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# A method for controlling the bandwidth of high-energy, few-optical-cycle laser pulses tunable from the visible to the near-infrared

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**Abstract.** In this work we report a new method for controlling the bandwidth of few-cycle optical pulses, using another femtosecond laser pulses chirped in a neon-filled hollow-core fibre. The observed bandwidth varies from 25 to 234 nm in the optical wavelength region 600–950 nm. The pulse energy has for the first time reached a sub-millijoule frontier at 1 kHz. The input pulses are positively chirped using a chirped-pulse amplifier to acquire the widths 32–56 fs at the entrance of the hollow fibre. Then the pulses are highly dispersed due to a self-phase modulation in a nonlinear medium (a neon gas) followed by a pair of chirped mirrors that compensate the dispersion. We have found that this scheme allows for direct tuning of the output-pulse bandwidth while varying the chirping of the input pulses under different neon-gas pressures. Our results can give an opportunity for controlling the interactions in strong electric fields on the ultrafast time scales and are crucial for regenerating attosecond X-ray pulses.

**Keywords:** ultrafast lasers, tunable femtosecond lasers, few-cycle lasers, self-phase modulation

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## 1. Introduction

As long ago as in 1960s it has been realized that lasers should have strong impact on the spectroscopy. To begin with, they offer intense light sources with high energy densities that surpass, by many orders of magnitude, those achieved earlier with incoherent light sources. Moreover, due to their bandwidth controlled from the vacuum UV to the far IR range, single-mode lasers allow for a very high spectral resolution inaccessible in frame of the conventional methods applied in the pre-laser epoch.

High intensity and spectral monochromatism of tunable lasers have initiated a new kind of spectroscopic methods, allowing detailed exploration of structures of ‘large’ atoms and molecules [1]. In many aspects, the laser absorption spectroscopy resembles the microwave spectroscopy, where carcinotrons or klystrons represent tunable coherent radiation sources, analogues of tunable lasers. In other words, the laser spectroscopy can transfer many advantages and techniques adopted from the microwave spectroscopy into the UV, VIS or even the IR spectral ranges [2]. The benefits of the absorption spectroscopy based on the tunable lasers include, in particular, the absence of need in monochromators, because the absorption coefficient and its frequency dependence can be determined directly from the difference between the intensities of the reference and transmitted beams. The detector noises can be often ignored because of high spectral power densities of lasers,

as a consequence of laser-beam collimation. Furthermore, long absorption paths can be analyzed using multiple back-and-forth reflections inside multiple-path absorption cells. Finally, it is possible to tune the laser wavelength very fast over the whole spectral region to match the molecular absorption. In this way rapid relaxation or transient absorption processes can be investigated, using short-pulse lasers with the time resolutions lying in the femtosecond domain [3, 4].

Novel developments of the femtosecond laser technology have enabled it to become a powerful tool in the fields of materials diagnostics and ultrafast spectroscopy [5–8]. Combining nonlinear optical techniques with the intense ultrashort pulses obtained from amplified solid-state lasers, one can generate widely tunable femtosecond light pulses ranging from the near UV to the IR [8–12]. Among many interesting developments, the nonlinear optical interactions with self-phase modulation, which generate supercontinuum in the optical region using two gas-filled microstructured hollow fibres, have attracted much attention of researchers [13–16]. Here essentially nonlinear pulse compression relies on the interaction of Kerr nonlinearity and quadratic dispersion [17]. These approaches are basically nonlinear and can produce increasing in the pulse bandwidth, which cannot be directly obtained with purely linear schemes [18]. According to this significant property, shorter transformation-limited pulses can be achieved, using a compression of pulses with nonlinear pulse-compression techniques [19]. Being widely applied in the nonlinear fibre optics, the pulse compression is based upon to the two different methods, a soliton effect and a fibre grating effect [20]. The fibre compressors are typically preferred in the VIS and near-IR optical ranges, while the compressors built on the soliton effect are often used in the IR range (1.3–1.6  $\mu\text{m}$ ) [21]. Meanwhile, both of the compressor types can be united for the IR wavelengths located in the vicinity of 1.3  $\mu\text{m}$ , to yield high compressions that offer some prospects for achieving the shortest possible pulses [22].

In 2008, Cirimi et al. [23] have used a complicated system to generate tunable few-optical-cycle pulses via a non-collinear phase-matching geometry optical parametric amplifier (OPA). Their construction contains two OPAs, the first of them delivering a phase-stable idler that experiences spectral broadening through a 3 mm-thick sapphire plate to generate supercontinuum spectra. The OPA is based on a BBO crystal enabling tunability of the output wavelength from 500 to 1600 nm by tuning the crystal angle. The main problem of the device is, however, that the pulse energy does not exceed 5  $\mu\text{J}$  [24]. Three years later, the same authors have modified their system and demonstrated a number of different OPA schemes which generate ultra-broadband pulses with the tunable wavelength (1000–3000 nm) and the pulse duration 8.5 fs, although the pulse energy has been 1–2  $\mu\text{J}$  only [25]. In 2012, J. Darginavicius et al. have demonstrated another configurations with the tunable wavelength and the energy as high as 100  $\mu\text{J}$ . One of these schemes works in the VIS range (530–720 nm) and the others can be tuned for the middle-IR and the near-IR ( $\lambda_{\text{MIR}} = 1.6\text{--}7.2 \mu\text{m}$  and  $\lambda_{\text{NIR}} = 0.9\text{--}1.6 \mu\text{m}$ , respectively), depending on the operational characteristics of the blue-pumped non-collinear phase-matching OPA. They have suggested using different crystals ( $\text{LiIO}_3$ ,  $\text{LiNbO}_3$  or BBO) in the non-collinear phase-matching OPA systems to enable differently tuned wavelength regions. Recently G. Cerullo et al. reported [26] on the development of ultra-broadband tuneable few-optical-cycle lasers based on the other OPA schemes. In this case the OPA system represents a so-called ‘degenerate’ OPA, in which the signal and the idler have the same frequency, or a non-collinear OPA. These systems are pumped with either fundamental frequency or second harmonics of an amplified Ti:sapphire laser (800 nm) having the repetition rate of 1 kHz. Tunability from the VIS to the IR is achieved with the crystals having relatively high refractive indices (e.g., periodically poled stoichiometric lithium tantalate)

and different thicknesses (1–3 mm). Nonetheless, the authors of Ref. [27] have claimed that the output pulse energy is still limited to the microjoule regime.

In this work, we suggest a new direct method for generating tuneable-bandwidth (from the VIS to the near-IR) laser pulses of which energies reach a sub-millijoule range for the first time. This is two orders of magnitude higher than the output of all the other complicated techniques known up to date [23–27]. The characteristics reported above have been achieved by generating super-continuum in a hollow fibre filled with neon gas. The wavelength tunability has been controlled with the gas pressure. We have also studied the effect of varying chirping of the pulses injected into the fibre on the bandwidth of the output pulses.

## 2. Experimental setup

An optical diagram of a table-top tunable ultrafast laser system is shown in Fig. 1. The amplification stage includes a regenerative kHz amplifier that produces high-power femtosecond pulses (2.5 W, 32 fs and the wavelength 800 nm). A 15 fs mode-locked Ti:sapphire seed oscillator (400 mW and 75 MHz at 800 nm) generates femtosecond laser pulses in the TEM<sub>00</sub> mode. The basic oscillator setup consists of pump beam mirrors, folded cavity mirrors, a pump beam-focusing lens, a pair of concave spherical mirrors aligned with the Ti:sapphire laser rod, an output coupler, metal-coated mirrors, a set of prisms used for the dispersion compensation, and a slit as a spectral tuning element.

The seed oscillator can be tuned from 780 to 820 nm. The oscillator is pumped by a green CW diode-pumped solid-state laser Opus (4 W and 532 nm; available from Laser Quantum). Since the output Ti:sapphire energy is limited to the nanojoule level, an additional amplification stage is needed. We have selected the chirped-pulse amplification since it offers the pulse energies of the order of millijoules at the kilohertz repetition rate [28]. It is worth noticing that the chirped-pulse amplification applied to the ultrashort solid-state laser pulses can raise the powers of the generated optical pulses up to gigawatt frontiers, thus making the technique favourite for the high-field lasers and the ultrafast spectroscopy [29–32]. Before chirped-pulse amplifying, a stretcher is used for stretching out the seed pulses temporally and spectrally in order to reduce the peak-power level and avoid destruction of the gain medium [33]. A strongly dispersive element is employed in the stretcher according to a standard two-path scheme, combining a single diffraction grating with a telescope formed by a flat mirror and a spherical mirror equipped with broadband dielectric coatings.

The chirped-pulse amplifier involves multiple passing through an amplifying gain medium located within an optical resonator. The latter comprises an optical switch for controlling the number of round trips and for reaching very high overall gains. The switch is composed of a Pockels cell combined with a quarter-wave plate and a thin-film polarizer. A Faraday isolator is used for separating the input and output pulses. It represents a passive optical device made of magneto-optic material, which preserves the linear polarization of light and rotates its polarization plane by 45° for the beam going in the forward direction and, due to a non-reciprocal character of that rotation, by additional 45° for the backward direction.

After passing through a Faraday isolator and changing the polarization from horizontal to vertical, the stretched pulses are injected into a regenerative amplifier at predetermined times, using a Pockels cell as an optical switch. Then, after a necessary number of round trips in the resonator, the energy of the laser pulse reaches its maximum value (higher than 3 mJ). The amplified laser pulses are released from the regenerative-amplifier resonator by a second optical

switch (i.e., a second Pockels cell). That is done by applying short bell-shaped high-voltage pulses to the Pockels cell that acts as a quarter-wave plate. The regenerative amplifier is pumped by a diode-pumped Q-switched green pulse Nd:YLF laser (the wavelength 527 nm, the energy 20 mJ, the duration 170 ns, and the repetition rate 1 kHz). It is a vertically polarized laser model DM20-527 available from Photonics Industries.

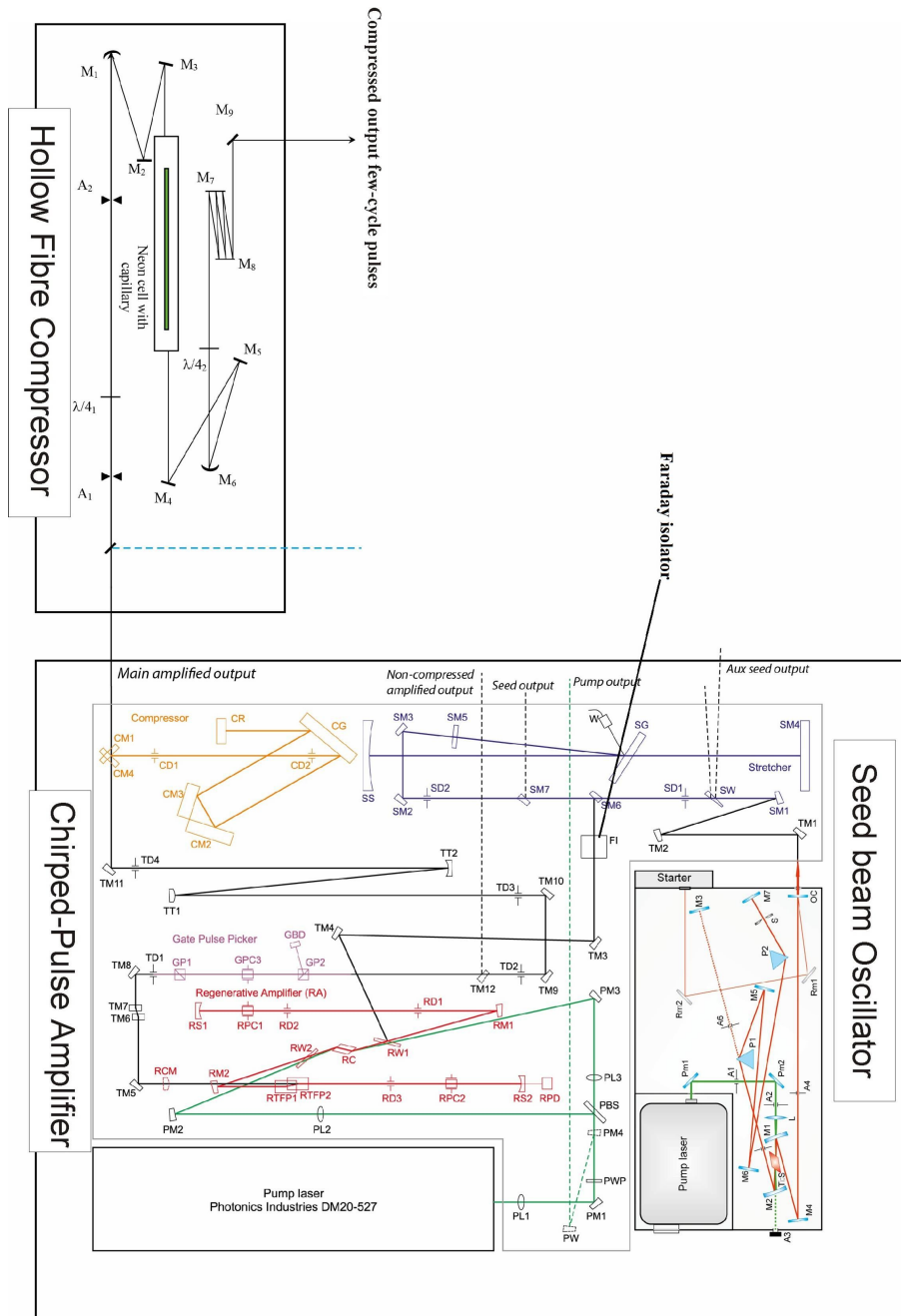
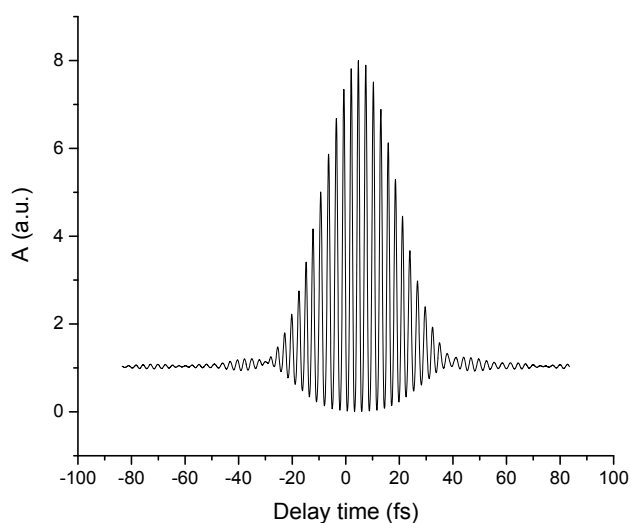


Fig. 1. A schematic setup of our tunable ultrafast laser system: RTFP – thin-film polarizer, RW – Brewster window, RPC – Pockels cells, RD – aperture, PD – photodiode, RC – Ti:sapphire crystal, GP – prism polarizer, CG – compressor grating, and SG – stretcher grating.

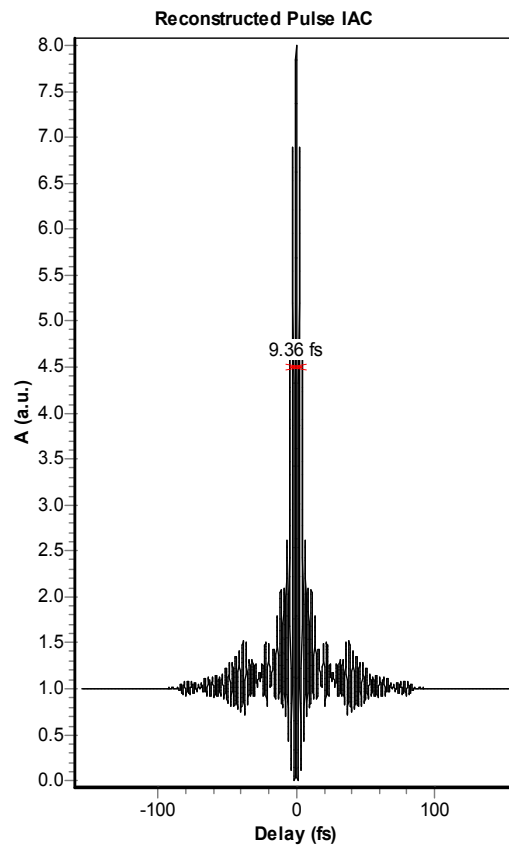
Having passed the regenerative amplifier, the laser pulse is sent to a gate subsystem consisting of two crossed polarizers and a third Pockels cell placed in between. This ‘pulse picker’ is employed to control the laser output by an external gate signal and to enhance the contrast. Finally, the pulse is compressed back to the femtosecond regime via a compressor. To prevent any damage of optical elements of the compressor, the beam is expanded by a telescope composed of mirrors TT1 and TT2. The compressor itself involves an input mirror, a diffraction grating, a mirror assembly, a ‘roof’ mirror assembly, and an output mirror CM4. The laser beam strikes the grating four times inside the compressor. Behind the compressor, the laser pulses reach the duration of 32 fs at the energy 2.5 mJ and the repetition rate 1 kHz. Then the amplified pulse is directed to a final stage to produce the few-cycle pulse regime. We call this stage as a ‘hollow-fibre compressor’. The mentioned compressor is composed of a one-metre hollow fibre filled with a neon gas, followed by a multilayer chirped-mirror compressor. With the aid of a concave mirror with the focal length  $f=1.2$  m, the laser radiation is focused into a fused-silica fibre with the inner diameter 250  $\mu\text{m}$ . The pulse spectrum gets broader via the self-phase modulation in the neon gas at a controlled pressure (up to 2.5 atm). Finally, the output beam is collimated by a concave mirror and then compressed after a six-path trip between the two chirped mirrors, thus reaching a few-cycle regime.

### 3. Results and discussion

The oscillator beam has been optimized in a mode-locked regime at 95 nm and the bandwidth 52 nm. The full width at half maximum (FWHM) of the pulse is found to be 18 fs at the energy 4 nJ (see Fig. 2). The wavelength of the oscillator beam can be tuned in the wavelength region 750–850 nm, using a variable optical slit and a couple of prisms. The Ti:sapphire emission is spread spatially and the wavelength can be tuned by varying the horizontal location and the width of the slit. It is worthwhile that the narrow bandwidth of the gain medium used by us restricts creating directly the few-cycle pulses. To avoid this fundamental limitation, we have employed an external spectral broadening due to the hollow fibre and the consequent pulse compressor (see Fig. 1). On the other hand, the amplified femtosecond pulses can be chirped in the compressor by changing the separation distance between the gratings. Then the chirped output pulses have the average power of 2.5 W at the repetition rate 1 kHz and the pulse energy 2.5 mJ.



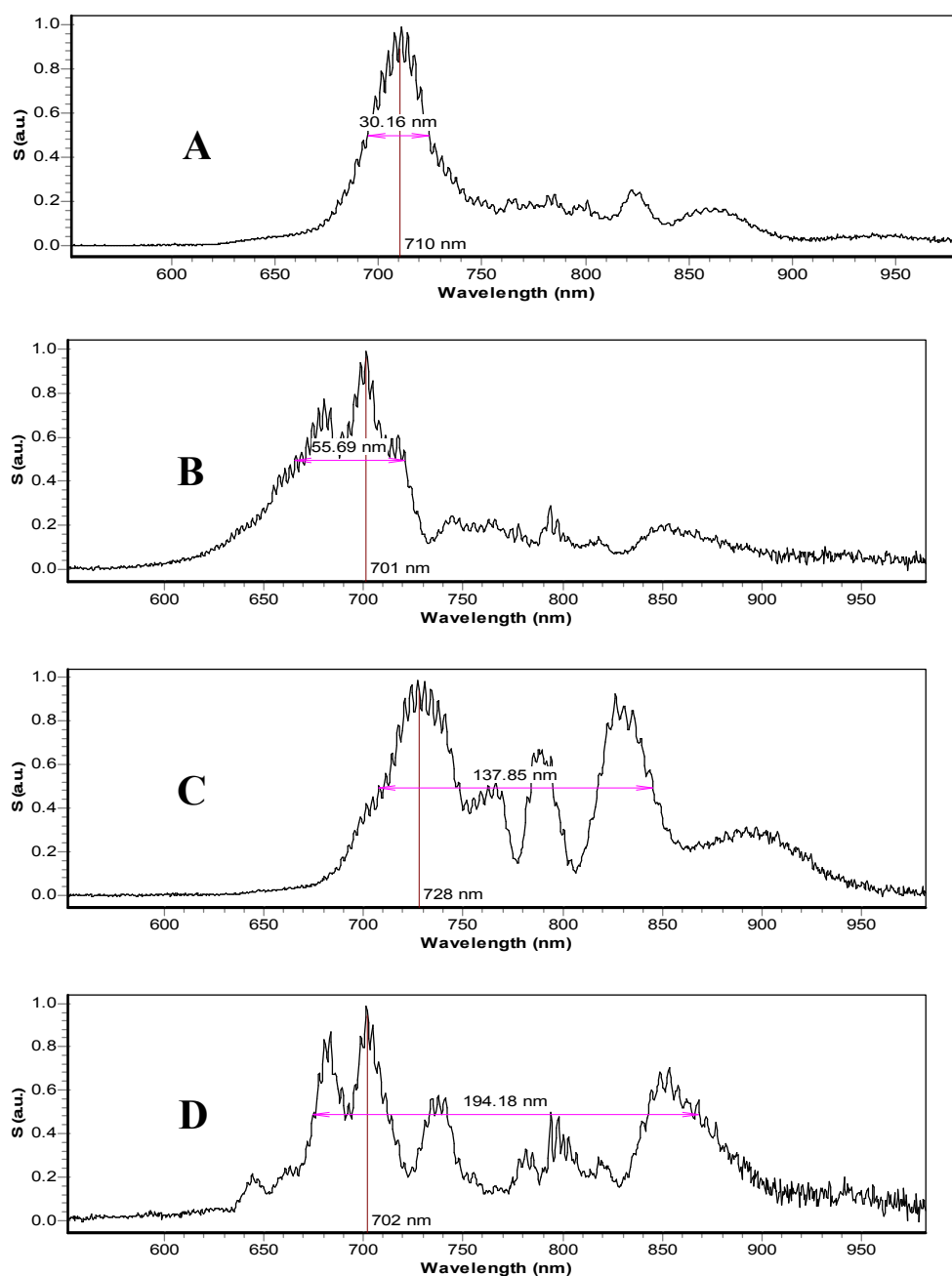
**Fig. 2.** Autocorrelation data measured for Ti:sapphire oscillator (18 fs at 795 nm, and the bandwidth 47 nm). A is the intensity.



**Fig. 3.** Output-pulse profile estimated using the intensity autocorrelation function: the FWHM value measured (9.36 fs) corresponds to the transformed limited pulses as short as 4.6 fs. IAC is the intensity autocorrelation function.

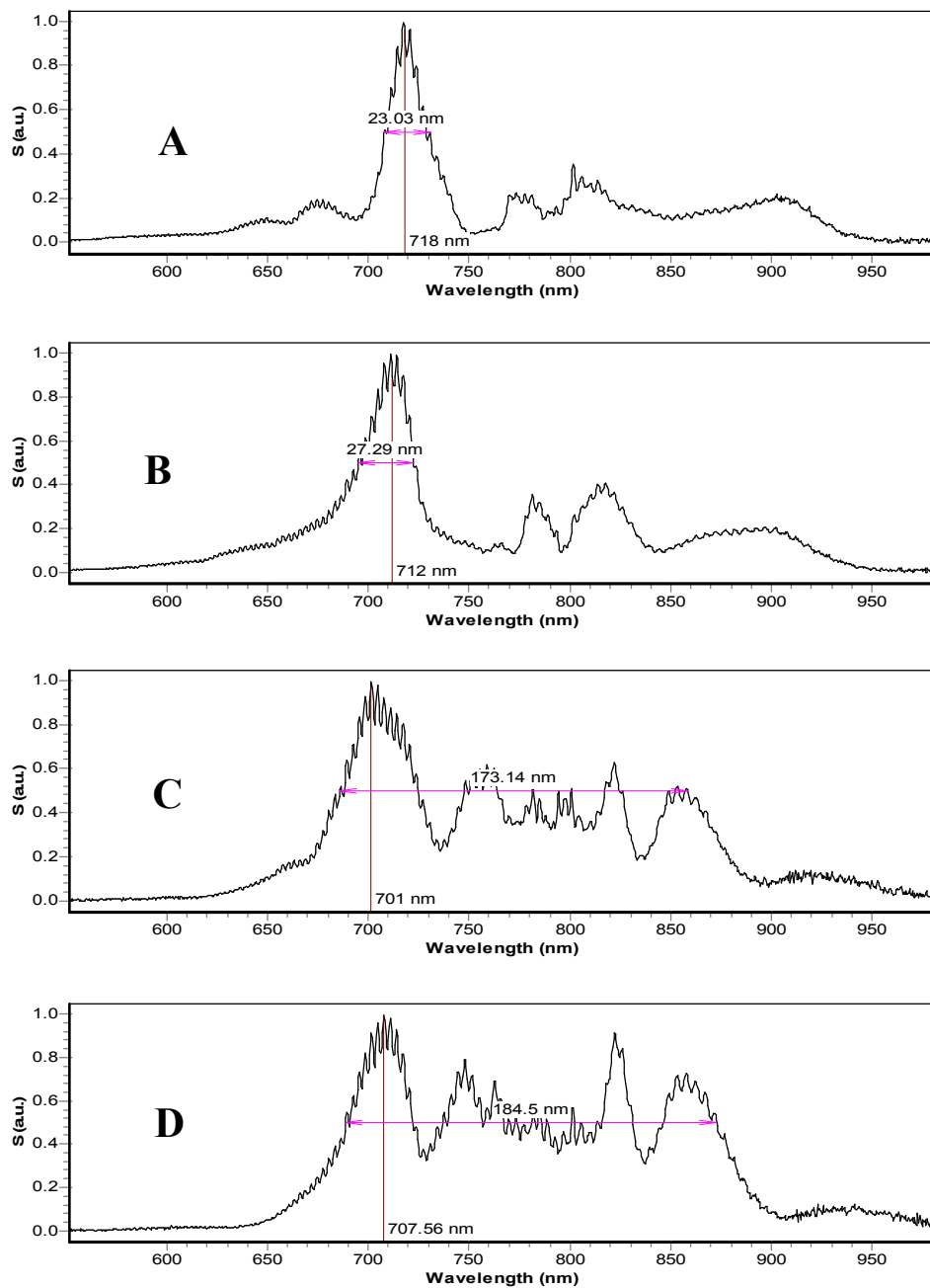
The properties of the ultrafast pulses at the output of our system are affected by many parameters, including the nonlinear phase shift due to generation of supercontinuum in the hollow fibre filled with neon gas, the pressure of that gas, and the efficiency of the regenerative-amplifier output. The final compressed pulses are found to reach the energies as high as 0.6 mJ at the repetition rate 1 KHz. Fig. 3 shows the autocorrelated intensity function for the ultrafast output, which has been measured with a so-called spectral phase interferometry for direct electric-field reconstruction. As seen from Fig. 3, the FWHM of the pulses is found to be 9.34 fs, which corresponds to the transformed limited pulses as short as 4.6 fs [34].

We have also studied the effect of varying amplified-pulse chirping (from 32 fs to almost 56 fs) on the final output of the system, under conditions of different neon pressures. Fig. 4 shows the variations in the output-pulse bandwidth and the peak wavelength occurring when the input-pulse chirping changes from 32 to 54 fs. Here the Ne pressure is kept fixed at 2.00 atm. The peak intensities are observed at the wavelengths 701–728 nm and the bandwidth changes from 30.16 to 194.18 nm. We have found that, under the given pressure, the position of the peak intensity is approximately fixed at 710 nm, with only slight changes of the order of  $\pm 2\%$ . On the contrary, the bandwidth changes by almost 6.5 times when the incident-pulse chirping increases from 32 to 54 fs.



**Fig. 4.** Output-pulse bandwidths and peak wavelength measured for the cases of different input-pulse chirping (a neon-filled hollow-core fibre at the pressure 2.00 atm): (A) 32, (B) 44, (C) 47 and (D) 54 fs.  $S$  is the spectral intensity.

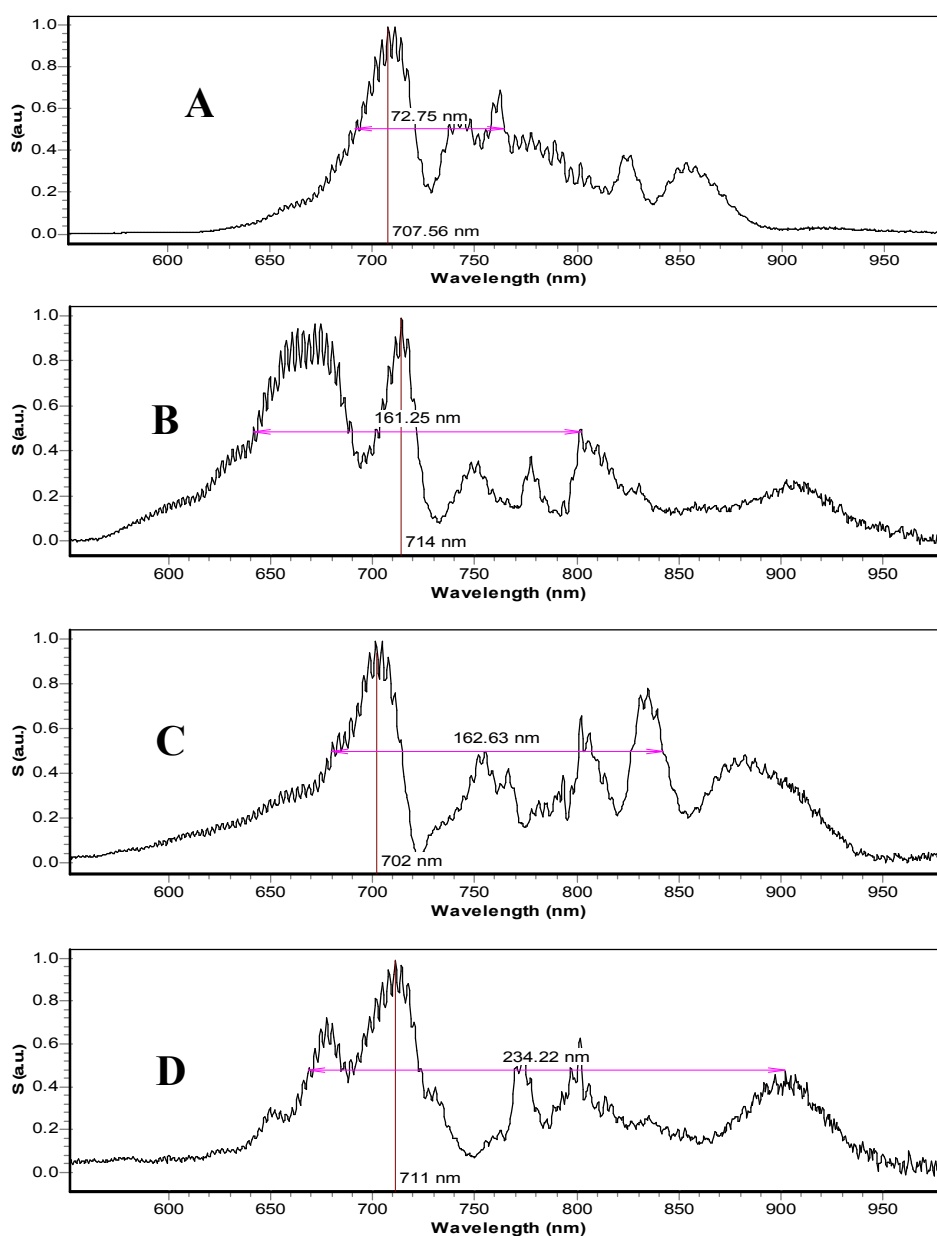
Fig. 5 shows variations in the bandwidth of the output pulse and its peak wavelength, which take place due to changing chirping of the input pulse (from 32 to 54 fs) under somewhat higher neon pressure, 2.25 atm, inside the hollow fibre. The peak intensities are again located in the wavelength region 701–728 nm and, as in case of the pressure 2.00 atm, the bandwidth changes from 30.16 to 194.18 nm.



**Fig. 5.** Output-pulse bandwidths and peak wavelengths measured for the cases of different input-pulse chirping (a neon-filled hollow-core fibre at the pressure 2.25 atm): (A) 32, (B) 47, (C) 50 and (D) 53 fs.

Finally, we have found that the bandwidths of the output pulses are much wider at still higher neon pressure (2.50 atm), as compared with the parameters obtained at 2.00 or 2.25 atm. Fig. 6 shows that the output bandwidth starts with 72.75 nm at the input-pulse chirping 32 fs and increases up to 234.2 nm at 54 fs. In this case the peak intensity is demonstrated to be approximately fixed near 707 nm, with only a small relative variation (0.5%). Notice that the bandwidth becomes very broad (almost 250 nm), beginning from the wavelength of 650 nm and ending at 900 nm (see Fig. 6).





**Fig. 6.** Output-pulse bandwidths and peak wavelengths measured for the vases of different input-pulse chirping (a neon-filled hollow-core fibre at the pressure 2.50 atm): (A) 32, (B) 37, (C) 47 and (D) 54 fs.

#### 4. Conclusion

Summing up the main results of the present work, we have demonstrated a new table-top setup and a corresponding technique that enable producing tunable, broad-band, ultrafast, few-cycle optical pulses in the wavelength region 600–950 nm. The pulses achieve high enough energies close to the sub-millijoule frontiers at the repetition rate 1 kHz. Notice that these characteristics have been obtained for the first time and with relatively simple methods. Our scheme reveals the ability to control the bandwidth from 23 to almost 250 nm. Our results seem to be very important since they facilitate a sufficient wavelength tunability greatly needed while investigating the transient

interactions of ultrafast pulses with different molecular-energy states. Moreover, the ultrafast tunable pulses can turn out to be very useful in generation of higher optical harmonics, which can be further used to create shorter pulses in the attosecond regime with shorter wavelengths covering the UV-to-X-ray range.

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**Анотація.** У цій роботі представлено новий метод керування шириною смуги пропускання оптичних імпульсів тривалістю у кілька періодів із використанням додаткових частотно модульованих фемтосекундних лазерних імпульсів, які поширюються крізь волокно з серцевиною, заповненою неоном. Показано, що ширина смуги пропускання змінюється від 25 до 234 нм для оптичного діапазону 600–950 нм, а енергія імпульсу сягає суб-міліджоулевої межі при 1 кГц. На вході волокна імпульси модулювали за частотою у прямому напрямку від 32 до 56 фс. Після цього імпульси ставали суттєво диспергованими самомодуляцією в нелінійному середовищі (газі неоноу), а після нього слідувала пара модульованих дзеркал, які компенсували дисперсію. Виявлено, що ця схема дає змогу прямо перестроювати ширину смуги пропускання вихідного імпульсу шляхом зміни модуляції вхідних імпульсів при різних тисках газу. Одержані результати вказують на можливість контролю взаємодій у сильних електричних полях за умови надшвидких процесів і мають вирішальне значення для відновлення аттосекундних рентгенівських імпульсів.