Creation of transform-limited 120 GW optical pulses using broadband supercontinuum generation in optical fiber

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A novel procedure to produce transform-limited optical pulses with high peak power using supercontinuum generation in an optical fiber has been developed. These pulses have been created using a mode-locked Ti:Sapphire oscillator to generate 18 fs pulses at 795.3 nm with energy of 4 nJ and bandwidth of 46 nm. A high power chirped pulse amplification was used to produce femtosecond pulses of 2.6 W at 32 fs for wavelength 800 nm. To achieve extreme pulse compression in the few-cycle regime, the 32 fs pulses have been injected through a hollow-fiber filled with neon gas to generate supercontinuum pulses then temporally compressor by multilayer-chirped mirrors. This arrangement enabled the generation of a five-octave-wide supercontinuum ultrafast pulses over a wide frequency range from 500 THz to 333 THz. That broad bandwidth has allowed to produce transform limited pulses of 6.01 fs time duration and 120 GW peak power that exceeds the previously observed value of 80 GW for similar pulses.
The observed results may give an opportunity to generate ultrashort pulses with extreme short optical wavelength using high harmonic generation that are needed for ultrafast spectroscopy in femtochemistry.

1. Introduction

Generation of ultra-short laser pulses is a crucial problem in laser physics. The overcome of such problem facilitates studying the ultrafast phenomenon associated with femtochemistry sciences[1]. The difficulty of time measuring itself comprises the exact determination of the extremely short-time periods using frequency standards. Accordingly, the improvements of femtosecond laser ensure remarkable attainments in the field of generation of laser frequency standards and the field of measurements of ultra-short time intervals. It is worth noting that, further increasing of pulse shortening results in intense light source with a huge increase in the laser pulse power which applied for numerous experiments which could not be done before with weak intensity[2]. These experiments include high harmonic generation (HHG)[3], femtochemistry[4], high field physics[5], generation of ultraviolet coherent attosecond laser pulses[6], and optical frequency metrology[7]. Moreover, the high intensity and spectral monochromasy of ultrafast lasers have introduced a new class of spectroscopic system, which permits exploration of the detailed structures of large atoms and molecules [8].

Since the invention of first laser by T Maiman[9] with burst duration of 1 ms and peak power of about 1KW on 1960, great efforts have been done for shortening pulse duration and increasing the laser power[10]. The majority of ultrafast lasers are divided into two categories. The first category depends on gain media that have quite suitable characteristics for diode-pumped high-power continues operation but cannot be applied for femtosecond pulse generation due to its rather narrow-amplification-bandwidth. For example, for Nd^{3+}:YAG pumped with high-power laser diodes, an average power up to 27 W for pulses of 20 ps has been achieved[11]. By using additive pulse modelocking technique, considerably shorter pulse duration of 1.5 ps has been achieved at lower output power of 20mW[12].

The second category of gain media are featured with a much broader-amplification-bandwidth, which facilitates shorter pulse durations below 0.5 ps but with smaller laser cross sections and considerably poorer thermal properties[12]. For instance, by applying a frequency-doubled solid-state laser or an argon ion as a pump source, Ti^{3+}:sapphire lasers have been capable to create short pulses with durations around 6 fs with average power of 300 mW[13]. These very short pulses have been achieved by using both of Kerr lens modelocking (KLM) and saturable absorber modelocking (SESAM) simultaneously while by using SESAM alone, longer pulse duration of 13 fs with 80mW average power has been reached[13]. By applying these techniques to Cr^{2+}:ZnSe crystal instead of Ti^{3+}:sapphire, significantly longer pulse durations of about 20 fs for wavelengths at midinfrared range of 2.2–2.8 μm with average power of several watts can be generated[14].

Actually, in 1990, Masataka et al. [15] created the first ultrashort transform limited pulses with a width of 17 psec and a spectral width of 0.21 nm. These pulses have reached output peak power of 90 mW at a 6-GHz repetition rate from a gain-switched distributed-feedback laser diode which was the highest peak power at that time[16]. A decade later, Kapteyn et al. group [17] successfully generated transform-limited 15-fs pulses and peak power of 1 W at repetition rate of 1 KHz by using a liquid-crystal deformable-mirror pulse shaper to correct high-order residual phase aberrations in a diode pumped laser system [17]. In 2008, Zaouter et al. [18] developed a compact double-stage ytterbium-doped-fiber chirped-pulse amplifier system to generate Transform-limited 270 fs, 340 MW pulses. Recently, Sergio et al. [19] have developed a gas-filled hollow-waveguide compression technique to create transform-limited 8 fs pulses peak power of 85 GW at a kilohertz-repetition-rate. They have injected 38 fs 1.5 mJ through an Ar gas filled hollow-waveguide compressor setup at a gradient pressure.

On the other hand, accurate determination of time domain waveform and pulse width is crucial to scientific research since they directly influence experimental results observed by use of ultrashort optical pulses. During the last three decades, many measurement methods for ultrafast pulse have been developed. The most commonly used methods are optical autocorrelation (AC) [20], spectral phase interferometry for direct electric-field reconstruction (SPIDER) [21], and frequency-resolved optical gating (FROG) [22]. Even though, the AC is direct and simple technique to consider autocorrelation width, but it fails to measure the waveform and phase of ultrafast pulses. Moreover, the AC measurements mainly depend on considering the intensity of the proposed pulse shape, thus ignoring any frequency dependent phase that may be present. This is important specially in case of measuring a transform limited pulse, or Fourier transform limit of a pulse, which known as the lower possible pulse duration for a given optical spectrum[23]. FROG represents a more advanced technique which offers a two dimensional measurement technique. Although, by using FROG, both of pulse waveform and phase can be retrieved from the constructed FROG trace, but a complicated computational mathematics algorithm is needed. On the other hand, SPIDER technique can be used to obtain directly both of the reconstruct pulse and the spectral phase waveforms. So that, SPIDER is considered to be more suitable for fast and accurate measurement of ultrafast optical pulses in in femtosecond regime[24].

In this paper, a new optical arrangement to generate transform limited ultrashort light pulses with high peak power and controlled broad-band spectrum has been represented. Moreover, a method to characterize both of the pulse intensity autocorrelation function and spectral phase of the observed transform limited pulses using SPIDER is demonstrated. The later was used to optimized the experimental parameters for best possible transform limited pulse.

2. Experimental setup

A schematic diagram for the generator of the transform limited ultrashort optical pluses is shown in Fig. 1. In this setup, A 15 fs mode-locked Ti: sapphire of at 800 nm with average power of 400 mW at repetition rate 75 MHz was used as a seed oscillator to create femtosecond laser pulses in TEM_{00} mode. The seed oscillator system composed of CW diode-pumped solid-state DPSS laser Opus (Laser Quantum) to pump the Ti:Sapphire oscillator with a green beam (532 nm) at 4 W. The seed setup contains pump beam mirrors to direct the green beam to Ti:Sapphire, a pump beam focusing lens to focus the green light into the laser rod, folded cavity mirrors, a pair of concave spherical mirrors aligned with the Ti: sapphire laser rod, an output coupler, a set of prisms used for dispersion compensation and a spectral tuning slit to tune the beam from 780 nm to 820 nm. The amplification phase was done using a high power chirped pulse amplification (CPA) with repetition rate 1 KHz. This stage produced 32 fs pulses with average power of 2.6 W at wavelength of 800 nm. To avoid damaging the gain medium, before that CPA amplification, a stretcher was used to stretch out seed pulses temporally and spectrally which reduces the peak power level. In the used stretcher a highly dispersive element composed of a single diffraction grating and a telescope that is followed by a flat mirror and a spherical mirror with broadband dielectric coatings. The CPA amplifier includes multiple passes within an amplifier gain medium that is sited within an optical resonator. The later contains an optical switch to control the number of round trips and to allow for a very high overall gain. The optical switch contains a Pockels cell, a quarter-wave plate and a thin-film polarizer. Then, a Faraday rotator was employed to separate input and output pulses. This is since a Faraday rotator represents a passive optical device that contains a magneto-optic material to change the polarization from horizontal to vertical by rotating the plane of the polarized light 45° in the forward direction. Afterwards, the stretched pulses were injected into regenerative amplifier at proper times using a optical switch by Pockels cell. After sufficient numbers of resonator round trips, the energy of the laser pulse achieves its maximum value of about 3 mJ. The formed high energy laser pulses are then released from the regenerative amplifier via a second Pockels cell as an optical switch. That was achieved by employing short bell-shaped high voltage electric pulses to the Pockels cell which works as a quarter-wave plate. A pulsed green 527 nm laser (diode-pumped Nd:YLF Q-switched green laser model DM20-527 , Photonics Industries) was used to pump the regenerative amplifier with energy of 20 mJ with duration of 170 ns at a high repetition rate of 1 kHz.
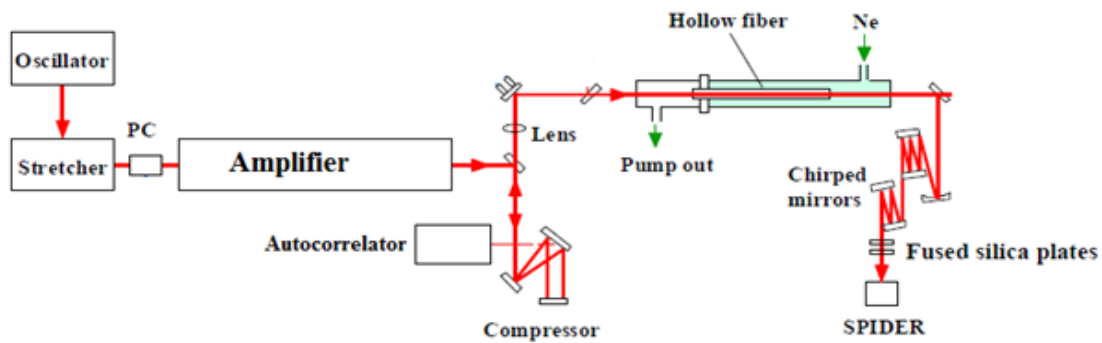
Fig. 1. Setup for the generator of the transform limited ultrashort optical pluses.

After the regenerative amplifier, the amplified pulses sent to a gate subsystem composed of two crossed polarizers and a third Pockels cell located between them. The later represents a pulse picker which is used to control the laser output though an external gate signal. The final amplified pulses are then compressed back to femtosecond regime via a compressor composed of a diffraction grating mirror assembly, "roof" mirror assembly, and output mirror. After the compressor stage, the laser pulses reach energy 2.6 mJ and duration of 32 fs at repetition rate of 1 kHz. Subsequently, the resultant pulses are directed to a final stage to produce the transform limited pulses. That final stage contains a hollow-fiber and multilayer chirped mirrors compressor. The hollow-fiber is composed of a one-meter optical fiber filled with neon gas. Using a concave mirror with focal length of 1.2 m, the laser radiation was focused into the inner of the fused silica fiber with diameter of 250 µm. The pulse spectrum was broadening via the self-phase modulation (SPM) which generates supercontinuum in the neon gas at controlled pressure from 2.0 to 2.5 atm. Finally, the output beam from the fiber is collimated using a concave mirror and then compressed again with six-beam-passes between two chirped-mirrors to achieve transform-limited regime.

3. Results and discussion

To achieve a transform-limited Gaussian pulse shape of the output pulse, both of the seeded and amplified pulses should be optimized. The oscillator beam was optimized in mode-locked at wavelength of 795.3 nm and bandwidth of 46 nm with energy of 4nJ/pulse for duration of 18 fs. Fig. (2) shows the seed signal bandwidth of about 46 nm measured with spectral phase interferometry for direct electric field reconstruction (SPIDER) where S is the signal intensity in arbitrary unites. On the other hand, the CPA amplified femtosecond pulses can be chirped in the compressor by changing the separation distance between the gratings. The CPA output pulses have average power of 2.6 watts at 1 kHz repetition rate and pulse energy of 2.6 mJ. Meanwhile, for a proposed optical pulse spectrum, the smallest possible pulse duration is observed when there is no chirp ("unchirped pulses"). This circumstance is similar to a constant instantaneous frequency, and (approximately) representing a constant spectral phase. For our system, the shortest ("unchirped pulse") accomplished pulse width of the amplifier is found to be 31.6 fs as shown in fig. (3A). The restriction of pulse compression is due to the limitation of the narrow bandwidth of the used laser gain media which found to be 47.42 nm for the amplified pulses as shown in fig. (3B).
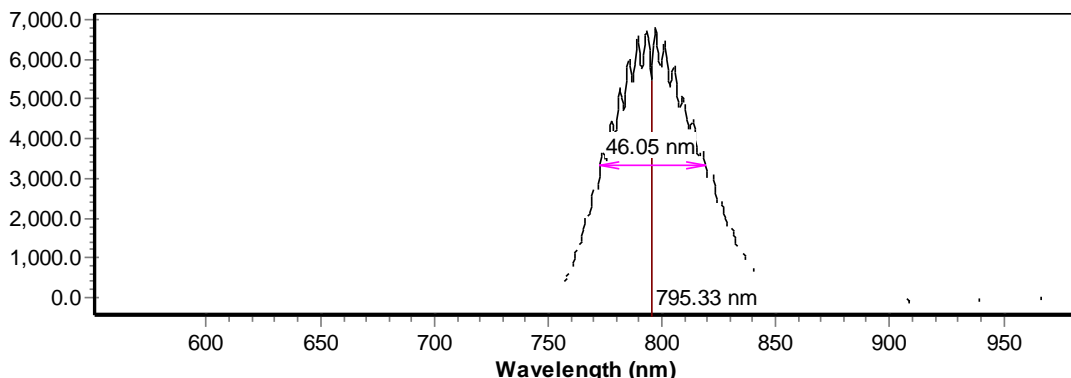

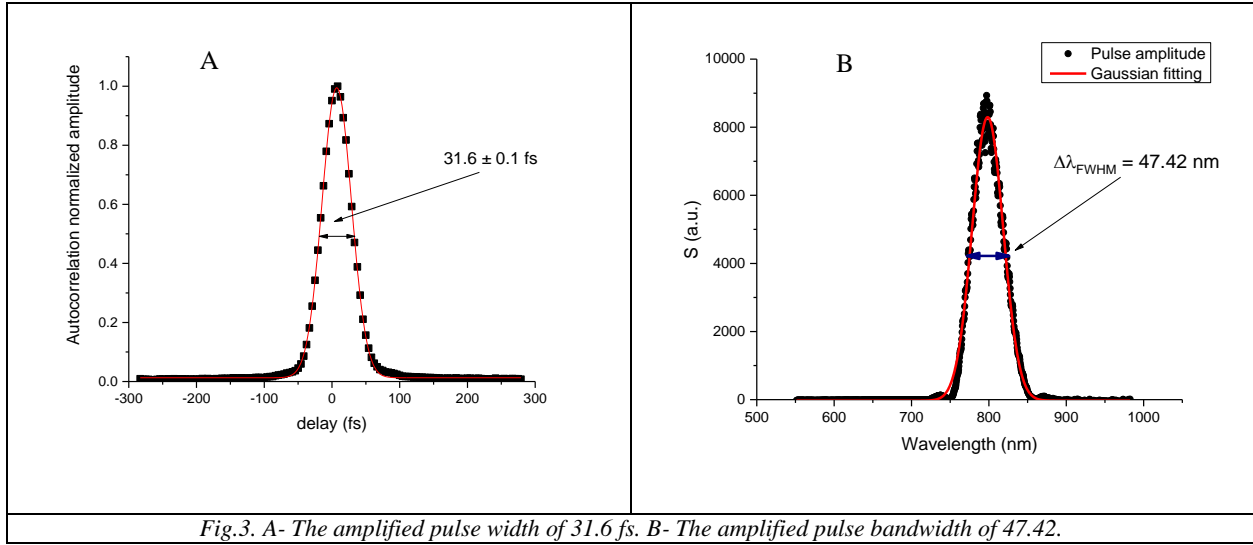*Fig.2 Spectral bandwidth of the 18 femtosecond seed laser beam.*

Fig.3. A- The amplified pulse width of 31.6 fs. B- The amplified pulse bandwidth of 47.42.

For achieving more pulse compression in the few-cycle regime, an external spectral broadening hollow-fiber followed by chirped mirror compressor was used. This facilitates the generation of a five-octave-wide supercontinuum ultrafast pulses extending from visible at 500 THz to the near-infrared at 333 THz in neon-filled capillary fiber as shown in fig. (4). The properties of the resultant output ultrafast pulses are affected by many factors include the pressure of the neon gas, the nonlinear phase shift due to SPM in the neon gas, and the pulse width of the input pulse before the fiber. The optimized final ultrafast pulses reached energy of 0.72 mJ at 1 KHz repetition rate, i.e average power of 720 mW.
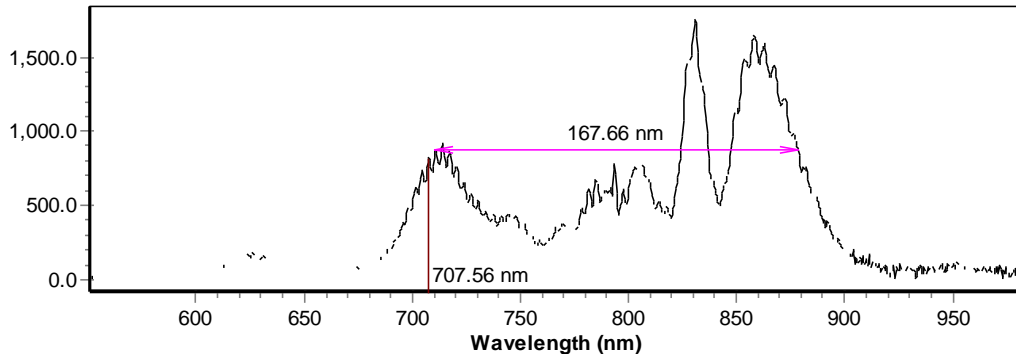

Fig. 4. Spectral bandwidth of the ultrafast pulses after the supercontinuum generation in the fiber.

On the other hand, spectral measurement of ultrashort pulses is crucial to deduce detailed information about the pulse temporal intensity-profile-function I(t). This is since the spectral wavelength intensity dependent, I(λ), or frequency dependent intensity, I(ν), is correlated to its temporal intensity, I(t), by the Fourier Transform. Thus frequency dependent phase can be "up-chirp" or "down-chirp" depends on whether the instantaneous frequency increases or decreases with time.

If an optical pulse is unchirped, the inverse Fourier transform of the square root of the spectral intensity delivers a precise representation of a "transform limited" pulse impartial of the actual shape of the optical pulse. If there are any traces of chirp in the actual pulse, this relation is not valid.

$$\mathbf{I}(t) = \left(F^{-1} \left\{ \sqrt{I(\nu)} \right\} \right)^2 \qquad (1)$$

Unfortunately, determination of whether the pulse there is transform limited cannot be verified by a direct procedure up till now[25]. In our procedure, the degree to which the observed optical pulse is transform limited can considered by assuming a proper form of the temporal pulse shape. This is done by comparing the experimental-measured time-band-width-product ($TBWP_{EXP}$) to theoretical-calculated time-band-width-product ($TBWP_{TH}$). The conventional determination of the time-bandwidth product is given as following[26];

$$TBWP = \Delta\upsilon_{FWHM} \, \Delta t_{FWHM} \qquad (2)$$

Where $\Delta\upsilon_{FWHM}$ and Δt_{FWHM} are the full width at half maximum of I(ν) and I(t) respectively.

Assuming Gaussian profile, the theoretical-calculated $TBWP_{TH}$ value is 0.44[26] while the experimental $TBWP_{EXP}$ is considered by measuring both of the $\Delta\upsilon_{FWHM}$ and Δt_{FWHM} for the observed optical pulses. The measurement of the degree to which the observed optical pulse is transform limited can be considered by

using the ratio of the $TBWP_{EXP}$ to $TBWP_{TH}$ which defined as $TBWP\%$ ratio as following;

$$TBWP\% = \left(1 - \left(\frac{TBWP_{EXP} - TBWP_{TH}}{TBWP_{TH}}\right)\right) \times 100 \quad (3)$$

Fig. 5 (A, B and C) describes the variation of transform bandwidth product ratio (TBWP %) with input pulse duration for neon pressures 2.0, 2.25 and 2.5 bars, respectively. In this figure, the pulse width of the input pulse before the fiber is varied from 32 fs to almost 56 fs for different neon pressures inside the hollow-fiber. The figure shows that the TBWP % linearly increased with the input pulse width. Furthermore, by increasing the neon gas pressure from 2.0 to 2.5 bar the TBWP % takes higher values with maximum of 95.65 % at 2.5 bar for input pulse duration of 54 fs.

Fig.5. The variation of transform bandwidth product ratio TBWP % with the input pulse using neon gas with pressure A – 2.0 bar B- 2.25 bar C- 2.5 bar.
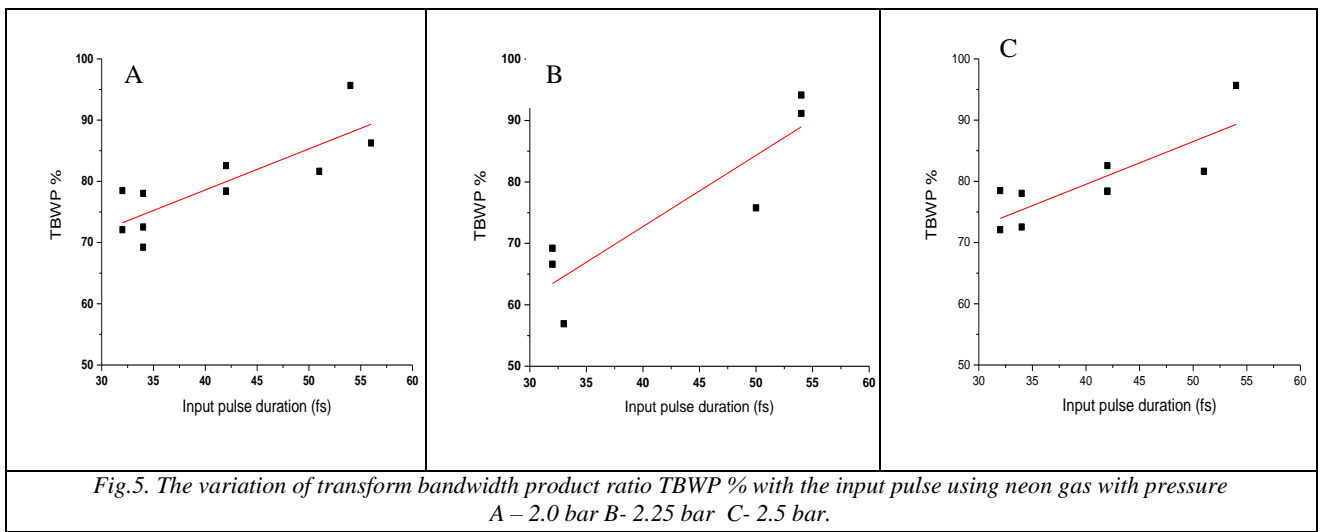
Fig. 6, shows the autocorrelation intensity function of the most optimized few-cycle pulses with TBWP % of 95.65 % and the experimental transform limited duration Δt_{FWHM} value of 5.76 fs measured using SPIDER. These measurements can be understood since the value of $TBWP_{TH}$ was considered to be 0.44 for Gaussian beam as given before and since the bandwidth $\Delta\upsilon_{FWHM}$ is 80.52 THz, then from (2), the theoretical Δt_{FWHM} = 5.76 fs which very close to our experimental finding of Δt_{FWHM} = 6.01 fs by 95.65 %. Then the peak power of the observed optical pulse is considered to be ~ 120 GW for pulse energy of 720 μJ and 6.01 fs pulse duration. The observed peak power value of 120 GW exceeds the previously observed value of 85 GW for similar pulses [19]. Moreover, the observed results show a recognized progress in the field of ultrafast pulses using a straight-forward technique in comparison with other more complicated methods[27, 28].

Fig 6. The transform limited pulse of 6.01 fs observed under optimized conditions.
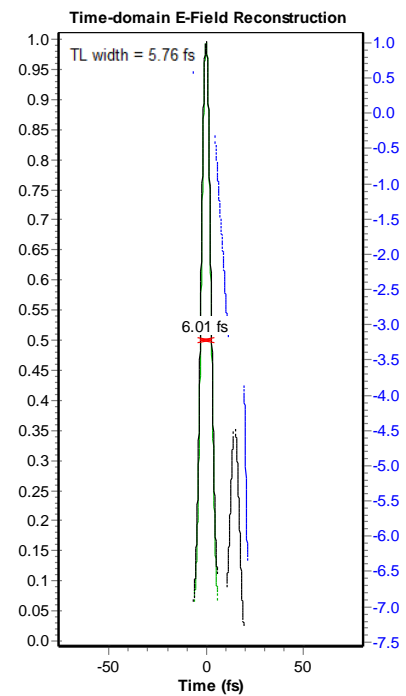
4. Conclusion

In the present work, I have demonstrated a new straight-forward table-top technique to create transform-limited optical pulses of 6.01 fs durations and 120 GW peak power. The generated pulses have a very broad spectral bandwidth from 500 THz to 333 THz. The experimental observed value of the time-band-width-product is consistent with the theoretical value for the formula which confirms the achievement to the transform limited duration for Gaussian profile. The achievement of high peak power value up to 120 GW is remarkable since it precedes the previously observed value of 85 GW for similar pulses. This method represents a simple and direct technique compare to other sophisticated methods. The observed results are very important since it generates very energetic few-cycle pulses which are needed to generate ultrashort wavelength in the X-UV. The later can be used to study the dynamics of ultrafast transient interactions in femtochemistry.

References

[1] A. H. Zewail, Angew Chem Int Ed Engl, **39**(Aug 4), 2586 (2000).
[2] P. Kryukov, Quantum Electronics, **31**(Feb), 95 (2001).
[3] B. Shan, S. Ghimire, Z. Chang, Physical Review a, **69**(Feb), (2004).
[4] D. Zhong, A. Zewail, Journal of Physical Chemistry a, **102**(Jun 4), 4031 (1998).
[5] Y. Li, X. Yuan, M. Xu, Z. Zheng, Z. Sheng, M. Chen, et al., Physical Review Letters, **96**(Apr. 28), 2006.
[6] J. Rocca, V. Shlyaptsev, F. Tomasel, O. Cortazar, H. Hartshorn, and J. Chilla, Physical Review Letters, **75**(Aug 7), 1236 (1995).
[7] H. Han, Z. Wei, J. Zhang, Y. Nie, Acta Physica Sinica, **54**(Jan), 155 (2005).
[8] W. Demtröder, Laser Spectroscopy: Basic Concepts and Instrumentation: Springer, 1996.
[9] T. H. Maiman, Stimulated Optical Radiation in Ruby, 1**87**, 493 (1960).

[10] P. G. Kryukov, Ultrashort-pulse lasers - IOPscience, 2001.
[11] G. Spühler, T. Südmeyer, R. Paschotta, M. Moser, K. Weingarten, and U. Keller, Applied Physics B, **71**, 19 (2000).
[12] G. Malcolm, P. Curley, A. Ferguson, Optics letters, **15**, 1303 (1990).
[13] D. H. Sutter, G. Steinmeyer, L. Gallmann, N. Matuschek, F. Morier-Genoud, U. Keller, et al., Optics Letters, **24**, 631 (1999).
[14] R. H. Page, K. Schaffers, L. D. DeLoach, G. D. Wilke, F. D. Patel, J. B. Tassano Jr, et al., Quantum Electronics, IEEE Journal of, **33**, 609 (1997).
[15] M. Nakazawa, K. Suzuki, Y. Kimura, Optics letters, **15**, 715 (1990).
[16] R. Tucker, U. Koren, G. Raybon, C. Burrus, B. Miller, T. Koch, et al., Electronics letters, **25**, 621 (1989).
[17] E. Zeek, R. Bartels, M. Murnane, H. Kapteyn, S. Backus, G. Vdovin, Optics Letters, **25**, 587 (2000).
[18] Y. Zaouter, J. Boullet, E. Mottay, E. Cormier, Optics letters, **33**, 1527 (2008).
[19] S. Carbajo, E. Granados, D. Schimpf, A. Sell, K.-H. Hong, J. Moses, et al., Optics letters, **39**, 2487 (2014).
[20] J.-C. M. D. Simoni, J. F. Joel, C. M. Ian, Francesco, Applied Optics, **24**(9), 1270 (1985).
[21] C. Iaconis, I. Walmsley, Quantum Electronics, IEEE Journal of, **35**, 501 (1999).
[22] D. J. Kane, R. Trebino, Optics letters, **18**, 823 (1993).
[23] A. M. Weiner, Optics Communications, **284**, 3669 (2011).
[24] E. R. S. London, L. Imperial College, S.-G. Alvaro, L. Imperial College, S. Thomas, L. Imperial College, et al., SPIE Optical Engineering + Applications, p. 958402, 2016.
[25] H. Fattahi, Broadband Seed Generation, Third-Generation Femtosecond Technology, ed: Springer, p. 45, 2016.
[26] K. C. C. Dienes, W. E. White, A. Sullivan, J. P. Heritage, R. S. Grant, K. X. Liu, et al., Optics Letters, **20**(8), 90406 (1995).
[27] D. Brida, C. Manzoni, G. Cirmi, M. Marangoni, S. Bonora, P. Villoresi, et al, Journal of Optics, **12**, 013001 (2010).
[28] G. Cerullo, C. Manzoni, D. Brida, CLEO: Science and Innovations, 2015, p. SF2M. 1.

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