°.

The gain was determined by sounding the gas stream with a beam from a low-power laser 6 in a direction perpendicular to that of the gas motion. The sounding was behind the end of the nozzle. The radiation of this CO_2 laser (power 2 W) passed through salt windows and through the stream, was gathered by lens 7, and was registered by the GeAu photoresistor 8. To exclude the influence of the spontaneous emission on the registered signal, diaphragm 9 was placed ahead of the lens. Control experiments have shown that the spontaneous emission of the investigated mixture lies beyond the sensitivity limit of the recording system. To take into account the influence of the possible instabilities of the sounding-laser beam power on the results, a fraction of the energy of this beam was diverted with the aid of a plane-parallel plate 10, was interrupted by chopper 11, and was registered by a second GeAu receiver.

The combustion process and the parameters of the combustible mixture were monitored with a calibrated piezoelectric pressure pickup 12 installed in the combustion chamber. By recording the pressure we were

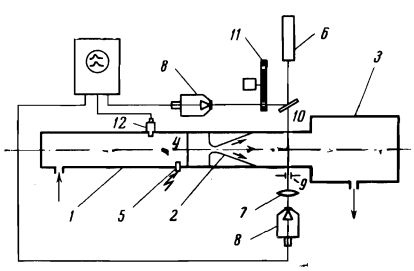

FIG. 1

able not only to determine the stagnation parameters, but also to follow their time variation. The stagnation temperature prior to the breaking of the membrane was calculated from the maximum value of the stagnation pressure. The combustion-product temperature calculated in this manner was in good agreement with the result measured temperature of the flames of acetylene-air mixtures[12].

One of the main advantages of the described procedure is that it is possible to register simultaneously the pressure and gain signals, so that in fact a single experiment yields a large amount of information concerning the dependence of the gain of the gas mixture on the stagnation parameters.

Figure 2a shows oscillograms of the pressure and of the gain of a 6% acetylene-air mixture. In this experiment, the temperature and the stagnation pressure of the combustion products at the initial instant prior to the breaking of the membrane were 2250°K and 22 atm, respectively. The amplification in this experiment was maximal and amounted to 8.4%, and while the gain coefficient was $k = 6 \times 10^{-3}$ cm^{-1}. From the oscillograms of the signals shown in Fig. 2a it follows that the amplication of the gas mixture begins immediately after the breaking of the membrane. The gradual decrease of the stagnation pressure in the compression chamber, due to the escape of the combustion products through the nozzle, leads to a rapid growth of the gain. The gain reaches a maximum at a combustion-chamber pressure on the order of 7–8 atm. The gain decreases gradually during the course of the subsequent expansion of the combustion products.

The oscillograms retained approximately the same shape in the entire series of experiments in which the acetylene concentration in the initial acetylene-air mixture was varied. In some experiments, however, we observed a curious picture of the gain signal, such as in Fig. 2b (in which, in contrast to Fig. 2a, amplification corresponds to a downward deflection of the beam) namely, the drop of the gain signal at the end of the expansion (arrow 1) was followed by a peaked increase of the amplification signal (arrow 2) and in turn by a rather abrupt decrease turning into absorption (arrow 3). The apparent reason for this effect is that as the combustion products expand through the supersonic nozzle the vacuum chamber becomes pumped by the combustion products, and at a certain instant of time the counterpressure results in a shock wave that propagates towards the nozzle. The passage of this wave through the section in which the sounding takes place causes first an increase of the gain signal, and then a sharp decrease that turns into absorption.

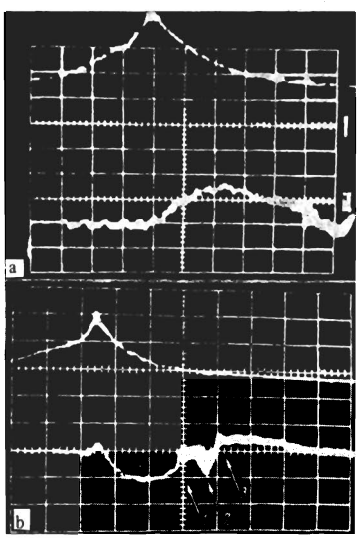

FIG. 2

The increase of the gain directly behind the front of the shock wave is due to the increase of the gas density was a result of the shock compression in the front of the shock wave while the molecules retain practically the same distribution over the vibrational levels as ahead of the shock-wave front. Subsequently, however, as a result of the increase in the translational temperature and density behind the front of the shock wave, the excited states relax and the translational temperature and density accordingly assume new values, leading to an underpopulation of the lower laser level, to a rapid decrease of the gain, and even to a change from amplification to absorption. In fact, the gain of light of frequency ν can be expressed in the form

$$k_\nu = \frac{Ac^2}{8\pi\nu^2\Delta\nu'}(n_2 - n_1)\,U\left[\frac{\Delta\nu(\ln 2)^{1/2}}{\Delta\nu'}, \frac{(\nu-\nu_0)(\ln 2)^{1/2}}{\Delta\nu'}\right]$$

where A is the Einstein coefficient for the spontaneous emission, c is the speed of light, ν_0 is the transition frequency at the line center, $\Delta\nu$ and $\Delta\nu'$ are the Lorentz and Doppler line widths, n_1 and n_2 are the populations of the upper and lower laser levels and U is the Voigt function. It is known that the Lorentz width is proportional to the density of the gas, whereas the Doppler width does not depend on the density. Therefore, in the initial stage of the escape process, when the level of the stagnation pressures is high enough, the spectral line has a Lorentz contour, and in this case the gain is practically independent of the pressure.

Some influence of the stagnation pressure on the gain can of course occur in this case, since the pressure affects the rate of the relaxation processes during the course of the expansion of the gas in the nozzle. Thus, the increase of the gain with increasing pressure after the start of the outflow is due to the fact that deactivation in the collisions of the upper laser levels comes into play at high pressures. At stagnation pressures on the order of 7–8 atm, the conditions for the amplification are optimal. At the end of the escape process, when the stagnation pressure decreases to 4–5 atmospheres, the gain decreases, as expected, and at such pressures the decisive role is assumed by the Doppler broadening of the spectral line and, in addition, at low pressures the populations of the lower level occurs are quenched in a section located closer to the throat of the nozzle.

Thus, when the stagnation pressure drops below 4–5 atm, a gradual decrease takes place in the inverted population, owing to the decrease in the density, which lead to a decrease in the gain if the line has a Doppler contour. Directly behind the shock-wave front, however, the gas becomes compressed, the density increases, and this, naturally, leads first to an increase in the gain. In fact, the ratio of the densities on the front of the shock wave can be estimated from the known expression for the limiting case of a strong shock wave $\rho_2/\rho_1 = (\gamma+1)/(\gamma-1)$. If we equate the adiabatic exponent to $\gamma = 1.3$, then the density ratio on the front of the shock wave equals 7.6. At the same time, the broadening of the Doppler contour as a result of the increase of the translational temperature when the gas passes through the front of the shock wave is approximately

1.6. It follows from these estimates that an increase of the gain should be observed at low stagnation pressures directly behind the front of the shock waves propagating in the inverted medium, owing to the increased density.

Subsequently, owing to the increase of the density and of the translational temperature of the gas, the rate of relaxation processes behind the shock-wave front increases and leads to an increase in the population of the lower laser level. Consequently the inversion vanishes and absorption is observed. Thus, in the design of gas dynamic lasers one must bear in mind the aforementioned effects, which are connected with the formation of shock waves in a stream of gas with inverted population.

The results of the measurements of the gain of the gas medium as a function of the concentration of the acetylene in the acetylene-air mixture are shown in Fig. 3 (dark circles). The concentration of the acetylene in the acetylene-air mixture was varied in the range from 4 to 13%. At lower concentrations, the mixture could not be ignited by our ignition source, and noticeable soot production set in at concentrations higher than 13%. All the experiments were performed at an initial mixture pressure 3.0 atm. The same figure shows a plot of the combustion temperature T aginst the composition of the acetylene-air mixture (curve) in accordance with the data of[12] and on the basis of our calculations from the measured value of the maximum pressure (light circles).

As follows from the diagram, for our nozzle the gain decreases with increasing acetylene concentration in the mixture, probably as a result of the increase of the concentration of the water vapor in the combustion products and the corresponding increase of the rate of deactivation of the asymmetrical valence vibration that leads to a decrease of the inversion. When the acetylene concentration was decreased below 4%, the gain should also decrease since the combustion temperature increases very rapidly, and this leads to a decrease in the populations of the asymmetrical valence and deformation vibrations and their difference.

Great interest attaches to a comparison of the obtained experimental values of k with the calculated ones. A method was proposed in[11] for calculating the level populations of different modes of vibrations of the CO_2 molecules in the mixtures $CO_2 + N_2 + Ag$ and $CO_2 + N_2 + H_2O$ as they are expanded in hypersonic nozzles. This procedure was verified by us earlier by comparing the calculated values of the gain with the experimental values obtained in experiments with an aerodynamic shock tube for the mixtures $CO_2 + N_2 + He$ (see[13]) and also for the mixtures $CO_2 + N_2 + H_2O$ containing up to 5% water vapor. Thus, the calculation procedure is not subject to any doubt, and to compare the results of the present experiments with the theory we calculated the gain coefficients as applied to the conditions of our experiments for combustion products of a stoichiometric acetylene-air mixture expanding in a supersonic nozzle; the equilibrium composition of the mixture was $CO_2:N_2:H_2O = 2:10:1$.

The results of the calculations are shown in Fig. 4 in the form of a plot of the gain against the distance from the nozzle throat at a stagnation pressure 7.5 atm and at three values of the stagnation temperature: 1500, 1750, and 2000°K. From the analysis of the presented data it follows that k depends very little on the stagnation temperature. The gain depends just as little on the stagnation pressure in the region 5–10 atm. This was

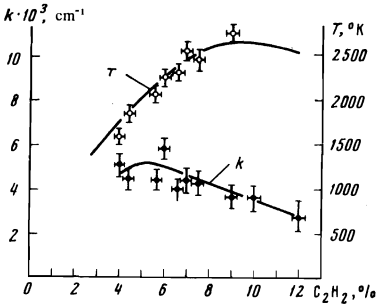

FIG. 3

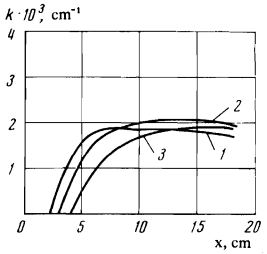

FIG. 4

to be expected, for at such stagnation pressures, as noted above, the spectral-line contour is determined mainly by the collision broadening, and the gain depends actually on the relative population inversion which in turn depends little on the stagnation pressure.

If we compare the experimental and calculated values of the gain we can conclude that the experimental results are 2–3 times higher than the calculated values. Such a discrepancy is unexpected and greatly exceeds the possible experimental errors or the uncertainty in the theoretical calculations. It seems to us that this discrepancy is of fundamental character and is due in all probability to the fact that the combustion process is not completed in the combustion chamber, but continues during the expansion of the reacting mixture in the supersonic nozzle, which in turn leads apparently to an increase in the population of the upper laser level.

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