Characterisation of the Carrier Envelope Phase of Few-Cycle Short-Wave Infrared Pulses

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Introduction

Carrier envelope phase (CEP) stabilized few-cycle optical pulses in the short-wave infrared (SWIR) spectral region have many applications in strong-field physics. One application of strategic importance to the user community is in the generation of coherent femtosecond soft x-ray (SXR) pulses in the water window (~280—530eV) where there exist many edges of important atoms such as carbon, nitrogen and oxygen [1].

High Harmonic Generation

A table-top method of producing radiation with such photon energy can be realised via a method called high harmonic generation (HHG) [2, 3], whereby the strong laser field ionizes and then accelerates an electron resulting in bursts of SXR photons as the electron's kinetic energy is transferred into electromagnetic radiation during recombination with the parent ion. The maximum kinetic and therefore photon energy obtained is proportional to the product of the laser intensity and the square of its wavelength. Due to phase matching considerations, the maximum intensity that can be utilised is limited; therefore it is necessary to use a driving wavelength of ~2 μm or longer to reach the water window [4, 5]. Since the efficiency of the generation process scales quite unfavourably with wavelength, one is required to use the shortest wavelength possible that can generate the desired photon energy. Using a few-cycle pulse reduces the number of free electrons generated from ionization, allowing the generation of higher photon energies than can be achieved when using a longer pulse.

The ionization, acceleration and recombination process in HHG occurs for every half-cycle of the laser field in which the field strength is sufficient to field-ionize the atom, resulting in a burst of SXR radiation every half cycle, and thus resulting in a spectrum consisting of a comb of odd harmonics. Since the ionized electrons are driven directly by the electric field, the maximum kinetic energy and hence photon energy upon recombination for each burst is determined by the peak strength of the electric field half-cycle immediately preceding emission as the electron was accelerated away and then back towards the parent ion. This maximum photon energy for each half-cycle is known as the half-cycle cut-off (HCO) [6].

The CEP, ϕ_0, of a pulse is defined as the phase offset between the maximum of the pulse's electric field and its intensity envelope. The highest HCO is achieved when the peak of the electric field coincides with the peak of the intensity envelope, i.e. a "cosine" pulse with $\phi_0 = 0$. In this scenario only a single HCO – the primary HCO – contributes to the maximum photon energy and hence the spectrum will consist of a broad continuum. In a few-cycle pulse, the intensity envelope varies almost as rapidly as the electric field; hence changes in the CEP will result in a reduced peak field strength and therefore a reduced primary HCO frequency. In the situation of a "sine" pulse, where $\phi_0 = \pi/2$ rad, two electric field half-cycles will contribute to the primary HCO and hence this spectral region will consist of fringes corresponding to two pulses separated by half a laser period [7].

CEP Stable Few-Cycle SWIR Pulse Generation

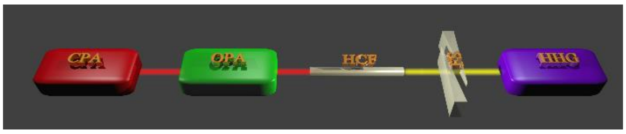

Figure 1: Schematic of optical setup. *The chirped pulse amplifier (CPA) laser delivers ~8mJ energy, ~30fs duration pulses with central wavelength of ~800nm. These pulses are used to pump an optical parametric amplifier (OPA) delivering ~1mJ energy, ~50fs duration pulses with a central wavelength of ~1.8μm in the idler wave. The CEP stable, SWIR output is spectrally broadened inside a gas filled hollow-core fibre (HCF) and then the spectral dispersion is compensated for using a pair of fused silica (FS) wedges before focusing into a gas target inside a vacuum chamber to drive the high harmonic generation (HHG) process.*

In attosecond pump-probe spectroscopy applications, a single probe pulse with a well-defined time-delay with respect to the pump pulse is desired. Since any measurement is averaged over an ensemble of pulses, it is therefore necessary to ensure that the CEP is controllable and held constant over the course of a single measurement. The first laser systems suitable for HHG that could achieve this were based on active stabilization of the CEP of the oscillator, typically via feedback to the pump intensity or cavity dispersion. A downside of this methodology is that measurement noise is also fed back to the oscillator. Mechanical methods to decouple vibrations in the laser increase long-term drift and degrade experimental results. An alternative method of generating CEP stable pulses is to use the idler wave from an optical parametric amplifier (OPA). The idler from the OPA is spectrally broadened and temporally compressed in a gas-filled hollow-core fibre followed by anomalous dispersion in a pair of fused silica wedges. The dispersion and CEP of the pulse is controlled by adjusting the insertion amount and hence material thickness of the wedge pair, as depicted in Figure 1.

CEP Measurement

The CEP of the pulse can be measured by measuring the HHG spectrum and extracting the frequency of the primary HCO [6]. We therefore measured the HHG spectrum as a function of wedge insertion. By repeating the measurement several times, statistics on the pulse CEP stability can be ascertained. Figure 2 shows the harmonic cut-off spectra as a function of wedge position for several of these scans. At present, the CEP is only passively stabilised via the OPA process and thus subject to noise due to variations in the pump intensity or phase shifts in the various beam paths inside the OPA, as is evident in the figure.

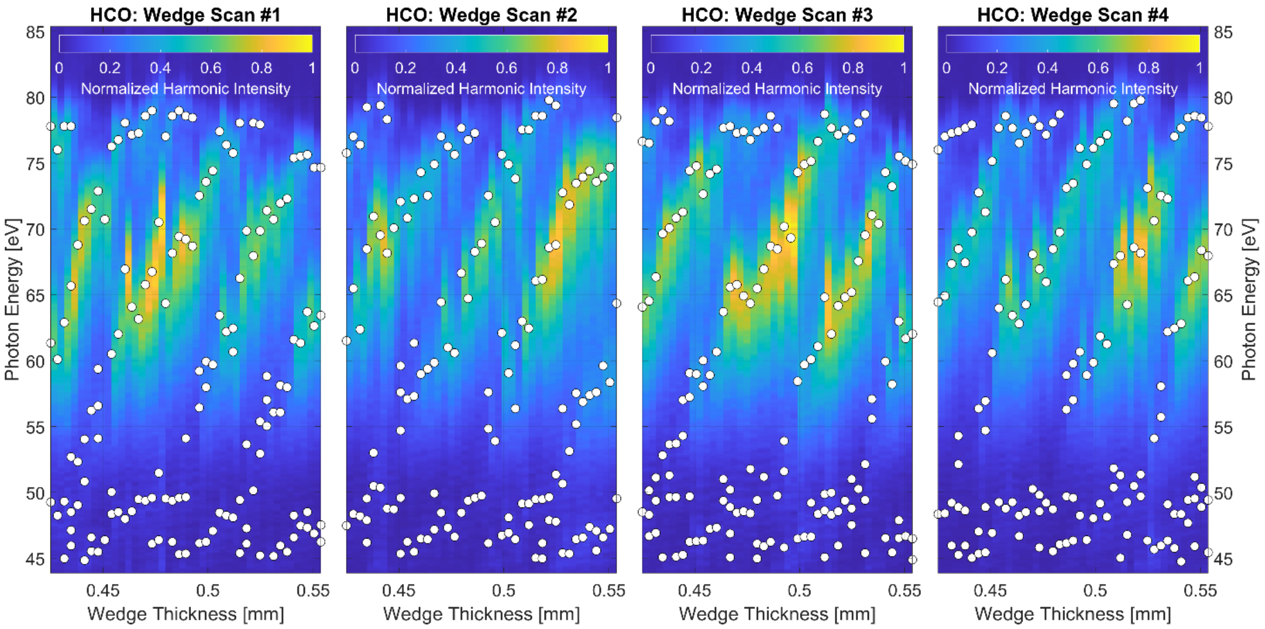

Figure 2: Half-cycle cut-off spectra. *Several scans of the half-cycle cut-off (HCO) spectra emanating from HHG with CEP stable few-cycle pulses. The CEP of the pulses is tuned by varying the insertion and hence material thickness of the fused silica wedges about the optimally compressed thickness. The on-axis integrated harmonic spectra are plotted in a false colour map as a function of additional wedge thickness for several repeat scans. The positions of the HCO (locally maximum intensity) are marked by the filled white circles.*

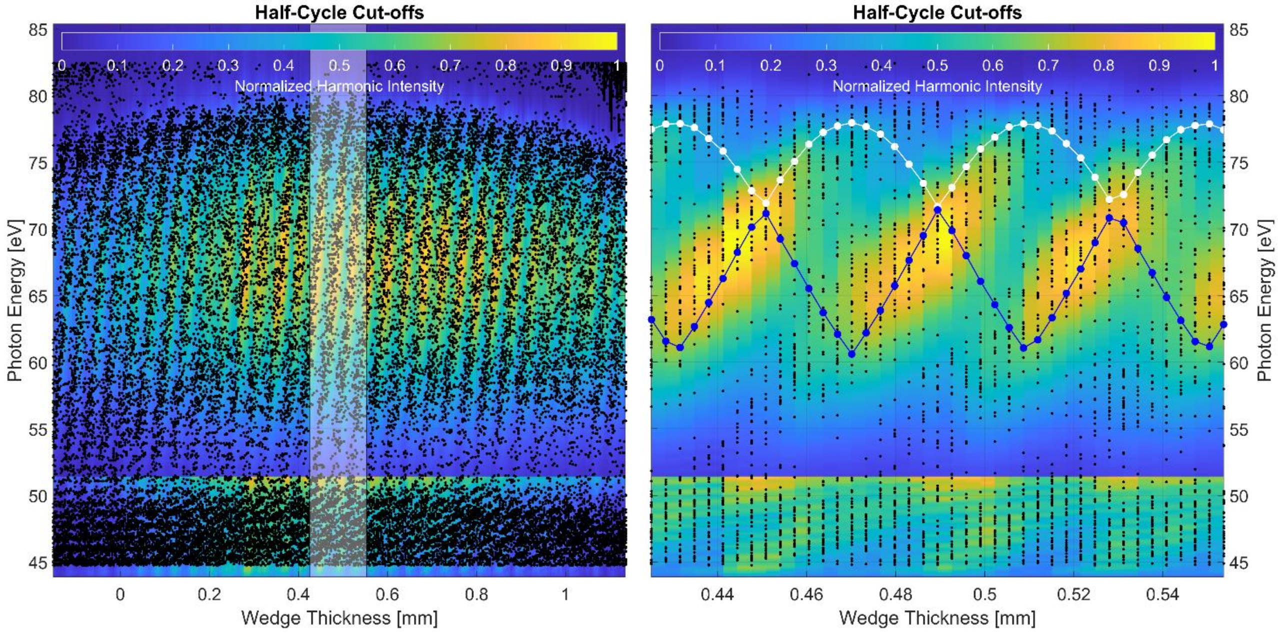

Figure 3: Averaged half-cycle cut-off spectra. Left*: False colour map of the harmonic spectra as a function of wedge thickness averaged over 25 repeat scans. The HCOs for each individual scan are marked by the black circles. The intensity of the lower energy harmonics has been scaled by a factor of x7.* Right*: Zoom in of the shaded white region marked on the left-hand plot. The individual HCOs are marked by the black dots. The white and blue dots/lines mark the theoretical primary (i.e. highest energy) and secondary HCOs calculated from the driving electric field.*

Figure 3 plots the HCO spectra averaged over 25 wedge insertion scans, demonstrating that the long time-scale average tracks the CEP phase introduced by the wedges. Therefore averaging over many measurements, as would be done in a pump-probe spectroscopy experiment, provides CEP sensitive measurements, as shown in Figure 3.

Conclusions

We have demonstrated the generation and passive CEP stabilisation of few-cycle SWIR pulses and used these to generate harmonics up to 100eV in argon, currently limited by the design of the flat-field XUV spectrometer. It was shown, via the measurement of HCO spectra, that it is possible to obtain CEP sensitive measurements from a passive stabilisation alone. We plan to implement active stabilisation to reduce the CEP noise and ensure CEP lock over longer acquisition time scales, and to implement a redesigned flat-field spectrometer capable of measuring up to 600eV. When combined with our few-cycle SWIR OPA [8] this should open up the potential to perform transient absorption x-ray spectroscopy in the water window.

References

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