

# Amplification of multi-gigawatt 3 ps pulses in an atmospheric CO<sub>2</sub> laser using ac Stark effect

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**Abstract:** The 3 ps pulses are amplified to ~20 GW peak power in a TEA CO<sub>2</sub> laser using ac Stark broadening. Demonstration of such broadband coherent amplification of 10 μm pulses opens opportunities for a powerful mid-IR source at a high-repetition rate.

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**OCIS codes:** (140.3470) Lasers, carbon dioxide; (190.5940) Self-action effects; (020.6580) Stark effect.

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## 1. Introduction

Progress on chirped-pulse amplification (CPA) in large bandwidth gain media has made possible to achieve multi-TW powers in ultra-short pulses around  $\lambda \sim 1 \mu\text{m}$  at a high-repetition rate [1]. Availability of such intense solid-state lasers has resulted in the development of new compact sources of X-rays and charged particles [2]. The maximum laser energy transferred to an electron in the laser field is determined by the ponderomotive potential, which scales as  $I\lambda^2$ , where  $I$  is the laser intensity. This relationship implies certain advantages of longer wavelength lasers for X-ray production via HHG [3] and laser particle acceleration and provides a strong motivation to develop TW-class lasers in the mid-IR range. As opposed to a solid state medium, a CO<sub>2</sub> gas laser -the only viable candidate for efficient generation of high-power mid-IR pulses- has a very high damage threshold (limited only by gas ionization at  $\geq 10^{12} \text{ W/cm}^2$ ) and, therefore can in principle be used without any CPA arrangement. However, amplification of a picosecond pulse in a CO<sub>2</sub> laser is complicated by the relatively narrow bandwidth of the CO<sub>2</sub> molecule for which the gain spectrum consists of discrete rotational lines. When the bandwidth of these lines is sufficiently broadened, they overlap filling the gaps in the spectrum that results in a quasi-continuous bandwidth across the branch ( $\sim 1.2 \text{ THz}$ ) suitable for amplification of  $\geq 1 \text{ ps}$  pulses. Thus 1-5 ps pulses can be amplified in high-pressure ( $\geq 10 \text{ atm}$ ) CO<sub>2</sub> lasers, when a collisionally broadened linewidth becomes approximately 30 GHz [4,5]. Unfortunately, technically it is extremely difficult to obtain a uniform glow discharge in a large aperture ( $>1 \text{ cm}$ ) module at a high pressure ( $\sim 10 \text{ atm}$ ). As a result output of a multiatmosphere CO<sub>2</sub> amplifier is limited because of small volume and the systems typically operate in a single-shot regime.

It has been long recognized that coherent effects, that appear when an active medium is illuminated by an intense pulse with a duration short compared to its relaxation time, play an important role in the dynamics of laser amplifiers [6–8]. In one of these mechanisms, the presence of a large electrical field associated with radiation connecting two energy levels of a molecule results in field broadening of the linewidth due to ac Stark effect [7,9,10]. Earlier we showed experimentally the demonstration of field broadening as an alternative mechanism to collisional broadening that allowed for generation of 40 ps pulses in a 2.5 atm CO<sub>2</sub> module [11]. The same mechanism has played a major role in providing the bandwidth necessary for recent picosecond pulse amplification to 15 TW level [12]. Here we show for the first time truly broadband amplification of 3 ps pulses in an atmospheric CO<sub>2</sub> laser when the bandwidth is provided not by collisions but predominately by the transient laser field itself via the ac Stark effect. The peak output power in a 1 Hz TEA CO<sub>2</sub> amplifier reached  $\sim 20 \text{ GW}$  in our present work.

## 2. Simulations of picosecond pulse amplification in an atmospheric CO<sub>2</sub> laser

Conventional TEA CO<sub>2</sub> lasers have a collisional linewidth of  $\Delta\nu_P \sim 3.5 \text{ GHz/atm}$  for an individual rotational line and the resulting gain spectrum is a comb of discrete lines separated by 55 GHz for the 10P-branch. This bandwidth limits the pulse length to  $\sim 1 \text{ ns}$ . The self-effect of electric field of the laser on the linewidth (field broadening) can be estimated using the Rabi frequency ( $\nu_R$ ), as  $\Delta\nu_F \approx \nu_R = 1.38 \times 10^7 \mu \sqrt{I}$ , where  $\mu$  is the CO<sub>2</sub> transition dipole moment in Debye, and  $I$  is the laser intensity in  $\text{W/cm}^2$  [10]. For the 10.6  $\mu\text{m}$  lasing transition (the 10P(20) line), the dipole moment is equal to 0.0275 D [13]. Therefore, the CO<sub>2</sub> laser rotational linewidth,  $\Delta\nu$  is a product of pressure broadening and the field broadening which

can be estimated by the following equation:  $\Delta\nu = \Delta\nu_p + \Delta\nu_F$ . Then a 10 $\mu\text{m}$  pulse with an intensity of 5 GW/cm<sup>2</sup> propagating in a 1 atm CO<sub>2</sub> amplifier will interact with a bandwidth equal to 30.3 GHz which is comparable to that obtained in the 10 atm amplifier. Therefore, a sufficiently intense pulse can generate the broad bandwidth in low pressure amplifiers opening the way for picosecond pulse amplification.

Regardless of line broadening mechanism, insufficient overlapping of these lines results in a residual modulation of the gain spectrum at 55GHz even at  $\Delta\nu \sim 30\text{-}40\text{GHz}$  [12]. When a short 3ps pulse propagates in an amplifying medium with a periodically modulated gain spectrum, some frequencies in the pulse bandwidth will not be amplified efficiently and the inverse Fourier transform for such a case results in a pulse train with a pulse separation equal to 1/55GHz, or 18.5ps. To study the effect of field broadening on short pulse amplification where the bandwidth is continually increasing with the 10  $\mu\text{m}$  field intensity, we have modeled amplification for the train of 3 ps pulses typically recorded in the present experiment [12]. In Fig. 1 we present the results of modeling of such a pulse train propagating in a TEA CO<sub>2</sub> amplifier using a density matrix code written by V. Platonenko [14]. In simulations we have fixed the  $g_0L$  product to be equal to 9, where  $g_0$  is the small signal gain and L is the amplification length and varied the level of the seeded pulse intensity in the range of 0.5-75 GW/cm<sup>2</sup>. Simulations confirmed that field broadened gain profile can support the 330 GHz bandwidth required for amplification of a 3 ps pulse. At a low intensity of the seed pulse of 0.5 GW/cm<sup>2</sup> (Fig. 1(a)), when  $\Delta\nu = 12$  GHz, the pulse train envelope is significantly broadened as shown in Fig. 1(b). By increasing I for a seed pulse from 0.5 to 6.5 GW/cm<sup>2</sup>, the pulse train envelope width shrinks by a factor of two. This shortening of the pulse train becomes more profound for an even higher intensity of 75 GW/cm<sup>2</sup> (see Fig. 1(f)), when the envelope duration is almost the same as that for the seed pulse train. However, this comes at a price of drop in a total gain due to strong saturation of the gain medium: the total net gain is 24, 10 and 3 for 0.5, 6.5, and 75 GW/cm<sup>2</sup>, respectively.

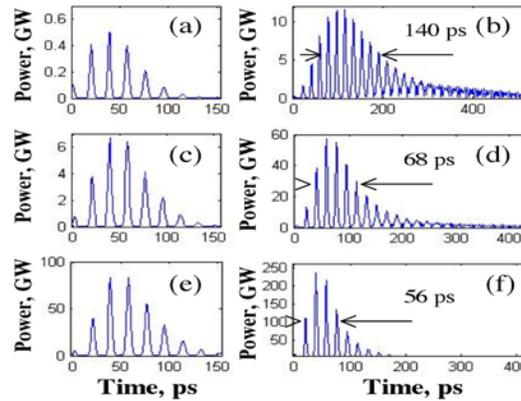


Fig. 1. Temporal profiles of the seed pulses with a peak intensity of 0.5 GW/cm<sup>2</sup> (a), 6.5 GW/cm<sup>2</sup> (c), and 75 GW/cm<sup>2</sup> (e) and the simulated pulses after amplification in a 3-m long TEA CO<sub>2</sub> amplifier with a small-signal gain of 3 m<sup>-1</sup> (b), (d) and (f), respectively.

For a picosecond pulse amplified in a CO<sub>2</sub> gain medium, the individual pulse width is limited by the bandwidth of the entire branch ( $\sim 1.2\text{THz}$ ) and the length of the pulse train envelope is limited by the bandwidth of the rotational line. According to theory [15], gain narrowing in an amplifier reduces the spectral width, and the broadening of the pulse train envelope can be estimated as:

$$\tau_p^2(z) = \tau_p^2(0) + (16 \ln 2) \ln G / \Delta\omega^2 \quad (1)$$

where  $\tau_p(0)$  and  $\tau_p(z)$  are the initial and final FWHM widths of the pulse train envelope,  $G$  is the total gain, and  $\Delta\omega$  is the gain bandwidth. For the  $0.5 \text{ GW/cm}^2$  case,  $\Delta\omega/2\pi = 12 \text{ GHz}$ ,  $\tau_p(0) = 50 \text{ ps}$  and  $G = 24$ , the expected gain narrowed pulse train envelope is  $\sim 122 \text{ ps}$  (FWHM). For the same initial width of the pulse train, the calculated value of the envelope width decreases to  $77 \text{ ps}$  (FWHM) with an increase of intensity to  $6.5 \text{ GW/cm}^2$ . Both these estimated values are in a good agreement with  $140 \text{ ps}$  and  $68 \text{ ps}$  (FWHM) deduced from simulated pulse trains in Fig. 1(b) and Fig. 1(d), respectively.

### 3. Experiments

Experiments have been carried out using the master oscillator–power amplifier (MOPA)  $\text{CO}_2$  laser system at the UCLA Neptune Laboratory which was recently upgraded to generate  $3 \text{ ps}$  pulses [12]. The main strategy behind the high-power picosecond  $\text{CO}_2$  MOPA system is to have two steps in amplification process. First, amplification of a weak seed pulse from nJ to mJ level in a multiatmospheric  $\text{CO}_2$  laser in which the bandwidth is broadened collisionally, and second, amplification of the mJ pulses to the Joule level in relatively low-pressure discharge modules. In the latter the field broadening mechanisms plays a major role in producing the bandwidth.

#### 3.1 Experimental set-up

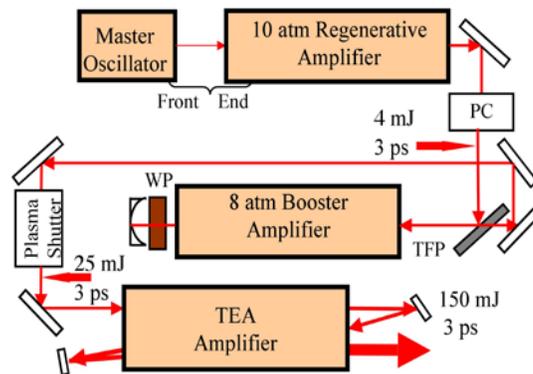


Fig. 2. A simplified scheme of 1 Hz  $\text{CO}_2$  laser MOPA chain: PC, CdTe Pockels cell; WP, half-wave plate; TFP, thin film polarizer.

Figure 2 shows the Neptune MOPA chain schematically. The front end of the  $\text{CO}_2$  laser chain includes a hybrid TEA master oscillator and a UV preionized 10 atm regenerative amplifier. The first stage of the laser system involves the production of a short  $10 \mu\text{m}$  seed pulse utilizing a  $\text{CS}_2$  Kerr modulator controlled by a  $3 \text{ ps}$   $1 \mu\text{m}$  pulse from a Nd:glass laser [16]. The second stage of the laser system is the amplification of the  $3 \text{ ps}$  seed pulse in a 10 atm TE  $\text{CO}_2$  regenerative amplifier having a gain volume of  $1 \times 1 \times 60 \text{ cm}^3$ . The total gain of  $\sim 10^7$  brings the  $\sim \text{nJ}$  seed pulse to  $\sim 10 \text{ mJ}$  level. Since injection mode-locking technique is used for this stage, an external CdTe Pockels cell selects a single pulse for further amplification. As expected, a  $3 \text{ ps}$  pulse train has been typically recorded in the output and its careful characterization is reported elsewhere [12]. Then, the  $4 \text{ mJ}$  pulse is amplified to  $\sim 40 \text{ mJ}$  in a 2-pass  $\text{CO}_2$  Booster amplifier which operates at 8 atm. Finally, this pulse is sent through a TEA  $\text{CO}_2$  amplifier (Lasermark 960) with a relatively large aperture ( $3 \times 4 \text{ cm}^2$ ) and a single-pass length of 1 m. The entire laser system operates at a repetition rate of 1 Hz.

### 3.1 Results on 3 ps pulse amplification at 1 atmosphere

A typical pulse profile of the Booster amplifier output is presented in Fig. 3(a). For this purpose a laser diode is gated in a nonlinear medium by the CO<sub>2</sub> laser pulse, allowing upconversion of the 10- $\mu\text{m}$  signal to the visible range so that the pulse duration can be measured with a streak camera (Hamamatsu C5680). Such a pulse train is sent through the TEA module for three-passes of amplification. The amplifier produces a small-signal gain of  $\sim 2\%/cm$  on the 10P(20) line. The CO<sub>2</sub> molecule linewidth  $\Delta\nu$  at the seed peak intensity of  $6.5 \text{ GW/cm}^2$  equals to  $\sim 34\text{GHz}$ . Note that the collisional linewidth in this laser is only  $\sim 3.5\text{GHz}$ . The 10- $\mu\text{m}$  beam is slowly expanding for an efficient energy extraction for the first two passes. For the third pass, the beam is converging to ensure the high field broadening and facilitate spatial separation of beams without inducing self-lasing. It is important that such arrangement causes transformation of initially Gaussian beam into a quasi-flattop beam with a constant intensity (field broadening) due to the strong gain saturation. Figure 3(b) shows a typical measured pulse profile after three passes of coherent amplification in the TEA module. The initially 140 ps FWHM pulse train during the amplification has slightly broadened by  $\sim 20$  ps. By applying Eq. (1) to the change in the envelope FWHM, one can extract an effective laser bandwidth of an individual rotational line averaged over the entire amplification length. The deduced bandwidth of  $\Delta\nu \sim 50 \text{ GHz}$  corresponds to a field broadening achievable at  $\sim 15 \text{ GW/cm}^2$ . It should be noted that decrease of the seed intensity by a factor of 4.7 resulted in a significant increase in number of 3ps pulses in a train ( $\sim 220$  ps FWHM). Extensive measurements demonstrated almost constant extracted energy for pulses with fluctuating envelope duration indicating possibility to increase power gain, if the number of pulses in a pulse train seeded in the TEA CO<sub>2</sub> amplifier can be reduced.

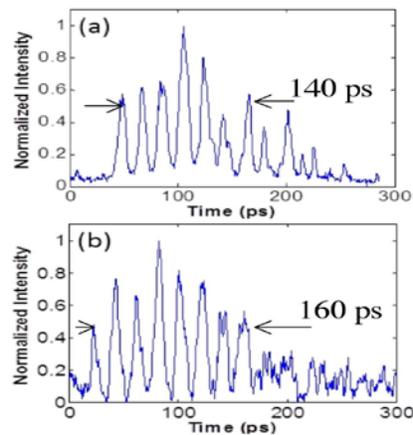


Fig. 3. Temporal profile of the 10  $\mu\text{m}$  3 ps pulses after amplification in the 8 atm Booster amplifier (a), 1 atm 3-pass CO<sub>2</sub> amplifier at an input intensity  $6.5 \text{ GW/cm}^2$  (b) as measured by the streak camera.

### 3.3 Amplification of picosecond pulses shortened by a plasma shutter

It is known that a rapidly expanding plasma of the optical breakdown in gases can screen an optical pulse effectively when the plasma reaches the critical density, which is equal to  $10^{19} \text{ cm}^{-3}$  for 10- $\mu\text{m}$  light. This technique has been applied successfully for truncating CO<sub>2</sub> laser pulses on the nanosecond [17, 18] and sub-nanosecond [11,19] time scales. In the present experiment, a plasma shutter cell is installed in the focus of a telescope before the TEA CO<sub>2</sub> amplifier. For the  $f/50$  focusing of the laser beam (spot size, 500  $\mu\text{m}$ ), the peak laser intensity

reaches  $1.7 \text{ TW/cm}^2$ . The threshold of the breakdown in the air for 3 ps 10- $\mu\text{m}$  pulses, as observed by a visible spark formation, is  $0.8 \text{ TW/cm}^2$ . The major process that leads to ionization is cascade ionization by electrons that have gained energy directly from laser field during collisions with neutral particles. In order to decrease this avalanche ionization threshold even further and to improve shot-to-shot reproducibility, the plasma shutter was filled with 3 atm of  $\text{N}_2$ . In this case the plasma screening reduced the seed pulse energy by  $\sim 30\%$ . Low losses in a laser-plasma for conditions when the observed breakdown threshold ( $\sim 0.25 \text{ TW/cm}^2$ ) is surpassed earlier during the pulse train can be attributed with low plasma densities achieved in avalanche ionization in the picosecond regime [20]. This observation points to refraction of the laser beam as the major mechanism of optical losses in a plasma with densities below the critical density [11]. Temporal profile of a truncated pulse after three passes of amplification is shown in Fig. 4(a). Its analysis reveals that the duration of the pulse train envelope is three times shorter than that obtained without the plasma shutter (see Fig. 3(b)).

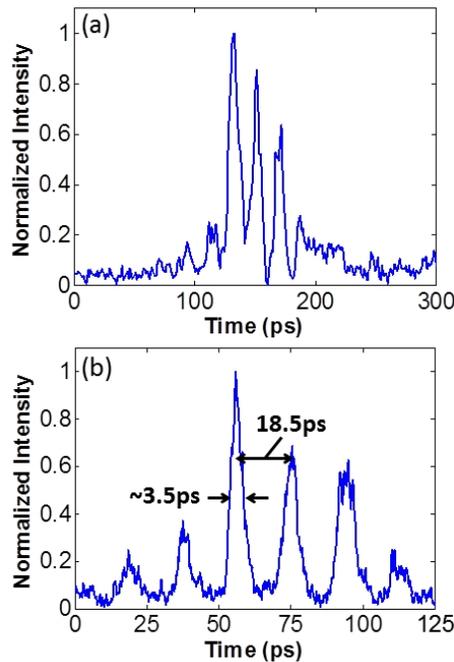


Fig. 4. Temporal profile of the 10  $\mu\text{m}$  pulses truncated in the plasma shutter after three passes of amplification in the TEA  $\text{CO}_2$  module measured by a streak camera at a speed of 100 ps/mm (a) and 20 ps/mm (b).

Figure 4(b) displays a typical pulse train recorded with the optical breakdown in plasma shutter at a maximum speed of the streak camera. The expected individual pulsewidth of 3 ps is near the resolution limit of the streak camera, therefore its measurement must take into account the instrumental function of the device. The latter is equal to 3 ps as measured using a sub-picosecond 532 nm glass laser pulse. Then the measured pulse width  $t_{\text{meas}}^2 = t_{\text{pulse}}^2 + t_{\text{instr}}^2$  is a product of the measured pulsewidth,  $t_{\text{pulse}}$  and the temporal resolution of the instrument,  $t_{\text{instr}}$ . The experimentally measured pulsewidth of 3.5 ps FWHM confirms that sufficient bandwidth for 3 ps pulses can be self-generated by strong laser fields in coherent amplification regime. The final energy in the pulse train in Fig. 4(b) reaches 150 mJ resulting in  $\sim 20 \text{ GW}$  peak power for the most powerful pulse in the pulse train. The total gain in the amplifier is around 10, after accounting for polarization losses. The laser intensity in the output beam reached  $70 \text{ GW/cm}^2$ . Further amplification of such an intense pulse, as

demonstrated by modeling in Fig. 1(e), 1(f) for the  $I = 75 \text{ GW/cm}^2$  case, will result in a much smaller net gain due to strong saturation of the  $\text{CO}_2$  transitions. However, reaching  $\sim 0.2 \text{ TW/cm}^2$  power predicted in simulations for extra 3 meters of coherent amplification can open possibility to generate a TW power  $10 \mu\text{m}$  pulses in a large-aperture TEA  $\text{CO}_2$  module.

#### **4. Summary**

We have demonstrated coherent amplification of  $10 \mu\text{m}$  3 ps pulses in an atmospheric  $\text{CO}_2$  laser in which the bandwidth is determined by strong field broadening of the lasing transition. Sufficient bandwidth for sustaining picosecond pulses achieved in a low-pressure  $\text{CO}_2$  active medium proves experimentally that incomplete overlap of the rotational lines is not a fundamental limitation for amplifying high-power short pulses in gases. This ac Stark based amplification regime can be applied to other gas lasers e.g. CO,  $\text{N}_2\text{O}$ , where the dipole moment of lasing transitions is much larger. It is shown that a plasma shutter can shorten the pulse train envelope prior to the amplifier allowing for a significant increase in a peak power gained in a TEA  $\text{CO}_2$  module.

It is important to note that TEA  $\text{CO}_2$  lasers, widely used for industrial applications, are capable of generating 1-100 J at a repetition rate of 10-1000 Hz. Therefore multi-GW mid-IR source running at 1 Hz can be scaled to 0.1-1TW power level and higher pulse repetition rate opening numerous applications. For example, recently a train of 3 ps pulses was successfully applied for acceleration of monoenergetic proton beams in an  $\text{H}_2$  gas plasma [21].

#### **Acknowledgments**

This work was supported by U.S. department of Energy grant DE-FG03-92ER40727.