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# Reaching white-light radiation source of ultrafast laser pulses with tunable peak power using nonlinear self-phase modulation in neon gas

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## HIGHLIGHTS

- A source of ultrafast white-light radiation has been developed.
- Observation of transform-limited pulses of 5.77 fs by compressing 32 fs pulses.
- Attainment of tunable peak power values varied from 57 GW up to 104 GW.

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## ABSTRACT

A source of white-light radiation that generates few-cycle pulses with controlled peak power values has been developed. These ultrafast pulses have been observed by spectral broadening of 32 fs pulses through nonlinear self-phase modulation in a neon-filled hollow-fiber then compressed with a pair of chirped mirrors for dispersion compensation. The observed pulses reached transform-limited duration of 5.77 fs and their peak power values varied from 57 GW up to 104 GW at repetition rate of 1 kHz. Moreover, the applied method is used for a direct tuning of the peak power of the output pulses through varying the chirping of the input pulses at different neon pressures. The observed results may give an opportunity to control the ultrafast interaction dynamics on the femtosecond time scale and facilitate the regeneration of attosecond pulses.



## 1. Introduction

Generation of an artificial white-light radiation source of ultrashort laser pulses is essential for applications such as: x-ray free electron laser X-FEL era (Adams et al., 2015), and applications of pulsed radiation implied by femtosecond-laser-induced high harmonic generation (HHG) (Feng et al., 2012; Cairns et al., 2009; Rocca et al., 1994; Hentschel et al., 2001). For ultrafast white light pulses, the electronic dynamics induced in molecules during the interaction with the pulse can be controlled by tuning the phase between the envelope and the field, which cannot be achieved by using conventional light sources (Mignolet et al., 2015).

Since the invention of the laser, it has been a dream for many scientists to generate bright source of white-light source with spectrum spans from the deep-ultraviolet to near-infrared, to employ it for state-selective chemistry (Judson et al., 1992). The phenomenon of white light continuum or supercontinuum (SC) generation that occurs during the propagation of a short intense laser pulse in a nonlinear optical medium has been known for more than three decades (Chin et al., 1999). SC generation is a process where laser light is converted to light with a very broad spectral bandwidth, whereas the spatial coherence usually remains high and the temporal duration ranges from femtosecond to nanosecond, depends on the seeding laser source (Alfano and Shapiro, 1970; Ranka et al., 2000). In 2000, Ranka et al. developed the first SC laser that could reveal anomalous dispersion at visible wavelengths from 500 to 1600 nm (Ranka et al., 2000). They injected 100-fs-duration pulses with only 8 kW of peak power at ~800 nm to induce nonlinear interaction in a 75-cm length of air–silica microstructure optical fiber. Even though, the observed results have represented a great success in comparison with previous work that has employed pulses with megawatt peak powers for generation of similar spectra (Pshenichnikov et al., 1994), the limitation of the observed peak power to 1.6 kW has represented a disadvantage.

Classically, high power broadband-radiation laser systems rely on complicated Ti:Sapphire amplifier to reach terawatt-frontiers

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(Pittman et al., 2002; Ito et al., 2003; Barty et al., 1996; Ishii et al., 2005). However, these systems have several problems such as pulse deformation which is caused by thermal load and thermal lensing inside the laser gain media (Shank et al., 1982). Consequently, it is very challenging to reach ultrashort laser pulses using such complicated systems for energies above one joule levels (Martinez et al., 1984).

In general, relatively long pulses can be compressed to very short ones in by two methods: linear-pulse compression and nonlinear-pulse compression (Fork et al., 1984). In the first technique, as soon as laser pulses are chirped, their periods can be shortened by reducing this chirp, i.e. by spectral-phase broadening. Thus, in linear pulse compression, a chromatic dispersion takes place if the laser pulses go through a dispersive optical element such as chirped mirrors, an optical fiber, a diffraction grating pair and a pair of prisms (Tomlinson et al., 1984; Alfano et al., 1986; Takeshi Kamiya and Masahiro Tsuchiya, 2006). The shortest possible pulse duration is governed by the dispersive compression limits and the optical bandwidth of the pulses. However, for the case of nonlinear pulse compression process, the optical bandwidth can be enhanced by supercontinuum, which can be generated by self-phase modulation (SPM) in nonlinear medium (Eggleton et al., 2000). Furthermore, after SPM, linear compression is employed to further compact the resulted chirped pulses (Agrawal et al., 2001).

The Kerr-nonlinearity-prompted intensity-dependent modification of the refractive index is a key physical parameter associated with supercontinuum generation. Therefore, most of the nonlinear effects of optical martials, such as optical fibers, originate from nonlinear refraction, the intensity dependence of the refractive index resulting from a significant contribution of optical susceptibility χ. Hence, the refractive index of a medium with a Kerr nonlinearity is given by Boyd (2008):

$$n = n_0 + n_2 I(t) \tag{1}$$

where $I(t)$ is the laser light intensity, $n_0$ is the field-free unperturbed refractive-index of the optical medium, $n_2=(2\pi/n_0)^2 \chi^{(3)}(\omega;\omega,\omega,-\omega)$ is the nonlinear refractive index at the optical frequency ω, $\chi^{(3)}(\omega;\omega,\omega,-\omega)$ is the third-order nonlinear-optical susceptibility (Shen et al., 1984).

The maximum possible SPM that can induce spectral-broadening of the laser pulse after passing through a distance L in a nonlinear material can then be estimated as (Hasegawa et al., 1973):

$$\Delta\omega(t) = \frac{\omega}{c} n_2 L \frac{I_0}{\tau} \tag{2}$$

where τ is the laser pulse width, c is the speed of light in vacuum and $I_0$ is the peak intensity of the pulse. Actually, SPM is accountable for spectral broadening of ultrashort pulses and the presence of optical-solitons in the anomalous-dispersion regime of the optical fibers. The third-order of susceptibility controls the nonlinear effects, which appear to be elastic, assuming that no energy is exchanged between the dielectric medium and the electromagnetic field. Another part of nonlinear effects results from stimulated-inelastic-scattering because the optical field transfers contribution of its energy to the nonlinear medium. In case of focusing of the laser beam into a hollow-fiber fused silica capillary filled with a noble gas, the observed optical spectrum will be broadened to both longer and shorter wavelengths due to nonlinear SPM in the gas. In fact, the hollow-fiber allows observing a large-mode-area, which is suitable for higher pulse energy. Moreover, the use of noble gases permits many advantages for SPM. For instance, in case of moderate pressures, these gases have purely third-order nonlinearity, and one can control the nonlinearity impact by varying the gas type and its pressure. Usually, the multi-photon ionization (MPI) is avoided by keeping lower laser intensity than the threshold of MPI, which is principally appropriate for femtosecond pulses. Consequently, the propagation of light wave along hollow-fiber can be determined as grazing incidence reflections at the dielectric inner surface. Taking into consideration the multiple internal reflections, only the fundamental mode can propagate in a suitably long hollow-fiber, whereas the higher order modes cannot propagate because of higher losses in this case. If a hollow-fiber with a capillary radius (a) is much larger than the beam wavelength and the lowest loss mode is $EH_{11}$ hybrid mode, its beam intensity profile as a function of the radial coordinate r given by Marcatili and Schmeltzer (1964):

$$I_0(r) = I_0 J_0^2(2.405 r/a) \tag{3}$$

where Jo is the zero-order Bessel function and $I_0$ is the peak intensity. It is worth noting that, even though higher-order modes maybe excited, mode refinement would take place anyway, according to the higher loss rate of $EH_{1m}$ with respect to fundamental mode. Mode selection in the capillary-fiber allows one to perform a spatial filtering of the input pulses.

In case of Gaussian pulse profile, if both of dispersion and self-focusing are ignored, the maximum output broadening spectrum that can occur after propagating a length of $l$ inside a capillary-fiber can be written as;

$$\delta\omega_{max} = 0.86 \gamma P_0 Z_{eff}/T_0 \tag{4}$$

where $P_0$ is the peak power; $T_0$ is the half-width (at the 1/e intensity point) of the pulse; $Z_{eff}=[1-\exp\alpha l]/l$, α is the imaginary part of the propagation constant; $\gamma$ is the nonlinear coefficient and is given by $\gamma=n_2 p(z)\omega_0/cA_{eff}$ [$n_2$ is given by Eq. (1)], where $n_2$ is the nonlinear index coefficient, $\omega_0$ is the laser central frequency; $A_{eff}$ is the effective mode area; $c$ is the light speed in vacuum.

A single-cycle optical pulse, the shortest possible waveform at a given wavelength takes place when the electric field within the envelope of an ultrafast optical pulse completes just one period before the pulse ends. In the near infrared region at around 0.8 μm, the duration of one optical cycle is approximately 2.7 fs. In this paper, a new method to control the pulse peak power of few-optical-cycle light pulses is described. In the proposed setup, the pulse compression is achieved by the nonlinear SPM in neon gas filled hollow-fiber and the output peak power value is found to be regulated by both the applied pulse duration and the neon gas pressure.

## 2. Experimental method

The ultrafast white-light radiation system is composed of a femtosecond oscillator, a regenerative amplifier and a final pulse compressor using neon gas as shown in Fig. 1. The oscillator laser is a mode-locked $TEM_{00}$ 800 nm Ti:sapphire with of 425 mW for pulses of 18 fs and repetition rate of 80 MHz. The basic oscillator setup consists of pump beam mirrors, a pump beam focusing lens, folded cavity mirrors, a couple of concave spherical mirrors lined up with the Ti:Sapphire laser rod, metal coated mirrors, an output coupler, a slit as a spectral tuning element compensation and a set of prisms to compensate the dispersion. The seed beam can be tuned from 780 nm to 820 nm range by moving the tuning-slit up and down. A 4 W CW solid-state diode-pumped DPSS laser Opus (Laser Quantum Ltd.) at wavelength 532 nm, was used to pump the oscillator. Additionally, the observed seed beam was stretched in temporal domain by using a stretcher. The stretcher is composed of a standard two-pass scheme with a single diffraction

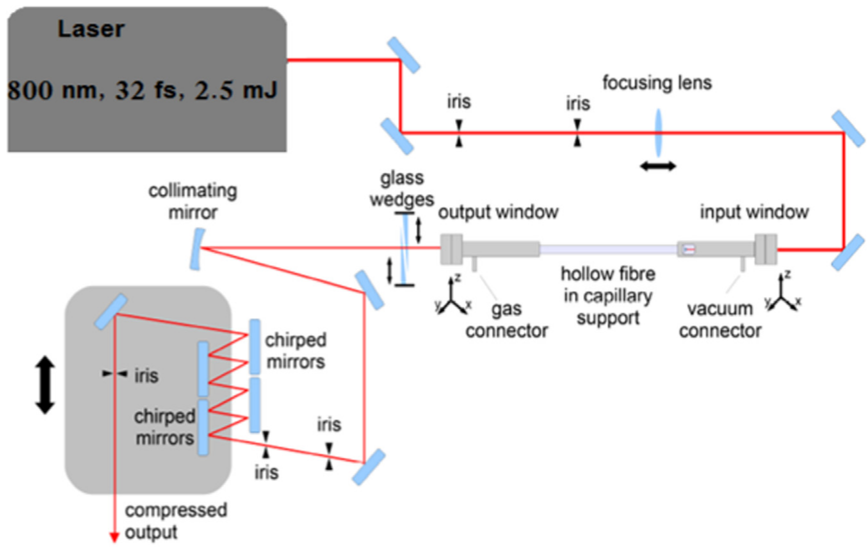


**Fig. 1.** Schematic of the ultrafast white-light radiation system with variable peak power.

grating, followed by a telescope formed by a flat mirror then a broadband spherical mirror with dielectric coatings to collimate the stretched beam. A Faraday isolator was used to change the beam polarization from horizontal to vertical. The stretched beam was injected into a regenerative amplifier (RA) by using an optical switch Pockels cell. The RA is a 1 kHz chirped pulse amplifier (CPA) that is used for the amplification stage to produce 32 fs pulses of 2.5 mJ at 800 nm.

After an essential number of beam round trips in the resonator, the energy of the observed pulse reached a value exceeding 3 mJ. The amplified pulses were directed out of the RA resonator using a second Pockels cell. The regenerative-amplifier pumping was achieved using a 1 kHz pulsed green laser at 527 nm via diode-pumped Nd:YLF Q-switched, (model DM20-527, Photonics Industries), to reach an energy of 20 mJ at 170 ns pulse duration of a vertically polarized laser light. Subsequently, the produced pulses were sent to a gate sub scheme consisting of two crossed-polarizers with a third optical switch (Pockels cell) placed between them. This pulse picker was used for controlling the laser output via an external gate signal to improve the contrast. Lastly, the pulse is compressed back to femtosecond regime using a pulse compressor. A telescopic mirror was used to expand the observed beam to prevent damaging the optical elements of the compressor. This final compressor was composed of an input mirror, then a diffraction grating, followed by a "roof" mirror assembly, then an output mirror. In that compressor, the laser pulses strike the grating four times. After this compression stage, the output pulses reach an energy of 2.5 mJ and a pulse duration 32 fs at repetition rate of 1 kHz.

The amplified laser pulses were directed to a final phase to yield the few-optical cycle pulses. These ultrashort pulses were achieved through a final compressor stage, which is consisted of a one meter hollow-fiber filled with neon gas. A final compressor for this stage composed of multilayer-chirped mirrors. By the use of a concave mirror with focal length of 1.2 m, the observed laser beam was focused into the hollow-fiber (250 μm inner diameter). The nonlinear interaction of the high intensity ultrafast pulses with the neon gas in the hollow-fiber, causes SPM that leads to super-continuum generation due to severe spectral broadening of the original optical pulses. The neon gas can be varied pressure up to 2.5 atm to control the observed spectral width. Finally, the observed laser beam was collimated by a concave mirror then formerly compressed using two multilayer-chirped mirrors with 6 round trips between them to achieve the few-optical-cycle region.

## 3. Results and discussions

The observed ultrafast pulses output features depend on many parameters, which include the nonlinear phase shift due to SPM in the neon gas, the gas pressure and the output pulse width after the amplifier. Moreover, the ratios of throughput efficiency of the fiber and the bandwidth of the pulses also affect the properties of the output beam.

Furthermore, the oscillator beam was optimized in mode-locked for pulses of duration 18 fs and bandwidth of 46 nm at central wavelength of 795.3 nm, which represent proper conditions for the seed femtosecond pulses as shown in Fig. 2A. There is an opportunity to tune the wavelength of the seed laser in the range of 750–850 nm using an adjustable optical slit and two prisms. The emission of the Ti:Sapphire can be spatially spread and the wavelength can be tuned by using a combination of energy-dispersive optics and a slit. Furthermore, the CPA pulses can be chirped by changing the distance between the gratings in the compressor, so that the pulse duration of these pulses can varied from 32 fs up to 54 fs.

Since the spectral phase of the observed ultrafast pulse is needed to be characterized and its intensity profile in time domain is needed to be measured, a spectral phase interferometry for direct electric field reconstruction (SPIDER) technique was employed to determine the obtained ultrashort pulses. SPIDER is a nonlinear self-referencing ultrashort pulse measurement technique based on spectral shearing interferometry (Kosik et al., 2005). It is important to note that, because of the narrow bandwidth of the laser amplifier gain media used, it is impossible to generate few-cycle pulses directly from a CPA amplifier. To overcome this restriction, an external spectral broadening was utilized using SPM in neon gas filled a hollow-fiber followed by a compressor composed of two multilayer chirped –mirrors as described earlier. By adjusting the neon gas pressures from 2 atm to 2.5 atm in the one meter hollow-fiber, the pulse duration of the observed pulse was optimized. The effect of temporal profile changes of the different given input pulses with duration from 32 to 54 fs on the output pulse are described in sequences for different neon pressures from 2 atm to 2.5 atm, for each in Figs. 3–5.

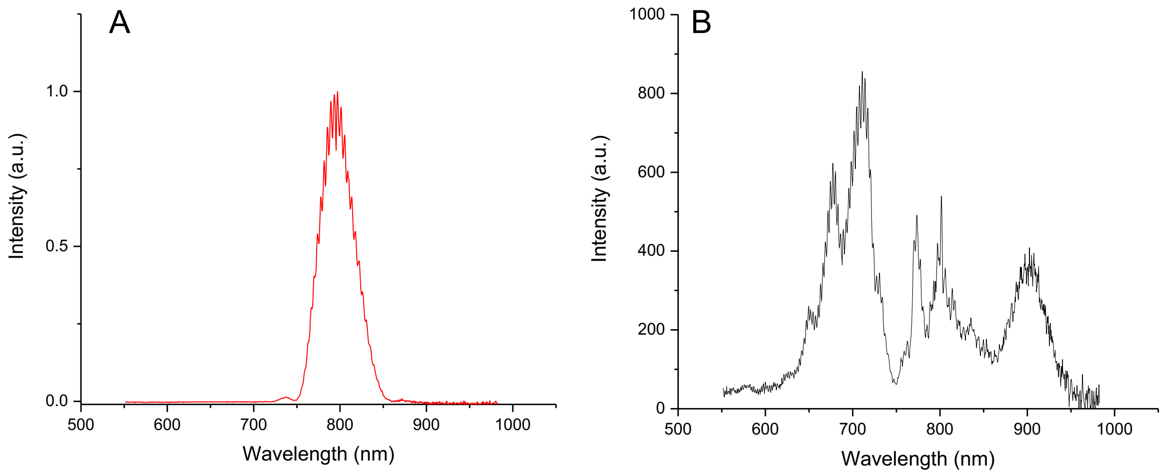


**Fig. 2.** (A) The 46 nm bandwidth of the Ti:sapphire oscillator at 795.3 nm and 18 fs pulse duration. (B) The 350 nm broadband width of the output pulses of 7 fs and energy of 600 μJ.

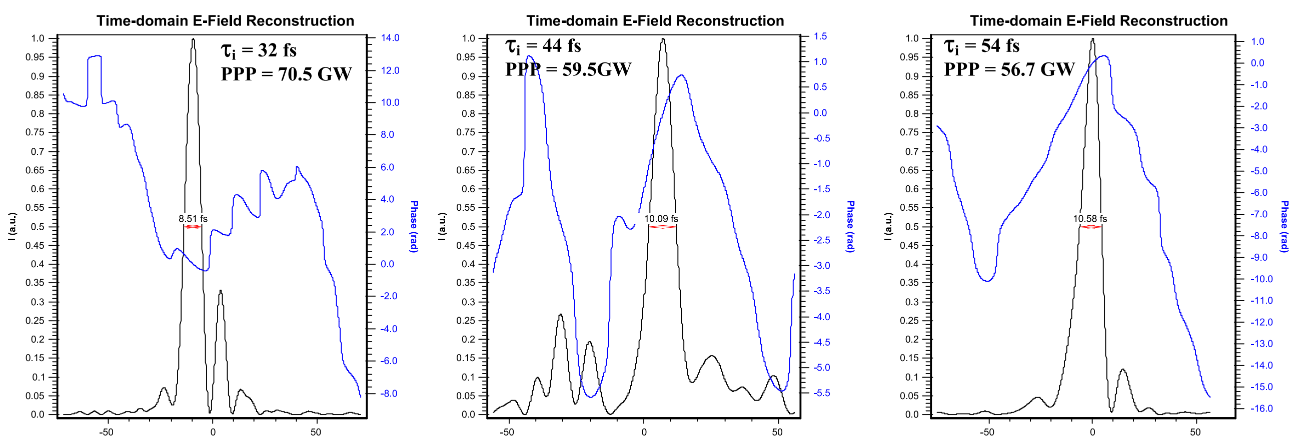


**Fig. 3.** The temporal profile change of the output pulses $\tau_i$ values of 32, 44, and 54 fs, respectively, at neon gas pressure of 2 atm. The compressed output pulse (black curve) and temporal phase (blue curve) of the compressed output pulses were measured using SPIDER. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

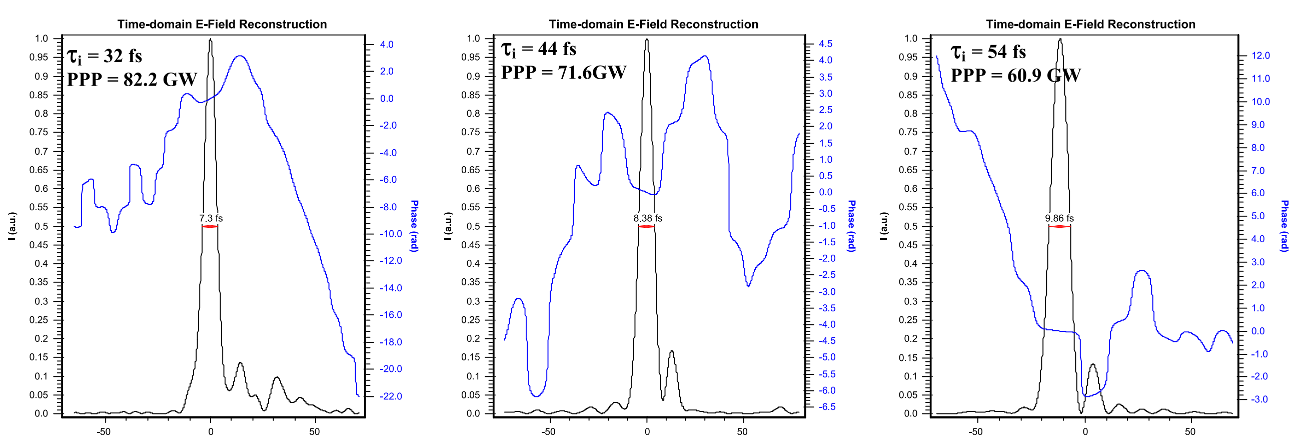


**Fig. 4.** The temporal profile change of the output pulses $\tau_i$ values of 32, 44, and 54 fs, respectively, at neon gas pressure of 2.25 atm. The compressed output pulse (black curve) and temporal phase (blue curve) of the compressed output pulses were measured using SPIDER. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

The time-domain electric field reconstructed pulse profile was observed by the SPIDER to monitor the pulse peak power of the generated ultrashort pulses. The pulse peak power value was varied by applying different pulse input pulse durations into the hollow-fiber at different neon gas pressures.

The Fourier limit of the measured pulses was characterized for each condition. During the optimization of the output pulses, the average peak power of the observed pulses reached values up to 0.1 TW. The pulse-to-pulse output power variations are about 2.5% for the final output after the compressor stage. Using a spectrometer, the output pulse was found to reach a broadband width of 350 nm as shown in Fig. 2B. This figure shows a measured spectral

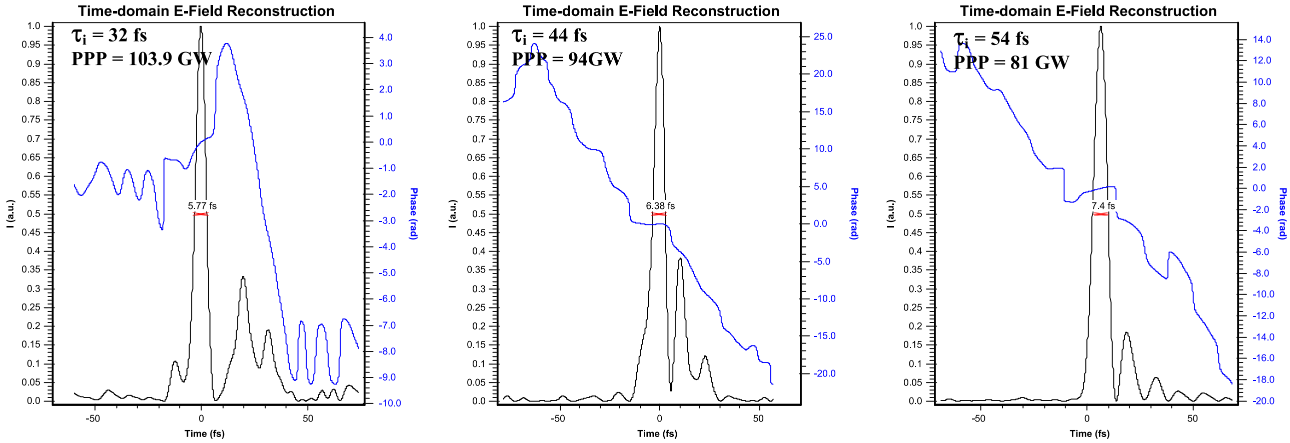


**Fig. 5.** The temporal profile change of the output pulses $\tau_i$ values of 32, 44, and 54 fs, respectively, at neon gas pressure of 2.5 atm. The compressed output pulse (black curve) and temporal phase (blue curve) of the compressed output pulses were measured using SPIDER. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

output from 600 to 950 nm using input pulses of 32 fs through neon gas filled hollow-fiber at pressure of 2.5 atm.

The observed ultrafast pulses were characterized for both the phase change and the pulse profile of these pulses in the time domain using SPIDER in the time domain. Fig. 3, shows the ultrafast pulses with durations from ca. 8.5 to 10.5 fs with the pulse peak power (PPP) values from 70.5 to 56.7 GW using different input pulse durations ($\tau_i$) gradually increasing from 32 to 54 fs at neon gas pressure of 2.0 atm. The figure reveals that the peak power is inversely proportional to the input pulse duration.

Fig. 4 describes the ultrafast pulses with different durations from about 7.3 to 9.8 fs with PPP measurements values from 80.2 to 60.9 GW for different $\tau_i$ values from 32 to 54 fs through neon gas at pressure of 2.25 atm. The observations demonstrate that the peak power values are inversely proportional to $\tau_i$ values but with an overall increment about 10% compared to the cases of relatively lower neon pressure of 2 atm.

Fig. 5, explores the ultrafast pulses with short durations from about 5.7 to 7.4 fs with PPP values from 103.9 to 81 GW for input pulse duration with values from 32 to 54 fs gradually at neon gas pressure of 2.5 atm. The observed pulses reached transform limited duration of 5.77 fs for 350 nm bandwidth (Chin, 2010). The obtained results reveal that the peak power values are inversely proportional to $\tau_i$ values with an average increment about 30% compared to the 2 atm cases.

Fig. 6, shows a summarized 2D contour representation of time-domain electric field reconstructed pulse profile with all of the observed PPP values from about 56.7 to 103.9 GW for different $\tau_i$ values in the range from 32 to 54 fs, under neon gas pressure values from 2.0 to 2.5 atm. The color pattern describes the output pulse from high peak power values (red) 103.9 GW to the lowest value (violet) at 56.7 GW.

The obtained results reveal the direct effect of increasing neon gas pressure and the input pulse duration on the output peak power values. Thus, the output pulse peak power values increases gradually with increasing the neon pressure from 2.0 to 2.5 atm. This observation can be understood since from Eq. (4) the optical bandwidth ($\delta\omega_{max}$) increases with the gas pressure i.e enhances the nonlinear SPM in the gas. Accordingly, increasing the bandwidth enables the compressor for more compression of the output beam

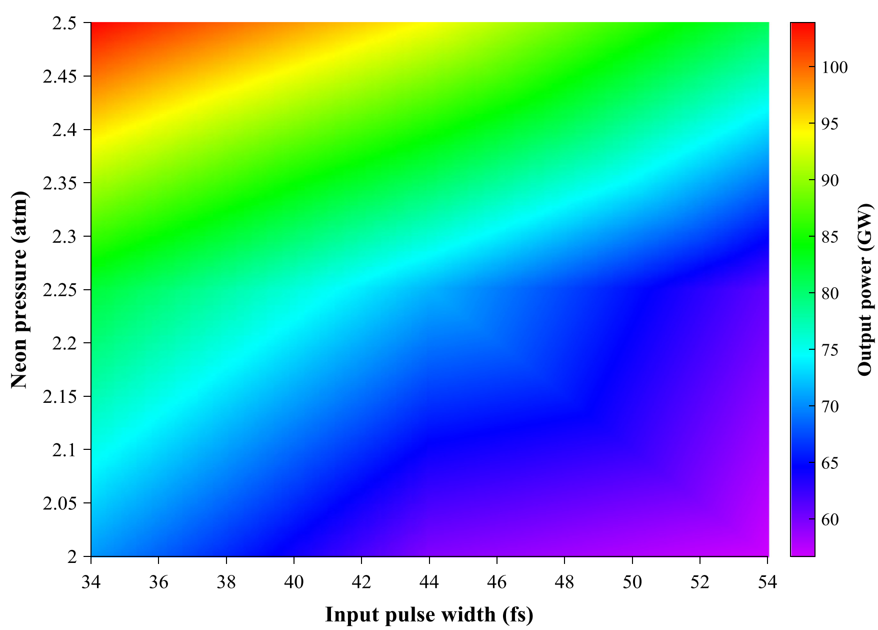


**Fig. 6.** A summarized 2D contour representation of time-domain electric field reconstructed pulse profile of the observed TLPPP values from 56.7 to 103.9 GW for different $\tau_i$ values in the range from 32 to 54 fs, and neon gas pressures from 2 to 2.5 atm. (For interpretation of the references to color in this figure, the reader is referred to the web version of this article.)

and results in short pulses with high peak power values. These results are in agreement with previously observed results, which showed that SPM increasing with neon pressure (Nisoli et al., 1996; Zhu et al., 2008; Bohman et al., 2008). Even though, the observed phenomenon of increasing the pulse-broadening due to increasing of the SPM with neon pressure is in agreement with previously observed results by others, the observed pulse-peak has reached a value of 103.9 GW which is higher than earlier observed value of 80 GW (Zhu et al., 2008).

However, the observed results revealed that the highest peak value of 103.9 GW was achieved at $\tau_i = 32$ fs without any additional chirping from the CPA compressor. It can be inferred from the results that short input pulse duration will also enhance the SPM. Detailed theoretical investigations are needed in the future studies for clear verification of the dynamics behind these ultrafast phenomena.

Moreover, the observed ultrafast pulses with high peak power values can be used to generate high harmonics in the prospective experiments. In these experiments, an extreme nonlinear process will be applied due to focusing the ultrafast pulses into a nonlinear medium (ex. neon gas). Thus, the medium-atoms driven by a strong laser field can emit high harmonics of the fundamental driving optical pulses, equal to some cutoff photon energy. The created harmonics can be of very high order, consistent with generated wavelengths in the soft x-ray region (Spielmann et al., 1997). Further, these high-harmonics produce beams of short-wavelength radiation of good spatial and temporal coherence, suitable to generate pulses with attosecond durations in gases (Antoine et al., 1996; Sansone et al., 2011). These extreme short pulses are very useful in pump-probe scheme which can be used to study molecules and atoms of interest where the 32 fs 800 nm pulses can be used as IR pump beam and the tunable white-light ultrashort pulses can be employed as probe beam for a range of transient states that can be detected by attosecond streaking method (Cirelli et al., 2015).

## 4. Conclusion

In conclusion, a method for production and control of an ultrashort white-light radiation source of transform-limited pulses with 5.77 fs and peak power reached ~0.1 TW at 1 kHz repetition rate has been developed. The characteristics of the observed pulses have demonstrated the capability of the developed method to vary the pulse peak power depending on both, the neon gas pressure inside the fiber and the input pulses durations of the CPA pulses.

This method of generation of few-cycle pulses with the feature of variable peak power will be exceptionally helpful in controlling laser-matter interaction dynamics. Moreover, the observed results are very useful in HHG, which could produce short pulses down to x-ray regime with very low pulse durations down to attosecond regime in the prospective future.

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