Generation of Coherent, Broadband X-Ray and Mid-IR Pulses in a Noble-Gas-Filled Hollow Waveguide

Jeremy Pigeon, Sergei Tochitsky and Chan Joshi

Neptune Laboratory, Department of Electrical Engineering, University of California, Los Angeles, California, 90095

Abstract. We discuss a proposed experiment on guiding of a high-repetition rate, 20 GW, CO_2 laser beam in a gas-filled hollow glass waveguide. Extended interaction of the 3ps pulses with a gas Kerr medium will be used for measurements of nonlinearity at 10 um, and supercontinuum generation followed by pulse compression. Feasibility of 3-5 keV X-ray production via HHG is presented.

Keywords: CO_2 laser, waveguide, nonlinear index, self-phase modulation, pulse compression
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1. INTRODUCTION

Recent work has demonstrated possible advantages of using a high power, mid-IR laser in studying the acceleration of ions [1] and discussed X-ray generation via HHG [2]. The main advantage of using a CO_2 laser can be attributed to the 10 um wavelength of the laser light. In the case of ion acceleration this long wavelength regime provides an opportunity to study over-dense laser plasma interactions in a gas jet or cell [1]. In the case of HHG, the 10 um wavelength could open an avenue to obtain 3-5 keV X-rays due to the quadratic scaling of the HHG cut-off with wavelength [2]. The CO_2 gain medium has a high damage threshold, determined by the ionization of the active medium on the order 10^{12} W/cm^2, which removes the necessity of pulse stretching so that direct amplification to very high peak power can be realized. However, amplification of picosecond pulses in a CO_2 gas medium is hindered by a relatively narrow bandwidth. A multi-atmosphere laser is suitable for picosecond pulse amplification but these devices often have a small discharge volume which limits the maximum pulse energy.

Recently 15 TW of peak power was demonstrated using the UCLA Neptune Laboratory's master oscillator power-amplifier (MOPA) chain [3] which relies on a multi-atmosphere final amplifier that can deliver a shot once every five minutes. However, there are many applications that would benefit from a system that operates at a high repetition rate, for which only atmospheric CO_2 amplifiers (TEA systems) are readily available. Here we first describe the development of a 1 Hz repetition rate, 20 GW, 3 ps CO_2 laser based on final amplification at 1 atm. Second, we propose an experiment to guide our high repetition rate laser in a commercially available, Ag/AgI coated hollow glass waveguide (HGW) for studies in nonlinear optics. The end goals of such an experiment is: first, to measure the Kerr index in noble gases and, second, to increase our peak power to 100 GW by using chirping caused by self-phase modulation and subsequent compression of our laser pulses. Finally we discuss the feasibility of X-ray production via high harmonic generation using a 10 um pump.

2. AMPLIFICATION OF PICOSECOND PULSES IN CO_2 LASERS

The main technological challenge in high power CO_2 laser development is that, at 1 atm of pressure, the CO_2 gain medium does not have sufficient bandwidth to support the amplification of picosecond pulses. Figure (1a) is a simulated CO_2 gain spectrum at 1 atm of pressure. At atmospheric pressure the bandwidth of the 10P branch is comprised of individual 3.5 GHz rovibrational lines separated by 55 GHz. Therefore, in order to amplify picosecond pulses in CO_2 one must rely on two broadening mechanisms: pressure and field broadening.

Pressure Broadening

The width of the individual rovibrational lines in the CO_2 gain medium is $\Delta\nu \approx 3.5\ GHz/atm$. At sufficiently high pressure, the mean free time in the gain medium is faster than the transition time for stimulated emission which results in collisional broadening of the rovibrational lines. Figure (1b) is a simulated CO_2 gain spectrum at 10 atm, here the rovibrational lines have been broadened to ~35 GHz providing a quasi-continuum over approximately 1.2 THz. The broadening in a laser gas mix consisting of CO_2, He and N_2 can be approximated as [4],

$$\Delta\nu_{pressure} = P\left(5.79\Psi_{CO_2} + 4.25\Psi_{N_2} + 3.55\Psi_{He}\right). \tag{1}$$

Where, in equation (1), Ψ_s is the partial pressure of the pertinent gas species. It should be noted that, at 25 atm, the gain medium loses its modulation and becomes a true continuum across 1.2 THz of bandwidth over the entire vibrational branch. Unfortunately it is technologically impractical to design a large aperture, discharge pumped amplifier which operates at greater than 10 atm of pressure [5].

Field Broadening

Field broadening arises when the amplified pulse is intense enough to perturb the energy levels of the quantum system resulting in a smearing of these levels, similar to the AC Stark Effect [6]. The bandwidth gained through field broadening can be expressed as [7],

$$\Delta\nu_{field} \approx \Omega_R = 1.38 \cdot 10^7 \mu\sqrt{I}. \tag{2}$$

In the above, Ω_R is the Rabi frequency, μ is the CO_2 transition dipole moment in Debye, and I is the laser intensity in W/cm^2. For the 10.6 um lasing transition, the dipole moment is approximately 0.0275 D [8]. The total broadened line width is then the sum of the contribution from pressure and field broadening,

$$\Delta\nu_{Total} = \Delta\nu_{Pressure} + \Delta\nu_{Field}. \tag{3}$$

At a seed intensity of 5 GW/cm^2 a 1 atm amplifier's gain spectrum becomes very similar to the 10 atm spectrum of figure (1b). Therefore, using a sufficiently intense seed one should be able to use atmospheric pressure CO_2 lasers to amplify picosecond pulses. Such atmospheric devices, TEA CO_2 lasers, are commercially available and can operate at a pulse repetition frequency of 1-100 Hz and an output energy of 1-100J.

Due to residual modulation in the gain spectrum, amplification of picosecond pulses at 10 atm results in a pulse train in the time domain. This pulse train contains 3 ps long pulses separated by 18 ps, a time constant related to the 1/55 GHz modulation in the frequency domain. Figure (1c) depicts simulation results of a typical output pulse train, generated by seeding a 10 atm device with a single picosecond pulse.

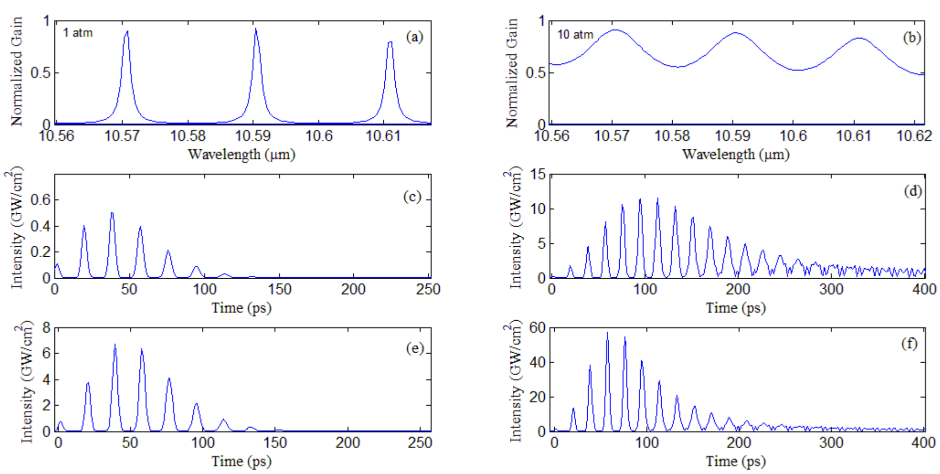

FIGURE 1. (a) Simulated CO_2 gain spectrum in the vicinity of the 10P(20) line at 1 atm and (b) at 10 atm of pressure. Temporal profiles of CO_2 laser pulses at 0.5 GW/cm^2 seeded into a 1 atm module (c) and corresponding output after 3 m of amplification (d). Temporal profiles of CO_2 laser pulses at 6 GW/cm^2 seeded into a 1 atm module (e) and corresponding output after 3 m of amplification (f).

Figures (1c-1f) summarize results of simulations showing the effect of field broadening in a 1 atm device. Figure (1c) and (1d) are the respective input and output pulse trains for a case when the input intensity is approximately 0.5 GW/cm^2. Figure (1e) and figure (1f) are the same simulation with the seed intensity scaled to 6 GW/cm^2. Due to the intense field depicted in figure (1e), the 3 ps pulse train of figure (1f) was amplified with negligible broadening.

3. NEPTUNE LABORATORY'S 20 GW, 3 PS, HIGH REPETITION RATE CO_2 LASER SYSTEM

We have recently developed a high repetition rate laser system which relies on a 1 atm device for final amplification. The system delivers 20 GW, 3 ps, 10 um pulses operating at a repetition frequency of 1 Hz [9]. The laser system consists of MOPA stages with two amplification regimes. In the first regime the pulse is amplified from nJ to mJ levels in 8 atm CO_2 amplifiers. In the final stage the pulse is amplified to Joule energies in a 1 atm booster amplifier where the laser field provides the bandwidth to sustain the pulse duration [9].

Figure (2) shows a typical pulse train after the final stage of amplification, as measured by a streak camera. Similar to modeling, the pulse train consists of ~3 ps pulses separated by 18 ps. Specific details regarding the system and the measurement presented in figure (2) are discussed elsewhere [9].

These results show that 3 ps pulses can be amplified in an atmospheric CO_2 laser, for which the bandwidth is predominantly provided by field broadening. Such a system, operating at a high repetition rate, lends itself to studies which are difficult or impossible to perform in a single shot experiment.

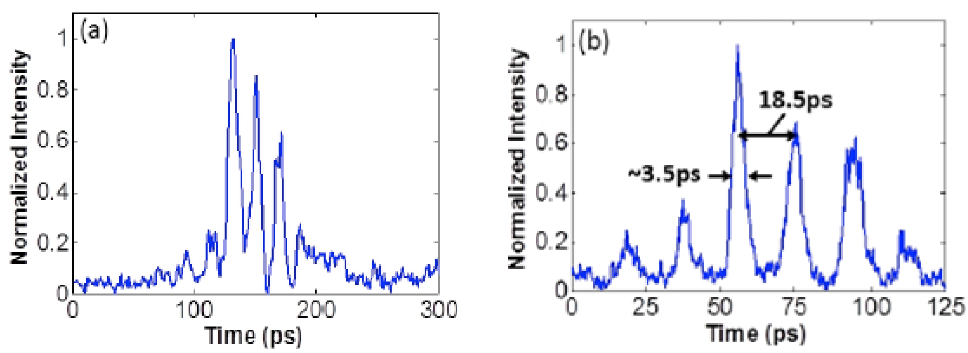

FIGURE 2. (a) A typical pulse train as measured by a streak camera and (b) zoomed image of the same data in (a) depicting a 3 ps pulse train separated by 18 ps.

4. NONLINEAR OPTICS IN A NOBLE GAS-FILLED HOLLOW GLASS WAVEGUIDE

As of late there has been a tremendous interest in ultrafast nonlinear optics, particularly in studies of broadband (supercontinuum) radiation generation in gas-filled hollow core fibers [14], laser filamentation in the air [21] and X-ray production via HHG [20]. The vast majority of these studies have been performed in the near-IR (0.8 – 1um). Despite the apparent interest in extending these studies to longer wavelengths, there have been no such experiments in the mid-IR except one very recent report on using a 0.8 um pumped OPA at 3.9 um for nonlinear optics studies [19].

Efficient Guiding of 10 um Light in a HGW

Commercially available, Ag/AgI coated hollow glass waveguides [23] have been explored as a method to guide high-power, short pulses at 10 um, the results of which have demonstrated 98% efficient guiding over 30 cm in air [10]. These results were obtained when the laser was properly matched to the EH_{11} mode of the waveguide for which the matching condition is $\frac{w_0}{a} = 0.64$. Here a is the radius of the waveguide and w_0 is the beam radius [11].

The first advantage of using a waveguide in studying nonlinear optics can be attributed to the fact that guiding the light removes uncertainty regarding the laser's intensity distribution and extends the interaction length well beyond that limited by diffraction. A second critical advantage of using a waveguide is that, when coupled to the EH_{11} mode, the laser maintains a linear polarization which is perpendicular to the axis of the guide [11]. This polarization preservation is crucial for the proposed experiment explained in the next section.

Proposed Measurement of Kerr Indices using a HGW

The standard way for determining the nonlinear index of refraction is measuring third/fifth harmonic yield in the medium of interest [12, 19]. However, due to the complicated temporal structure of the CO_2 pulse train, a simple measurement of harmonic yield is ambiguous. In order to measure the nonlinear index of refraction in noble gases, we propose to use a method of polarization rotation, namely to use a gas-filled HGW as a Kerr cell placed between two crossed polarizers. In this scheme, depicted in figure (3), the 10 um laser acts to rotate the polarization of a 630 nm probe beam. Upon rotation, the 630 nm probe is leaking through an analyzer, originally set to minimize the transmission of the probe, and this signal is sent to a streak camera to measure the temporal structure of the probe. It is important to note that the temporal structure carried by the 10 um pump pulse is transcribed unto the probe signal through this process.

After the probe's temporal distribution is measured by the streak camera, we now have several pulses of different intensity which can be individually fit to Malus's law:

$$T(t) = Asin^2\left[2.34 \cdot 10^6 \left(\frac{\pi L}{\lambda}\right) n_2 I(t)\right]. \tag{3}$$

In the above, A is the attenuation of the probe beam, L is the length of the waveguide, $n_2$is the nonlinear index of refraction in cm^2/W and I is the laser intensity in W/cm^2 [13]. On each shot we will measure the transmission of the probe as a function of pump intensity and extract n_2 as a fitting parameter. Preliminary analysis show that a 5% change in the probe's transmission results in a ~7% change in n_2. It should be mentioned that recently measured values of n_2 at 0.8 um are quoted with uncertainties on the order of 10% [22]. After averaging the nonlinear index over many shots, we will fit the index as a function of pressure from ~100-300 torr, noting that the nonlinear index should vary linearly with pressure.

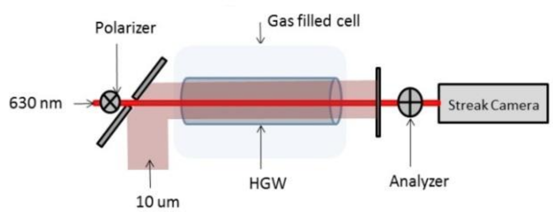

FIGURE 3. Schematic for a proposed experiment to measure the nonlinear index of refraction.

In order to ensure that we measure the transient birefringence related only to the nonlinear index of refraction we must operate at a plasma free condition. We will perform these experiments in the vicinity of 100 torr to ascertain that we are free of avalanche ionization and we will ensure that our pump power remains well below the critical power of self-focusing yet guide the beam using the HGW. It should be noted that this measurement can be easily extended to very high intensities on the order of $10^{12} - 10^{13}\, W/cm^2$ where a possible saturation of n_2 may be detected [22].

5. PROPOSED 100 GW, HIGH REPETITION RATE CO_2 LASER SYSTEM BASED ON CHIRPING AND COMPRESSION

With knowledge of the nonlinear index of refraction for noble gases at 10 um we plan to study spectral broadening from self-phase modulation in a HGW. The goal of such a study is to chirp and compress our picosecond pulses in order to increase their peak power. This technique is often used at 1 um in order to produce pulses shorter than those limited by the gain bandwidth [14] but has never been demonstrated in the mid-IR. In the picosecond regime one may approximate the increase in the pulse's time bandwidth product by [15],

$$\Delta f \Delta\tau \approx \sqrt{1+\left(\frac{2\pi n_2 I_0 L}{\lambda}\right)^2}. \tag{4}$$

For $I_0 = 6\,TW/cm^2$, $n_{2Xe} = 8\cdot 10^{-19} cm^2/W$ [16] and $L = 200\ cm$ we calculate an increase in bandwidth by a factor of 5 for an initially 3 ps, 10 um pulse. Figure (4a) is a plot of the input and output pulse spectrum after 2 meters of propagation in Xenon. It is important to note that propagation through noble gases, like most materials, will place a positive linear chirp on the pulse virtue of the gas's positive group velocity dispersion (GVD) in the spectral range of interest [15]. Although the calculation above was performed as an illustrative example, recent numerical simulations using the generalized nonlinear Schrödinger equation have found that the broadening induced by self-phase modulation in noble gases can be substantial in the mid-IR [16].

If the chirp induced on the pulse is linear then one could envision a scheme to compress our pulses by passing the radiation through a material with a negative GVD. An example of such a material in the mid-IR is NaCl; the insert of figure (1a) is a plot of the group velocity versus wavelength for NaCl which shows the negative GVD of the material. If we further assume that the level of spectral broadening is on the order of a factor of five then we should be able to increase our peak power from its current level of 20 GW to ~ 100 GW. One critical feature of this scheme is that both the HGW and the NaCl plate have very high transmissions, 98% and 92%, respectively [10]. Figure (4b) is a block diagram of this set-up.

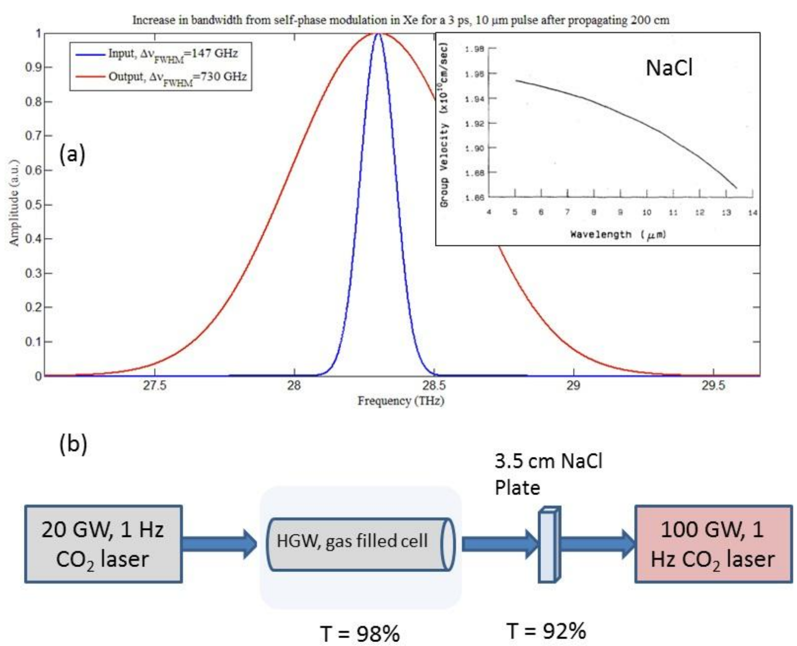

FIGURE 4. (a) Input and output spectral profile for a 3 ps pulse after self-phase modulation in 200 cm of Xenon gas, insert is the GVD of NaCl (data taken from [17]) and (b) block diagram of a scheme to increase our peak peak power from 20 GW to 100 GW through chirping and compression.

6. FEASIBILITY OF X-RAY GENERATION VIA HHG IN THE MID-IR

The need for X-rays for a myriad of scientific applications has driven research in X-ray sources for decades. One avenue for such a source is high harmonic generation (HHG). The novelty of HHG is that, using a relatively compact, femtosecond laser, one can produce a coherent beam of X-ray radiation [20]. X-ray photons produced via HHG have a characteristic cut-off energy which can be calculated as,

$$E_{max} = I_p + 3.17U_p. \quad (5)$$

Where I_p is the ionization potential of the atom and U_p is the ponderomotive energy of the laser given as,

$$U_p = E_0^2/4\omega^2. \quad (6)$$

Equations (5) and (6) imply that there is a certain advantage in using long wavelength drive lasers in HHG [2]. With a 10 um pump, intensity of $1 - 5 \cdot 10^{14} W/cm^2$ yield harmonic cut-offs of 3 – 5 KeV, respectively.

Although the scaling of maximum X-ray energy is favorable, numerical simulations have shown that the microscopic efficiency of HHG scales as $\lambda^{-5.5\pm0.5}$ [18]. This very rapid drop in microscopic efficiency is attributed to electron wave-packet spreading in the continuum and represents the most significant challenge in using a 10 um laser to produce X-rays via HHG. The microscopic efficiency can be compensated through phase matching between the harmonics and the pump laser, typically performed by increasing the pressure of the medium. However, a recent experimental investigation using a 3.9 um pump laser has shown that, in order to have phase matching, one must produce HHG at 30 atm of pressure [20]. This finding indicates that phase matching at long wavelengths may prove to be a serious technological challenge.

7. CONCLUSIONS

Although high power CO_2 lasers provide certain advantages in studies related to advanced acceleration and to radiation generation, present CO_2 laser technology is limited by low repetition rate, multi-atmosphere final amplifiers. Here we have shown that nonlinear optical phenomena, mainly field broadening and self-phase modulation, can provide a viable route to extend 0.1 – 1 TW CO_2 lasers to a high repetition rate, all without the need for chirped pulse amplification. We have demonstrated the promise of field broadening through the development of a 1 Hz, 20 GW CO_2 laser system which relies on an atmospheric final amplifier. The future work will be focused on using self-phase modulation, chirping and compression to increase the peak power of this system at a high repetition rate.

8. ACKNOWLEDGEMENTS

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