

Optics Letters

Single thin-plate compression of multi-TW laser pulses to 3.9 fs

SZABOLCS TÓTH,^{1,†} ROLAND S. NAGYMIHÁLY,^{1,†*}  IMRE SERES,¹ LEVENTE LEHOTAI,² JÁNOS CSONTOS,¹ LÁSZLÓ TAMÁS TÓTH,¹ PRABHASH PRASANNAN GEETHA,¹ TAMÁS SOMOSKÖI,¹ BARNA KAJLA,¹ DÁNIEL ABT,¹ VIKTOR PAJER,¹  ARNOLD FARKAS,¹ ÁRPÁD MOHÁCSI,¹ ÁDÁM BÖRZSÖNYI,¹ AND KÁROLY OSVAY²

¹ELI ALPS Research Institute, ELI-HU Non-Profit Ltd., Wolfgang Sandner utca 3, 6728 Szeged, Hungary

²National Laser-initiated Transmutation Laboratory, University of Szeged, Dugonics tér 13, 6720 Szeged, Hungary

*Corresponding author: roland.nagymihaly@eli-alps.hu

[†]These authors contributed equally to this work

Received 13 October 2022; revised 18 November 2022; accepted 22 November 2022; posted 22 November 2022; published 16 December 2022

Post-compression of 12-fs laser pulses with multi-TW peak power from an optical parametric chirped pulse amplification (OPCPA) system was performed by using a single thin fused silica plate in a vacuum. By optimizing the input pulses in both spatial and temporal domains, after compression with customized chirped mirrors, we achieved pulses as short as 3.87 fs, in combination with 12-mJ energy. The spatio-spectral quality of the post-compressed pulses was thoroughly analyzed. The generated 1.4-cycle pulses pave the way for next generation attosecond and particle acceleration experiments. © 2022 Optica Publishing Group

<https://doi.org/10.1364/OL.478253>

State-of-the-art laser systems are able to generate few-cycle laser pulses with tens of mJ of energy, providing peak intensities in the range of 10^{18} – 10^{19} W/cm² [1]. Generation of isolated attosecond pulses from gaseous and solid-state media requires driver pulses composed of only few-to-single electric field oscillations, while the phase difference between the carrier wave and the pulse envelope (carrier-envelope phase, CEP) is kept stable [2,3]. Recent studies show that few-cycle laser pulses may significantly improve the efficiency of the particle acceleration, as well [4,5]. Hence, there is an increasing demand on laser pulses with octave spanning bandwidth and tens to hundreds of mJ of pulse energy, providing peak intensities above 10^{20} W/cm² on the target. Since the output spectrum of laser systems is limited by the amplification stages and the stretcher-compressor arrangements, third-order nonlinear optical phenomena are often used in a controlled way to increase the available spectral bandwidth, and to obtain much shorter pulses after dispersion compensation. This process is called post-compression, which can be performed in hollow-core fibers, multipass cells, or multiple thin plates. The hollow-core fiber technique was efficiently used in the last decade to obtain few-to-single-cycle pulses with Ti:Sa-based driver sources in the sub-mJ to few-mJ output energy range [6,7]. Multipass cells are extremely efficient by combining them with a few-hundred-fs Yb-based driver lasers, which

can provide output pulses from few tens of fs even down to 6 fs [8–10]. Energy scalability is, however, the weak point of the hollow-fiber and the multipass cell techniques due to damage threshold and ionization limitations with already <20-fs-long driver pulses.

Efficient post-compression of linearly polarized pulses down to few-cycle duration was demonstrated by spectral broadening through a sequence of solid thin plates [11–14], where the intensity in the nonlinear medium typically reaches 2–5 TW/cm². The key of this technique is the controlled utilization of self-focusing and diffraction repetitively, while the nonlinear medium is kept thin enough not to introduce filamentation and significant dispersion. In connection to energy scaling, simulation and experimental work has been already conducted by using thin plastic films [15]. Plastics, however, are rather easily exposed to thermal damage, limiting their application to single-shot or 1-shot/min operation. Using solid thin plates with high-energy pulses at reasonably high repetition rates up to 1 kHz seems to be a good candidate for energy scaling of post-compression and to obtain single-cycle pulses with multi-TW driver sources. Increasing the transmitted pulse energy to the tens of mJ range requires significant efforts regarding the necessary vacuum environment and spatial beam characteristics. A recent study has already demonstrated that, with a reasonably good beam profile, pulses with even multiple joules of energy can be compressed from 23 fs to approximately 10 fs [16].

In our experiment, the SYLOS experimental alignment (SEA) laser system was used as the driving source. The system, based on optical chirped pulse amplification stages only, produces 45-mJ pulses with <12-fs full width at half maximum (FWHM) duration at 852-nm central wavelength and at 10-Hz repetition rate [1]. The partially compressed, 0.5-ps-long laser pulses enter the vacuum beam transport system, so that the B-integral due to the vacuum window is negligible. Prior to the experimental stage, the pulses are fully compressed by chirped mirrors (PC1705, Ultrafast Innovations).

Inside the chamber, the pulses are focused by a concave mirror of 4-m focal length (Fig. 1, FM1), after which they are reflected

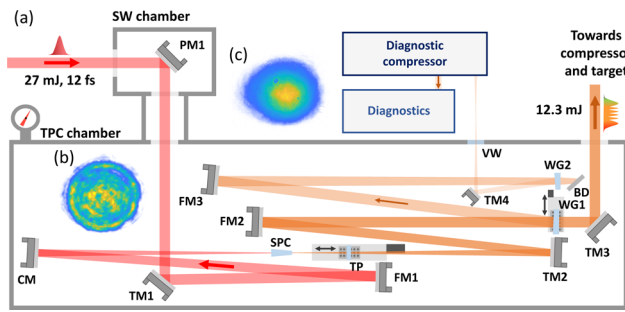


Fig. 1. (a) Schematic view of the experimental setup with a single thin plate. PM1, piezo-controlled turning mirror; TM1–TM4, enhanced silver flat turning mirrors; FM1–FM3, focusing mirrors; TP, thin fused silica plate; SPC, spatial filtering glass cone; VW, thin vacuum window; WG1 and WG2, fused silica wedges; BD, beam dump; CM, chirped mirror. (b) Input NF and (c) spatially filtered intensity distribution on the thin plate.

from a chirped mirror (CM1512, Ultrafast Innovations) with $+100\text{-fs}^2$ group delay dispersion (GDD; Fig. 1, CM) to reach final compression. Upon our first attempt to compress the input pulse propagating through a $500\text{-}\mu\text{m}$ -thick fused silica (FS) plate installed in an intermediate field, we experienced not only spectral broadening but also a distorted spatial beam profile. The inhomogeneities shown in Fig. 1(b) were more pronounced in the spectrally broadened beam, while the spectral content also changed significantly along the beam cross section. To improve the beam profile prior to compression, the input beam was spatially filtered by a glass cone with entrance and exit diameters of 1 mm and 0.23 mm , respectively (Fig. 1, SPC). By using the deformable mirror at the end of the SEA laser, we optimized the focal spot at the tip of the cone. This way, a Gaussian intensity distribution was achieved with a reasonably high, 50% transmission [Fig. 1(c)].

After spatial filtering, the pulses are propagating through a thin FS plate as the nonlinear medium, which has a thickness of $500\text{ }\mu\text{m}$ and AR coating on both sides for the spectral range of $550\text{--}1150\text{ nm}$. The spectrally broadened pulses are collimated by a concave mirror of 4-m focal length (Fig. 1, FM2), after which they are propagating to the next vacuum chamber for final compression. Compression is achieved by using a pair of custom tailored complementary chirped mirrors (PC2007, Ultrafast Innovations) optimized for the spectral range of $550\text{--}1200\text{ nm}$, and propagation through 2.7 mm of FS. During the alignment of the setup, a $4''$ -sized FS wedge is moved into the beam path (Fig. 1, WG1). The uncoated front surface reflection at close to 0° angle of incidence (AOI) of WG1 is then refocused by a concave mirror of 3-m focal length (Fig. 1, FM3), and the convergent beam is reflected from another FS wedge's uncoated front surface at close to 0° AOI (Fig. 1, WG2). The still convergent and two times sampled beam is then sent out of the vacuum chamber through a 1-mm -thick FS vacuum window AR coated for the $500\text{--}1200\text{-nm}$ spectral range. The beam is re-collimated in air by a concave mirror with a focal length of 1 m . Finally, the pulses are compressed by a pair of PC2007 type chirped mirrors and FS, identical to those in the main beam.

Input pulses in the TPC chamber were characterized by using the chirp scan technique in the TPC chamber. The dispersion of the pulses was tuned by using the acousto-optic programmable dispersive filter (DAZZLER, Fastlite) in the OPCPA system.

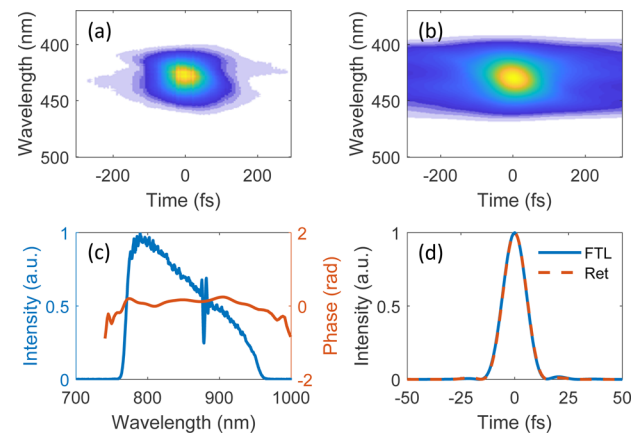


Fig. 2. (a) Measured and (b) retrieved chirp scan traces, (c) measured spectral intensity and retrieved phase, (d) reconstructed pulse (dashed orange line) compared to the FTL intensity distribution (solid blue line) in the case of input pulses.

Retrieved pulses were as short as 11.6 fs , practically equal to the Fourier transform limited (FTL) duration (Fig. 2).

The broadened spectrum was measured with an imaging spectrograph (MISS-Large-Broadband, FemtoEasy), and linear spectrometers in the visible and near infrared ranges (Broadcom Qmini and Qred types), which were coupled by joint fibers (Fig. 3). All three devices are calibrated in both wavelength and intensity. The beam at the measurement point was coupled to the imaging spectrograph directly, and to the linear spectrometers by placing a piece of lens paper as a diffuser in front of the fiber entrance. Spectral broadening was optimized by changing the position of the TP. The visible and near infrared spectra were matched manually in the spectral region around 950 nm in intensity. As a comparison, the 2D spectrum of the driver laser pulses was recorded in the laser laboratory, also for the spatially filtered chirped pulses in the diagnostics after the TPC chamber at full energy [Figs. 3(a) and 3(b)]. Signatures of diffraction in the 2D spectrum originate from the edges of a not sufficiently large enough mirror in the diagnostic line, which slightly cuts the edge of the low-intensity part of the beam [Fig. 3(c)]. Detection of the imaging spectrometer is limited to 1100 nm on the red side of the spectrum. The combination of the visible and NIR spectrometers, however, was able to detect the complete broadened spectrum, which spans from 500 to 1350 nm at the 10^{-3} intensity level [Fig. 3(d)]. The broadest spectrum was achieved when TP was 43 cm from the focus of FM1, which represented a peak intensity of 7.4 TW/cm^2 on the plate. The mechanisms responsible for the spectral broadening were further analyzed by an in-house developed $3 + 1\text{D}$ numerical simulation code [17]. Based on the good agreement between the broadened spectrum obtained from the simulation and from the experiment, we are convinced that the new spectral components are generated by self-phase modulation and self-steepening, accompanied by weak self-focusing, while ionization in the TP is negligible [Fig. 3(c)].

Spatial intensity distribution of different spectral components was analyzed by measuring the FF with a CCD camera (CMOS-1.001-Nano, Cinogy Technologies). The measured FF profiles with different bandpass filters (types 65177, 65179, 65184, 87824, Edmund Optics) are shown in Fig. 4 together with the full bandwidth profile.

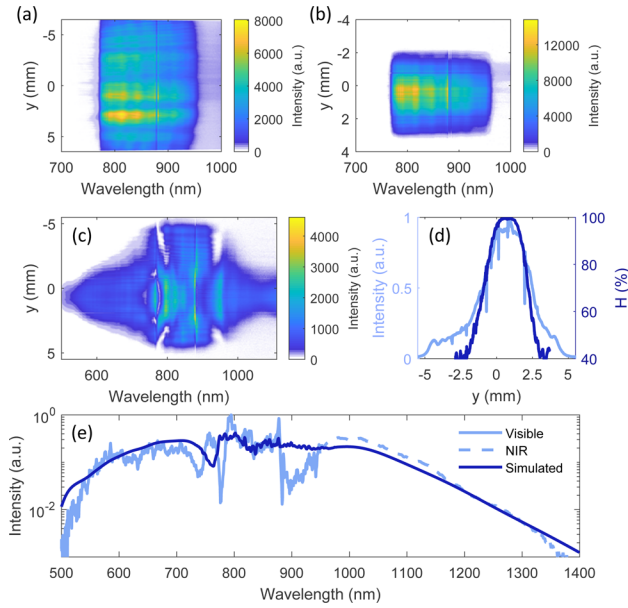


Fig. 3. (a) 2D spectrum measured at the laser exit of the sampled pulses in the vertical direction (b) without and (c) with spectral broadening on a linear scale. (d) Integrated beam profile (light blue) and spectral homogeneity (dark blue). (e) Linear spectrum measured by the combined visible and NIR spectrometers (solid and dashed light-blue lines) compared to the result of numerical simulation (solid blue line).

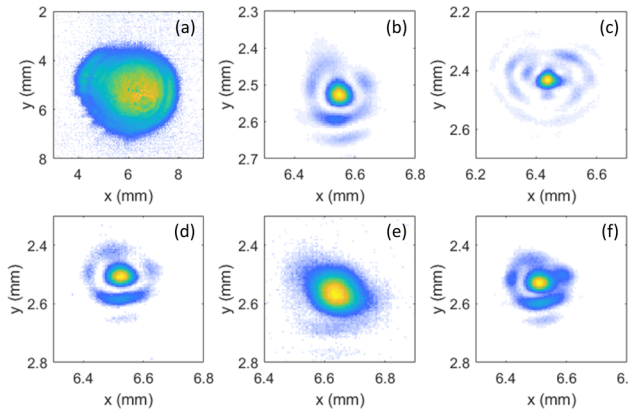


Fig. 4. (a) Measured near-field profile in the image plane of the thin FS plate, far-field profiles with (b) 766-nm, (c) 800-nm, (d) 905-nm, (e) 1050-nm bandpass filters, and (f) with full bandwidth at the focus of FM3.

The bandpass filters are designated with their central wavelength. The near field of the spectrally broadened pulses was measured in the image plane of the TP, meaning that the spatial distribution of each spectral component was visualized just after the nonlinear propagation [Fig. 4(a)]. It needs to be noted that the detector cuts the spectral components above 1100 nm. This means that the total bandwidth profile misses the intensity of the spectral components above the detection limit.

Temporal characterization of the compressed pulses was performed by using a customized D-scan device (D-cycle NIR, Sphere Ultrafast Photonics) designed for the wavelength range of 500–1250 nm. Calibration of the device is made in such a

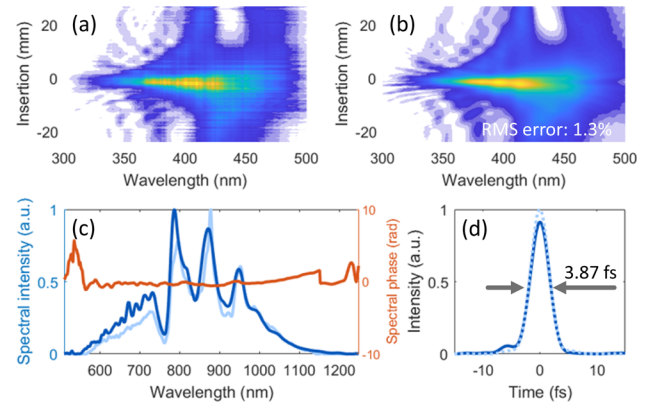


Fig. 5. (a) Measured and (b) retrieved D-scan traces. (c) Measured (light blue) and retrieved (blue) fundamental spectrum with retrieved spectral phase. (d) FTL (dashed light blue line) and retrieved (dark blue line) temporal intensity profile of the compressed pulse.

way that a 1-mm-thick FS vacuum window and 1 m of propagation in air is subtracted from the measurement, resulting in a compressed pulse at the sampling point in the vacuum chamber. The central part of the beam was coupled to the measurement device by using an iris filtering the outer edge of the beam. Results of the D-cycle measurement are showing a compressed pulse duration of 3.87 fs with a retrieval error of only 1.3% rms, very close to the FTL duration of 3.74 fs (Fig. 5).

The sharp spectral features were not possible to resolve by the D-cycle as efficiently as in the case of the imaging and linear spectrometers (Fig. 3). Nevertheless, the agreement between the measured and retrieved D-cycle traces is excellent. The temporal profile shown in Fig. 5 was found to be reproducible on a day-to-day basis after minor realignments of the setup. The retrieved spectral phase of the compressed pulse shows no high order residual phase distortions. Based on our simulations, this feature can be explained by the following: a well-defined amount of self-steepening and positive GDD introduces negative third-order dispersion (TOD), which adds up to the negative TOD of the PC2007 type CMs. In total, this amount of TOD exactly compensates for all the TOD that the pulse accumulates during propagation. Hence, our configuration balances all these phase contributions efficiently, providing compressed pulses with a very high, 92.1% energy content in the main intensity lobe [Fig. 5(d)].

Robustness of our experimental configuration is provided by the vibration damped floor system under the laser and experimental laboratories, also by the exceptional passive pointing stability of the SEA laser. Consequently, without using any beam stabilization after the laser system, we could use the experimental equipment without any fine tuning for hours. Pulse energy after collimation in the diagnostic setup was measured to be 14.2 μ J on average, where an rms of 1% and a PTP of 7.1% was recorded for 1000 shots. Accounting for the reflection losses up to the measurement point, the pulse energy before WG1 was calculated to be 12.4 mJ. This turns into a pulse energy of 12 mJ after compression in the main beam, and an overall optical efficiency of 89% without the spatial filtering, and 44% with spatial filtering. We also verified long-term operation of our setup with spectral and energy stability measurements, where the pointing stability of the laser was also monitored simultaneously (Fig. 6).

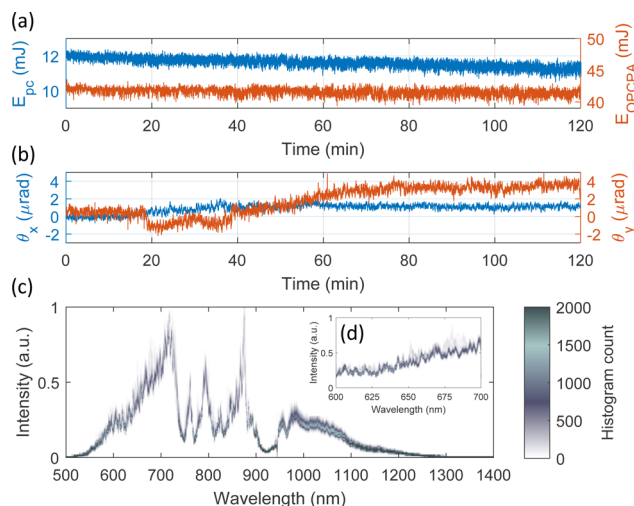


Fig. 6. (a) Measured energy stability of the spectrally broadened pulses in the diagnostic bench (blue) and at the output of the laser (orange). (b) Pointing stability of the laser in the horizontal (blue) and vertical (orange) directions. (c) Spectral stability, where all central cross sections are composed of a histogram of the intensity counts from individual shots. (d) Enlarged view of the spectral stability.

Long-term energy stability was measured to be 2.4% rms over 2 hours of operation, while a slow drift in the beam pointing of the laser was observable in both the horizontal and vertical directions [Fig. 6(b)]. FTL of the measured spectrum over the same operational period was kept in the range of 3.0 ± 0.1 fs, which represents a high long-term spectral stability.

In conclusion, we experimentally realized a thin plate post-compression arrangement for >2-TW input pulses with 12-fs duration. Spatial filtering before spectral broadening was a key step to obtain compressed pulses with high spatio-spectral quality. Spatio-spectral and focal point characterization was also performed on the post-compressed pulses. The generated 1.4-cycle pulses with 3-TW peak power are used for ion acceleration and subsequent neutron generation, expecting significantly enhanced particle yields compared to that with the initial 12-fs driver pulses. These pulses are the most energetic single-cycle pulses ever obtained, to the best of our knowledge. Energy scaling of the current setup is possible at least up to 100 mJ at the input.

Funding. ELI ALPS (GINOP-2.3.6-15-2015-00001); National Research, Development, and Innovation Office of Hungary through the National Laboratory program (NKFIH-476-4/2021, NKFIH-877-2/2020); Nemzeti Tehetség Program (NTP-NFTÖ-22-B-0077).

Acknowledgments. We greatly acknowledge the help of Sphere Ultra-fast Photonics in the temporal characterization.

Disclosures. The authors declare no conflicts of interest.

Data availability. Data underlying the results presented in this paper are not publicly available at this time but may be obtained from the authors upon reasonable request.

REFERENCES

1. S. Toth, T. Stanislauskas, I. Balciunas, R. Budriunas, J. Adamonis, R. Danilevicius, K. Viskontas, D. Lengvinas, G. Veitas, D. Gadonas, A. Varanavicius, J. Csontos, T. Somoskoi, L. Toth, A. Borzsonyi, and K. Osvay, *J. Phys. Photonics* **2**, 045003 (2020).
2. S. Kühn, M. Dumergue, S. Kahaly, S. Mondal, M. Füle, T. Csizmadia, B. Farkas, B. Major, Z. Várallyay, E. Cormier, M. Kalashnikov, F. Calegari, M. Devetta, F. Frassetto, E. Månsson, L. Poletto, S. Stagira, C. Vozzi, M. Nisoli, P. Rudawski, S. Maclot, F. Campi, H. Wikmark, C. L. Arnold, C. M. Heyl, P. Johnsson, A. L'Huillier, R. Lopez-Martens, S. Haessler, M. Bocoum, F. Boehle, A. Vernier, G. Iaquaniello, E. Skantzakis, N. Papadakis, C. Kalpouzos, P. Tzallas, F. Lépine, D. Charalambidis, K. Varjú, K. Osvay, and G. Sansone, *J. Phys. B: At. Mol. Opt. Phys.* **50**, 132002 (2017).
3. S. Mondal, M. Shirozhan, N. Ahmed, M. Bocoum, F. Boehle, A. Vernier, S. Haessler, R. Lopez-Martens, F. Sylia, C. Sire, F. Quéré, K. Nelissen, K. Varjú, D. Charalambidis, and S. Kahaly, *J. Opt. Soc. Am. B* **35**, A93 (2018).
4. D. Guénot, D. Gustas, A. Vernier, B. Beaupaire, F. Böhle, M. Bocoum, M. Lozano, A. Jullien, R. Lopez-Martens, A. Lifschitz, and J. Faure, *Nat. Photonics* **11**, 293 (2017).
5. P. K. Singh, P. Varmazyar, B. Nagy, J. G. Son, S. Ter-Avetisyan, and K. Osvay, *Sci. Rep.* **12**, 8100 (2022).
6. T. Nagy, P. Simon, and L. Veisz, *Adv. Phys.: X* **6**, 1845795 (2021).
7. T. Nagy, M. Kretschmar, M. J. J. Vrakking, and A. Rouzée, *Opt. Lett.* **45**, 3313 (2020).
8. M. Hanna, X. Délen, L. Lavenue, F. Guichard, Y. Zaouter, F. Druon, and P. Georges, *J. Opt. Soc. Am. B* **34**, 1340 (2017).
9. P. Balla, A. Bin Wahid, I. Sytceyich, C. Guo, A.-L. Viotti, L. Silletti, A. Cartella, S. Alisauskas, H. Tavakoli, U. Grosse-Wortmann, A. Schönborg, M. Seidel, A. Trabattoni, B. Manschwetus, T. Lang, F. Calegari, A. Couairon, A. L'Huillier, C. L. Arnold, I. Hartl, and C. M. Heyl, *Opt. Lett.* **45**, 2572 (2020).
10. L. Daniault, Z. Cheng, J. Kaur, J.-F. Hergott, F. Réau, O. Tcherbakoff, N. Daher, X. Délen, M. Hanna, and R. Lopez-Martens, *Opt. Lett.* **46**, 5264 (2021).
11. C.-H. Lu, Y.-J. Tsou, H.-Y. Chen, B.-H. Chen, Y.-C. Cheng, S.-D. Yang, M.-C. Chen, C.-C. Hsu, and A. H. Kung, *Optica* **1**, 400 (2014).
12. M. Stanfield, N. F. Beier, S. Hakimi, H. Allison, D. Farinella, A. E. Hussein, T. Tajima, and F. Dollar, *Opt. Express* **29**, 9123 (2021).
13. B. Zhu, Z. Fu, Y. Chen, S. Peng, C. Jin, G. Fan, S. Zhang, S. Wang, H. Ru, C. Tian, Y. Wang, H. Kapteyn, M. Murnane, and Z. Tao, *Opt. Express* **30**, 2918 (2022).
14. M.-S. Tsai, A.-Y. Liang, C.-L. Tsai, P.-W. Lai, M.-W. Lin, and M.-C. Chen, *Sci. Adv.* **8**, eabo1945 (2022).
15. S. Y. Mironov, J. Wheeler, R. Gonin, G. Cojocaru, R. Ungureanu, R. Banici, M. Serbanescu, R. Dabu, G. Mourou, and E. A. Khazanov, *Quantum Electron.* **47**, 173 (2017).
16. J. I. Kim, Y. G. Kim, J. M. Yang, J. W. Yoon, J. H. Sung, S. K. Lee, and C. H. Nam, *Opt. Express* **30**, 8734 (2022).
17. L. Lehotai, S. Tóth, V. Pajer, J. Csontos, Á. Börzsönyi, K. Osvay, and R. S. Nagymihály, in *Optica High-brightness Sources and Light-driven Interactions Congress 2022*, Technical Digest Series (Optica Publishing Group, 2022), paper HF3B.4.