Generation of 1.5 cycle pulses at 780 nm at oscillator repetition rates with stable carrier-envelope phase

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Abstract: We demonstrate a spectral broadening and compression setup for carrier-envelope phase (CEP) stable sub-10-fs Ti:sapphire oscillator pulses resulting in 3.9 fs pulses spectrally centered at 780 nm. Pulses from the oscillator with 2 nJ energy are launched into a 1 mm long all-normal dispersive solid-core photonic crystal fiber and spectrally broadened to more than one octave. Subsequent pulse compression is achieved with a phase-only 4f pulse shaper. Second harmonic frequency resolved optical gating with a ptychographic reconstruction algorithm is used to obtain the spectral phase, which is fed back as a phase mask to the shaper display for pulse compression. The compressed pulses are CEP stable with a long term standard deviation of 0.23 rad for the CEP noise and 0.32 rad for the integrated rms phase jitter. The high total throughput of 15% results in a remaining pulse energy of about 300 pJ at 80 MHz repetition rate. With these parameters and the ability to tailor the spectral phase, the system is well suited for waveform sensitive photoemission experiments with needle tips or nanostructures and can be easily adapted to other sub-10 fs ultra-broadband Ti:sapphire oscillators.

1. Introduction

In contrast to atomic gases [1] and bulk materials [2], pulse energies of hundreds of pJ suffice to enter the extremely interesting strong-field regime of photoemission in the case of metal nanostructures [3–8] and metal-like 2D materials [9]. The reason for this is near-field enhancement in the case of nanostructures and a vanishing or small bandgap for 2D materials. Studying field-driven electronic processes with higher temporal resolution is not only of fundamental interest, but may also allow the creation and control of electronic devices consisting of metallic nanostructures, semiconductors or 2D materials, driven at optical frequencies [10–12]. Precise control, however, requires a high contrast between adjacent field extrema within the waveform, in other words pulses ideally as short as a single optical cycle with high repetition rates for easily detectable signals. In particular for needle tips, only CEP-stable single-cycle pulses will allow the creation of clear, isolated attosecond electron pulses. These isolated electron pulses require a single time window for emission, which leads to full suppression of the multi-photon peak structure in the energy spectra of the emitted photo-electrons [13].

Two-cycle pulses with energies of a few nJ can currently be provided by commercial, CEP-stabilized Ti:sapphire oscillators, making them an ideal initial source to extend to the sub-two-cycle regime. CEP-stabilization by an f-2f interferometer requires octave-wide spectra. Such systems usually provide pulses with sub-10 fs duration and energies of 2-3 nJ. At these low output energies, a high nonlinearity is required in order to broaden the spectrum and support shorter pulse durations. Solid-core photonic crystal fibers (PCFs) can provide this [14, 15] although a
few other systems, such as bulk diamond, can be used under tight focusing conditions [16].

Besides spectral broadening, shortening of the pulse duration requires an appropriate compression scheme. One option that combines spectral broadening and pulse compression in a fiber is soliton self-compression, which has been shown to generate pulses with 4.6 fs duration at the fiber output [17]. However, soliton-self compression is very sensitive to the precise fiber length and input pulse parameters and produces asymmetric pulse shapes with pedestals. In the case of all-normal dispersive photonic crystal fibers, spectral broadening is less sensitive to the exact pulse parameters, and works easily also for broad initial spectra. It has also been shown numerically that spectral broadening in the normal dispersion region is more stable against phase and intensity fluctuations of the input pulses [18]. However, this comes at the cost of external pulse compression to reduce the pulse duration.

For external pulse compression many different methods have been developed. Using chirped mirrors alone, pulse durations of 5.0 fs have been reported [19]. Ti:sapphire oscillators with pulse durations close to 10 fs at lower repetition rates of tens of MHz can provide higher initial energies. For example, starting with 16 nJ pulse energy and 9 fs long pulses at 24 MHz repetition rate, 4.0 fs has been demonstrated using just a standard single-mode fiber [20]. By spectral decomposition and individual dispersion compensation using chirped mirrors, followed by recombination, the spectral range has been extended, yielding pulses as short as 1.0 fs [21] in amplified systems – in principle also applicable to oscillators. It is problematic, however, that both the waveguide dispersion and the additional nonlinear phase need to be accounted for. Hence accurate dispersion compensation is only possible with a custom broadband chirped mirror design, especially in the case of strong waveguide dispersion, as is the case in solid-core fibers. Another typical approach relies on 4f pulse shapers to correct almost arbitrary spectral phase distortions and to reshape the intensity spectrum. This approach, together with an all-normal dispersive fiber, could provide 4.5 fs pulses with phase-only shaping, and 3.64 fs pulses if the intensity spectrum is additionally flattened. Although reshaping the intensity spectrum can reduce the transform limit, it also reduces the throughput significantly. In [22], only 45 pJ pulse energy remained.

In this manuscript, we report efficient, sub-4-fs pulse generation with stable CEP and a remaining pulse energy of about 300 pJ at an overall throughput of 15%. Our approach relies on an all-normal dispersive solid-core photonic crystal fiber with zero-dispersion wavelength of 800 nm and small core diameter of 1.5 µm, allowing more than octave-spanning spectra with low phase distortion for 1 nJ pulse energy coupled into the fiber. A prism-based pulse shaper with a phase-only spatial light modulator and chirped-mirror-based post-compression, covering a bandwidth from 500 to 1050 nm, is used to compress the output pulses to 3.9 fs duration. Finally, we investigate the CEP stability of the setup with respect to the seed oscillator used.

2. Experimental setup

In the experiment we obtain nearly single-cycle pulses by compressing the output of a CEP stable Ti:sapphire oscillator (Venteon Pulse:One PE) delivering 6.1 fs pulses at 800 nm with 6 nJ energy at 80 MHz repetition rate. The experimental setup is depicted in Fig. 1. Half of the laser output is used to stabilize the CEP and track its slow drift. For the stabilization we use a home built (in-loop) f-2f interferometer, which provides the feedback that drives an acousto-optic modulator (AOM) to adjust the power of the pump laser and thus the CEP. We monitor the slow CEP drift with an out-of-loop f-2f interferometer, but do not actively correct this drift at the moment.

The other half of the oscillator output is first broadened in a solid-core PCF and then compressed using a prism-based 4f-pulse shaper followed by chirped mirrors. Prior to broadening the pulse duration is optimized using chirped mirrors (Laserquantum DCM 7) followed by a fused silica wedge pair. A reflective telescope expands the beam to a 1/e² intensity radius of \( w = 4 \) mm. The resulting 2 nJ pulses are launched into the fiber using a 90-degree off-axis parabola with protected silver coating and 15 mm effective focal length. An identical parabola is used to collect
Fig. 1. Schematic layout of optical setup. Laser source: CEP-stabilized Ti:sapphire oscillator with dispersion management by a chirped mirror pair (CMP) and a wedge pair (W). Beam splitter (BS) supplies CEP-lock and experiment. Broadening stage: Coupling of expanded beams from telescope (T1) to solid-core photonic crystal fiber (PCF) with fiber output collimation by off-axis parabolic mirror (OAP) and beam diameter reduction by second telescope (T2). 4f-setup: prisms (P) and concave mirrors (CM) generate the Fourier-plane for spectral phase correction by a spatial light modulator (SLM). Post-compression: additional dispersion compensation by double-angle chirped mirror pairs (DA-CMP) before pulses are sent to diagnostics.

and collimate the light at the output of the PCF. Both off-axis parabolae are mounted on 3D translation stages along with the fiber, which can be tilted. After careful optimization we typically achieved at least 50% transmission through the PCF, including the launch efficiency. Before entering the 4f-pulse shaper [23] the beam waist is reduced to $w = 1.2$ mm to minimize the material dispersion introduced by the prisms in the shaper.

Pulse shaping requires access to the spectral phase and intensities, which are available in the Fourier-plane of the 4f-setup. For this the pulse is first decomposed into its spectral components by a SF10 prism. After this a concave mirror (focal length $f = 750$ mm, placed at distance $f$ away from the prism) focuses the individual spectral components on to a computer controlled liquid crystal spatial light modulator (Jenoptik SLM-S 640), which is placed at position 2f. For our application a single display SLM is sufficient and allows shaping of the spectral phase. The spectral components are then recombined by the inverted sequence of components. Material dispersion caused by the prisms and SLM display result in long output pulses and would require strong correction by the shaper. However, post-compression by double-angle chirped mirrors (UltraFast Innovations PC70) can compensate for most of this dispersion and can be fine-tuned by varying the insertion length of the prism. In this way the required corrections by the shaper are minimized, although the chirped mirror bandwidth limits the spectral range from 500 nm to 1050 nm. The shaper geometry is designed such that this wavelength range exactly covers the active area of the SLM display. The throughput of the shaper is around 30% which, combined with the 50% throughput of the broadening stage, results in about 300 pJ at the output of the compression setup.

The diagnostics consists of an optical spectrum analyzer, a home-built second harmonic generation frequency resolved optical gating (SHG-FROG) device and an f-2f interferometer, allowing us to measure the intensity spectrum, the pulse duration and the CEP-stability.
3. Spectral broadening in a small mode area solid-core photonic crystal fiber

We used a fused silica solid-core PCF with a pitch $\Lambda = 1 \, \mu m$ and a hole diameter $d = 0.54 \, \mu m$, designed to provide small normal dispersion in the vicinity of 800 nm. Figure 2(a) shows a scanning electron micrograph (SEM) of the PCF end-face and Fig. 2(d) the group velocity dispersion $\beta_2$ of the PCF calculated using the empirical model described in [24]. Achieving sufficient spectral broadening to support single cycle pulses without imprinting strong spectral phase distortions is a challenging task, requiring careful choice of fiber length.

This was done with the help of numerical simulations based on the generalized nonlinear Schrödinger equation (GNLSE) for an effective mode area of $A_{\text{eff}} = \pi (\Lambda - d/2)^2$ and including both the shock effect and the delayed Raman-response [18]. Furthermore, we measured the phase and amplitude profile of the pulse at the fiber input by spectral phase interferometry for direct electric-field reconstruction (Venteon SPIDER) and used it in the numerical simulations. To validate the numerical code, in Figs. 2(b) and 2(c) we compare measured and simulated spectra after propagation through 2 mm of PCF, plotted as a function of input energy. Simulation and experiment agree quantitatively: both the bandwidth and most spectral features are well reproduced. Furthermore, they show that, even for input energies as low as 0.3 nJ, the spectrum already covers more than one octave at 20% of the peak spectral intensity.

In Figs. 2(e) and 2(f) the spectral and temporal evolution of a pulse with energy 1 nJ are plotted as a function of position along the PCF. The white dashed lines indicate the spectral range supported by the pulse shaper. The pulse reaches maximum spectral broadening within less than 1 mm, after which it rapidly stretches in time, the peak power drops and no further spectral broadening is possible. Numerical simulations predict that after 1 mm propagation, the output pulse duration is around 20 fs. We selected this length as it is practically manageable and gives an acceptable phase distortion.
4. Feedback mechanism and pulse compression result

Compressing the pulses with the shaper requires precise knowledge of the spectral phase. We obtained this from the SHG-FROG trace measured after the compression setup, and retrieved with the ptychographic algorithm [25]. FROG is set up in a non-collinear geometry using only reflective optics and a 5 µm thick beta-barium borate (BBO) crystal cut for maximum second harmonic efficiency at 600 nm. The cutting angle was chosen also to support the short wavelength spectral region down to 500 nm. The ptychographic method is more robust in reconstructing complicated traces than the generalized projection based approach [26], especially as it allows the use of a fixed intensity spectrum during the retrieval, which is easily measured separately with a spectrometer. The retrieval with fixed spectrum has been applied in estimating the spectral phase used for the feedback. The pulse duration is first optimized by changing the prism insertion position while the phase mask is deactivated. By doing so we can obtain a pulse duration of around 22 fs, but with considerable remaining spectral phase distortions and a complex temporal structure. The remaining phase distortions can be reduced by feeding back the retrieved spectral phase to the spatial light modulator. FROG does not directly provide the correct sign for the time axis and thus the spectral phase. By testing both possibilities, this ambiguity can be removed as only the correct sign results in compressed pulses.

The resulting trace after the feedback is given in Fig. 3(a) with its corresponding reconstruction in Fig. 3(b). The experimental trace was obtained via merging two measurements with different integration time. For the second one, the spectrum in the Fourier plane of the shaper was reduced to avoid overlapping this with the high frequency components of the fundamental spectrum (1050-1200 THz). In order to obtain a comparison between measured and retrieved spectrum, we do not fix the spectrum in the retrieval as done for the feedback. Both the measured spectrum and FROG trace agree well with the retrieved ones as shown in Figs. 3(a)-(c), however because of the complexity and the fine features of the trace the FROG error is 2.2% (on a 1024x1024 grid).

Spectral phase distortions are strongly reduced by the pulse shaper, resulting in a pulse duration

\[ \tau = 3.9 \, \text{fs} \]
of 3.9 fs (intensity full width at half maximum) with low pedestals shown in Fig. 3(d). As the spectral wings are yet not well compressed by the feedback scheme, the pulse duration is still significantly longer than the Fourier-limited duration of 2.8 fs. Nevertheless the measured pulse duration is among the shortest reported for oscillator-based systems and to our knowledge the shortest for phase-only shaping.

The theoretical limit, which we estimated following the model described in [27] and considering the pixel spacing of the SLM, the spectral separation in the Fourier plane and the actual spectral shape, is 3.0 fs and therefore slightly larger than the transform limit. The theoretical estimate indicates that with our setup we could achieve even shorter durations, however, this is difficult to measure with a single SHG-FROG trace due to spectral overlap and low signal at the spectral wings of the FROG trace. In principle this could be overcome by selecting partial, but overlapping, spectral windows in the Fourier-plane of the shaper. Although these spectral windows could be measured separately with SHG crystals optimized for each bandwidth and carefully combined to yield a better phase estimate, which is beyond the scope of this work.

5. Carrier-envelope phase stability

The frequency-resolved CEP noise of the shaper is measured using a two-arm interferometer and compared to the oscillator noise, which is recorded by a robust single-arm PPLN-based f-2f interferometer. In both cases the second harmonic is provided by a 1 mm thick PPLN crystal (Covesion PPLN MSHG-1064). As the PCF output is already octave-spanning, no further spectral broadening is required, but adjusting the intensities of the fundamental and second harmonic components requires a two-arm design to obtain a strong enough beat note signal. In the case of the direct oscillator output, the PPLN slightly broadens the spectrum and directly gives a good mixture in the out-of-loop interferometer. An avalanche photo diode (MenloSystems APD 210) and electronic spectral analyzer (Rohde and Schwarz FSL18-B7) is used to measure the phase noise spectrum around the beat note, set to $f_{CEO} = 6$ MHz in this measurement.

![Fig. 4. Single-sided phase-noise spectrum (a) and integrated phase-noise starting from 200 kHz (b).](image)

Figures 4(a) and 4(b) show the phase-noise spectrum and the integrated phase-noise starting from 200 kHz to 20 Hz for the oscillator, fiber and shaper with and without active phase mask. The signal-to-noise ratio of the beat note at 100 kHz resolution bandwidth is 29 dB for the oscillator measurement and about 24 dB for all other measurements, which limits the evaluation of the integrated phase noise to upper frequencies of few hundred kHz. The lower signal-to-noise ratio after the shaper is due to the lower power of the second harmonic generated in the PPLN. Up to 200 kHz, all the noise spectra provide a sufficient signal-to-noise ratio, which was thus chosen as a common starting point for measuring the integrated phase jitter. It can be seen that
the oscillator reference and the direct fiber output show the same noise characteristics determined by the CEP-lock. Thus, neither the fiber nor the two-arm interferometer significantly affect the CEP noise. For soliton-based broadening and compression the situation would be different, because this process is very sensitive to energy fluctuations of the seed pulses, especially for long fibers and input pulse durations above 25 fs [28]. In our case the fiber and the input pulse duration are short and only self-phase modulation takes place. This, together with saturation of the spectral broadening for energies around 1 nJ as seen in Figs. 2(b) and 2(c), makes the system robust against pulse energy fluctuations. The shaper, in contrast, contributes to the low frequency noise below 300 Hz with an additional 100 mrad. The corresponding sharp edge of the integrated phase noise in Fig. 4(b) is present in all traces and gets enhanced and broadened for the shaper. As neither the fiber nor the display add noise in this frequency range, the noise source has to be present in all parts of the system. Such a common noise source would be vibrations, which are enhanced by the additional elements and long beam paths in the shaper. Air flow can additionally add noise, as the shaper is not enclosed at the moment. To our knowledge this is the first frequency-resolved noise measurement showing the origin of added noise components by a 4f pulse shaper and broadening stage – results that may be useful for designing similar setups.

![Fig. 5. Slow CEP noise after oscillator (a) and shaper (b), each with an integration time of 50 ms.](image)

For measuring slow, integrated CEP-noise we use the spectral interference method [29], which is independent of intensity noise and is implemented in a stable single-arm scheme for both the oscillator and the shaper. We limit our analysis to short term noise as it is the key quantity in the achievable CEP stability and cannot be corrected. Drifts with durations up to about 20 seconds are removed from the data using a moving mean filter. Currently we cannot actively correct long term drifts, however this can be achieved by a motorized wedge in the oscillator. The residual phase-noise follows a Gaussian distribution, as shown in Fig. 5. The standard deviation of the CEP fluctuations for the active pulse shaper is $\sigma = 0.23$ rad. Thus the whole setup increases the oscillator noise ($\sigma = 0.11$ rad) by about 100 mrad. Similar values for the short term fluctuations have been obtained for longer time windows.

Both the slow integrated, and fast frequency-resolved CEP noise, provide a useful benchmark for estimating the expected CEP-stability, depending on the specific application. Measuring electron energy spectra for example requires integration times of 10-100 ms for each electron energy bin, resulting in minutes of measuring time for each energy spectrum. Applications with very short time windows or lock-in measurements, on the other hand, are sensitive to particular noise frequency components determined by the time window or the lock-in bandwidth.
6. Conclusion

In conclusion, we have demonstrated a CEP-stable setup providing 3.9 fs short laser pulses with about 300 pJ pulse energy at 80 MHz repetition rate from a standard Ti:sapphire oscillator with an initial pulse duration of 6.1 fs by using a 1 mm long all-normal dispersive PCF and a phase-only pulse shaper. The pulse duration corresponds to 1.5 optical cycles at a central wavelength of 780 nm and is pedestal- and side-band-free. Stitched FROG measurements for various spectral bandwidths may even allow pulse durations approaching the theoretical limit of 3.0 fs. The broadened spectra cover one octave for energies as low as 1 nJ coupled to the fiber, which is crucial, as CEP-stable few-cycle oscillators usually provide only a few nJ of pulse energy. The efficient phase-only shaping permits residual pulse energies sufficient for CEP-dependent measurements at nanostructures or 2D-materials with sub-two-cycle pulses and thus enhanced temporal resolution.

Measurements of CEP stability showed that essentially only low frequency noise is added by the shaper. This additional noise is of order 100 mrad in both a frequency resolved noise measurement and a slow, integrated CEP-stability measurement and can be reduced by fully enclosing the setup and reducing vibrations. This procedure for noise characterization and the shown results can be used to estimate the CEP-noise in many different experimental situations and should help in the design of other pulse shaping setups.

As only self-phase modulation is relevant for the spectral broadening, the setup could be easily modified for various sub-10 fs Ti:sapphire oscillators by adapting the fiber length and core size to match the initial pulse duration and energy. For example, up to 1 nJ output energy could be achieved via single-pass amplification of the oscillator output [30], and using a slightly shorter fiber with a twice larger core. In conclusion we believe that the presented system is a promising extension for other oscillator systems.

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References


