Synchronised pulses generated at 20 eV and 90 eV

for attosecond pump probe experiments

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The development of attosecond pulses across different photon energies is an essential precursor to performing pump-probe attosecond experiments in complex systems, where the potential of attosecond science¹ can be further developed^{2,3}. We report the generation and characterization of synchronised XUV (90 eV) and VUV (20 eV) pulses generated simultaneously via high harmonic generation. The VUV pulses are well suited for pump-probe experiments that exploit the high photoionisation cross-section of many molecules in this spectral region⁴, and the higher photon flux due to the higher conversion efficiency of the high harmonics generation process at these energies⁵. We temporally characterised all pulses using the attosecond streaking technique⁶ and the FROG-CRAB retrieval method⁷. We report 576 ± 16 as pulses at 20 eV and 257 ± 21 as pulses at 90 eV. Our demonstration of synchronised attosecond pulses at different photon energies, and inherently jitter-free due to the common-path geometry implemented, offers unprecedented possibilities for pump-probe studies.

The production of isolated attosecond pulses (IAP) in the extreme ultraviolet (XUV) region (30-150 eV) with an intense near-infrared (NIR) femtosecond laser is nowadays a robust process. The generation of IAP via high harmonic generation¹ (HHG) requires temporally gating the high harmonics emission to a single burst by using one of several techniques. Most common are polarization gating⁸, double optical gating⁹, ionization gating¹⁰ and amplitude gating¹¹. However, the

use of two XUV pulses in attosecond pump-probe experiments is usually inhibited by the low pulse energy and small interaction cross-section with matter. This shortcoming can be mitigated by employing the driving NIR field in conjunction with the XUV pulse. Using the longer NIR field, however, degrades the time resolution, and the high NIR field strength applied perturbs the attosecond dynamics under study.

To overcome these limitations, and to extend the attosecond science experimental possibilities, there is a considerable interest in developing high power attosecond sources both at higher¹² or lower^{13–18} energies with respect to the standard XUV region. For lower photon energies, in the range 4–7 eV, low order nonlinear optical processes have been used to generate pulses in the range of 3–18 fs^{14–16,19}, but sub-femtosecond duration have not yet been attained. HHG offers a promising route to attosecond VUV pulses^{13,17}. HHG has been successfully exploited^{13,20} to generate sub femtosecond pulses with photon energies in the range 20–40 eV using polarisation gating. In both these earlier works, however, the attosecond pulses were available only at one specific photon energy at a time. Furthermore, the polarisation gating has been shown theoretically to lead to elliptically polarised VUV pulses²¹ which may be unsuitable for certain experiments. So despite impressive developments, a robust way to independently produce attosecond pulse pairs at different photon energies suitable for a pump probe experiment has not yet been reported.

Here we present results on the generation and characterisation of pulses in the vacuum ultraviolet (VUV) region (20 eV) synchronised with IAP in the XUV (90 eV) using the amplitude gating technique. We employed in-line interaction regions in a pair of pulsed gas targets^{22,23} to simultaneously generate XUV and VUV pulses with a single NIR pulse. Such implementation provides a convenient spatial and temporal superposition of the generated pulses and minimizes the time jitter of the two pulses due to the common-path geometry. As shown in the main part of Figure 1 and in insets (f-g), the VUV and XUV pulses were selected from the high harmonic spectrum of two different gas targets using band-pass spectral filters. After spectral selection, temporal delay between any pair of pulses can be introduced with the two part mirror assembly²⁴. The temporal characterisation of the pulses

was performed by attosecond streaking⁶ by selecting the XUV and NIR, or the VUV and NIR pulses pair. The successful streaking traces recorded assure synchronisation between the NIR field and each pulse generated in both the XUV and VUV range. Using a single NIR pulse for the production of attosecond pulses at both photon energies, guarantees intrinsic synchronisation between all generated pulses.

The HHG process was driven by carrier envelope phase (CEP) stable few cycle (3.5 fs) NIR laser pulses centred at 790 nm, focused into two in-line pulsed gas jets, as shown in Figure 1.

Figure 1 | Geometry for the production of simultaneous attosecond XUV and VUV pulses. The NIR pulse is focused in two in-line Kr and Ne gas targets. The radiation propagates collinearly and is spectrally filtered with different foil filters mounted in a row, as shown in inset (e), to allow the selection of any combination of photon energies by appropriate alignment of the filter assembly, as shown in the main figure and in insets (f)-(g). The transmitted radiation is then focused in front of an electron time of flight spectrometer with a 2-part mirror providing the time delay between the selected pulses (the inner part of this concave mirror can be moved precisely with respect the outer annular part using a piezo stage). Insets (a)-(d) show the spectral properties of the high harmonics produced and of the filters.

The first pulsed gas target was operated with Kr (gas density 10^{16} cm^{-3}) which we found to be the most efficient noble gas for VUV harmonics generation. The second pulsed gas target was Ne (gas density 10^{17} cm^{-3}) better suited to the XUV energy region due to its higher ionisation potential. The position of the two targets with respect to the laser focus, and with respect to each other, was determined by the phase-matching conditions of each spectral region of interest. Position scans of the jets along the propagation axis (see supplementary material) showed that the optimum position for the production of the XUV pulse was 7 mm downstream from the focal plane of the laser. The VUV yield showed a much weaker dependence on the jet position and was placed 3 mm downstream from the laser's focal plane.

Thin foil filters with different spectral transmission properties closely spaced in a custom-designed mount (see inset (e) of Figure 1) were positioned into the beam using a motorised translation stage to select the NIR, VUV and XUV pulses. The filter mount was positioned so that the beam was intercepted by two adjacent filters, leading to side-by-side co-propagating pulses. The 3 mm aperture of each foil filter is much bigger than the wavelengths under consideration and therefore does not perturb the transmitted wavefront. Hence, even if spatially separated, the beams reaching the focussing two-part mirror can be considered as originating from the same original wavefront before the apertures, thus avoiding temporal smearing in the focus.. A Kapton foil (7.5 μm thick) rejected the high harmonics radiation providing the NIR pulse for the attosecond streaking experiment. A Zr foil (200 nm) rejected the NIR and lower photon energies, thus filtering the XUV pulse, while a Sn foil (200 nm) filtered the 11th and 13th harmonic of the NIR pulse providing a VUV pulse centred at 20 eV. The spectral properties of the Sn and Zr filters used in the experiment are shown in insets (a-b) of Figure 1. Other foil filters can be installed to filter the high harmonics spectrum at different photon energies, for example with germanium (25 eV) or indium (15 eV). The streaking setup²⁴ used for the pulses temporal characterisation includes a MoSi multilayer two-part focusing mirror with a piezo adjustment of the inner part of the mirror relative to the outer part to provide the time delay between any pulse pair selected by the filters. The two time-delayed

pulses were focused onto the effusive gas target positioned at the entrance of an electron time-of-flight spectrometer. Photoelectron spectra were recorded as a function of time delay between the XUV/VUV pulses and the NIR pulse.

The propagation of the NIR laser pulse through the first gas jet could potentially disrupt attosecond XUV pulse generation in the second jet. To quantify such an effect, XUV-NIR attosecond streaking experiments were performed with the Kr gas jet (VUV) either on or off, and the results compared.

The gas target used to obtain the photoelectron replica of the XUV pulse was Ne. The results are shown in Figure 2.

Figure 2 | XUV pulse characterisation: The isolated attosecond pulse in the XUV has been characterised with the Kr jet off (left column) and on (right column). The measured traces (a) and (b) are well reproduced by the retrieved ones (c) and (d). The temporal characteristics (e) and (f) are preserved. The spectrum (blue line) is presented together with the spectral phase (red line) in (g) and (h). The shaded areas are the $\pm\sigma$ error bars

The traces have been analysed with the LSGPA algorithm²⁵ and the error bars obtained from the last signal matrix of the algorithm²⁶, giving a pulse duration of 272 ± 25 as when the Kr gas jet was off (Figure 2 left column), and 257 ± 21 as with it on (Figure 2 right column). The retrieved traces reproduce the features of the measured ones. The two measured XUV pulse durations are comparable within experimental error demonstrating the in-line implementation of the two gas jets does not perturb significantly the XUV attosecond pulse.

In Figure 3 we present the results obtained by filtering the high-harmonics spectrum with a 200 nm tin foil providing 15% transmission at 20 eV. The band-pass of the tin filter overlaps with the 11th and 13th harmonics of the NIR field. The characterisation of these pulses was also performed by attosecond streaking, confirming the validity of this technique in the little explored low photon energy regime¹³. Photoelectron spectra obtained by focusing the VUV pulse into an Ar effusive gas target in presence of the NIR field were recorded as a function of delay.

Figure 3 | VUV pulse characterisation: The photoelectron wavepackets generated with VUV radiation filtered by a tin foil were streaked with the few cycle IR pulse in a similar way as in **Figure 2**, with the Ne gas jet off (left column) and on (right column). The retrieved traces (c) and (d) reproduce the measured ones (a) and (b). The temporal intensity profiles (e) and (f) exhibit pre and post pulses as expected by selecting two distinct harmonics, but the FWHM is comparable in the two cases and agrees with the pulse shape obtained from the TDSE simulations (dashed black line, FWHM=575 as). The amplitude (blue) and phase (red) of the retrieved spectra is shown in (g) and (h). The shaded areas are the $\pm\sigma$ error bar.

The traces have been analysed with the LSGPA algorithm to provide a more reliable retrieval in presence of satellites pulses compared to the PCGPA algorithm^{25,27}. These satellites arise from the selection of two discrete harmonics in the transmission spectral window provided by the tin filter. The measured traces 3(a,b) are comparable with the retrieved ones 3(c,d). The temporal profile shown in 3(e,f) is well reproduced by the one obtained from time-dependent Schrödinger equation (TDSE) simulations²¹. The duration of the central peak is 585 ± 31 as with the Ne gas jet off (the second moment of the temporal intensity distribution is 950 ± 30 as), and 576 ± 16 as with it on (second moment 860 ± 45 as), in good agreement with the theoretical value from the TDSE

simulations of 575 as (second moment 940 as). From this agreement we infer that the energy dependence of the atomic photoionization amplitude and phase of argon²⁸, used as gas target for our streaking experiment, has little influence to the pulse duration under these experimental conditions. Due to the propagation of the VUV radiation through the Ne gas jet the signal with the jet on is weaker and noisier than when it is off, but the attosecond duration of the main peak is preserved in both cases, with similar overall contrast ($41\pm14\%$ when the Ne jet was off, $38\pm13\%$ with the jet on).

In conclusion we have demonstrated the generation of two synchronised and collinear attosecond pulses at two different photon energies of 90 eV (XUV) and 20 eV (VUV). These pulses can be used together for attosecond pump-probe experiments. Our compact experimental set-up uses two in-line high harmonics generation gas targets independently optimised for each photon energy. VUV and XUV pulses are selected using metallic foil filters and pulse delay is achieved using a two-part mirror assembly. The common path geometry does not require active stabilisation to achieve the necessary attosecond stability for pump-probe experiments. The pulses were measured using the attosecond streaking technique and pulse durations of 257 ± 21 as in the XUV, 576 ± 16 as at 20 eV were obtained. We confirmed that the presence of the second gas jet does not alter significantly these pulse durations. The VUV energy per pulse at 20 eV was measured to be 0.5 nJ (see supplementary material) at generation and will scale to higher values by increasing the laser pulse energy above the 0.4 mJ we had available. This first demonstration of a set-up that is able to provide the pulses for attosecond pump-probe experiments opens up a range of new scientific opportunities in attosecond science.

Methods

A chirped pulse amplification laser system (Femtolasar GmbH, Femtopower HE CEP) provided pulses at 1 kHz, 28 fs with up to 2.5 mJ per pulse **centred at 790 nm**. A portion of this energy (1.1 mJ) was used in a Ne filled, differentially pumped hollow core fibre (HCF)²⁹ and pulses were compressed to

3.5 fs duration to implement amplitude gating in the HHG process. The carrier-envelope phase (CEP) of the pulses was locked after the fibre output with a feedback system to the oscillator pump³⁰ with a residual CEP fluctuation of less than 250 mrad rms (single shot). The compressed laser pulses of 400 μ J energy were focused with a 75 cm focusing mirror (Rayleigh length 15 mm) onto two in-line pulsed gas targets. The Kr gas target for the VUV pulse generation was positioned 3 mm after the focus and the Ne gas target for the generation of the XUV pulse generation was positioned 7 mm after the focus, ensuring the selection of the short-trajectory component of the HHG emission via phasematching.

The enhancement of high harmonics generation boosted by below-threshold harmonics²³ has not been seen in this experiment. This enhancement was seen when the low harmonics (3-7 eV) of the first gas target were optimised, with the high harmonics (>20 eV) not produced efficiently. In the experimental conditions of our work the Kr harmonics above 20 eV are still produced efficiently, and therefore we expect the low harmonics not to be strong enough to enhance the XUV generation process in the second jet.

Photoelectron energies were measured with a time of flight spectrometer²⁴ with a 4 mm diameter entrance aperture positioned 19 mm from the interaction region. To improve the collection efficiency a set of electrodes in the TOF nozzle was used to achieve a 9 V accelerating potential for the low energy electrons that were then directed to the drift tube (44 cm). This electrostatic lens has been calibrated with Simion[®] simulations together with experimental ATI and VUV spectra. Each photoelectron spectrum was integrated for 3 min (1.8×10^5 laser shots).

The LSGPA algorithm was run, for the VUV dataset, on a 1024x370 grid and the FROG error was 1.9% and 5.4% with the Ne gas jet off and on respectively. For the XUV dataset the algorithm was run on a 512x401 grid and the FROG error was 4.8% and 5.8% with the Kr jet off and on respectively.

The TDSE simulations for the HHG process are based on a soft core potential model²¹. The parameters for the potential are adapted such that ionisation occurs from the lowest p -orbital which is aligned along the laser polarisation direction and matches the ground-state energy of krypton. A

VUV pulse is obtained by applying the transmission data of Sn³¹. Dispersion data³² is only available up to 21 eV and has been extrapolated for the missing energy. Applying such dispersion causes only a small change in the VUV pulse duration and shape.

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Author contributions

D.F. and T.W. contributed equally as first authors. D. F and T.W. performed, with contributions from W.A.O., the streaking experiments. J.W.G.T. and J.P.M. conceived and designed the experiment. P.M.-H. and D.J.W. performed the absolute photon flux calibration. J.H. and M.L. provided the TDSE calculations and theoretical support. D.F. performed the analysis of the data. D.F., J.W.G.T. and J.P.M. wrote the manuscript with contributions from T.W., T.R.B., J.H. and W.A.O. All authors discussed the results and the analysis of the data.

Competing financial interest

The authors declare no competing financial interests.