Few-cycle pulse generation by double-stage hybrid multi-pass multi-plate nonlinear pulse compression

Anne-Lise Viotti$^{1,2}$, Chen Li$^1$, Gunnar Arisholm$^3$, Lutz Winkelmann$^1$, Ingmar Hartl$^1$, Christoph M. Heyl$^{1,4,5}$, and Marcus Seidel$^{1,*}$

$^1$Deutsches Elektronen-Synchrotron DESY, Notkestr. 85, 22607 Hamburg, Germany
$^2$Department of Physics, Lund University, P.O. Box 118, SE-221 00 Lund, Sweden
$^3$FFI (Norwegian Defence Research Establishment), P. O. Box 25, NO-2027 Kjeller, Norway
$^4$Helmholtz-Institute Jena, Fröbelstieg 3, 07743 Jena, Germany
$^5$GSI Helmholtzzentrum für Schwerionenforschung GmbH, Planckstrasse 1, 64291 Darmstadt, Germany
$^*$Corresponding author: marcus.seidel@desy.de

Abstract
Few-cycle pulses present an essential tool to track ultrafast dynamics in matter and drive strong field effects. To address photon-hungry applications, high average power lasers are used which, however, cannot directly provide sub-100fs pulse durations. Post-compression of laser pulses by spectral broadening and dispersion compensation is the most efficient method to overcome this limitation. Here, we demonstrate a notably compact setup which turns a 0.1GW peak power, picosecond laser into a 2.9GW peak power, 8.2fs source. The 120-fold pulse duration shortening is accomplished in a two-stage hybrid multi-pass, multi-plate compression setup. To our knowledge, neither shorter pulses, nor higher peak powers have been reported to-date from bulk multi-pass cells alone, manifesting the power of the hybrid approach. It puts, for instance, compact, cost-efficient and high repetition rate attosecond sources within reach.

Few-cycle pulses have pushed the frontiers of nonlinear optics far beyond the perturbative regime. The (temporary) detachment of weakly bound electrons from the nuclei by strong fields leads to the creation of large electric dipole moments [1]. The atomic polarization is switched by few-cycle pulses on sub-femtosecond timescales without prior distortions of the interacting matter [1]. Many unique applications emerged, most prominent, the generation of coherent extreme ultraviolet or X-ray radiation and its temporal confinement to attosecond durations [2]. This, in turn, enabled tracking of ionization dynamics and performing electron microscopy with highest temporal and spatial resolution [3,4]. Beyond that, few-cycle pulses prospectively enable PHz bandwidth signal processing in semiconductors, dielectrics and novel quantum materials [5,6]. Initial few-cycle sources relied on broadband laser gain media that are difficult to scale in average power [1]. However, high pulse repetition rates are important to achieve good signal-to-noise ratios despite the low efficiencies of extremely nonlinear processes or limitations caused by Coulomb interactions after ionization [1]. The advance of ultrafast lasers in the past years to substantially higher average powers [7], has allowed to overcome the repetition rate short-coming of few-cycle sources, but has also imposed the challenge to reduce the inherent pulse durations of power-scalable lasers from hundreds or thousands of femtoseconds to the sub-10fs regime. One approach to accomplish this is optical parametric chirped pulse amplification [8]. It provides wavelength tunability and excellent pulse contrast but is a relatively inefficient, complex method. Alternatively, nonlinear spectral broadening and pulse post-compression present a direct, cost-efficient path to the few-cycle regime [9]. In particular, the multi-pass cell (MPC) spectral broadening technique has combined large pulse compression factors, i.e. the input to output pulse duration ratios, and high power efficiencies in an outstanding manner [10,11,12]. Recently, several few-cycle pulse generation schemes by means of MPCs have been reported [13,14,15,16,17]. However, all experiments were based on gas-filled MPCs which require hundreds of $\mu$L of pulse energies as well as a chamber that needs to be evacuated and refilled with up to several bars of nonlinear gas. In contrast, bulk material based few-cycle or even single-cycle pulse generation was demonstrated in the past years by the multiple plate continuum approach [18,19,20]. We have recently shown that combining the multiple plate and the bulk MPC techniques can clearly overcome the compression factors that are achievable by the methods alone in a single stage [21,22]. Here, we apply this novel hybrid approach to demonstrate more than hundred times duration reduction of powerful ultrashort pulses, that is from the picosecond regime to 8.2fs FWHM duration. Moreover, we report the first bulk-based MPC that delivers sub-10fs pulses with multi-GW peak powers.

The compression setup was based on an Yb:YAG laser and two spectral broadening stages (Fig. 1a). The laser and the first MPC stage (MPC 1) were similar to the setup reported in ref. [21]. The front-end of the amplifier was improved, which led to 15% more pulse energy than in [21] and pulses with down to 1ps FWHM duration. The amplifier emitted laser bursts every 100ms with a variable number of pulses and a 1MHz pulse repetition rate. We adjusted the number of pulses to the dynamic range of our measurement devices and typically worked with 150 - 200 pulses per burst. MPC 1 consisted of two quarter-wave stack dielectric mirrors with 200nm radius of curvature (ROC) and five 1mm thin anti-reflection coated silica substrates. The sixth plate used in ref. [21] mainly introduced additional chirp without lowering decisively the 43fs Fourier transform limit (FTL) of the MPC 1 output spectrum (Fig. 2 blue line). After 68 reflections from chirped mirrors with -200fs$^2$ group delay dispersion (GDD), the
In MPC 1, the five silica plates near the cavity center formed a weak waveguide. Therefore, the beam size in the center was larger compared to the linear case. Details are provided in ref. [21]. In contrast, MPC 2 hosted only two silica plates which were located closer to the MPC mirrors than to the beam center. Consequently, the Kerr media merely added to the refractive power of the MPC mirrors causing a smaller beam waist. Nonlinear mode-matching was hence akin to gas-filled MPCs [23]. We had to separate the 1 mm thin silica plates in MPC 2 by about 22 cm to preserve the compressibility of the pulses. The spectrum measured after 7 roundtrips of the 75 µJ, 46 fs pulses is plotted in Fig. 2 (red line). The corresponding 7.4 fs FTL was enabled by octave-spanning chirped mirrors (CMs, Laseroptik) with 200 mm ROC, which strongly reduced the net dispersion per pass in MPC 2. To suppress the GDD oscillations inherent to single broadband CMs, an MPC mirror pair with complementary dispersion design was used.

We characterized the compressed pulses by second harmonic frequency-resolved optical gating (FROG) with a 10 µm thin BBO crystal cut at θ = 29°. The dispersion-free FROG setup is described in ref. [24]. The shortest pulse duration we retrieved was 8.2 fs FWHM (Fig. 3), corresponding to more than 120 times overall reduction of the pulse duration taking the feasible 1 ps pulses from the amplifier as reference. A pair of glass wedges (Fig. 3) was used to find the best compression point. We compared the retrieved pulse durations from multiple FROG traces at different wedge positions (Fig. 3b) and obtained very good consistency of the results, such that we infer a ±0.2 fs uncertainty of the 8.2 fs duration. To our knowledge, only bulk-MPCs with at least twice as long pulses were reported before [23, 26]. We determined a pulse energy of 56 µJ after MPC 2. The corresponding 75% transmission of the stage included three bounces off silver mirrors. To minimize the reflection losses of the Kerr media, we placed the silica plates at Brewster’s angle into MPC 2. Assuming 97.2% and 99.6% reflectivity of the silver and chirped mirrors, respectively, we deduce an average Fresnel loss of 0.5% per silica-air interface. This shows that polarization rotation due to out-of-plane propagation in the MPC is a minor concern. We attribute this to the tenfold ratio between MPC length and Herriott-pattern diameter.

---

**Figure 1:** a. Two-stage pulse compression setup. Both MPC mirror pairs are separated by circa 38 cm. The compressor and MPC 2 mirrors were chirped. The silver mirrors are denoted by Ag. All other mirrors were quarter-wave stacks. Thin-film polarizers (pol) were used. b. A 200 mm beam ROC at one MPC mirror was assumed and the ROC after one pass was predicted by ABD matrices for different beam sizes. For mode-matching in presence of self-focusing, the beam radius on the mirrors in MPC 1 was reduced by about 30% (intersection blue and black lines) and increased in MPC 2 by circa 15% (intersection red and black lines) in relation to Kerr lens-free mode-matching. c. ABD matrix calculations of beam sizes in MPCs 1 and 2 for mode-matching in presence (nonlinear) and absence (linear) of the Kerr effect.

---

**Table 1: Results of the $M^2$-measurements.**

| $M^2_a$ | amplifier | MPC 1$^a$ | MPC 1$^b$ | MPC 2$^{a,c}$ |
|--------|-----------|-----------|-----------|---------------|
| $M^2_a$ | 1.16      | 1.43      | 1.28      | 1.45          |
| $M^2_b$ | 1.13      | 1.56      | 1.32      | 1.58          |

$^a$ 128.5 µJ at MPC 1, $^b$ 96.5 µJ at MPC 1, $^c$ detection up to 1.1 µm
The CM reflectivities were calculated from the broadened spectrum and the mirror design. However, we measured 76.3% transmission of a 12 roundtrip Kerr medium free MPC while we predicted 80.3% transmission from the theoretical reflectivity, implying an average 0.2% difference per pass. Nevertheless, the >99% reflectivity of the CMs is an advantage over (enhanced) silver mirrors, which have been so far used in all MPCs for sub-10 fs pulse generation. We note that the CM design exhibits a 0.6% lower reflectivity at 1030 nm than at the wings of the spectrum after MPC 2. This helps to remove several percent of the residual narrow band radiation emitted by the Yb:YAG amplifier from the compressed pulses. In fact, the autocorrelation traces of Fig. 3 show that a side pulse with 1-2 ps delay from the main peak is suppressed by 5 dB in comparison to pulses after MPC 1 which is also due to the peak power enhancement of the main pulse. From the pulse energy, the FROG retrieval, which covered a 700 fs delay range, and the autocorrelation measurement over a 10 ps range, we derive a peak power of about 2.9 GW which surpasses the present bulk-MPC record of 2.5 GW.

Owing to the small net dispersion per pass, we could readily broaden the pulse spectra to fully cover the CM reflectance band from about 0.6 µm to 1.4 µm by reducing the plate distance and increasing the number of passes in MPC 2. An experiment was conducted with 1 ps pulses from the laser, 45 fs, 61.5 µJ pulses from MPC 1, 12 roundtrips in MPC 2 and 12 cm distance between the two Kerr media. This yielded an octave-spanning spectrum with a single-cycle FTL (violet line in Fig. 2). However, a FROG measurement showed that it is not possible to compress the pulses close to the spectrum’s FTL by the CMs we used. We attribute this to spatio-temporal couplings that arose from increased intensities in the Kerr media. Tailored CMs could compensate for the characteristic bulk-broadening phase. Alternatively, the use of thinner Kerr media like in the multiple plate continuum method promises to push achievable durations in MPC 2 toward the single-cycle regime.

Figure 4 compares the experimental results (red lines) with SISYFOS simulations of MPC 2. The shortest pulses attainable for two 1 mm thin silica plates were computed in the course of the seventh roundtrip through MPC 2 omitting the need for post-compression (blue and black lines in Fig. 4). The net anomalous dispersion was about -10 fs² per pass in the simulations. The CM compressor and the glass wedges required in our setup indicate, however, that the experimental net dispersion per pass was closer to 0 fs². We attribute the small difference to the imprecise knowledge of the CM mirror dispersion which we did not measure. Nevertheless, the overall agreement between experimental and simulated spectra and pulse shapes is very good. We investigated if the GDD oscillations exhibited by a single CM are detrimental for pulse compression. The blue lines in Fig. 4 show the simulation results under consideration of both complementary mirror designs, whereas the black dashed lines show the results for considering only the averaged reflectivity and GDD of the CM pair. Only minor differences in spectrum and compressed pulse shapes are visible, and thus we conclude that the GDD oscillations of the CMs only marginally influenced the compression results. For the most part, the simulation methods are described in ref. 21.

Owing to
shorter input pulses, the Raman response of silica was included in addition to the Kerr effect. Reflectivity and GDD used in simulations were blue-shifted from the CM design by 2 THz owing to slightly lower deposition rates close to the curved mirror edges. The FROG retrieval from MPC 1 and a fundamental Gaussian were used as pulse and beam shapes, respectively. The simulated pulse energy was set to 33.4 µJ in order to match the experimental intensities in the Kerr media. First, the beam area in mode-matched MPCs scales with the M^2 factor (here 1.5). Second, Brewster’s angle of incidence results in a beam area 45% larger than for normal incidence and a 21% longer optical path through the silica plates which was also taken into account.

We eventually measured the spectral homogeneity of the experimental output beam shown in Fig. 5a with a 4f-imaging spectrograph [22, 21]. Despite Brewster’s angle orientation of the Kerr media, the horizontal (x-) and vertical (y-) beam axes exhibited a very good > 96% spectral homogeneity of the output beam as usual for MPC compression (Fig. 5b). The determined M^2 values were nearly identical to the ones after MPC 1 (Table 1).

In conclusion, we have turned a ps laser into a few-cycle light source by a sub-m^2 footprint two-stage hybrid multiplate MPC setup that yielded a record-high more than 120-fold pulse duration shortening. The demonstrated multi-GW peak power is well suited for high harmonic generation and probing other strong field phenomena. With better phase control over the attainable octave-spanning spectra and the carrier-envelope offset, a compact MHz rate attosecond source is in reach.

Acknowledgements. We thank Cord Arnold (Lund University) and Tobias Groß (LASEROPTIK) for fruitful discussions and DESY (Hamburg, Germany), a member of the Helmholtz Association HGF, for the provision of experimental facilities. A.-L. V. acknowledges support from the Swedish Research Council (Vetenskapsrådet grant No. 2019-06275).

References

[1] T. Brabec and F. Krausz, “Intense few-cycle laser fields: Frontiers of nonlinear optics,” Reviews of Modern Physics 72, 545–591 (2000).

[2] I. Orfanos, I. Makos, I. Lioutos, E. Skantzakis, B. Förä, D. Charalambides, and P. Tzallas, “Attosecond pulse metrology,” APL Photonics 4, 080901 (2019).

[3] M. F. Ciappina, J. A. Pérez-Hernández, A. S. Landsman, W. A. Okell, S. Zherebtsov, B. Förä, J. Schötz, L. Seiffert, T. Fennel, T. Shaaraan, T. Zimmermann, A. Chacón, R. Guichard, A. Zair, J. W. G. Tisch, J. P. Marangos, T. Witting, A. Braun, S. A. Maier, L. Roso, M. Krüger, P. Hommelhoff, M. F. Kling, F. Krausz, and M. Lewenstein, “Attosecond physics at the nanoscale,” Reports on Progress in Physics 80, 054401 (2017).

[4] S. Mikaelsson, J. Vogelsang, C. Guo, I. Sytcevich, A.-L. Viotti, F. Langer, Y.-C. Cheng, S. Nandi, W. Jin, A. Olofsson, R. Weissenbinder, J. Mauritsson, A. L’Huillier, M. Gisselbrecht, and C. L. Arnold, “A high-repetition rate attosecond light source for time-resolved coincidence spectroscopy,” Nanophotonics 10, 117–128 (2021).

[5] S. Y. Kruchinin, F. Krausz, and V. S. Yakovlev, “Colloquium: Strong-field phenomena in periodic systems,” Reviews of Modern Physics 90, 021002 (2018).

[6] I. Jiménez-Galán, R. E. F. Silva, O. Smirnova, and M. Ivanov, “Sub-cycle valleytronics: control of valley polarization using few-cycle linearly polarized pulses,” Optica 8, 277 (2021).

[7] J. Zuo and X. Lin, “High-Power Laser Systems,” Laser & Photonics Reviews 16, 2100741 (2022).

[8] F. J. Furch, T. Witting, M. Osołodków, F. Schell, C. P. Schulz, and M. J. J. Vrakking, “High power, high repetition rate laser-based sources for attosecond science,” Journal of Physics: Photonics 4, 032001 (2022).

[9] T. Nagy, P. Simon, and L. Veisz, “High-energy few-cycle pulses: post-compression techniques,” Advances in Physics: X 6, 1845795 (2021).

[10] J. Schulte, T. Sartorius, J. Weitenberg, A. Verneleken, and P. Russbuekdlt, “Nonlinear pulse compression in a multi-pass cell,” Optics Letters 41, 4511 (2016).

[11] A.-L. Viotti, M. Seidel, E. Escoto, S. Rajhans, W. P. Leemans, I. Hartl, and C. M. Heyl, “Multi-pass cells for post-compression of ultrashort laser pulses,” Optica 9, 197 (2022).

[12] M. Hanna, F. Guichard, N. Daher, Q. Bournet, X. Délen, and P. Georges, “Nonlinear Optics in Multipass Cells,” Laser & Photonics Reviews 15, 2100220 (2021).

[13] P. Balla, A. Bin Wahid, I. Sytcevich, C. Guo, A.-L. Viotti, L. Silette, A. Cartella, S. Alisauskas, H. Tavakol, U. Grosse-Wortmann, A. Schönberg, M. Seidel, A. Trabattoni, B. Manschwetus, T. Lang, F. Calegari, A. Couairon, A. L’Huillier, C. L. Arnold,
I. Hartl, and C. M. Heyl, “Postcompression of picosecond pulses into the few-cycle regime,” Optics Letters 45, 2572 (2020).

[14] M. Müller, J. Buldt, H. Stark, C. Grebing, and J. Limpert, “Multipass cell for high-power few-cycle compression,” Optics Letters 46, 2678 (2021).

[15] P. Rueda, F. Videla, T. Witting, G. A. Torchia, and F. J. Furch, “8 fs laser pulses from a compact gas-filled multi-pass cell,” Optics Express 29, 27004–27013 (2021).

[16] L. Daniault, Z. Cheng, J. Kaur, J.-F. Hergott, F. Réau, O. Tcherbakoff, N. Daher, X. Délen, M. Hanna, and R. Lopez-Martens, “Single-stage few-cycle nonlinear compression of millijoule energy Ti:Sa femtosecond pulses in a multipass cell,” Opt. Lett. 46, 5264–5267 (2021).

[17] S. Hädrich, E. Shestaev, M. Tschernajew, F. Stutzki, N. Walther, F. Just, M. Kienel, S. P. Jójárt, Z. Bengery, B. Gilicze, Z. Várallyay, A. Börzsönyi, M. Müller, C. Grebing, A. Klenke, D. Hoff, G. G. Paulus, T. Eidam, and J. Limpert, “Carrier-envelope phase stable few-cycle laser system delivering more than 100 W, 1 mJ, sub-2-cycle pulses,” Optics Letters 47, 1537 (2022).

[18] M. Seidel, F. Pressacco, O. Akcaalan, T. Binhammer, J. Darvill, N. Ekanayake, M. Frede, U. Grosse-Wortmann, M. Heber, C. M. Heyl, D. Kutyakhow, C. Li, C. Mohr, J. Müller, O. Puncken, H. Redlin, N. Schirmel, S. Schulz, A. Swiderski, H. Tavakol, H. Tümmermann, C. Vidoli, L. Wenthaus, N. Wind, L. Winkelmann, B. Manschwetus, and I. Hartl, “Ultrafast MHz-Rate Burst-Mode Pump–Probe Laser for the FLASH FEL Facility Based on Nonlinear Compression of ps-Level Pulses from an Yb-Amplifier Chain,” Laser & Photonics Reviews 16, 2100268 (2022).

[19] M. Hanna, L. Daniault, F. Guichard, N. Daher, X. Délen, R. Lopez-Martens, and P. Georges, “Nonlinear beam matching to gas-filled multipass cells,” OSA Continuum 4, 732 (2021).