

Short pulse formation in a TEA CO₂ laser using a CO₂–N₂–H₂ gas mixture

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Abstract. The use of an optimal CO₂–N₂–H₂ gas mixture with a high concentration of hydrogen (30%–40%) was shown to allow the formation of high-power TEA CO₂-laser pulses with a base duration of ~200 ns whose energy and peak power significantly exceed the parameters of pulses obtained in binary CO₂–H₂ mixtures. Considering all the parameters, the helium-free 56% CO₂–14% N₂–30% H₂ gas mixture at a pressure of 0.7 atm is optimal for the generation of short high-power pulses in the TEA CO₂ laser. A small addition of nitrogen ([CO₂]/[N₂] ~ 10–12) to the binary CO₂–H₂ mixture not only substantially increases the pulse energy and peak power (by a factor of 3–3.5) but also shortens their duration at half-maximum with retention of the total (base) duration.

Keywords: TEA CO₂ laser, short pulses, additions of hydrogen.

1. Introduction

The problems of shaping of short high-power TEA CO₂-laser pulses without the so-called nitrogen tail are of considerable interest to researchers in the field of laser photochemistry, laser isotope separation, remote gas analysis, nonlinear optics, and nonlinear laser spectroscopy. The most obvious way of solving this problem involves cutting the undesirable nitrogen ‘tail’ of a typical TEA CO₂-laser pulse using a special optical shutter. Despite the apparent simplicity, this approach necessitates the use of complex expensive optical elements, such as an electrooptical shutter [1] or a plasma pulse clipper [2, 3], a laser-triggered spark gap [3], a synchronisation device, etc. A CO₂ laser scheme with a self-modulation of intracavity losses was proposed in Ref. [22] for generating pulses free from the radiation ‘tail’ inherent in CO₂ lasers.

At the same time, there exists a rather well-known technique of removing the ‘tail’ of TEA CO₂-laser radiation pulses based on the use of gas mixtures with a reduced nitrogen content or without nitrogen at all [4]. In this case, no additional optical elements are required. However, the

total pulse energy lowers significantly, thereby making topical the problem of selecting the optimal gas mixture composition and the excitation mode to achieve high energy laser parameters. Investigations of nitrogen-free CO₂–H₂ and CO₂–He mixtures [5, 6] showed that replacement of helium with hydrogen results in the increase in the energy and peak power of the output laser pulses and reduces their duration. The addition of deuterium also results in an increase of the energy of TEA CO₂-laser pulses [7, 8], which is attributed to the additional pumping of the upper laser level by deuterium. However, gas mixtures with additions of H₂ appear to be more attractive for practical use because of the high cost of deuterium.

The effect of hydrogen on the parameters of TEA CO₂-laser radiation was studied in papers [9–11]. In particular, small additions of hydrogen to the gas mixture of the laser were shown to improve its parameters and the volume discharge stability, and to increase the gain in the active medium. Higher output laser energies [9–11] and peak powers [9, 11] can be achieved at higher molecular gas densities and a higher energy input. At the same time, the degradation of the working mixture can be significantly reduced [12–15] because hydrogen acts as a gaseous catalyst [13].

The increase in the small-signal gain in hydrogen-containing mixtures is caused by the increase in the relaxation rate of the lower 01⁰ laser level of CO₂ molecules, which is ~20 times higher for hydrogen than for helium [16]. However, hydrogen is also responsible for the deactivation of the upper 00¹ laser level [16], which lowers the gain [8] and the energy of laser pulses [17] at high concentrations of H₂. In the light of the above discussions the measurement of the optimal hydrogen concentration in the gas mixture of a TEA CO₂-laser is important for the efficient operation of the laser. The aim of this work is to study the effect of additions of hydrogen on the formation of output laser pulses and the service life of a tunable TEA CO₂ laser.

2. Experimental setup

We used in experiments a tunable repetitively pulsed TEA CO₂ laser described in detail in papers [18, 19]. A characteristic feature of this laser is the combination of an intense symmetric UV preionisation of the active volume, the optimal delay of the onset of the main discharge (~800 ns), and a bipolar low-inductance pulsed voltage generator (PVG). This allowed us to expand the range of operating pressure in molecular gas mixtures [18]. The active laser medium was excited by a high-voltage current pulse with a duration at half-maximum of ~240 ns. A homogeneous volume dis-

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charge of size $2\text{ cm} \times 3\text{ cm} \times 73\text{ cm}$ (0.44 L) was produced in the laser discharge cell in the $\text{CO}_2 - \text{N}_2 - \text{H}_2$ molecular gas mixture at a pressure up to 1 atm. The stable selective laser cavity 1.3 m in length was formed by a 100 mm^{-1} reflection diffraction grating operating in the autocollimation regime and a semitransparent mirror (a plane ZnSe plate with a transmittance of $\sim 75\%$). The transverse mode structure of a laser beam was controlled with an intracavity aperture of diameter from 8 to 30 mm.

The shape of output laser pulses was recorded with a fast photon-drag Ge detector and recorded with a C9-27 digital storage oscilloscope. The pulse energy was measured with an IMO-2N power meter.

For every selected value of the pressure and composition of the gas mixture, five oscilloscope traces of the TEA CO_2 -laser pulses were recorded with simultaneous measurements of the pulse energies. The measured energies and the stored oscilloscope traces were used to determine the average values and the standard deviations of the total pulse energy E , the peak power P , the energy E_1 contained in the initial 100-ns long interval (in the leading peak), the relative energy distribution $\xi = E_1/E$, the total pulse width at levels of 0.5 (τ_1), 0.1 (τ_2), and 0.01 (τ_3) of the amplitude on the peak output laser power P , and also the pulse duration τ_4 accounting for 99% of the total pulse energy E .

Note that the system for recording the temporal TEA CO_2 -laser pulse shapes employed did not allow us to resolve the temporal fine structure arising from the self-mode-locking and yielded smoothed pulse shapes. In the context of our consideration of the effect of additions of hydrogen on the energy characteristics of laser pulses and the service life of the laser, this approach is legitimate, in our opinion, because the obtained dependences reflect the integral characteristics of laser pulses and were measured by averaging over several pulses.

3. Effect of H_2 on the parameters of output laser pulses

To produce short TEA CO_2 -laser pulses and investigate the influence of hydrogen on their energy and temporal parameters, we chose the $4\text{CO}_2 - \text{N}_2$ molecular gas mixture with additions of hydrogen. This mixture was chosen on the basis of preliminary experiments [18] and the data of Ref. [21]. Fig. 1 shows the oscilloscope traces of single output laser pulses obtained with the $4\text{CO}_2 - \text{N}_2 - \text{H}_2$ gas mixture for the optimal pressure $p = 0.7\text{ atm}$, which illustrate the variation in the shape of laser pulses with increasing hydrogen concentration. Fig. 2 depicts the experimental dependences of the energy and time parameters of the output laser pulses on the hydrogen concentration in this gas mixture for a pressure $p = 0.7\text{ atm}$.

One can see from Figs 1 and 2, that the addition of hydrogen to the mixture up to 10% resulted in the increase in the output pulse energy by a factor of 1.8 (from 2.4 to 4.3 J). At higher hydrogen concentration, the pulse energy decreased. A similar result was observed in Refs [17, 20], where the optimal hydrogen concentration was 3%–5%. In Ref. [8], the small-signal gain increased with increasing hydrogen concentration to 10%. The peak output power also increased by a factor of 1.6 (from 35 to 56 MW) with increasing hydrogen concentration from zero to 20%, and then decreased. The energy E_1 depended on the hydrogen concentration similarly to the peak output power and had a

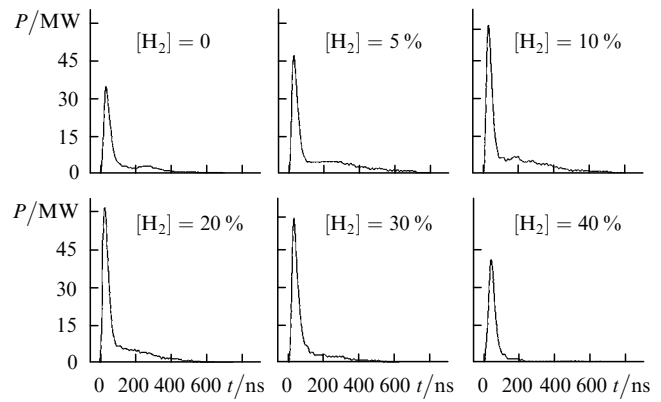


Figure 1. Oscilloscope traces of output laser pulses obtained with a gas mixture of the $4\text{CO}_2 - \text{N}_2 - \text{H}_2$ composition for different hydrogen concentrations and a total pressure $p = 0.7\text{ atm}$.

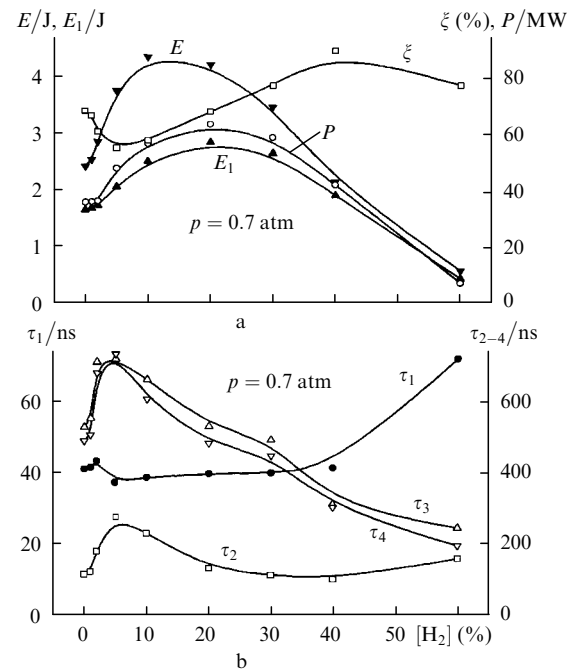


Figure 2. Experimental dependences of the energy (a) and time (b) parameters of output laser pulses on the hydrogen concentration in a gas mixture of the $4\text{CO}_2 - \text{N}_2 - \text{H}_2$ composition.

maximum at $[\text{H}_2] = 20\%$.

The parameter ξ shows the dependence of the ‘peak/tail’ energy distribution on the hydrogen concentration. One can see from Figs 1 and 2a that the increase in the pulse energy upon the addition of hydrogen (5%–10%) is accompanied by the increase in the nitrogen ‘tail’ intensity and the increase in its relative contribution to the pulse energy. However, a further increase in the hydrogen concentration up to 40% resulted in a significant suppression of the ‘tail’ accompanied by a weak decrease in the peak pulse power. This is due to the fact that large additions of hydrogen cause the deactivation of the upper laser level. In this case, the process described above favours the shaping of short high-power TEA CO_2 -laser pulses.

The pulse duration at half-maximum τ_1 (Fig. 2b) remained invariable ($\sim 40\text{ ns}$) in a broad range of hydrogen concentrations. As the hydrogen concentration was increased from zero to 5%, the total pulse durations (τ_3 and τ_4)

increased, achieved a maximum (~ 700 ns) and then decreased monotonically with increasing hydrogen concentration. The dependences of τ_3 and τ_4 on the hydrogen concentration are virtually identical. The increase in the total pulse duration upon small additions of hydrogen confirms the appearance of the nitrogen ‘tail’. The pulse width τ_2 , determined at a level of 0.1 of the peak power, also increased at first to achieve a maximum (~ 270 ns) at the hydrogen concentration of 5%, and then decreased to ~ 100 ns.

We also determined the effect of additions of hydrogen on the service life of the TEA CO₂ laser under study. The time variation of the average output power of the laser operated in the quasi-sealed-off regime with a single filling at a pulse repetition rate $f = 0.5$ Hz was investigated for all above-considered 4CO₂-N₂-H₂ gas mixtures for the optimal pressure $p = 0.7$ atm and the hydrogen concentration between zero and 60%. The maximum average output power was observed for the hydrogen concentration of 10%, which is consistent with the dependence of the energy of single output laser pulses (Fig. 2a). Fig. 3 shows the experimental dependences of the laser service life on the hydrogen concentration for a pressure $p = 0.7$ atm and a pulse repetition rate $f = 0.5$ Hz, which were determined from the oscilloscope traces of the average output laser power for different reductions of the output power. One can see that the laser service life is minimal for small additions of hydrogen (5%–10%).

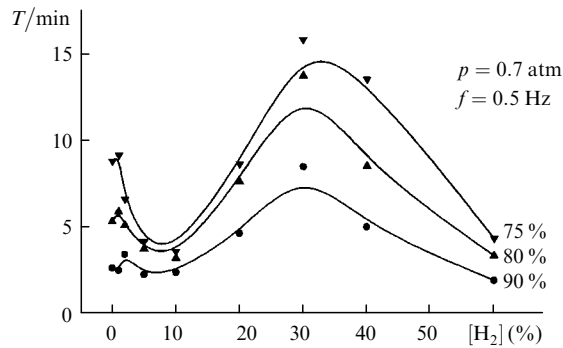


Figure 3. Experimental dependences of the laser service life T on the hydrogen concentration in the gas mixture with a composition 4CO₂-N₂-H₂ in the quasi-sealed-off regime, which were plotted for different output-power levels (75, 80, and 90%) relative to the initial output power.

The maximum laser service life was reached for the hydrogen concentration of 30% (the mixture composition was 56%CO₂-14%N₂-30%H₂). Note that this mixture composition at the optimal pressure $p = 0.7$ atm yielded output pulses with a total energy $E \approx 3.5$ J, a peak power $P = 58$ MW, an energy $E_1 = 2.6$ J, and $\xi \sim 80\%$ (Fig. 2a). For the mixtures investigated, this is close to the highest values of these parameters for pulses with $\tau_1 = 40$ ns and $\tau_2 = 109$ ns (Fig. 2b).

4. Effect of N₂ on the parameters of output laser pulses

It is known that the reduction of the relative nitrogen content in the gas mixtures of TEA CO₂ lasers lowers the total output pulse energy and shortens the pulse duration [21]. As the nitrogen content is increased (up to a certain limit),

the opposite effect is observed, primarily due to the nitrogen pulse ‘tail’.

Fig. 4 shows the experimental dependences of the energy and time parameters of the output laser pulses on the $[N_2]/[CO_2]$ partial-pressure ratio in the range from zero to 0.25 for CO₂-N₂-H₂ gas mixtures for a fixed hydrogen concentration (30%) and a fixed mixture pressure ($p = 0.7$ atm). As expected, the energy and the peak power of the output laser pulses increased with the relative nitrogen content, the peak power not reaching its maximum in the $[N_2]/[CO_2]$ -ratio variation range studied here. In our earlier work [19], the maximum of the peak output power of the laser with a nonselective cavity was observed for the mixture with a composition CO₂:N₂:H₂ = 1:1:0.1.

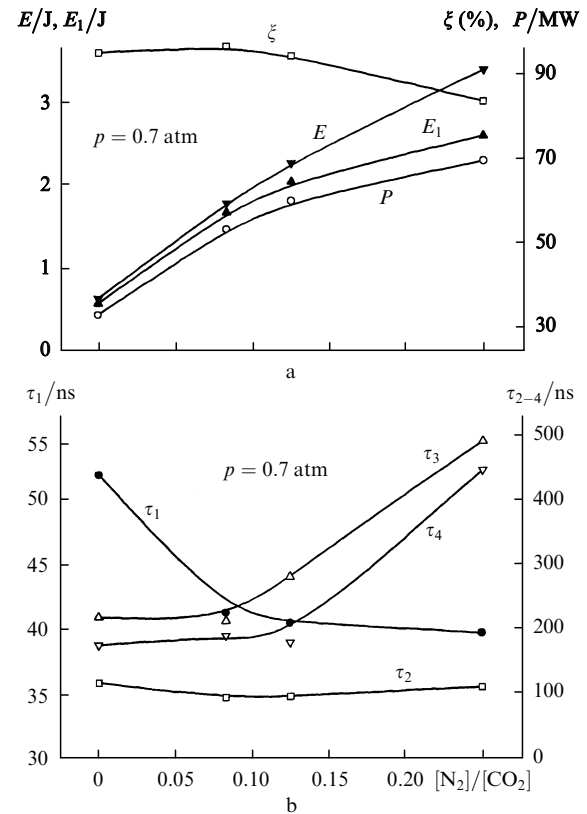


Figure 4. Experimental dependences of the energy (a) and time (b) parameters of output laser pulses, obtained with the use of the CO₂-N₂-30%H₂ gas mixture, on the $[N_2]/[CO_2]$ partial-pressure ratio.

One can see from Fig. 4 that, beginning with $[N_2]/[CO_2] = 0.125$, the total pulse energy increases much faster than the peak power and the energy E_1 , while the ratio ξ somewhat decreases, which is indicative of the appearance of a weak nitrogen ‘tail’. This is also confirmed by the increase in the total pulse durations τ_3 and τ_4 with increasing $[N_2]/[CO_2]$. The pulse duration τ_2 remained virtually constant in the range of variation of $[N_2]/[CO_2]$. The pulse duration τ_1 in the absence of nitrogen was ~ 52 ns; upon the addition of small amounts of nitrogen, it dropped sharply to 40 ns (by a factor of ~ 1.3).

In Ref. [5], a nitrogen-free gas mixture of the CO₂:H₂ = 2:1 composition proved to be optimal for maximising the energy and peak power of short output TEA CO₂-laser pulses without a nitrogen ‘tail’. In our experiment, this is close to the CO₂:H₂ = 7:3 mixture composition for

$[\text{N}_2]/[\text{CO}_2] = 0$. One can see from Fig. 4 that as the ratio $[\text{N}_2]/[\text{CO}_2]$ increases from zero to 0.125 upon the addition of nitrogen, the parameters ξ , τ_2 , τ_3 , and τ_4 determining the shape and duration of output pulses remain virtually invariable, while the duration τ_1 decreases. However, the pulse energy and peak power increase by factors of 3–3.5 even for so small an addition of nitrogen. Therefore, upon the shaping of short output TEA CO_2 -laser pulses with the use of $\text{CO}_2 - \text{H}_2$ mixtures, small additions of nitrogen for a partial-pressure ratio $[\text{CO}_2]/[\text{N}_2] \sim 10 - 12$ causes the shortening of the pulse duration at half-maximum, which is accompanied by a significant increase in the pulse energy and peak power without the formation of a nitrogen ‘tail’.

5. Effect of He additions on the parameters of output laser pulses

It is interesting to compare the above results obtained for helium-free gas mixtures with the laser-pulse parameters observed upon the addition of helium to the optimal gas mixture. At this stage of research, the laser emitted the TEM_{00} mode in the $9P(20)$ line when the diameter of the intracavity aperture was 10 mm. The discharge laser cell was filled with a gas mixture of the $\text{CO}_2 : \text{N}_2 : \text{H}_2 = 56\% : 14\% : 30\%$ composition at a pressure $p_0 = 0.5$ or 0.7 atm; helium was added to the mixture in portions till the total pressure amounted to $p = 1.3$ atm.

Fig. 5 shows the dependences of the energy and time parameters of output laser pulses on the total mixture pressure p raised by additions of He to the above four-component gas mixture at a constant partial pressure of the helium-free mixture $p_0 = 0.5$ atm. Raising the pressure of the gas mixture by adding helium resulted in a reduction of the total pulse durations τ_3 and τ_4 (by 50%) and in some increase of ξ . However, in this case, the pulse duration τ_1 increased from 43 to 54 ns (by 25%). This can be caused by the decrease in the gain in the active medium due to the spectral line broadening with increasing the total pressure upon the addition of He to the laser mixture optimised in the hydrogen content.

A similar picture was observed earlier [19], when the output pulses of the above laser with the use of the $\text{CO}_2 : \text{N}_2 : \text{H}_2 = 1:1:0.1$ mixture had not only shorter duration at half-maximum than with the use of the $\text{CO}_2 : \text{N}_2 : \text{He} = 1:1:3$ mixture, but also had higher energy and peak power. A significant difference in pulse durations at half-maximum was also observed in Ref. [6]: about 50 ns for a $\text{CO}_2 - \text{H}_2$ mixture and 75 ns for a $\text{CO}_2 - \text{He}$ (the component density ratio was not specified in Ref. [6]). Along with the results of our latest measurements, all these experimental data confirm the fact that helium-free TEA CO_2 -laser gas mixtures with additions of hydrogen produce output pulses of shorter durations than helium-containing gas mixtures.

The increase in the gas mixture pressure upon the addition of helium to the initial four-component mixture resulted in a reduction of the output pulse energy and peak power in our experiment (Fig. 5a), though the maximum P and E_1 values were observed for a total pressure $p = 0.7$ atm. Note that for an initial partial mixture pressure $p_0 = 0.7$ atm, the maximum values of P and E_1 were also observed at a total pressure $p = 0.7$ atm (i.e., in a helium-free mixture), which is optimal for the above mixtures and the laser described here.

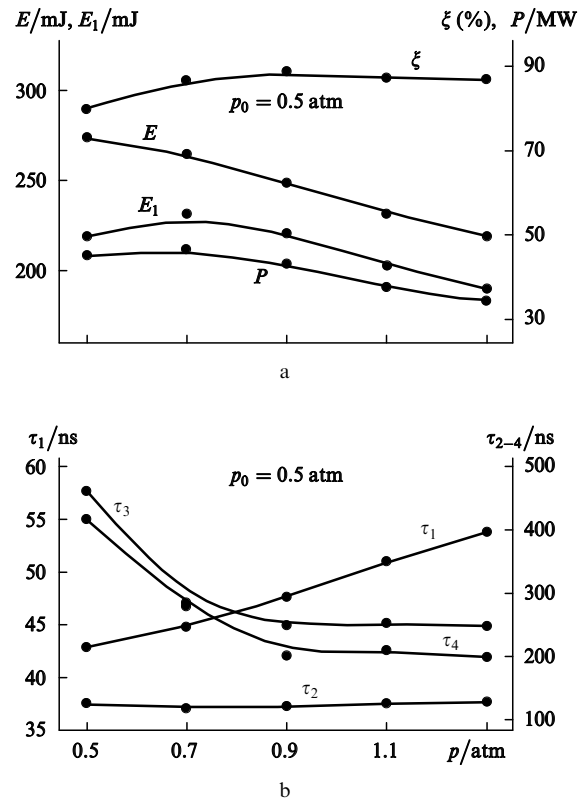


Figure 5. Experimental dependences of the energy (a) and time (b) parameters of output laser pulses on the total pressure p obtained with the use of the four-component ($\text{CO}_2 : \text{N}_2 : \text{H}_2$):He = (56 : 14 : 30) : [He] gas mixture.

6. Conclusions

The following conclusions can be drawn from the experimental results obtained in our work. The addition of hydrogen to the molecular gas working mixture of the $\text{CO}_2 : \text{N}_2 = 4 : 1$ composition results in a significant increase in the energy and peak power of output TEA CO_2 -laser pulses. The maximum output pulse energy, peak power, and laser service life in the quasi-sealed-off regime were observed for the H_2 contents of 10, 20, and 30%, respectively. The minimal total pulse duration and the maximum fraction of energy in the leading peak were observed for the H_2 concentration of 40%. The use of $\text{CO}_2 - \text{N}_2 - \text{H}_2$ gas mixtures with a high density of hydrogen (30%–40%) results in a significant suppression of the ‘tail’ of output pulses.

Small additions of nitrogen to the $\text{CO}_2 - \text{H}_2$ gas mixture cause the shortening of the pulse width at half-maximum accompanied by a significant increase in the energy and peak pulse power without formation of the nitrogen ‘tail’. Upon the addition of helium to the optimal $\text{CO}_2 - \text{N}_2 - \text{H}_2$ gas mixture, the energy and peak power of output laser pulses decreased and their duration at half-maximum increased.

The helium-free $\text{CO}_2 : \text{N}_2 : \text{H}_2 = 56\% : 14\% : 30\%$ gas mixture at a pressure of 0.7 atm is optimal, according to a set of its parameters, for the production of shortened laser pulses with a high energy, high peak power, and long service life of the TEA CO_2 laser outlined above.

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