CEP stable 1.6 cycle laser pulses at 1.8 µm

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Abstract: By using the novel approach for pulse compression that combines spectral broadening in hollow-core fiber (HCF) with linear propagation in fused silica (FS), we generate 1.6 cycle 0.24 mJ laser pulses at 1.8 µm wavelength with a repetition rate of 1 kHz. These pulses are obtained with a white light seeded optical parametric amplifier (OPA) and shown to be passively carrier envelope phase (CEP) stable.

OCIS codes: (320.5520) Pulse compression; (190.4970) Parametric oscillators and amplifiers.

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1. Introduction

Engineering light forms is one of the major challenges in the ultrafast optics community [1]. Expanding those capabilities from established Titanium Sapphire (Ti-Sa) wavelengths towards the infrared spectral range will be particularly useful to study ultrafast energy redistribution in the condensed phase like in the case of correlations in liquid water where the hydrogen-bonded network dephases on a sub-50 fs time scale [2]. Aiming to investigate electron correlations [3–5], the time scale of interaction has to be decreased to the sub fs time scale which is accessible via the process of high harmonic generation (HHG) [6–8].

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Furthermore, CEP stable few-cycle laser sources have been the key breakthrough for generating isolated attosecond (asec) pulses [1,9,10]. Those are generated upon recombination of laser field-driven, tunneled and accelerated electrons within two thirds of an optical cycle of the electric field oscillation [11].

Currently, CEP stabilized few-cycle laser pulses are mostly generated using ~800 nm Ti-Sa lasers, providing the basis for the synthesis of isolated asec pulses by HHG in gases [3]. CEP stabilization allows one to control the waveform of the electric field under the intensity envelope with asec precision. Using this asec control, one can restrict the generation of cut-off XUV photons within only one half-cycle leading to isolated pulses with durations near 100 asec [9,10]. Furthermore, in view of increasing the cut-off energy, it has been numerically simulated that few-cycle driving pulses enable self–phase matching in HHG where the group velocity mismatch of XUV and IR driving field is balanced by the harmonics emission phase [12]. This emission phase is determined by the phase of the recolliding electron wave packet and thus depends on varying parameters of the driving field throughout the gaseous target [13]. Alternatively, the approach of quasi-phase matching where HHG takes place in a short hollow fiber can be utilized as well [14]. Complementary to few-cycle driving laser pulses for generation of isolated asec fields, a variety of optical gating techniques has been successfully demonstrated to overcome difficulties in pulse compression [15, 16]. However, one advantage of few-cycle driving fields lies in their reduced amount of ionization, i.e. reduced depletion of the neutral gas target, before the main peak of the electric field can generate the highest energy XUV photons.

Presently, asec laser pulses are generated using XUV photons in the spectral range of 50-110 eV [9,10]. Further reduction of duration requires shorter XUV wavelengths. With HHG having a cut-off at Ip+3.17Up (Ip is the ionization potential of the atom and $Up \sim I \times \lambda^2$, I is the laser intensity and λ the wavelength of the driving laser field) [11], it is evident that further reduction of the duration of the asec pulses requires CEP stabilized few-cycle laser pulses at wavelengths longer than 800 nm [3]. In this letter, we report a simple approach for generation of 1 kHz sub-millijoule CEP stabilized 1.6 cycle laser pulses at 1.8 µm wavelength. The basic idea is to utilize narrowband high power parametric amplification in type-II beta barium borate (BBO) nonlinear crystal followed by subsequent spectral broadening in a HCF. Compression is achieved by linear propagation through appropriate bulk material like FS.

2. Experimental section

To generate intense CEP stabilized IR few-cycle laser pulses, different approaches have been investigated. They are; (1) the optical parametric chirped-pulse amplifier (OPCPA) providing sub-mJ 15.1 fs laser pulses at 2.1 µm [17], pulse self-compression by filamentation at ~2 µm [18,19], and the generation of CEP stabilized 1.5 µm few-cycle laser pulses by DFG (difference frequency generation) of few-cycle 800 nm laser pulses followed by broadband type II parametric amplification [20]. None of these approaches has been able to generate sub-millijoule sub-two cycle CEP stabilized laser pulses.

Recently, we have demonstrated a novel scheme to compress infrared laser pulses to sub-two optical cycle duration [21]. Briefly, non CEP stabilized 1.8 µm laser pulses with duration of 73 fs from a 100 Hz superfluorescence seeded OPA were spectrally broadened via nonlinear propagation in argon filled HCF. Subsequent dispersion compensation was performed by linear propagation through a 3 mm thickness (FS) window. It exhibits anomalous dispersion (zero dispersion around 1.3 µm) causing the group velocity dispersion (GVD) to be negative over the entire spectral range of the broadened spectrum. Unexpectedly, third order dispersion (TOD) being positive for all materials has not limited pulse compression with the FS window. Numerical modeling revealed that in addition to self-phase modulation (SPM) as the main driver for spectral broadening in the HCF, self-steepening also occurs causing the trailing pulse edge to become steeper than the leading one, enforcing an asymmetric temporal envelope shape. Again, this temporal asymmetry leads not only to an asymmetric power spectrum but, more important for subsequent compression with bulk material, also to an asymmetry of the spectral phase. This phase asymmetry largely balances

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the TOD of FS in the range from 1.4 – 2.2 µm [22]. The positive second order phase due to SPM is cancelled by the negative GVD of the anomalous dispersive FS. Similar approaches to balance the effect of self-steepening during nonlinear propagation in single mode fibers by grating pairs have been discussed already in mid 80's [23, 24] and were recently implemented to compensate TOD in an Yb fiber laser oscillator [25]. However, above mentioned schemes were demonstrated with multi-cycle pulses.

Pulse characterization was performed using a home-built all reflective SHG-FROG (second harmonic generation – frequency resolved optical gating [26]) which was also used for the experiments described in this paper. The design took into account all considerations for characterization of few-cycle pulses, like for instance phase matching and temporal smear, as described by Baltuška et *al.* [27]. A detailed description of the SHG-FROG apparatus can be found in Ref [21]. Confirmation of the CEP stability was carried out with a collinear f-2f interferometer [28]. A 0.25 mm thick type-I BBO (θ = 23°) was used as doubling crystal in combination with transmission polarizer (Thorlabs, LP-NIR) to project the blue spectral part of the supercontinuum generated in the HCF and the frequency doubled fundamental onto the same polarization axis.

Besides the increase of repetition rate to 1 kHz, the hereinafter described setup fulfills two important requirements towards isolated asec pulse generation, (i) we employed CEP self stabilization by means of white light seeded OPA [29] and (ii) demonstrated the capability of generating quasi single-cycle pulses of the driving laser, i.e. 1.6 cycle intensity FWHM duration which corresponds to 9.5 fs. CEP control of few-cycle pulses is necessary to generate isolated attosecond pulses [9, 10].

Figure 1 shows the schematic layout of the experimental setup comprising high energy OPA (upper part) and pulse compression stage (lower part). OPA pumping is carried out by a non CEP stabilized kHz Ti-Sa amplifier delivering 3.8 mJ, 35 fs pulses at 800 nm center wavelength (KMLabs, Boulder, USA) which is split into four paths, represented as dashed lines with corresponding numbering. The splitting ratio for the various paths is indicated in Fig. 1 as percentage rate for the reflected part of the beam. All nonlinear BBO crystals in the OPA are cut for type-II phase matching at θ = 28°. Passive CEP stabilization is achieved via (DFG) of the seeding white light continuum in path 1 (gray line in Fig. 1) carrying the same CEP fluctuations as the split fraction of the pump beam in path 2. As a result of DFG via three wave interaction in the first BBO 1, three beams, pump, Signal and Idler herein after identified by the subscripts p, s and i, emerge from the crystal. Conservation laws for energy ($\omega_p = \omega_s + \omega_I$) and momentum ($\vec{k}_p = \vec{k}_s + \vec{k}_I$) have to be fulfilled with ω denoting the center frequency and $\vec{k}$ the wave vector. The latter requirement is also referred as the phase matching condition

$$\Delta\vec{k} = \vec{k}_s + \vec{k}_I - \vec{k}_p = 0 \tag{1}$$

which becomes increasingly difficult to satisfy as the bandwidth increases to support shorter pulses. Violating the phase matching condition causes temporal and spatial chirp, but can be minimized using non-collinear interaction with tilted pulse fronts. This approach has been used by several research groups to generate ultra broadband laser pulses [30, 31].

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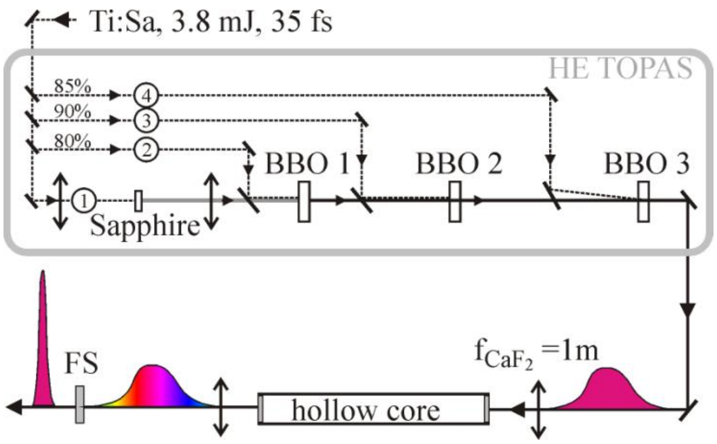

Fig. 1. Schematic of the pulse compression experimental setup. Ti-Sa laser pulses with 35 fs duration and 3.8 mJ of energy are frequency shifted to 1.8 µm using a white light seeded Optical Parametric Amplifier consisting of three parametric amplification stages. Output pulses with energy of 650 mJ and 35 fs duration at 1.8 µm is coupled to a hollow-core fiber and subsequently compressed solely by a FS window to 1.6 optical cycles.

Because of the challenges associated with broadband parametric amplification, we developed an approach based on narrowband OPA systems delivering high peak power infrared laser pulses (HE-TOPAS from Light Conversion) followed by spectral broadening in a HCF. Generally, OPA techniques enable passive CEP stabilization because of conservation of the offset phase during parametric interaction [29]:

$$\phi_p = \phi_s + \phi_I + \pi/2 \tag{2}$$

From Eq. (2), it is evident that for equally fluctuating CEP of ϕ_p and ϕ_s, ϕ_I is intrinsically CEP stable [29]. The broadband seeding Signal generated through SPM in the Sapphire window of path 1 has the same, random, CEP as the pump beam. For the Idler after BBO 1 this phase cancels resulting in an Idler with constant CEP as long as the path length between seed and pump is maintained. To facilitate this we have modified the commercial OPA such that the Idler rather then the Signal is amplified after BBO 1 [32]. The effect of this is that interferometric stability is only required for the first gain stage between arms one and two in the Fig. 1. The CEP of the Idler is fully defined after BBO 1 and any changes in path length after this shifts the, still random, phase of the Signal while leaving the Idler unaffected. In the commercial OPA, DFG in BBO 1 is carried out in a non-collinear interaction. Considering Eq. (1), it can be seen that this causes the pump, Signal and Idler pulses to be spatially spread. Moreover, Idler pulses exhibit a spatial chirp if the intersecting pump and Signal beams contain more than a single frequency. Thus, we rearranged this stage to be collinear and selected the Idler by means of dichroic optics. This Idler beam was then amplified further in BBO 2 and 3 whereby pumping in BBO 3 is carried out under a small angle (~0.5°) to separate Signal and Idler spatially in front of the HCF. A CaF_2 plano-convex lens with 1 m focal length is used to couple the light into the HCF being supported on an aluminum V-groove. Both fiber ends are attached to closed gas cells with 1 mm thickness, anti-reflection coated CaF_2 windows to minimize material dispersion. At the output, the laser beam was collimated using a f=1 m concave silver mirror. The HCF is evacuated and filled with argon at a constant pressure of 1.6 Atm.

3. Results and discussion

At the output of the OPA, after BBO 3, 650 µJ of pulse energy at 1.77µm was available at a repetition rate of 1 kHz. Pulse intensity duration of 35 fs at full width half maximum has been characterized using SHG-FROG. The collimated beam exhibits an $1/e^2$ intensity width of

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about 10 mm. In comparison with our previous experiments using superfluorescence seeded OPA [19], we now start with already shorter input pulses (35 fs vs 73 fs) for spectral broadening in the HCF. During propagation of the laser pulses in the HCF, the duration at the output remains almost unchanged. The broadened spectra extend from 1.15 µm to 2.15 µm and the measured spectral density is shown in Fig. 2(c) (gray line). An asymmetric shape with a wider extension towards the blue spectral side is observed due to self-steepening during nonlinear propagation. The spectral sensitivity of the IR spectrometer (NIR 256 Ocean optics) was precisely determined with a broadband calibration lamp.

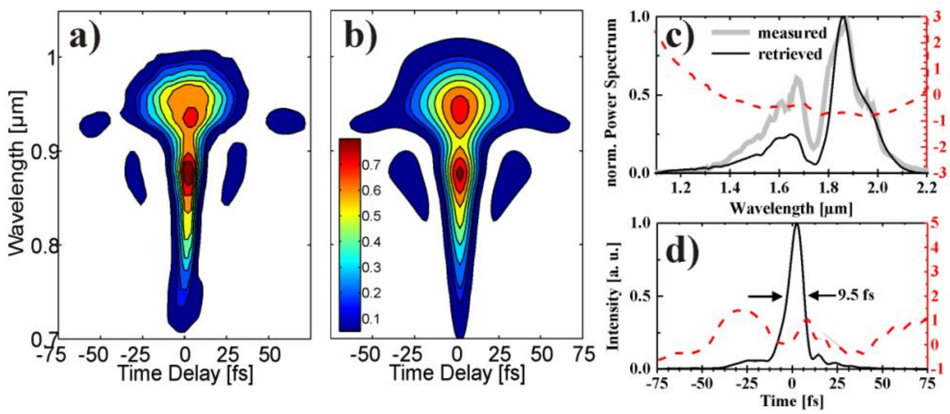

Fig. 2. Results of pulse compression. Measured (a) and retrieved (b) SHG-FROG traces of the 1.6 cycle pulse after compression with 1.5 mm FS. Lowest contour line at 4% intensity value is followed by successively increasing steps of 10%. (c): measured (gray) and retrieved spectral intensity (black) and phase (dashed red) with corresponding time domain representation in (d).

Flattening the spectral phase with 1.5 mm FS leaded to 1.6 cycle laser pulses with 240 µJ pulse energy without detecting any superfluorescence or remaining pump beam background after the HCF. Good agreement down to the 4% intensity level between measured and retrieved spectrograms can be verified in Fig. 2(a) and 2(b), respectively. Excellent spatial properties were observed at the output due to spatial cleaning provided by HCF [33]. In addition, it should be noted that less FS was used to compress the pulses than in our previous experiment with 73 fs input laser pulses (1.5 mm vs 3 mm). The amount of dispersion to compensate depends on the ratio between input and output pulse duration. In Fig. 2(c), based on the SHG-FROG reconstruction, the corresponding retrieved power spectral density (black) and the spectral phase (dashed red) after passing through the FS window are presented. Considering that our laser pulses have almost an octave spanning spectra, the agreement between measured and retrieved spectral density is very good. Prior to SHG-FROG reconstruction, the measured spectrogram is filtered according to reference [34] in order to take into account the spectral distortions of the SHG process as well as the spectral response of the VIS-NIR spectrometer (Model USB2000, Ocean Optics) in the FROG apparatus.

Temporal intensity and phase of the 9.5 fs pulse are presented in Fig. 2(d) as solid black and red dotted curves. Residual uncompensated higher order spectral phase in Fig. 2(c) leads to the non-transform limited temporal shape. Still, the main peak carries 83% of the total pulse power and will be an excellent tool to study strong field processes like HHG, nonsequential double ionization, and above threshold ionization. Since full control over the electric field is desirable in attosecond science and in general for strong field phenomena, we confirmed passive CEP stabilization which is a necessary prerequisite for controlling the position of the electric field underneath of the intensity envelope. Though not visible on the linear scale of the measured spectrum of Fig. 2(c) (gray curve), a small fraction of the spectrum extends into the NIR where it can interfere with the SHG of the strong IR part of the spectrum. This allowed us to measure the f-2f interferogram without additional need for a

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white light generation stage. From the f-2f interferogram presented in Fig. 3 a RMS jitter of 350 mrad for the CEP has been characterized.

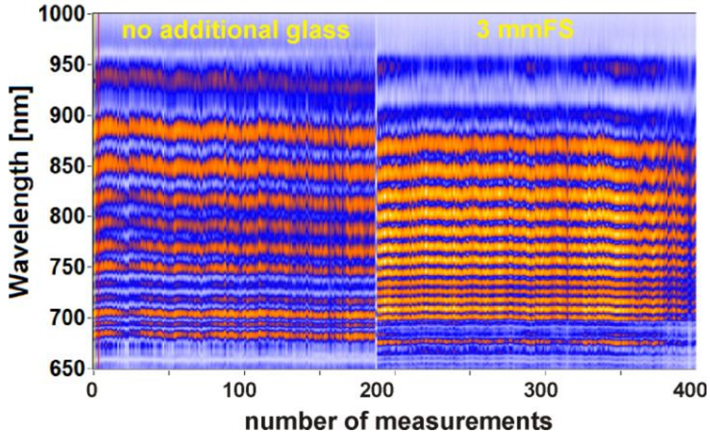

Fig. 3. Passive carrier envelope phase stability after the hollow-core fiber. Each measurement is averaged over 10 laser shots. The reduced fringe spacing on the right side (3 mmFS) compared to the left one (no additional glass) corresponds the group delay of 3 mm FS introduced before frequency doubling.

We believe that 350 mrad represents an upper bound for the true CEP fluctuations from our OPA. Wang et *al.* recently demonstrated strong intensity to CEP coupling in a HCF. They show that a 1% change in intensity results in a 128 mrad shift in the measured CEP at 800nm despite no change in the CEP of the actual waveform [35]. Due to the nonlinear nature of the OPA process, energy fluctuations are unavoidable. We measured 6% energy fluctuation after the HCF (3.5% after the OPA). This energy fluctuation in the HCF translates into spectral phase fluctuations in particular in the wings of the broadband spectra, causing the f-2f interferogram to over report CEP errors. This artifact has been also discussed by Fuji et *al.* for few-cycle IR laser pulses generated using OPCPA [36]. Pointing instability into to the HCF also contribute to the measured variation in CEP. In our case, it should be possible to reduce energy fluctuation in the HCF and the associated CEP jitter by implementing beam pointing stabilization as described in [37]. However, the measurement of Fig. 3 clearly shows that the CEP stability is preserved even if temporal reshaping of the pulse intensity envelope during nonlinear propagation in terms of self-steepening occurs.

4. Conclusion

In conclusion, we have reported a simple and robust experimental approach to generate CEP stabilized sub-millijoule 1.6 cycle laser pulses at 1.8 µm wavelength. Future improvements to this source will include beam pointing stabilization as well as the integration of a white light seeded high energy OPA to the 100 Hz 100 mJ Ti-Sa laser system of the Advanced Laser Light Source (INRS-EMT) to have sub-5 mJ Idler laser pulses to couple to the HCF [32]. This will be the ideal IR laser source for HHG and to extend attosecond pulse synthesis in the sub-keV spectral range.

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