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Controlling water-window high-harmonic generation with sub-cycle synthesized waveforms

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Abstract. We present the first results concerning synthesizer-driven high-harmonic generation that reach the water-window region. This approach holds the promise of offering greater spectral tunability in the generation of isolated attosecond pulses and at the same time of achieving higher photon-flux, required for attosecond-resolved soft X-ray transient absorption experiments.

1 Introduction

Understanding the ultrafast mechanisms that govern the interaction of light and matter in complex systems, such as polyatomic molecules or crystalline solids, is expected to play a fundamental role in the development of light-driven green technologies in fields such as energy generation and storage[1] or chemistry[2]. In fact, the efficiency of converting light into other forms of energy (electrical, chemical, thermal, etc.) often depends to a large extent on dynamical electronic processes on attosecond (as) and femtosecond (fs) time scales [3]. Time-resolved soft X-ray absorption spectroscopy (XAS) is one of the most powerful experimental methods for observing electron dynamics in complex systems since it returns information on the electronic structure (such as chemical bonds) and local geometric structure for the different atomic species. However, the extension of XAS to extreme time scales is not trivial. One of the most promising approach for the development of XAS setups with as/fs resolution consists in the upconversion of ultrashort laser pulses via high-harmonic generation (HHG). HHG allows for the production of as/few-fs soft X-ray pulses with excellent spectrotemporal stability and intrinsic synchronization with widely tunable pump pulses. The main challenge of soft X-ray HHG is the limited conversion efficiency of the process, which results in low photon fluxes. Moreover, HHG often spans a very large spectral region, for instance the full water-window (280-520 eV). This, although an advantage when measuring the absorption edges of different elements simultaneously, often results in a very low flux integrated over each individual absorption edge region. Therefore, the major challenge for soft X-ray HHG sources is to achieve higher efficiencies and/or spectral tunability in order to increase the number of photons in the spectral region of interest. In our group we have recently developed the technology of coherent synthesis of multiple ultrashort pulses obtained via OPAs, the parametric waveform synthesis (PWS)[5], in order to generate pulses with sub-cycle duration, controllable electric field with non-sinusoidal waveforms and energies in the order of 0.5 mJ. These waveforms, given the central wavelength of about 1200-1400 nm (depending on synthesis parameters) and the almost 2-octave spanning bandwidth, are a novel tool to try to circumvent the scaling laws of HHG driven by sinusoidal fields[4], that limit its conversion efficiency in the water-window region.

2 Tunable attosecond pulses in the water-window

Here we report the first results concerning the generation of water-window HHG via sub-cycle waveforms, probably corresponding to isolated attosecond pulses (IAPs). In previous works we already demonstrated that via the PWS it is possible to generate IAPs without gating[5] and to control the spectro-temporal characteristics of the IAPs in the < 200 eV range[6]. Our PWS setup features two spectral channels, spanning the 650-1000 nm range (NIR) and the 1200-2200 nm range (IR). By controlling the relative-phase (RP) between NIR and IR and the overall carrier-envelope phase (CEP) a manifold of non-sinusoidal waveforms can be created. We observed the HHG spectra while scanning the CEP and RP values. The generating medium is helium in a $\sim$ 0.5 mm long gas cell with up to 10 bar of backing pressure. The measurement were taken following this procedure: after optimizing spatiotemporal overlap of NIR and IR pulses, the NIR beam is blocked and the HHG parameters (gas cell position, gas pressure, fine adjustment of iris aperture) are optimized for the IR channel pulse (0.5 mJ of energy at OPA-channel output, 8 fs duration) only in order to achieve the highest spectral intensity

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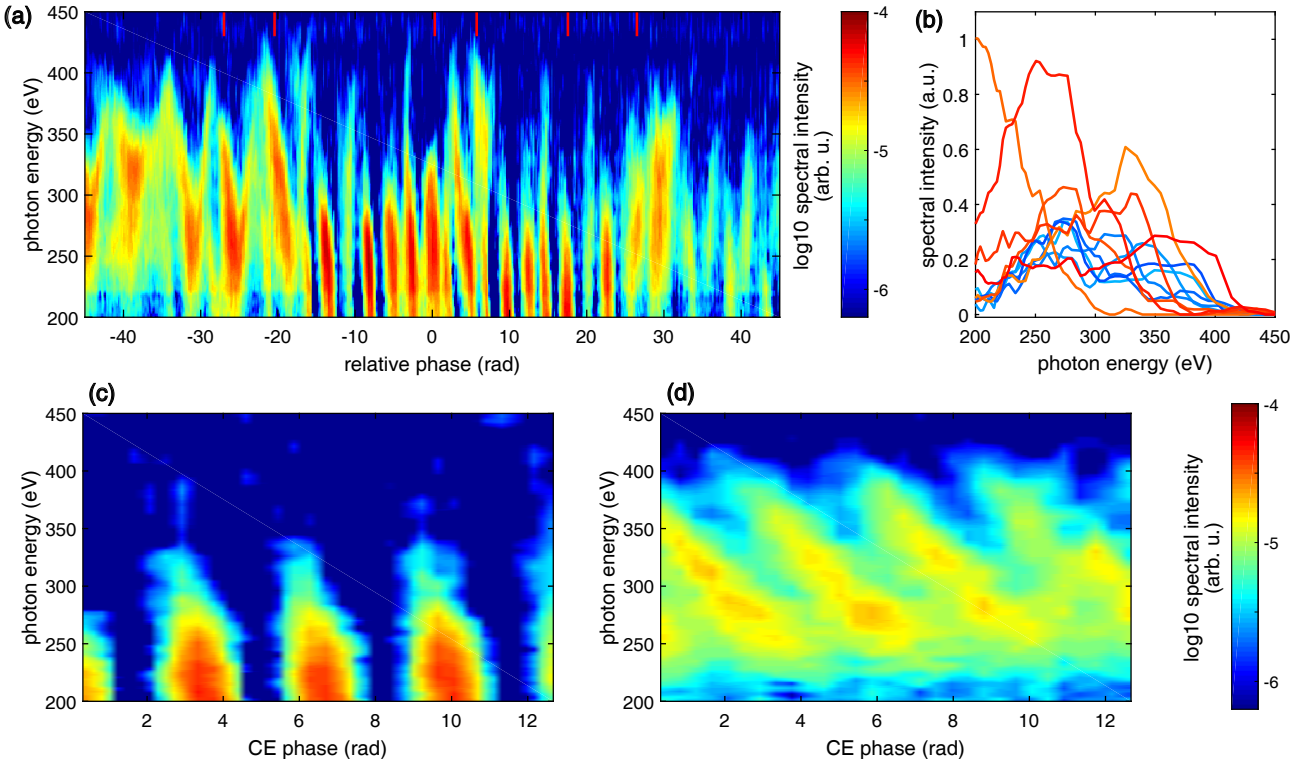

Figure 1. High-Harmonic spectra generated in helium with 10 bar of backing pressure and 2 seconds integration time. The spectra are Jacobi-corrected, transfer functions of grating and CCD are not applied. A 200 nm thick aluminum filter is used to block the IR. (a) Scan of the relative phase (RP) for a fixed CEP value, showing continua of various central photon-energies and bandwidths. (b) In red colour scale: selected HHG spectra corresponding to different RP values, marked with red ticks on the top of (a); in blue scale: IR-only HHG spectra corresponding to different CEPs covering a π range. (c) CEP scan for one particular RP value. (d) CEP scan of IR-only.

and cutoff; afterwards the NIR pulse (0.1 mJ of energy at OPA-channel output, 7 fs duration) is released and the RP scan is performed for while the CEP is actively stabilized. In the scan trace, shown in Fig. 1(a), it is clearly visible the effect of the RP on the HHG spectrum that spans the 200-420 eV region. The HHG intensity is suppressed or enhanced depending on the RP value if compared with the IR-only spectrum. The HHG spectrum is as well reshaped by the precise RP value, resulting in a tunable peak emission between 200 and 350 eV (the lower limit is dictated by the grating used in this measurement) and in maximum photon energies up to $>$ 400 eV. In this scan, it appears that particular values of CEP and RP deliver a higher photon flux with respect to the IR-only case (Fig. 1(b)). This experiment is currently being repeated for a broad range of different macroscopic conditions (gas type/pressure, gas-cell position, focusing condition), in order to obtain a comprehensive picture. We also performed CEP scans for particular values of RP and for the IR pulse alone. As shown in Fig. 1(c,d), while the IR pulse alone produces HHG for any CEP value, the synthesized waveform completely suppresses HHG for certain CEPs, suggesting the presence of IAPs. This is the first time that synthesizer-driven HHG is shown to reach the water-window region.

References

[1] Bräm, O. et al., Phys. Chem. Chem. Phys. **14**, 7934–7937 (2012).
[2] Baldini, E. et al., ACS Photonics **5**, 4, 1241–1249 (2018).
[3] Valentini, A. et al., Phys. Chem. Chem. Phys. **22**, 22302-22313 (2020).
[4] Jin, C. et al., Nat Comm. **5**, 4003 (2014).
[5] Rossi, G.M. et al., Nat. Phot. **14**, 629–635 (2020).
[6] Yang, Y. et al., Nat. Comm. **12**, 6641 (2021).