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Extending laser plasma accelerators into mid-IR spectral domain with the next-generation ultra-fast CO₂ laser

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Abstract. Expanding the scope of relativistic plasma research to wavelengths longer than $\lambda \approx 0.8\text{--}1.1\mu\text{m}$ covered by conventional mode-locked solid-state lasers would offer attractive opportunities due to the quadratic scaling of the ponderomotive electron energy and critical plasma density with λ . Answering this quest, a next-generation mid-IR laser project is being advanced at the BNL ATF as a part of the user facility upgrade. We discuss the technical approach to this conceptually new 100-TW, 100-fs, $\lambda = 9\text{--}11\mu\text{m}$ CO₂ laser BESTIA (Brookhaven Experimental Supra-Terawatt Infrared at ATF) that encompasses several innovations applied for the first time to molecular gas lasers.

BESTIA will enable new regimes of laser plasma accelerators. One for example is shock-wave ion acceleration from gas jets. We review ongoing efforts to achieve stable, monoenergetic proton acceleration by dynamically shaping the plasma density profile from a hydrogen gas target with laser-produced blast waves. At its full power, 100-TW BESTIA promises to achieve proton beams at energy exceeding 200 MeV.

In addition to ion acceleration in over-critical plasma, the ultra-intense mid-IR laser BESTIA will open new opportunities in driving wakefields in tenuous plasmas, expanding the landscape of Laser Wake Field Accelerator (LWFA) studies into unexplored long-wavelength spectral domain. Simple wavelength scaling suggests that a 100-TW CO₂ laser beam will be capable to efficiently generate plasma “bubbles” thousand times bigger in volume compared to a near-IR solid state laser of an equivalent power. Combined with a femtosecond electron linac available at the ATF, this wavelength scaling will facilitate study of external seeding and staging of LWFA.

1. Introduction

Generating energetic beams of electrons and positive ions from laser-produced plasmas is the mainstay of strong-field laser research focused on developing compact particle accelerators for scientific, industrial and medical applications. Strong progress in this research area has been achieved largely by using solid-state lasers operating at wavelengths $\lambda = 0.8\text{--}1.1\mu\text{m}$ that fall in the near-IR spectral range. To ensure efficient laser energy conversion into particle beams, laser beams are compressed to a few oscillation cycles of the laser electromagnetic field and focused to ultra-relativistic intensities. We call laser intensity relativistic when a free electron reaches relativistic energy over a single oscillation in the laser field, which is characterized by the normalized field strength parameter $a_0 = eE/m\omega c \geq 1$, where E is the laser’s electric field, e and m are, correspondingly, the electron charge and mass, c is the speed of light, and $\omega = 2\pi c/\lambda$ is the laser’s frequency. State of the art lasers used in forefront plasma accelerator experiments typically provide $a_0 \sim 5$ or more in as short as $\tau \sim 5\text{--}10\lambda/c$ pulses. Pushing solid-state laser

technology to extreme a_0 and τ parameters is one of the main thrusts of the laser-driven particle acceleration research.

Is the intensity scaling of near-IR lasers the only direction for progressing the strong-field research? It is well acknowledged that the increase of the laser's wavelength would offer opportunities in scaling plasma processes due to enhanced plasma response mainly manifested through a quadratic dependence of the electron's ponderomotive energy and the critical density on λ . Indeed, the electron's ponderomotive energy $\Phi = e^2 E^2 / 4m\omega^2$, which is the cornerstone of any plasma acceleration process, is proportional to λ^2 . Thus, a 10- μm laser will achieve the same electron quiver velocity at 100 times lower intensities than its 1- μm counterpart. This advantage is countered by the capability of shorter-wavelength beams to tighter diffraction-limited focusing. It should be kept in mind, however, that although lasers of equal power but different color may end up producing the same a_0 , a laser with ten-times-longer wavelength can do it over a hundred times larger area and potentially thousand times bigger volume (focus area multiplied by the Rayleigh range or the laser pulse duration, whichever is shorter), thus exhibiting proportionally higher hot-electrons' yield.

Another aforementioned wavelength-sensitive plasma parameter is its critical electron density $n_c = \omega^2 m \epsilon_0 / e^2$. Plasmas with electron density $n_e > n_c$ impede the laser beam propagation by reflecting or absorbing its energy within a thin skin layer and converting it into the hot electrons' energy, which is the necessary condition for generating charge-separation electrostatic fields suitable for accelerating heavy charged particles – ions. We see that n_c is proportional to λ^{-2} , and it reaches 10^{19} cm^{-3} at $\lambda=10 \mu\text{m}$; such plasma density can be attained by ionization of atmospheric pressure gases. In Section 2.1, we address the production of monoenergetic ion beams by shock-wave ion acceleration (SWA) [1] from over-critical gas jets readily achievable with CO_2 lasers. We review ongoing efforts and recent findings in optimizing this method by dynamic shaping the gas target density profile with laser-produced blast waves. The predicted scaling of ion-beam energy with the laser strength, $\sim a_0^{3/2} - a_0^2$ [2], characteristic of SWA, promises to achieve $\sim 200 \text{ MeV}$ ion beams of significance for cancer therapy at $a_0 \approx 10$.

Mid-IR lasers open a convenient opportunity for exploring a broad variety of laser/plasma interaction regimes by simply adjusting the ionized gas density, tuning it from strongly over-critical necessary for ion acceleration to deeply under-critical required for laser wakefield electron acceleration (LWFA). Expanding the landscape of LWFA studies, ultra-intense CO_2 laser will open distinctively new regimes and research directions, as we review in Section 2.2. Simple wavelength scaling suggests that, in a blow-out regime, a multi-TW CO_2 laser will be capable of efficiently generate plasma “bubbles” thousand times bigger in volume as compared to a near-IR solid state laser of a comparable power [3]. It is conceivable that such an over-sized plasma cavity can accept a bunch produced by a conventional RF linac, or from an intra-cavity virtual micro-photocathode, to facilitate studies of external seeding and staging of LWFA, or for achieving ultra-low electron beam emittance. Bigger plasma bubbles can also support multi-nano-Coulomb accelerated bunch charges, much higher than is afforded so far with solid-state laser drivers.

Benefits from expanding the landscape of strong-field laser interactions deeper into the mid-IR range discussed in Section 2 validate our ongoing efforts in developing BESTIA (Brookhaven Experimental Supra-Terawatt Infrared at ATF) - the next-generation, ultra-fast CO_2 laser that is designed to deliver 100-TW, 100-fs laser pulses at $\lambda=9-11 \mu\text{m}$. The technical approach to this laser encompasses several innovations applied for the first time to molecular gas lasers. This includes a solid-state OPA, isotopic gas amplifiers, the chirped pulse amplification (CPA) technique, and a patented method of femtosecond pulse post-compression. This laser project is discussed in Section 3. The laser will be offered to ATF users in 2018. Focused to $a_0=10$, the BESTIA's beam will empower new regimes of particle accelerators ranging from multi-MeV, monoenergetic ions at super-critical plasma density to high-current, externally seeded, GeV-class LWFAs in low-density plasmas.

2. Application of a mid-IR laser for plasma accelerators

2.1. Over-critical Plasma.

Successful methods of generating ion beams with lasers require strongly over-critical, steep plasma density ramps that favor efficient conversion of laser energy to hot electrons of relativistic energies within a very small volume on the order of $100\text{-}1000 \lambda^3$. Researchers usually achieve such plasma conditions experimentally by focusing laser beams onto a thin metal- or plastic- films. Ponderomotively forced to escape the laser's focus, hot electrons produce strong electrostatic fields due to charge separation, particularly along the target's rear surface where an electrostatic sheath forms. Within these sub-millimeter thick sheaths, ions can be accelerated to multi-MeV energies. This is the physical principle of the Target Normal Sheath Acceleration (TNSA) method [1, 2], the most commonly used in laser ion acceleration research.

The main trend in enhancing the ion energy by this method is reducing the target thickness with simultaneous reduction of the laser pulse length and the increase of its intensity and contrast. Pushing to extremes, a target can be so thin that the entire ionized material will be moved by the laser's radiation pressure producing a monoenergetic ion bunch [3, 4].

An alternative method of realizing a monoenergetic proton acceleration regime has been accomplished recently using terawatt CO₂ lasers and gas targets of near-critical density. Collisionless, electrostatic shocks launched into the plasma by laser hole-boring and localized heating has been identified as the main acceleration mechanisms in this so called Shock Wave Acceleration (SWA) of ions [5]. Upstream ions partially are reflected at the shock's front to twice the velocity of the driving shock [6, 7]. Monoenergetic ion beams had been experimentally produced by this method by Palmer *et al.* at ATF [8] and by Haberberger *et al.* at UCLA [9].

The localized deposition of laser energy needed for generating electrostatic shocks requires a steep density profile rising to overcritical density within several laser wavelengths. Such a steep profile is problematic to achieve technically in gas jets, which are typically limited to sub-millimeter density gradients. For such an overly long density ramp, the laser self-focuses and bores a channel, as shown in Fig. 1a. All of the laser energy is expended in the under-critical region, and no forward shock is formed. Simulations shown in Fig. 1b and 1c indicate the critical importance of sharpening plasma profiles within a gas target. Simulations presented in Fig. 1 use the 2D PIC code EPOCH. The plasma was initialized with 30 particles per cell of cold ions H⁺ and electrons, in cells of size $\lambda/50$. The CO₂ laser had $a_0=1.4$, $w_0=65$ mm at focus, pulse length $\tau=5$ ps (FWHM), and was linearly polarized.

This theoretical finding has been successfully demonstrated now in the ATF's user experiment [10] where the plasma density profile is steepened across a supersonic gas jet by a hydrodynamic blast wave imposed by an earlier, weaker laser pulse preceding the intense one. This pre-pulse deposits its energy to the gas around the focal volume and drives an expanding blast wave. This creates a ~ 100 μm thick gas density spike followed by a hydrodynamic depression region at the front edge of the target. This sharp density front is targeted by the main pulse that produces a collisionless electrostatic shock to accelerate the background ions.

The sharpness and stability of a density front has been further improved using an auxiliary Nd:YAG laser to launch a hydrodynamic blast wave from a laser focus placed on a metal foil at the edge of the supersonic gas nozzle [11]. The experimental setup is shown in Fig. 2a. A piece of stainless steel foil is mounted near the 1-mm-diameter orifice of the gas jet nozzle, and a 5-ns, 70-mJ Nd:YAG laser pulse is focused on it. The laser-ablated material is ejected from the foil surface at supersonic velocity, launching a hydrodynamic blast into the gas jet before the CO₂ laser pulse arrives. The produced density spike is ~ 6 times the local density over a length of < 30 μm . Fig. 2b shows a sample interferogram of the hydrodynamic shock 30 ns after the YAG laser pulse, the approximate time when a main CO₂ laser is delivered. See ref. [8] for details of our plasma optical probing technique. A 3-ps CO₂ pulse of 3-5 J energy focused ~ 1 mm above the nozzle orifice with a spot radius $w_0 = 60$ μm and maximum $a_0 = 1.4$

within the gas jet produces a typical proton energy spectrum shown in Fig. 2c. The spectrum exhibits a quasi-monoenergetic feature at 1 MeV.

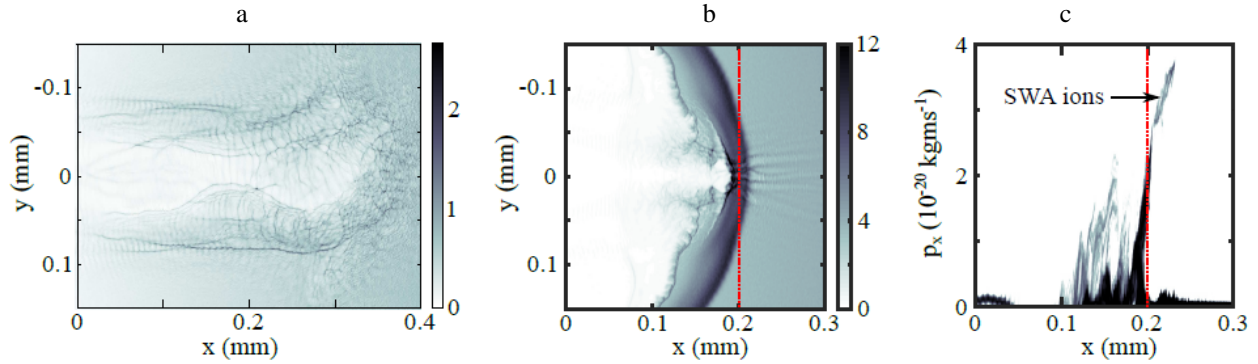


Figure 1. 2D PIC Simulations of laser interaction with an over-critical gas jet (gray scale is in n_c): a) a long linear density profile rising from 0 to $2n_c$ over 0.8 mm linear ramp, b) the same initial profile pre-modulated by a prepulse driven blast-wave, and c) proton p_x - x phase space showing shock-wave acceleration of an ion population to high energies for the case b). A red dashed line marks a position of the shock at $t=10$ ps, right at the end of the laser interaction with the jet.

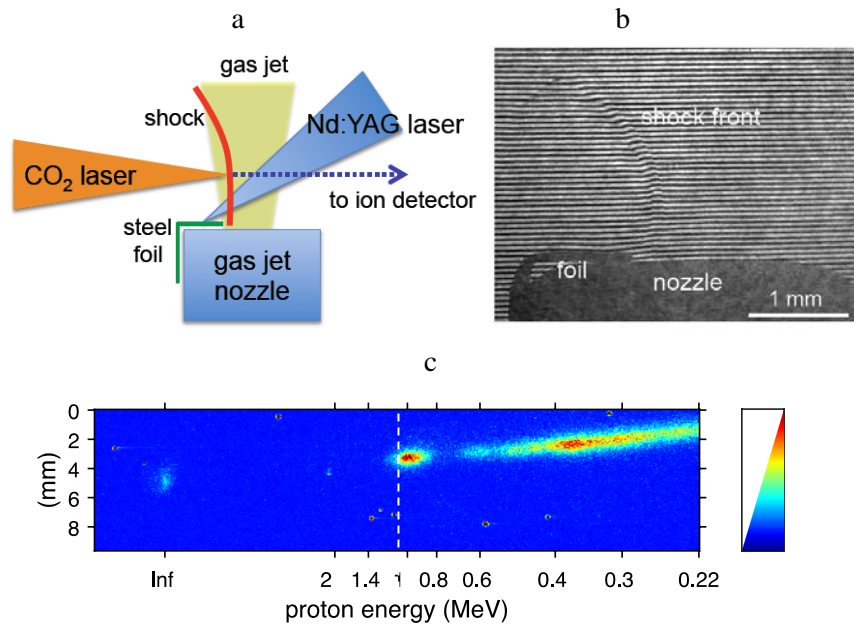


Figure 2. a) The experimental setup for a “two-color” SWA experiment; b) Plasma interferometry produced with a 3-ps optical probe at $\lambda=0.53 \mu\text{m}$ (2^{nd} harmonic of a Nd:YAG laser) shows a blast wave produced by a 70-mJ Nd:YAG beam focused on a tip of a foil; c) A luminescent screen of the magnetic spectrometer shows proton spectrum (to the right from a dashed line); a zero-deflection point is naturally marked by the plasma’s x-rays propagating through the spectrometer’s pinhole.

Reproducibility and consistency of produced proton spectra enables scanning the maximum proton energy versus CO_2 laser energy while maintaining the same gas and blast wave parameters, as shown in Fig. 3. The solid line represents the theoretical maximum proton energy $E_{max}=4I/n_c c$ estimated for hole-boring SWA under assumption that a collisionless electrostatic shock propagates with the doubled hole boring velocity and the background protons are reflected by a shock front at the double shock velocity [8, 12], which agrees with the experimental data. Assuming this close-to-linear scaling of the ion’s energy with the laser’s intensity, one can project that the 170-MeV $\sim 10\%$ energy-spread proton beams can be obtained

at a CO₂ laser intensity $\sim 10^{18}$ W/cm², attainable at 100TW peak power ($a_0=10$). Recently reported simulations [13] support this expectation.

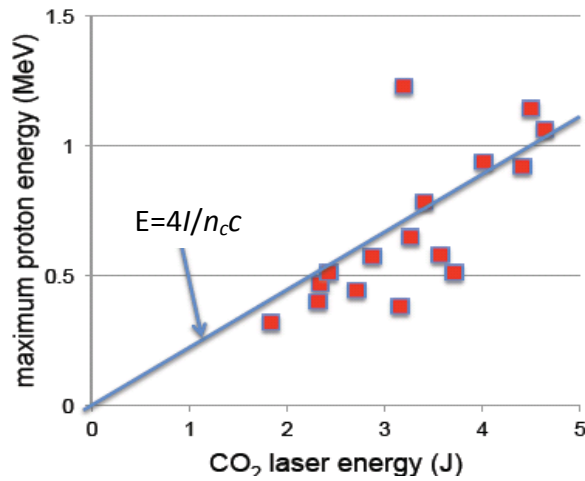


Figure 3. Dependence of the maximum proton energy upon the CO₂ laser energy.

Such compact laser-plasma ion accelerators will find a host of applications in nuclear physics and medicine, including radiation therapy. Gas jets used by the SWA method offer an added convenience of an easily renewable target as well as selecting pure ion species for acceleration, both of these features being important for realizing practical ion sources of high-repetition-rate, since they remove the limit on the pace of target renewal.

In addition to producing high-energy mono-energetic ions, the forespoken SWA could be simultaneously a source of powerful THz radiation. A high-intensity CO₂ laser beam produces copious number of hot electrons that escape the laser's focus at relativistic energies. Such a violent change in the electron's energy at the picosecond time-scale produces a single-cycle THz radiation. Due to their non-ionizing nature, good spatial resolution, and ability to penetrate several millimeters of biological tissue, such high peak power THz radiation is useful for studying structural transitions on the molecular level, or detecting cancerous cells [14].

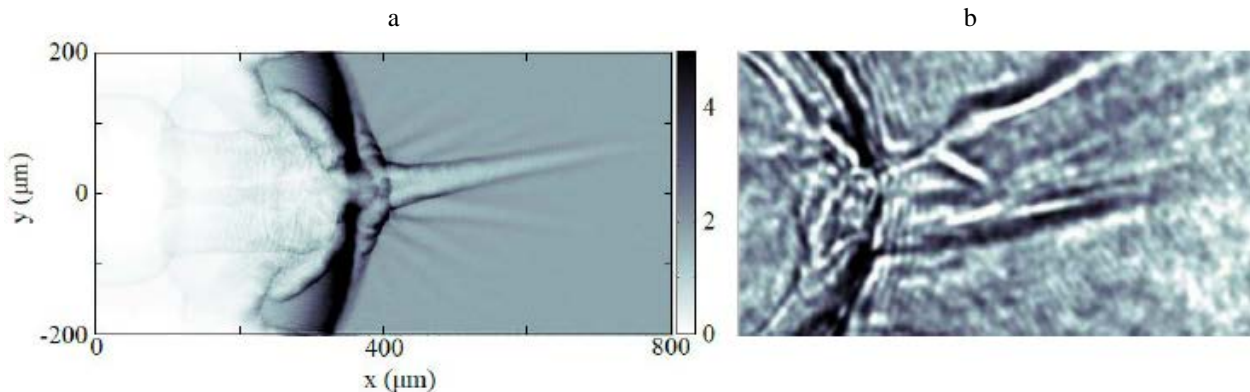


Figure 4. a) 2D PIC simulation of the interaction showing fast-electron driven filamentation in hydrogen, gray scale is measured in n_c ; b) Shadowgraphy image of fast-electron filaments obtained with a 3-ps optical probe ($\lambda=0.53 \mu\text{m}$) at 25 ps delay after the CO₂ laser interaction with a hydrogen gas jet at $7 \times 10^{18} \text{ cm}^{-3}$ gas density, which corresponds to $1.5 n_c$ electron density for full-ionized gas; the size of the image fragment is equivalent to the frame size in Fig. 4a.

Figure 4b illustrates our direct experimental evidence of hot electrons' currents visible due to their filamentation caused by transverse beam instabilities, verified by simulations (Fig. 4a) as well produced under similar assumptions and input parameters as our Fig.1. To obtain these images, the hydrogen plasma density in simulations and backing gas pressure in experiment has been reduced ~ 3 times to compare with the case on Fig.1 resulting in proportional reduction of the H_2 gas density in the center of the jet from $2 \times 10^{19} \text{ cm}^{-3}$ to $7 \times 10^{18} \text{ cm}^{-3}$. Also the gas jet diameter is expanded from 1 mm to 2 mm, and simulations are done for a later moment after the laser interaction when a filament is more visible. The observed currents are strong enough to exceed the threshold for beam filamentation, which is of the order of 100 kA for the plasma densities and time-scales we explored.

Analytical estimates based on the deposition of laser energy into the hot electron ensemble imply the generation of $\sim 1 \mu\text{C}$ charge. Assuming a 5 ps ponderomotive electron acceleration time, we obtained a 200 kA current. The power of the radiation emitted by the particles is given by the Larmor formula, $P = \frac{2}{3} c^3 \ddot{d}^2$, where $\ddot{d} = \sum e \ddot{x}$, i.e., the power of the emitted radiation is proportional to the square of the number of the particles and their acceleration. This leads to calculated 10 GW peak power in THz radiation, with an associated magnetic field ~ 10 MG, and a total radiated energy ~ 50 mJ, which favorably compares with best results reported with other methods, including solid state laser interaction with solid targets [15, 16].

2.2. Under-critical Plasma.

The pre-eminent criterion for many accelerator applications is a space-efficient design. Lasers offer the most compact configuration for electron accelerators as well. Laser Wake Field Accelerators (LWFAs), wherein a high-intensity laser pulse creates a relativistic plasma wake that can trap and accelerate electrons from the plasma at very high gradient, up to 100 GV/m [17], three orders-of-magnitude greater than that of conventional RF accelerators, have been recognized as a promising approach towards compact and affordable next-generation energy-frontier particle accelerators. The latest generation of a few-centimeters-long plasma accelerators operating in the "blow-out", or the otherwise-called "bubble" regime [18], already demonstrated GeV-class beams with parameters competitive with that of RF accelerators, including a micron-sized electron bunch and emittance, 100 pC charge, and sub-percent energy spread. All these achievements have been attained so far using near-infrared lasers ($\lambda=0.8\text{-}1.1 \mu\text{m}$) that deliver multi-terawatt peak-powers in femtosecond pulse format.

It has been theorized that an increase in laser wavelength may offer certain advantages due to the potential of inducing stronger ponderomotive excursions of plasma electrons, which is crucial in launching collective plasma-electron motion, i.e. plasma wakes. A straightforward wavelength scaling applied to the LWFA suggests that more efficient electron-acceleration can be achieved at lower plasma densities with 10- μm lasers. As a result, blowout structures, or "bubbles" can be efficiently produced at plasma densities below 10^{16} cm^{-3} . This implies smaller acceleration gradients, but simultaneously longer acceleration distances, thereby providing the same net energy gain for an equal laser peak power. The linear dimensions of the plasma-bubble structure also grow in proportion to λ , approximately tenfold, so supporting higher accelerated charges estimated by $N_{\text{mono}} \approx \frac{1.8}{k_0 r_e} \left(\frac{P}{P_{\text{rel}}} \right)^{\frac{1}{2}}$ that can reach more than 3 nC for a 100 TW-class CO_2 laser, in contrast to just hundreds of pC with a solid-state laser. This estimate agrees with recent simulations where 5.7 nC bunches are trapped and accelerated by crossing two CO_2 laser pulses with $a_0=2$ in in plasma of $3 \times 10^{16} \text{ cm}^{-3}$ electron density [19].

Simultaneously, having a bigger size plasma bubble simplifies achieving controlled external charge injection, providing a convenient test-bed for exploring beam loading effects and precise phasing into the wake of preformed femtosecond electron bunches injected from conventional accelerators, such as with the photocathode linac available at ATF [7].

Such a long-wavelength LWFA regime could not be explored so far because of the lack of multi-TW, mid-IR laser sources with a pulse length sufficiently short to drive a plasma wake to a competitively high field-gradient. The parameters soon to be achieved with the ATF CO₂ laser BESTIA open up the possibility to perform LWFA experiments at 10 μm . In addition, the ATF has undertaken capillary-discharge experiments where channeling of the CO₂ laser light was demonstrated [20]. Thus, the ATF not only possesses a viable laser driver for LWFA, but has a verified means to confine it over many Rayleigh lengths.

The prospects of using a 60 TW CO₂ laser driver for LWFA has been explored by 3D PIC simulations at [21] with the demonstrated possibility of a 2 GeV net energy gain over the 60 cm dephasing length for a 200-MeV electron bunch injected into a $3.2 \times 10^{15} \text{cm}^{-3}$ low-density plasma channel. These capabilities, in combination with the available femtosecond electron injector (ATF RF linac), make the potential LWFA ATF's experiment a viable complement to the ongoing plasma-accelerator research at world-class PW laser facilities.

3. BESTIA - next-generation ultrafast CO₂ laser

After discussing prospects of utilizing multiterawatt pico- and femtosecond mid-IR lasers for laser-plasma interactions at different physical regimes and applications, we address here our plans in developing this next-generation laser technology. High-pressure carbon dioxide (CO₂) lasers currently are the prime tools for generating terawatt-peak-power 10-micron radiation. Presently, Neptune Laboratory at UCLA [22] and the ATF at BNL [23] are the two best-known research facilities in the world that support strong-field physics experiments with CO₂ lasers. Achieving nearly a hundredfold gain in peak power, compared with the ATF's present laser, will require implementing methods never before applied to IR gas lasers. One of them is the CPA technique [24], which is critically important for operating mode-locked near-IR solid state laser systems but never applied to molecular lasers.

Compared to solid-state lasers, nonlinear effects in the active medium of gaseous amplifiers are weak, so supporting the attainment of terawatt peak power by directly amplifying picosecond pulses. However, nonlinear pulse distortions in transmissive optics will become critical for the next-generation ultra-intense CO₂ lasers that will operate above tens of terawatts. We can use the CPA technique for mitigating this issue.

In this conceptually new laser system named BESTIA (Brookhaven Experimental Supra-Terawatt IR at ATF), a laser pulse will be amplified to ~ 70 J energy in two high-pressure CO₂ amplifiers isotopically enriched with 50% of Oxygen-18 [25]. The amplified 100-ps laser pulse will be compressed in a grating compressor to 2 ps, limited by the spectral width of the CO₂ gain spectrum (9R branch). At this stage, we will achieve 25 TW peak power.

Detailed modeling and optimization of the amplifier configuration using our in-house developed code 'co2amp' [26] show that despite the fairly smooth gain spectral envelope due to the combined broadening by pressure and the effects of isotope mixing, the exponential enhancement of the pulse's spectrum over the long amplification distance results in its narrowing around the maximum of the 9R branch, along with considerable high-frequency modulation by the rotational structure. This last effect, in turn, causes modulation on the temporal envelope of the amplified pulse. However, CPA compression is hardly affected; This prediction is being confirmed in recent proof-of-principle experiments at the ATF [27] that demonstrated for the first time the feasibility of CPA in a CO₂ laser.

The 2-ps, 25-TW CO₂ laser pulse will be further compressed to the duration of few optical cycles, so exploiting the effect of spectrum broadening via self-phase modulation in a nonlinear medium. A patent-pending nonlinear pulse compressor will provide an output beam at the 100-TW level and down to 100 fs (three laser cycles) pulse duration [28]. The process's modeling is illustrated with Fig.5.

An intense 2-ps pulse with the assumed Gaussian temporal and spatial distributions transmitted through a Ge wafer will be a subject to the Kerr effect, which simultaneously induces the pulse's frequency

modulation and self-focusing. The most intense central portion of the pulse, which experiences the strongest Kerr-focusing, will be selected with a spatial filter and recollimated. This central portion of the pulse has nearly linear frequency chirp that allows recompression in a grating compressor down to 60 fs, which equals just two laser cycles. In spite of considerable energy loss on filtering, the peak power will reach the 100 TW level.

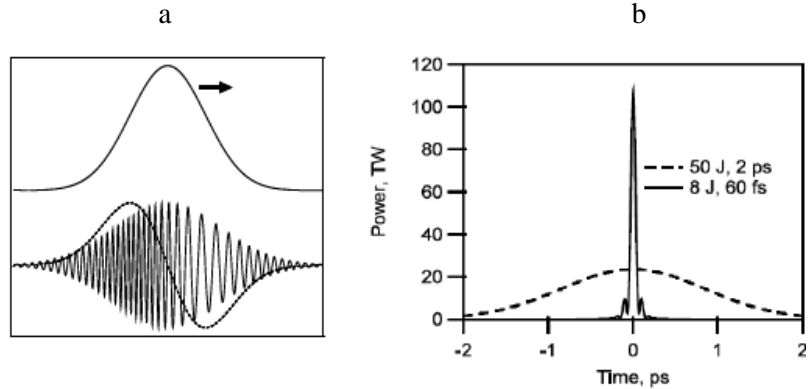


Figure 5. Simulation of a the nonlinear femtosecond pulse compression: Transmitted through a Ge wafer, the pulse experiences frequency modulation with a close to nearly linear frequency chirp observed around its peak (a); experiencing strong self-focusing, this central portion of the pulse will be filtered through a pinhole and recompressed (b).

We plan to complete the 25-TW 2-ps CPA CO₂ laser in 2017 and equip it with a nonlinear 100-fs compressor in 2018. BESTIA will be offered to users as a part of the ATF-II facility that additionally provides two radiation shielded experimental areas for strong-field physics research (Fig.6). In one experimental hall, the BESTIA laser beam will be brought to interaction with femtosecond, multi-MeV electron bunches in vacuum or plasma enabling advances studies in LWFA, ICS, IFEL, etc. The second experimental hall will be dedicated to laser interaction with gas- and material- targets for Ion Acceleration, THz, Trojan Horse, and other user-inspired experiments.

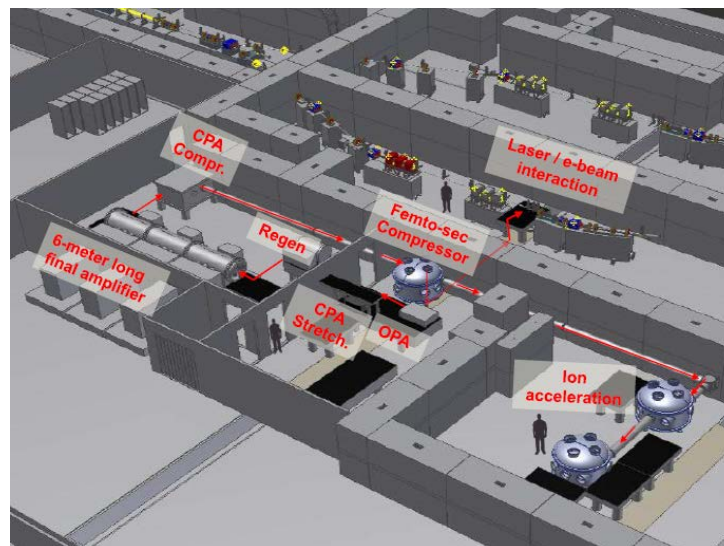


Figure 6. A fragment of a floor plan of the ATF-II facility shows the 100-TW CO₂ laser BESTIA and laser experimental areas

4. Conclusions

We recently started a program of upgrading the ATF CO₂ laser to the 100-TW level that we expect to complete within three years. The availability of such a laser will open up invaluable opportunities for innovative research in strong-field physics and the design of advanced accelerators within a range of parameters not accessible previously with mid-IR lasers. Some examples of those are the laser acceleration of ions from a gas jet to energies suitable for medical applications, the generation of few-MeV gamma-rays via high-harmonic Compton scattering, and the realization of new regimes of LWFA as well as the 1 GeV/m inverse FEL- acceleration, not discussed in this paper.

The 0.2 Hz repetition rate of Marx generators used to energize the BESTIA's final amplifier will define the overall repetition rate of this laser system. Potentially, a terawatt-class CO₂ laser can be configured to operate at higher repetition rates of 10-100 Hz, practical for many applications in medicine, material study and national security.

Acknowledgements

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