High flux table-top soft X-ray source driven by sub-2-cycle, CEP stable, 1.85 µm 1 kHz pulses for carbon K-edge spectroscopy

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We report on the first table-top high flux source of coherent soft X-ray radiation up to 400 eV, operating at 1 kHz. This source covers the carbon K-edge with a beam brilliance of (4.3 ± 1.2) × 10^{15} photons/s/mm^2/strad/10% bandwidth and a photon flux of (1.9 ± 0.1) × 10^7 photons/s/1% bandwidth. We use this source to demonstrate table-top X-ray near edge fine structure spectroscopy at the carbon K-edge of a polyimide foil and retrieve the specific absorption features corresponding to the binding orbitals of the carbon atoms in the foil.

X-ray absorption fine structure (XAFS) and near edge X-ray absorption fine structure (NEXAFS) spectroscopy are well established methods for retrieving structural information about the composition of solid state materials and soft matter. The water window spectral range between 284 eV and 543 eV is of special interest as it contains the K-shell absorption edges of the biological building blocks: carbon (284 eV), nitrogen (410 eV) and oxygen (543 eV). Up until recently only facility scale light sources have been capable of generating coherent water window radiation: synchrotrons with a high degree of spatial coherence and hundreds of femtoseconds pulse durations, and X-ray free electron lasers with a high degree of spatial coherence and femtosecond temporal resolution [1]. High harmonic generation (HHG) [2, 3] offers an attractive alternative approach since it is realizable on a small table-top scale and is capable of generating fully coherent radiation, i.e. femto- to atto- second and possibly even zeptosecond pulse durations. The ability to generate coherent water window radiation from HHG is extremely exciting as it would bring ultra- short time resolution to structural probing with a table top method.

HHG is most commonly driven by Ti:sapphire sources at 800 nm with the highest achievable photon energy, the so called cutoff, scaling linearly with the laser intensity and quadratically with the driving wavelength [4]. While the water window range is reachable with such sources via non-phase-matched HHG [5], the contradicting requirements of increasing the cutoff with higher laser intensity while avoiding excessive ionisation, severely limits the achievable flux in the water window. A solution to this dilemma is to use a source with a similar peak intensity and pulse duration, but at much longer emission wavelengths in order to exploit the quadratic wavelength scaling of the HHG cutoff. A drawback of such an approach is the unfavourable single atom response scaling of harmonic yield with λ^{-9} [6] which can however be mitigated, to a large extent, through high gas-pressure phase matching [7]. This concept was demonstrated by reaching a 1.6 keV cutoff when driving with a mid-IR laser system [8]. Despite this cutting-edge result, the 20 Hz repetition rate and stability of the system have thus far proved insufficient for applications, thereby underlining the need for significant improvements of the laser parameters.

We find that while high X-ray flux can be achieved through phase-matched HHG driven by kHz or higher repetition rate long-wavelength sources, achieving sufficient intensity and carrier to envelope phase (CEP) stability of the driver laser is an essential key both for producing attosecond pulses and for generating reproducible X-ray spectra from each laser pulse and throughhout an X-ray measurement.

Currently at the kHz level and with long wavelength drivers, the lower end of the water window at 300 eV was reached using a Ti:sapphire pumped optical parametric amplifier (OPA) at 1.5 µm [9] however no photon flux was reported. A Ti:sapphire based CEP-stable optical parametric chirped pulse amplifier at 1.6 µm [10] has demonstrated a modest photon flux of ≈ 20 photons/eV/s at the carbon K-edge. Recently, a Ti:sapphire pumped OPA at 1.3 µm was reported to generate radiation with a flux of 10^9 photons/s/1% bandwidth at 200 eV and 10^7 photons/s/1% bandwidth at the cutoff at 300 eV [11]. This system was used to demonstrate absorption spectroscopy of features up to 250 eV however not at the carbon K-edge or beyond.

In this Letter we report on the first X-ray absorption measurements at the carbon K-edge from a high-flux water window source providing radiation up to 400 eV that is based on HHG with a 1 kHz, 2-cycle source at 1.85 µm with CEP stability. CEP control is essential as it ensures shot-to-shot reproducibility of the X-ray spectrum and cutoff. This prevents cumulative errors in absorption measurements due to shot by shot changes of the spectral shape and amplitude, and importantly it is the basis of many of the attosecond pulse generation techniques. Moreover, our cutoff extends to 400 eV, i.e. more than 100 eV beyond the carbon K-shell edge. Such spectral extent is crucial for real applications in

XAFS and NEXAFS spectroscopy which require measurement of the pre- and post- edge structures over a range of a few tens of percent of the absorption edge location and not just the edge itself. Our system is based on a cryogenically cooled two-stage Ti:sapphire amplifier system which generates 7 mJ, 40 fs laser pulses, with a central wavelength of 800 nm at a repetition rate of 1 kHz with single shot peak to peak stability of 0.8% rms over 40 min. Wavelength conversion is achieved using a commercial white-light seeded three-stage OPA (HE-TOPAS, Light Conversion Ltd.). We modify the final stage of the OPA to operate in a non-collinear configuration to mitigate spectral dispersion acquired through dichroic filters (needed in the collinear configuration). Operating the OPA at an idler wavelength of 1.85 µm, results in 45 fs duration pulses with 1 mJ pulse energy.

The 1.85 µm pulses are focused using a f = 500 mm calcium fluoride lens into a 0.7 m long, 400 µm diameter hollow-core fibre which is filled with argon at 1.2 bar for spectral broadening [12]. The residual dispersion is largely compensated for by propagation through a 3 mm RG1000 filter (Schott). The compressed pulses are measured with a home built second harmonic generation frequency resolved optical gating (FROG) setup. The measurement is shown in Fig. 1 and we obtain a pulse duration of 11.2 fs at FWHM which is close to the fourier transform limit of 10.3 fs. Upon compression, the sub-2-cycle pulses have an energy of 0.46 mJ and a single shot peak-to-peak stability of 1% rms over 40 min.

With the white-light seed originating from the pump pulses, the idler is expected to be self CEP stable [13]. The CEP is monitored using a homebuilt f-2f interferometer, using a reflection from the RG1000 filter. The setup takes advantage of the more than octave spanning spectrum generated in the HCF to give access to the required f-2f frequencies. The broad spectrum is frequency doubled in a 100 µm thick barium borate crystal (BBO) and then interfered inline with the fundamental, resulting in CEP-dependent spectral fringes. Figure 2a shows a measurement of the sub-2-cycle pulses overall single-shot CEP jitter of 316 mrad over a duration of

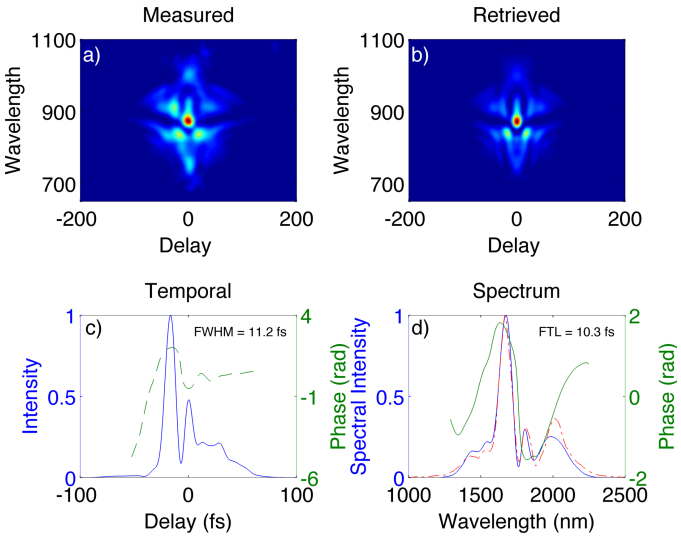

Fig. 1. a) Experimental second harmonic FROG trace, together with the retrieved trace (b); the FROG error was 0.012. The extracted temporal intensity profile (c) exhibits a 11.2 fs FWHM pulse duration. The spectra are shown in (d) with the retrieved spectrum (blue) matching well with the measured spectrum (red).

6 min. In order to improve this value, we implemented an electronic feedback system with a piezoelectric actuator which adjusts the pump-to-idler timing in the OPA to compensate for slow drifts of the average CEP offset. Figure 2b shows the dramatic improvement when activating the feedback loop measuring an overall single-shot CEP jitter of 88.8 mrad over one hour.

Bright soft X-ray flux and a 400 eV cutoff in the water window is achieved by focusing the CEP-stable sub-2-cycle pulses with an f = 100 mm silver-coated curved mirror into

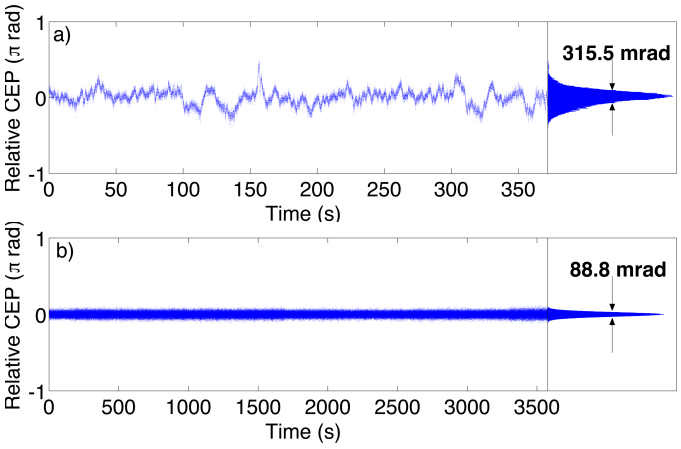

Fig. 2. f-2f Measurement of the 1.85 µm sub-2-cycle pulses for the passively stabilized system in a) and when activating the electronic feedback loop in b). The stability increased dramatically from 315.5 mrad over 360 s to 88.8 mrad over 3650 s.

our HHG target. The target consists of a 1.5 mm outer diameter, 0.5 mm inner diameter aluminium tube sealed at one end, with the laser focused through a 300 µm drilled hole. The target region is differentially pumped to achieve low ambient pressures and to avoid reabsorption of the radiation. This enables backing pressures in the target of up to 7 bar. The harmonic spectra are resolved with a home-built X-ray spectrograph consisting of a flat field grating with 2400 lines/mm (Hitachi) and a cooled X-ray CCD camera (Princeton PIXIS-XO). We estimate the resolution of our X-ray spectrometer as 0.25 eV at 300 eV.

As a first step, we optimized the harmonic flux by scanning the target backing pressure from 0 to 7 bar of neon. Figure 3 shows the result of integrating the HHG spectrum, for a given backing pressure, for 5 seconds in steps of 0.25 bar per spectrum. The figure illustrates that we achieve a cutoff far beyond the 284 eV carbon K-edge and that the highest yield is reached together with the highest cutoff at a backing pressure of 3.5 bar.

Having determined the optimum pressure for our target, we investigated CEP control over the spectral shape and measured the X-ray beam brilliance and photon flux. Figure 4 shows spectra which were taken for two distinct CEP values. We clearly see the importance of CEP control in order to avoid averaging over varying pre and post edge structures from shot to shot while integrating a NEXAFS spectrum.

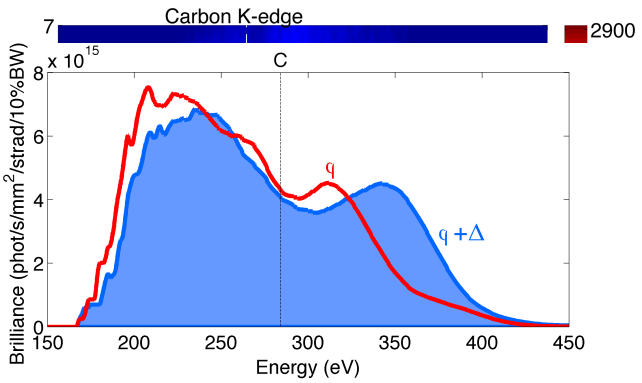

Fig. 4. HHG spectra for two specific driving pulse CEP values illustrating a clearly visible spectral amplitude modulation at the pre- and post- carbon edge (dotted line) labelled C at 284 eV)

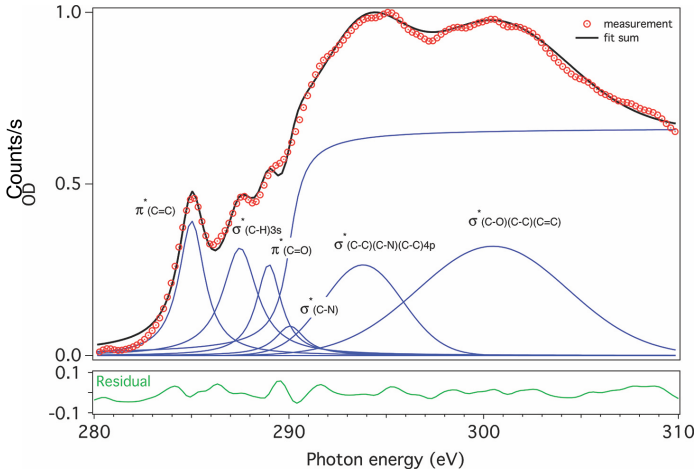

Fig. 5. NEXAFS measurement of a 200 nm free-standing polyimide foil (red circles). A peak fit with known transitions (blue) from Ref. 15 agrees very well (black curve) with the measurement.

We determine a beam brilliance of (4.3 ± 1.2) × 10^{15} photons/s/mm²/strad/10% bandwidth and a photon flux of (1.85 ± 0.12) × 10^{7} photons/s/1% bandwidth at 300 eV. Our photon flux is around twice as high than previously reported, moreover this flux is achieved for the first time from a CEP stable source which presents a significant step towards femto- and atto- second transient absorption spectroscopy in the water window spectral range. Contrasting our source with Ref. [11] we achieve brighter X-rays despite our longer driver wavelength, our lower driver pulse energy, and in a simple effusive target without the need for special X-ray capillaries.

Finally, we demonstrate the applicability of our table- top X-ray source to carbon K-edge NEXAFS spectroscopy of a 200 nm free-standing polyimide film. Figure 5 shows the absorption spectrum and the peaks around the carbon K-edge. The data was extracted from a single 5 minute measurement. We can clearly identify the individual fine structure peaks with the known orbitals in polyimide and the data is in agreement with state of the art synchrotron [15] and incoherent X-ray [14] data.

In conclusion, we present the first high-flux table-top source of coherent X-ray radiation in the water window which reaches a photon flux of (1.85 ± 0.12) × 10^{7} photons/s/1% bandwidth at 300 eV with a cutoff at 400 eV. The X-ray source is driven by a 1 kHz, sub-2-cycle CEP- stable laser system at 1.85 µm with demonstrated control over the X-ray spectral shape and cutoff. We demonstrate the excellent performance of our source with a first NEXAFS spectrum, recorded with only 5 minutes of integration at the carbon K-edge of a polyimide film and identify the fine structure peaks of the carbon binding orbitals. The ability to generate fully waveform controlled water window harmonics with sufficient pre- and post- carbon K-edge spectral coverage presents a crucial step towards time resolved core-shell X-ray absorption spectroscopy with attosecond resolution.

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