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LONG-WAVE IR TERAWATT LASER PULSE COMPRESSION TO SUB-PICOSECONDS*

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Abstract

We report an experiment and simulations on post-compression of 2 ps, 0.15 TW CO_2 laser pulses to 480 fs, ~0.25 TW by means of a self-phase modulation accompanied by a negative group dispersion in KCl and BaF_2 optical slabs. In addition, down to 130 fs fine pulse structure, but at lower conversion efficiency, has been observed through self-compression in a bulk NaCl crystal. The obtained results surpass by far previous achievements in the ultra-fast long-wave IR laser technology.

INTRODUCTION

Recent years have seen a growing interest in applying long-wave infrared (LWIR) lasers ($\lambda > 4$ µm) to strong-field physics research. This comes as the realization of benefits from the wavelength scaling for such applications as electron acceleration in plasma wake fields, ion beams generation from gas jets, intense gamma sources via inverse Compton scattering, energy transport through air, etc. The extension of experimental studies of these and other phenomena into the LWIR spectral domain has been enabled primarily by a CO_2 laser technology capable to producing picosecond pulses at $\lambda \approx 10$ µm and several-joule energies [1, 2]. However, the continuing advancement of most demanding applications, such as realization of the blow-out regime at a low plasma density, calls for down to few optical cycles sub-picosecond LWIR pulses at the multi-terawatt peak power.

The duration of an ultra-short laser pulse measured at the FWHM, τ_{FWHM}, is normally limited by the spectral bandwidth of the laser gain, which does not exceed ~1 THz for a CO_2 laser potentially allowing to attain τ_{FWHM}=500 fs [3]. However, the present-day CO_2 laser systems are restricted to a few-picosecond minimum pulse duration due to the spectral narrowing under a high amplification. Sub-picosecond post-compression of such pulses amplified to the several terawatt peak power can be explored through self-frequency modulation caused by the Kerr effect in an optical material followed by dispersive compression with diffraction gratings, a chirped mirror, or a bulk material with a negative group velocity dispersion. This idea has been elaborated in several theoretical and experimental studies, mainly for the purpose of improving characteristics of near-IR solid state lasers [4, 5]. Some recent works address also LWIR lasers at $\lambda \approx 4$ µm [6] and $\lambda \approx 10$ µm [7-9]. We report here the experimental results on post-compression of the 2-ps, terawatt-class CO_2 laser pulses to sub-picoseconds using optical materials transparent in the $\lambda \approx 10$ µm spectral range.

PRINCIPLES OF POST-COMPRESSION

The Kerr effect in an optical material accounts for an intensity-dependent change in the refractive index $\Delta n = n_2 I(t)$, where n_2 is the nonlinear refractive index and $I(t)$ is the laser intensity. The self-induced phase shift across a laser pulse after its propagation through a material of a length L_1, that we call a spectral stretcher, is given by $\Delta\phi(t) = -\left(\frac{\omega_0}{c}\right) n_2 I(t) L_1$ where ω_0 is the central carrier frequency of the pulse. The induced frequency sweep over the laser pulse envelope is

$$\Delta\omega(t) = \frac{d(\Delta\phi)}{dt} = -\left(\frac{\omega_0}{c}\right) n_2 L_1 \frac{dI}{dt}.$$

As a top of a Gaussian pulse can be approximated with a parabola, its derivative makes a linear chirp in frequency (see Fig. 1). Then, the maximum frequency excursion, $\Delta\omega_{max}$, can be estimated from

$$\Delta\omega_{max} = \pm\left(\frac{\omega_0}{c}\right) n_2 L_1 \frac{I_0}{\tau}, \quad (1)$$

where I_0 is the peak intensity and τ is the half-width of the parabola base or ~$0.6\tau_{FWHM}$ of a Gaussian laser pulse.

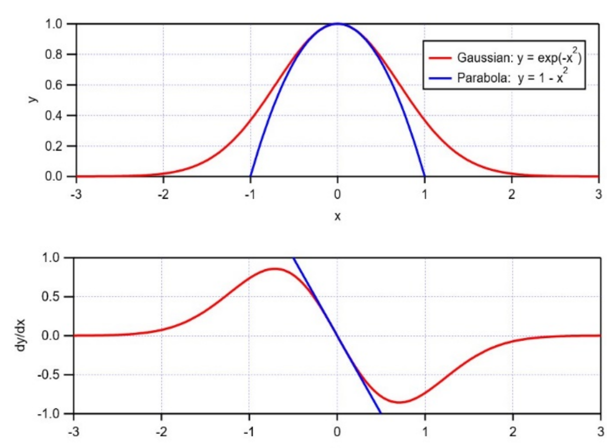

Figure 1: Illustration of the validity of a linear approximation for the frequency chirp induced by a Kerr effect on a Gaussian laser pulse, where x, y and dy/dx have the following physical meaning: $x = t/\tau$, $y = I/I_0$ and $dy/dx \sim dI/dt \sim \Delta\omega$.

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The maximum compression of a spectrally broadened, frequency chirped pulse will be achieved when we introduce a relative delay to the leading portion of the chirped pulse such that the tail of the pulse catches up the head. Since the nonlinear index n_2 is generally positive, a negative group dispersion is required in the compressor. For this, we can use a grating compressor. An alternative option is using a dispersive optical material of the length L_2 chosen according to the equation

$$L_2 = \frac{\tau/2}{D\Delta\lambda_{max}}, \tag{2}$$

where D is a chromatic dispersion parameter that characterizes a group dispersion and $\Delta\lambda_{max} = \frac{2\pi c \Delta\omega_{max}}{\omega_0^2}$. This brings us to the condition for the maximum pulse compression that relates the parameters of a stretcher (n_2, L_1) with those of a compressor (D, L_2) for given initial parameters of a laser pulse $(\omega_0, \tau_{FWHM}, I_0)$:

$$n_2 L_1 \times D\, L_2 \approx \frac{0.1\omega_0 \tau_{FWHM}^2}{\pi I_0}. \tag{3}$$

Such simplified argumentation is applicable under the condition that a stretcher and a compressor functions can be separated between different elements.

NUMERICAL SIMULATIONS OF CO_2 LASER POST-COMPRESSION

Modelling a Kerr-assisted post-compression becomes much more involved when the material has both attributes for pulse compression including a measurable nonlinearity and a group dispersion. As the chirped pulse compresses and different spectral components gradually come in overlap further enhancing the self-phase modulation, which is still in progress, the combined process becomes more complex and generally requires numerical modelling. Earlier numerical simulations [7] suggested the possibility of compressing 2.5 ps, 500 GW CO_2 laser pulses down to 250 fs with the peak power raised up to 2.2 TW using nonlinear and dispersive properties of bulk NaCl crystals. Our simulations conducted for a flat-profile CO_2 laser beam propagating through a 10-cm NaCl slab with parameters $n_2 = 3.5\times10^{-20}$ m^2/W and D=22.5 fs/(nm×km) demonstrate the maximum pulse compression to 80 fs with simultaneous intensity increase by ~11× at the optimum initial energy fluence on a sample of 0.48 J/cm^2 (Fig. 2). They also show the criticality of satisfying optimum conditions for achieving the best pulse compression; Just a 10% deviation from the optimum initial intensity results in a twice reduced intensity in a compressed pulse. Over-compression at higher intensities leads to spectral irregularities and severe pulse distortions with its splitting into satellite pulses.

Such simulations conducted also for other nonlinear materials, including KCl and BaF_2, have been used as a guidance to selecting test samples for our experiments on pulse compression reported in next Sections.

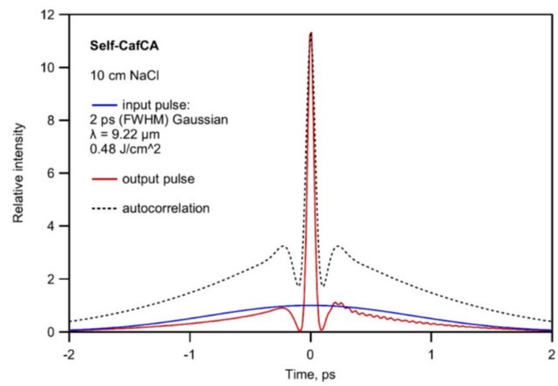

Figure 2: Simulated pulse compression for a 9.2 µm, 2-ps Gaussian beam propagating through a 10-cm NaCl slab; blue - initial pulse; red - compressed pulse; dashed – autocorrelation function of the compressed pulse.

PULSE COMPRESSION TESTS IN A SINGLE NaCl SLAB

The optimum regime identified by our simulations for the 10-cm NaCl slab is readily accessible with a 2 ps LWIR ATF laser that provides up to 5 TW peak power in a quasi-flattop λ=9.2-µm beam [2]. A collimated beam with up to 0.6 J/cm^2 fluence corresponding to the peak intensity ~300 GW/cm^2 was directed onto a plane-parallel 10-cm optically polished slab cut from a NaCl mono-crystal. After passing the slab, the laser beam was sent to diagnostics that included a joulemeter, a beam profile monitor, a spectrometer with ~8 nm resolution and a single shot autocorrelator (AC) of ~50 fs resolution. With the AC, we directly observed fine femtosecond structure down to 170 fs FWHM when the input energy fluence is close to the predicted regime for the optimum compression (see Fig. 3). In order to arrive from the width of an AC function, τ_{ac}, to the pulse duration, τ_{FWHM}, we use a relation τ_{FWHM} =0.746 τ_{ac} based on the exact numerical solution obtained for our particular pulse format. This gives us τ_{FWHM} =126 fs. No compression was observed with the laser fluence below 300 mJ/cm^2, whereas its increase beyond 500 mJ/cm^2 results in a notable degradation of the compression where a fine single peak splits into a more complex pattern as has been predicted by simulations. The observed spectral broadening in the compressed pulse $\Delta\lambda_{max}$ =70 nm is also in a reasonable agreement with the model.

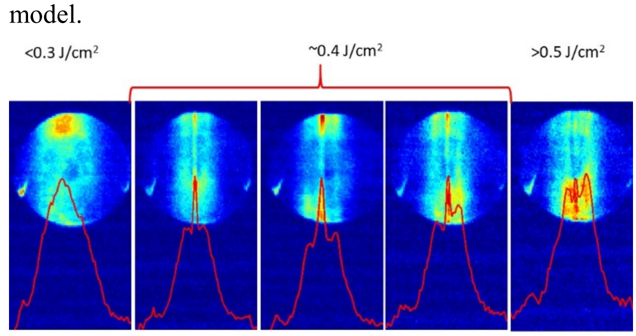

Figure 3: Raw AC images obtained with a single 10 cm thick NaCl slab at a variable input fluence.

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However, simultaneously with the pulse compression, we observed a nonlinear absorption, micro-filamentation and colour centers formation that affect the conversion efficiency at laser fluences of 400 mJ/cm^2 and above (see Fig. 4).

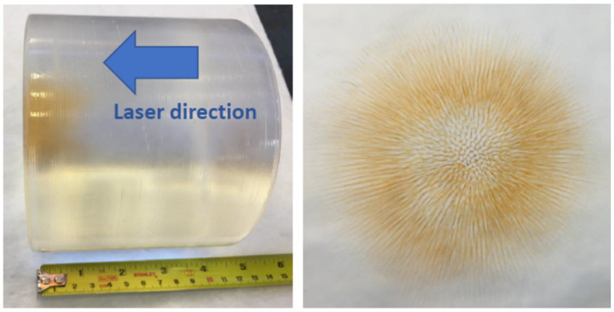

Figure 4: Colour centers formation with micro-filament traces observed after exposing a NaCl slab to CO_2 laser pulses of over 500 mJ/cm^2 fluence.

TWO-STAGE PULSE COMPRESSOR

To abate limitations of a single-element post-compressor design, we investigated other available materials for a more selective control over a process of the frequency chirping and dispersion compression. For a spectral stretcher stage, we choose KCl that has nearly the same n_2 index (3.4×10^{-20} m^2/W) as NaCl but twice smaller dispersion D = 12.4 ps/(nm×km); as per the compression stage, BaF_2 appears to be a better choice with its relatively small nonlinearity, n_2=1.7×10^{-20} m^2/W, and a high dispersion, D = 52.0 ps/(nm×km). For a better control of the intensity distribution in the laser beam, we selected a small portion of the amplifier output beam by filtering it through a 12.9-mm Teflon aperture and allowing several meters of a free space propagation before hitting the post-compressing slabs. This produces a high-quality quasi-Gaussian beam of 10 mm FWHM with up to 0.5 J energy, permitting a better correlation between experimental results and simulations.

With both KCl and BaF_2 slabs of the identical geometry (75 mm diameter and 50 mm thick), with no AR coating, spaced by 20 mm, we observed a well-defined compression as is illustrated by Fig. 5. The best compression to 290 fs has been observed at the input fluence 430 mJ/cm^2. Further increase in the laser energy causes reduction in the peak pulse intensity due to nonlinear absorption. As only the 5-mm diameter axial portion of the beam exiting the BaF_2 slab has been probed with the AC crystal, we applied our benchmarked computer model to estimate the pulse duration integrated over the entire quasi-Gaussian beam upon compression. The result of this processing is shown in Fig. 6. We see that the pulse compression averaged over the entire beam amounts to τ_{FWHM} =480 fs with the peak power increase from 0.15 TW to 0.25 TW.

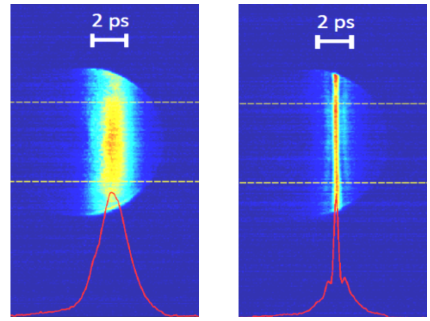

Figure 5: Raw AC images obtained at 450 mJ/cm^2 incident laser peak fluence without post-compression (left) and with a two-element KCl+BaF_2 compressor (right).

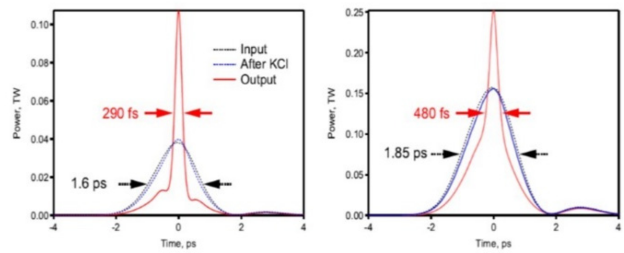

Figure 6: Results of numerical modelling of the pulse compression in the KCl+BaF_2 configuration at the conditions corresponding to the optimum compression (see Fig. 5, right); left – input and output temporal profiles of a 5 mm dia. central portion of a laser beam; right – the same but integrated over the entire beam.

CONCLUSION

Our obtained initial results surpass by far those previously achieved with the ultra-fast laser technology in the LWIR spectral domain. Experiments will continue towards further improving the efficiency of the energy conversion to femtoseconds by optimizing the post-compressor staging. We expect that our continuing material search and optimization of the beam and the setup geometry will lead to a practical system for an effective femtosecond conversion of the entire LWIR laser output at a multi-terawatt level as is required for advanced plasma accelerators and other strong-physics research and applications.

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