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Picosecond CO2 Laser Filamentation in Air

Chan Joshi UNIVERSITY OF CALIFORNIA LOS ANGELES

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## ABSTRACT

Very-long-range guiding and distant projection of high-energy laser pulses and highaverage power beams are of interest for many potential DoD applications. Earth's atmospheric trasmittance favors the long-wavelength infrared (LWIR) window from 8 to 14  $\mu$ m, however diffraction proportional to the wavelength results in strong beam spreading in space in this spectral range. We have performed proof-of-concept experiments on nonlinear filamentation at ~10  $\mu$ m by propagating ~1TW 3 ps CO<sub>2</sub> laser pulses in laboratory air. The main experimental observation is that an intense CO<sub>2</sub> laser beam is self-guided in a macroscopic single filament of  $\leq$ 1 cm diameter over many Rayleigh lengths. We call such a single filament a *megafilament* because it confines an ~1TW laser pulse with several Joules of energy and its cross-section is 10<sup>4</sup> times larger than a typical near-IR single filament. In this study we demonstrated self-guiding of an ~1TW CO<sub>2</sub> laser pulse in air over at least 32 meters (~20 Rayleigh ranges).

We have measured the temporal dynamics of the laser pulse self-guided in air. It was found that an initial 10  $\mu$ m macropulse consisted of a leading ~1 TW picosecond pulse and one or more weaker postpulses is modified during nonlinear propagation in the atmosphere. The leading self-channeled pulse is shortened from 3.5 ps to 1.8 ps and the postpulses separated by 25 ps are strongly screened by plasma produced via the avalanche ionization mechanism. Taking into account such pulse shortening, the clamped intensity in the *megafilament* was measured to be ~1TW/cm<sup>2</sup> much smaller that the tunnel ionization threshold of O<sub>2</sub>/N<sub>2</sub>.

By comparing these results with simulations, we find that in addition to Kerr self-focusing and diffraction, the self-channeling of LWIR pulses with an intensity of 1 TW/cm<sup>2</sup> can only be explained by considering plasma formation below the tunnel ionization threshold due to many-body Coulomb induced ionization in air. This idealized theoretical microscopic description of air as a homogenous atmospheric pressure gas allowed to reproduce main experimental observables.

We also conducted the first interferometric measurements of atmospheric plasmas produced by a filamented multiterawatt picosecond  $CO_2$  laser beam in air and found two distinct regimes of ionization. For  $\geq 10$  TW/cm<sup>2</sup> laser intensities, plasma dynamics is dominated by growth of a few discrete ~100 µm diameter plasmas formed on aerosol particles as well as self-focusing of multifilamented laser beam. Avalanche ionization seeded by these processes results in strong attenuation of postpulses. For a smaller intensity around 1TW/cm<sup>2</sup> and a single picosecond pulse, avalanche ionization seems to be limited to low plasma densities below 10<sup>15</sup> cm<sup>-3</sup> sensitivity of our interferometer.

The results of research supported by this grant were published in 8 papers in refereed journals, 1 chapter in the Book and also were reported at 12 National and International conferences.

**Project Title:** Picosecond CO<sub>2</sub> Laser Filamentation in Air , PI Prof. Chan Joshi, co-PI Sergei Tochitsky, AFOSR Grant Number FA9550-16-1-0139

**Final report.** Very-long-range guiding and distant projection of high-energy laser pulses and high-average power beams are of interest for many potential DoD applications. Low-loss propagation of radiation in air over long distances is naturally limited to few transparency windows in the mid-IR range of spectrum from 3-14  $\mu$ m. The main goal of this program is to perform proof-of-concept experiments on nonlinear filamentation at 10  $\mu$ m by propagating TW 3 ps CO<sub>2</sub> laser pulses in laboratory air.

In particular, four years ago we have proposed to investigate the following:

- 1. Measure nonlinear Kerr coefficient  $\chi_3$  of air and air constituencies at 10  $\mu$ m using sideband generation by a 10 GW/cm<sup>2</sup> CO<sub>2</sub> laser beatwave guided in a gas filled hollow waveguide. Determine contribution of H<sub>2</sub>O at to the nonlinear refractive index of air.
- 2. Study nonlinear self-focusing of 3 ps multi-TW CO<sub>2</sub> laser pulses in laboratory air with a beam propagation distance of 20-30 m.
- 3. Conduct measurements of spatial, spectral and temporal characteristics of a high-power CO<sub>2</sub> laser beam self-guided in air.
- 4. Investigate laser-plasma parameters for an ionized air using time resolved green pulse interferometry and other diagnostics.
- 5. Model atmospheric propagation of high-peak power picosecond CO<sub>2</sub> laser pulses and compare results of numerical simulations with experiments.

In the following report we will show that practically all proposed tasks have been successfully accomplished. It should be noted that numerical modeling of nonlinear propagation has been conducted by the University of Arizona group of Prof. J. Moloney (funded by AFOSR separately) in close collaboration with the UCLA group.

**Measurements of n<sub>2</sub> in air.** As a natural first step in our program on picosecond CO<sub>2</sub> laser filamentation in air, in Y1 we have made measurements of Kerr coefficients of air and major air constituents around 10  $\mu$ m [1,2]. We have used collinear four-wave mixing (FWM) in gases of 200 ps long beat-wave obtained by our CO<sub>2</sub> laser operating on two lines. The master oscillator-power amplifier CO<sub>2</sub> laser system at the UCLA Neptune laboratory is capable of producing 3-200 ps long pulses with a high-peak power at a pulse repetition frequency of 1 Hz. For this experiment, a beat-wave was produced by combining radiation from the 10P(20) (10.59  $\mu$ m) and 10R(16) (10.27  $\mu$ m) CO<sub>2</sub> lines. The laser beam was focused in a ~2 m long gas filled cell. Generated in the gas sidebands were frequency resolved by a monochromator and analyzed by a HgCdTe detector. The set-up allowed for the detection of FWM signals that were 10<sup>-7</sup> relative to the pump.

To avoid the ionization contribution, manifested by drop in a FWM signal at pressures above 630 Torr, all  $n_2$  measurements were performed in high purity gases at pressures less or equal to 380 torr. The measured values for an effective nonlinear coefficients scaled to an atmospheric pressure are 4.5 for N<sub>2</sub>, 8.4 for O<sub>2</sub> and 5.0 for dry air (in units of  $10^{-19}$  cm<sup>2</sup>/W). Since the magnitude of the electronic nonlinear response is expected to decrease for longer wavelengths, these measurements indicate the importance of the delayed molecular nonlinearity at long wavelengths. The asymmetry we have observed in the FWM spectrum in Fig.1 indeed points to the fact that the molecular response plays an important role. To study this further we have performed FWM measurements in Ar, Kr and Xe, gases that have a comparable

electronic nonlinearity as air but that lack a molecular response. Measurements of FWM in Kr and Xe produced a comparable FWM spectrum as that produced in air, however the FWM sidebands were symmetric and were generated with lower yield [3,4]. These measurements represent the first study of Kerr nonlinearity of molecular and atomic gases at long-wavelength infrared and a dedicated paper was published in a Phys. Rev. A [5]. In Table 1 we summarize results of our n<sub>2</sub> measurements in both molecular gases  $O_2$ ,  $N_2$  and air. Measurements of the effective nonlinear index at 10  $\mu$ m are comparable with values measured at 0.8 and 2.4  $\mu$ m.



*Figure 1. FWM* spectrum obtained in laboratory air at an intensity of ~15 GW/cm<sup>2</sup>.

		J	
Gas Species	$\lambda = 0.8 \ \mu m \ [16]$	$\lambda = 2.4 \ \mu m \ [18]$	$\lambda = 10.6 \ \mu m$
	$n_{2,eff}(10^{-19}  cm^2/W)$	$n_{2,eff}(10^{-19}  cm^2/W)$	$n_{2,eff}(10^{-19}  cm^2/W)$
$N_2$	$3\pm0.7$	$3.2\pm0.5^{b}$	$4.5\pm0.9$
O2	$8\pm2.2$	$6.4\pm0.8^{b}$	$8.4 \pm 1.3$
Air	$4 \pm 1.1^{a}$	-	$5.0 \pm 0.9$

Table 1: Effective nonlinear refractive indices for major air constituents scaled to 1 atm

<sup>a</sup>Calculated from measurements of N<sub>2</sub>, O<sub>2</sub> and Ar.

<sup>b</sup>Calculated from measurements in S. Zahedpour, *et. al.* using  $n_{2,eff}=n_2 + n_{2,rot}[18]$ .

Our measurements of nonlinear refractive index of atomic and molecular gases at 10  $\mu$ m using fourwave mixing [2,5] gave a value of the Kerr coefficient of air as 5±0.9 cm<sup>2</sup>/W. This corresponds to the critical power for self-focusing (P<sub>cr</sub>) in air in the range of 300-400 GW. It should be noted, however, that the value of the critical power for self-focusing based on the measured n<sub>2,eff</sub> will change for the much shorter ~3 ps pulses considered for laser filamentation in air. The main unknown here is related to a complicated interplay between the instantaneous and delayed contributions to the molecular response related to long-wavelength laser-matter interactions.

While upgrading the UCLA high-power CO<sub>2</sub> laser system to generate a quasi-single ~3 ps pulse, we decided to stage initial experiments using a ~1 TW power (P~2-3P<sub>cr</sub>) CO<sub>2</sub> laser system at Brookhaven National Laboratory (BNL). At BNL due to the use of carbon dioxide isotopes, the laser pulse consists of a leading TW pulse and one or more low-power postpulses separated by a 25 ps period, with the total pulse energy up to ~5.2 J contained in a 5 cm diameter beam. Based on direct measurements of n<sub>2</sub> of air via analysis of a nonlinear self-focusing of a ~3 ps, terawatt power beam of the ATF BNL CO<sub>2</sub> laser

system in air described below and theoretical modeling the atmospheric propagation the 10 micron nonlinear refractive index of air is close to  $\sim 4 \times 10^{-19} \text{ cm}^2/\text{W}$  [6-8].

**Self-guiding of TW picosecond CO<sub>2</sub> laser in air.** The main results in the Y2-Y3 AFOSR program came from a detailed study of spatial, temporal and spectral characteristics of a terawatt  $CO_2$  laser beam self-guided in air performed at BNL using an experimental set-up in Fig.2.



**Figure 2.** Experimental set-up. The TW CO<sub>2</sub> laser beam was focused by a mirror, M(f'=11m) in air with a focus located at z=0 and intercepted at a distance z=2 to 32 m by a pair of NaCl wedges, W each of which produce two reflected beams with ~4% of the incident energy. Two energy meters E1 and E2 were used to measure energy before and after propagation, respectively. These beams were analyzed by a pyroelectric array of a Pyrocamera and other diagnostics. A grating spectrometer could be replaced by odd-harmonic diagnostic comprised of broadband pass filters (BBPF) centered at  $3\omega-3.41 \mu m$ ,  $5\omega-2.04 \mu m$ , or  $7\omega-1.46 \mu m$  of the CO<sub>2</sub> laser and either a calorimeter detector, Det. for  $3\omega$  or a Golay cell for  $5\omega$  and  $7\omega$  measurements. Alternatively the CO<sub>2</sub> laser pulse was sent to a streak camera or a single-shot autocorrelator for pulse profile measurements. Right hand panel describes the visualization technique via sum frequency mixing of a red diode laser and 10  $\mu$ m beams in a AgGaS<sub>2</sub> nonlinear crystal (AGS). The typical measured CO<sub>2</sub> laser pulse consisted of a leading ~TW power picosecond pulse and postpulses separated from the first pulse by a 25 ps period.

As shown in Fig. 2, the laser beam after the focus was intercepted by a pair of NaCl wedges that allowed for simultaneous measurements of the spatial and temporal or spectral properties of the beam during its propagation in air by using a two-dimensional IR array and a streak camera (or an autocorrelator) or a grating spectrometer (or a set of narrow-band IR filters with an energy meter). We found that for pulses with the peak power, P below the critical power of self-focusing in air, P<sub>cr</sub>, the laser beam diverged from the focal plane of the mirror where the full width at half maximum (FWHM) spot size and corresponding Rayleigh length (Z<sub>r</sub>) were measured to be  $\sim 2.3$  mm and 1.6 m, respectively. For CO<sub>2</sub> laser shots with a peak power above ~870 GW in the first pulse, this energetic laser pulse was suddenly seen to be self-guided producing an  $\sim 5$  m long visible plasma channel (with an estimated electron density around  $10^{15}$ -10<sup>16</sup> cm<sup>-3</sup>) followed by a many meter long light channel with no visible plasma or fluorescence emission from air (see Fig.3). For higher powers (≥1 TW), this ~1 cm diameter channel persisted from the curved mirror focus to z=32 meters (20Zr) limited only by the available laboratory space [6-11]. We call such a single filament a *megafilament* because it confines an ~1TW laser pulse with several Joules of energy and its cross-section is 10<sup>4</sup> times larger than a typical near-IR single filament.



**Figure 3.** Measurements of spatial evolution of the  $CO_2$  laser beam propagating in air in the range of distances from z = 0 to z = 32 m (red dots) from the nonlinear focus of the mirror, M. Black diamonds indicate laser size measurements in the focus obtained by a thermosensitive paper. The blue lines show the calculated propagation of the Gaussian beam in vacuum. Typical spatial beam profile of the self-channeled ~ 1 TWCO<sub>2</sub> laser at z = 11.22 m, z=22.4 m, and z=32 m measured by a two-dimensional pyroelectric array is shown on a side inset.

In Fig.4 we depict how the temporal structure of the train of picosecond pulses generated by a CO<sub>2</sub> laser, shown in the right hand panel in Fig.2, evolves during propagation in the atmosphere. Here for the self-guided pulse with a power exceeding 1TW, we detected a significant drop in energy contained in the postpulses after the first 5 meters of propagation. Recall that a plasma could only be seen over this distance. Clearly, these postpulses are strongly attenuated by the plasma produced by avalanche ionization over a short distance  $(2-3Z_r)$ . Further downstream of the focus, at z=11.2 m, only the single leading pulse was detected.

The self-guided pulse is expected to undergo pulse shortening, because both the head and the tail of the pulse have  $P < P_{cr}$  and therefore, experience significant diffraction losses. As shown in Fig 4b, by using an autocorrelation diagnostic with a time resolution of 0.2 ps, at z=11.2 m we measured a drop in the FWHM pulse length from 3.5 ps to 1.8 ps when the laser power increased from below to above the self-guiding threshold. Such pulse shortening is the reason why, despite some energy losses, the clamped intensity of ~1TW/cm<sup>2</sup> and P/Pcr>1 was sustained in the *megafilament* over almost the entire measurable propagation distance.



**Figure 4.** Temporal evolution of a picosecond  $CO_2$  laser pulse self-guided in air. a) Recorded temporal evolution of the  $CO_2$  laser pulse train at different positions in space (indicated by a schematic) while propagating in the atmosphere as measured by a streak camera with a time resolution of ~10 ps. b) Temporal profile of the initial 3.5 ps  $CO_2$  laser pulse without (P=476 GW) and with (P=1.14 TW) self-guiding in air, as measured by a single-shot autocorrelator with a temporal resolution of 0.2 ps at z=11.2 m.

For near-IR terawatt femtosecond pulses, the filamentation regime is usually characterized by the formation of a weakly ionized plasma channel followed by the second propagation stage in which the laser field is below the ionization threshold and diffraction is only balanced by the Kerr effect. In our experiment, the clamped intensity of 10<sup>12</sup> W/cm<sup>2</sup> in the filament was significantly below the tunnel ionization threshold of O<sub>2</sub>/N<sub>2</sub> (~10<sup>13</sup> W/cm<sup>2</sup>). Note that for long-wavelength radiation, multiphoton ionization should not play any significant role, since the Keldysh parameter is ~0.1. However, at this longer wavelength and relatively long pulse duration, impact ionization of neutrals by free electrons produced by the Excitation Induced Dephasing (EID) via many-body Coulomb effects enhances the plasma formation [12]. These freed electrons are also accelerated by the strong laser field that eventually results in avalanche generation. We have calculated the electron density build-up due to these processes. We find that during the first pulse the electron density rapidly increases to  $\sim 5 \times 10^{12}$  cm<sup>-3</sup>. Such a low density plasma would be not visible but plasma density further cascades to  $\sim 10^{16}$  cm<sup>-3</sup> during the postpulse (see Fig.5a) sufficient to form a visible plasma channel. The relatively low plasma density  $\geq 10^{12}$  cm<sup>-3</sup> created by EID effectively decreases the molecular polarizability during the 3.5 ps laser pulse sufficiently to arrest Kerr self-focusing and provide for self-balancing mechanism to channel light in the LWIR megafilament. Note that this novel theoretical model [12,13] was developed by our collaborators from the University of Arizona lead by J.M. Moloney.



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**Figure 5.** a) The calculated electron plasma density (solid line) versus time, the dash line showing the intensity profile of the incident double-pulse. Comparison between calculated and measured in the experiment b) spatial beam profile and c) pulse energy as a function of propagation distance in air.

This many-body description of air as a homogenious gas at the atmospheric pressure is idealized but provided a very useful self-consistent micropscopic theory to model propagation of a 3.5 ps TW CO<sub>2</sub> laser pulse in the laboratory air. A good agreement is obtained between the UPPE calculated and the measured in the experiment laser beam profile and pulse energy shown in Fig. 5b and 5c, respectively. Here red lines are calculated parameters as a function of propagation distance, the blue diamonds being the experimental data. Further analysis shows that *the megafilament* arises dominantly from the main first pulse when, as in the experiment, an incident pulse train is considered. The postpulses are effectively screened by an avalanche ionized air which is consistent with experimental data in Fig. 4a.

We also measured spectral content of the 10  $\mu$ m *megafilament* in air. Results of the on-axis spectral broadening measurements in the 8-14  $\mu$ m window are presented in Fig.6. Here, the conversion efficiency dropped dramatically from 2x10<sup>-3</sup> at 11  $\mu$ m to 2x10<sup>-5</sup> at 13.5  $\mu$ m. We also observed a significant asymmetry in the broadened spectrum favoring the red shifted light. This could be attributed to a delayed molecular contribution in n<sub>2</sub> for air favoring the red side as was observed in 10  $\mu$ m four-wave mixing study [5]. Thus the efficient SC generation with a short pulse CO<sub>2</sub> laser predicted numerically [14] is suppressed in the experiment by a competing third-order nonlinear effect. To estimate the possible role of  $\chi^3$  induced odd-harmonic generation on CO<sub>2</sub> laser self-guiding in air, we measured energy inside the *megafilament* contained in the harmonics: third-3.41  $\mu$ m, fifth-2.05  $\mu$ m, and seventh-1.46  $\mu$ m. The practically dispersionless infrared atmospheric window should make the harmonic generation process rather efficient. In the experiment the 3rd harmonic generation efficiency inside the *megafilament* reached  $\sim$ 4x10<sup>-4</sup>. Thus the spectral broadening is affected by harmonic generation in air in the range of 10<sup>-3</sup>-10<sup>-4</sup>.



Figure 6. Spectral broadening of an initial 3.5 ps  $CO_2$  laser pulse self-guided in air recorded at z=22.4 m.

These pioneering results on self-channeling of a TW picosecond  $CO_2$  laser beam in the atmosphere have triggered a lot of interest in the filamentation community and beyond. The study was published in a January issue Nature Photonics [8] and was also featured in the News and Views section of the same journal. The material was reported at major conferences, CLEO [6], SPIE [7], DEPS [9,10], COFIL2018 [11], OSA Nonlinear Optics [15], and IEEE RAPID [16] including invited talks [7,11]. We have published an invited paper in a special issue of Journal of Optical Society of America dedicated to Laser Filamentation and its applications [17]. We have written a chapter on LWIR filamentation in a book which will be published this summer [18]. Our collaboration with Arizona group was very productive and several aspects of long-range propagation of a TW  $CO_2$  laser in the atmosphere were reported in OSA journals [19,20].

**Modification of the UCLA Neptune CO<sub>2</sub> laser.** Propagation of a high-power CO<sub>2</sub> laser pulse in air strongly depends upon its temporal profile. Also a single picosecond pulse with a high nanosecond contrast is required for such experiments but it is very difficult to obtain due to modulated gain medium in a high-pressure CO<sub>2</sub> amplifier. The main strategy to mitigate this effect causing pulse splitting with a periodicity of tens of picoseconds is to increase the seed pulse energy into a chain of CO<sub>2</sub> amplifiers. During Y1-Y3 we explored two different approaches to increasing the energy of the seed pulse. The first plan was to install in Y2 a new, more energetic front end based on fiber lasers. However, due to complexity and limited funding (DOE Phase II SBIR) this system developed by Agiltron Inc. (MA) never reached the required level of seed energy.

In the second approach, in house we explored potential of a seeded 10  $\mu$ m OPA pumped by a home built CPA Nd:glass laser system. In this context a novel method for a long-wave infrared difference frequency generation (DFG) using a near-infrared laser and a Raman shifter was experimentally tested and characterized [21,22]. As shown in Fig. 7a, the picosecond pump laser at 1061 nm is split and sent through separate arms of the optical set-up. In the first arm, there is a cell filled with Raman active material C<sub>6</sub>D<sub>6</sub>, whose Raman frequency of 944.3 cm<sup>-1</sup> coincides with the 10P(20) line of the CO<sub>2</sub> laser. To generate a 10  $\mu$ m pulse the first Stokes sideband was then mixed in a DFG GaSe crystal with the 1 $\mu$ m radiation from the second arm.



**Figure 7.** a) Experimental set-up for 10  $\mu$ m DFG using a 1 $\mu$ m Nd:glass pump laser and Raman shifter. BS and BC are beam splitter and beam combiner, respectively. b) Measured 10  $\mu$ m idler spectra when the output is maximized (blue)) and slightly detuned from phase-matching to match the 10P(20) CO<sub>2</sub> laser transition at 10.59  $\mu$ m.

Strong nonlinearity of liquid C6D6 resulted in a significant spectral broadening of the 1  $\mu$ m pulse. We have shown experimentally that in the transient Raman regime, the conversion efficiency and frequency control can be accomplished by having a small negative chirp in a pump pulse for compensation of the spectral shift caused by self-phase modulation in the nonlinear Raman medium [22]. With this method, we demonstrated the generation of picosecond 3  $\mu$ J pulses of 10  $\mu$ m light. However, substantial broadening of the Stokes component resulted in a bandwidth far from transform limited for picosecond pulses (see Fig. 7b) that reduced the spectral density of the generated seed pulse and may reduce the efficiency of amplification in a CO<sub>2</sub> laser. To avoid this drawback, we tested an alternative scheme for LWIR OPA in which a low-energy 10  $\mu$ m picosecond pulse sliced from a nanosecond long CO<sub>2</sub> laser pulse was used as an idler seed in the same DFG/OPA crystal. This method provides tunability of the idler in the entire 9-11  $\mu$ m CO<sub>2</sub> laser spectral range and allowed for generation of a transform limited pulse for a picosecond 10  $\mu$ m seed [23,24]. We have tested both a single and double stage OPA in GaSe. The energy of 3-5  $\mu$ J was recorded in the experiment. However, amplification of such pulse in a high-pressure CO<sub>2</sub> amplifier indicated that spectral bandwidth for such a pulse was much broader than

that of an  $\sim$ 1 THz bandwidth in the amplifier and as a result stable injection mode-locking was not achieved. We had to come back to a semiconductor switching technique, in which a 3 ps Nd:glass laser pulse modulates reflectivity of Ge slab, to produce close to transform limited  $\sim$ 3 ps seed pulse which is sliced from a 100 ns long TEA CO<sub>2</sub> laser pulse by using reflective and transmissive switches.

While working on the 10  $\mu$ m seed source, using ONR MURI funding we have procured a large aperture 10 atm CO<sub>2</sub> amplifier to be used as a booster amplifier in the UCLA Neptune CO<sub>2</sub> laser chain. Acquisition and successful commissioning of this new amplifier allowed us to modify the way the short seed pulse is amplified and build a chain of multipass high-pressure CO<sub>2</sub> amplifiers schematically shown in Fig. 8. The most important feature of the system is a high nanosecond contrast of the order of  $10^4$ - $10^5$  achieved due to significantly reduced level of ASE in multipass amplifier scheme as opposed to a regenerative amplifier scheme used in the past. A typical pulse profile of a 2 mJ pulse as measured by a picosecond streak camera is presented in Fig. 8. Although there are still 2-3 postpulses in the structure of an ~60 ps long macropulse, the first leading pulse has the highest peak power and suitable for experiments on nonlinear atmospheric propagation. Such a pulse was successfully amplified in a large aperture e-beam controlled CO<sub>2</sub> module called MARS and energies in the range of 30-60 J are extracted in a 3" diameter output beam after three passes. These multiterawatt power 10  $\mu$ m pulses were focused in air and applied for plasma density measurements described below.



*Figure 8.* Simplified optical schematic of a picosecond  $CO_2$  laser chain built at the UCLA Neptune Laboratory and a typical pulse profile as measured by a streak camera for 2 mJ pulse output of the Booster amplifier.

Laser-plasma studies in a CO<sub>2</sub> laser ionized air. In our previous experiments on filamentation in air at BNL we have observed that a self-guided TW 10  $\mu$ m beam has two distinguished zones. As shown in Fig. 3, an approximately 5 m long plasma channel (where the visible fluorescence of N<sub>2</sub>/O<sub>2</sub> was observed) was followed by a light channel with no visible plasma formation. In Fig. 9 we show that a ~4 meter long plasma channel was recorded when a 30-60 J multuterawatt CO<sub>2</sub> laser pulse was focused in laboratory air by a Cu mirror with a long focal length f=7.2 m. For a low power unamplified pulse, the measured laser spot size in the focus was around 1.8 mm corresponding to the Rayleigh length of ~1m. For high-power shots, the focus was shifted upstream by approximately 1.5 meters indicating that Kerr self-focusing affects the beam propagation and is responsible for upstream position of the nonlinear focus. For each shot spatial profile was measured by the Pyrocamera and laser energies before and after the filament by a calorimeter.



*Figure 9.* A CCD camera image of the many meter long plasma formed in air by focusing of the multiterawatt  $CO_2$  laser beam at the UCLA Neptune Lab.

We have built a Michelson interferometer to probe the plasma channel formed in air transversely by using an~10 ns long 532 nm green laser pulse. This diagnostic allowed to record a 2D density map of the plasma and its evolution in time. Also by moving the nonlinear focus position in respect to the green probe beam we could gain an insight into longitudinal plasma dynamics along the channel. From earlier studies of CO<sub>2</sub> laser induced breakdown of air one can estimate the recombination limited atmospheric plasma lifetime to be in the 1-3 us time window. In Fig. 10a we present a typical interferogram recorded when a multiterawatt CO<sub>2</sub> laser beam was producing the intensity  $\geq 10$  TW/cm<sup>2</sup> sufficient to tunnel ionize O<sub>2</sub> in air. As opposed to homogenious plasma expected from the tunnel ionization of oxygen, by probing right after the CO<sub>2</sub> laser pulse we detect a few discrete plasma balls with a diameter  $\sim 100 \mu m$  and the estimated plasma density between  $5 \times 10^{18}$  and  $10^{19}$  cm<sup>-3</sup>. For statistically significant number of laser shots, in a  $\sim$ 1 cm diameter plasma channel, the number of such plasma balls was estimated to be  $\sim$ 50 cm<sup>-3</sup>. It is known that aerosol particles or dust particles of different size 1-100 µm are always present in unfiltered air and can affect the plasma formation [25]. We can estimate that for 10 µm light, the optically thick particles (i.e. particles with x=2r/ $\lambda$ >1, where r is the particle radius) will be particles with a diameter of 20 microns and larger. Detecting only of  $\geq 100 \mu m$  diameter microplasmas, when spatial resolution of the diagnostic was at least 10 times higher, could be explained by evaporation of smaller size particles and producing microclouds of a hot vapor. Similar family of localized discrete plasma balls were recorded along the entire plasma channel even the integrated in time fluorescence brightness changed significantly as seen in the plasma image in Fig.9. The longitudinal variation of number of plasma balls was statistically insignificant.

Thus, at such high laser intensities  $\geq 10$  TW/cm<sup>2</sup>, the plasma is transversely very inhomogeneous and dominated by ionization occurred on aerosol particles as well as formation of small self-focused microfilaments due to transverse break-up of the laser beam in multifilamentation regime (P/P<sub>cr</sub>>10). Electrons generated by these processes effectively seed avalanche ionization and plasma density is growing on the many picosecond time scale approaching the critical plasma density for 10 µm light. We have recorded a 50-90% drop in energy of the laser pulse propagating through such a plasma channel. The latter seems to be mainly related to screening of 10 µm postpulses in avalanche ionized air as was observed in our previous experiment [8]. Applying the same interferometric diagnostic to a part of 10 µm postfilament behind the plasma channel, in which the CO<sub>2</sub> laser intensity in the diverging beam dropped to ~1TW/cm<sup>2</sup> and the pulse length may be shortened to 2 ps, no signature of a high-density plasma was detected. The plasma density below 10<sup>15</sup> cm<sup>-3</sup> was not detectable even when a small angle Mach-Zehnder interferometer configuration was built and tested in the experiment.



Figure 10. Green pulse interferograms taken at a) 5 ns and b) 300 ns after the multiterawatt CO<sub>2</sub> laser pulse.

For several applications, e.g. guiding of electrical discharges or microwaves in the atmosphere, it is critically important to generate a large diameter channel in air with a long lifetime of 0.1-1 ms. Since atmospheric plasma recombination time is many orders of magnitude faster, this can be achieved only by creating a neutral density channel on laser axis when depression of neutral particles is formed due to hot hydrodynamically expanding gas. We explored time dynamics of plasma and hot neutrals on a longer time scale using our interferometric diagnostic. As an example, in Fig.10b we show a single fringe interferometric image of air taken 300 ns after the CO<sub>2</sub> laser pulse. Here it is apparent that described above small discrete plasma balls expand in time and now can have a millimeter scale diameter interacting with each other. Plasma density measurements indicate that high temperatures and high densities can be produced in the region near the surface of the aerosol particle. Expansion of hot plasma acts like a piston, driving a shock wave into the ambient air. This shock wave, such as that shown in Fig.10b, can additionally heat up gas and contribute to additional electron production via thermal ionization. We observed a very complicated and violent dynamics of how these discrete plasmas and individual shocks coalesce producing a neutral density channel that exist over a long  $\sim 100 \ \mu s$  time scale. We currently study this long-term dynamics of neutral density channel in air using green laser interferometry and other diagnostics. The results of this study along with findings on plasma dynamics in a 10  $\mu$ m filament formed in air by a multiterawatt CO<sub>2</sub> laser pulse will be reported [26].

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