



Post-doctoral one-year research project, funded by GPR Extreme Light from October 1st 2024

Subject: Attosecond metrology of spatio-temporally tailored XUV pulses and nano-scale coherent imaging

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"Harmonics and Applications" team from CELIA laboratory is dedicated to production, characterization and applications of coherent attosecond (1 as = 10^{-18} s) XUV radiation. The XUV source is produced by high order harmonic generation (HHG) in noble gases. It possesses unique properties enabling to track attosecond electronic dynamics in matter and investigate nano scale coherent imaging. The first goal of the research project aims at measuring the attosecond pulse train from spatio-temporally tailored XUV emission. In a second step, the tailored XUV beam will be used to perform coherent imaging of nano structured surfaces. We use the CELIA's ECLIPSE CPA Ti:Sapph Laser delivering pulses of 35fs, with a fraction of the available energy (up to few tens of 10 mJ) at a repetition rate of 10 Hz. We have already refined and developed spatial, spectral and temporal shaping of the intense attosecond XUV source, based on the interaction of this laser with a noble gas jet. The XUV tailoring is obtained via the shaping of the driver laser and of the interaction geometry with the gas jet. The harmonic yield is characterized by an XUV spectrometer with a vertical spatial resolution in the far field and XUV wave front sensing methods that are used to reconstruct the size and position of the sources associated to individual harmonics. Following previously identified causes of spatio-temporal couplings in HHG sources [Catoire 2016], we have therefore explored some methods to finely control their spatial properties and mitigate these couplings. We demonstrated the possibility of achieve self-focusing of harmonics without XUV optics [Quintard2019], to perform tunable spectral selection via spatial filtering [Veyrinas2021]. In a joined experiment with Anne L'Huillier's (Nobel prize in Physics 2023) team at the Lund Laser Center (Sweden) we have demonstrated that the effect of chromatic aberrations in a refocused broadband harmonics beam are limited when shaping the IR driving laser to get a radial flat top intensity profile at focus [Veyrinas2023]. Simulations (see figure 1) shows that attosecond pulse distortion are largely suppressed when harmonics are generated with a spatially shaped Flat-top laser beam.

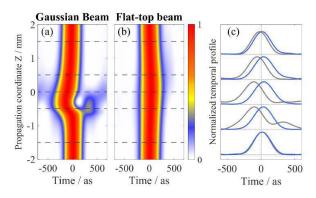


Figure 1: Calculated evolution of the on-axis attosecond pulse profile (H11 – H19) with propagation near XUV focus. Harmonics are generated in a gas jet with a Gaussian beam (a) or a spatially shaped flat-top beam (b). (c) Temporal profiles of the pulses generated with the Gaussian beam (grey) or the flat top beam (blue) at specific positions indicated by the dashed lines (from [Veyrinas2023]).

In an even more recent campaign at CELIA hosting a team from King's College London, we have extensively mapped out the harmonic source chromatic aberrations evolution while scanning the generating medium over the confocal range for both Gaussian and shaped Flat-top laser beams. The next step, that is one of the objective of the post-doctoral project, is to directly characterize the attosecond pulse train and measure the influence of controlled chromatic aberrations. This will be done using the so-called FROG-CRAB technique [Mairesse2005] that is derived from the seminal RABBITT method that was used by Pierre Agostini (Nobel prize in Physics 2023) for the first measurement of an attosecond pulse train [Paul2001]. An additional refinement will allow us to be sensitive to the possible change in the attosecond profile along the propagation axis. Those investigations are of major interest in attoscience for 3 main reasons. First, it would provide the first direct experimental evidence of attosecond distortion due to chromatic effects. Second, it would interrogate the limitations of the standard RABBIT-like methods that are insensitive to time structure inhomogeneities. Furthermore, when using the Flat-top shaping, it should be possible to visualize for the first time the double attosecond pulse structure associated to the long and short quantum paths contributing to harmonic generation within the single atom response.

In a second step, the ability to finely control the spatial properties of the harmonic beams and measure their wave front will also be used to image nano-structured surfaces. Alternative methods to standard coherent diffraction imaging approaches [Huijts 2020], such as phase sensitive ptychography or plenoptic 3D imaging will be investigated with XUV light. The goal is to resolve structures down to 100 nm along the surface and a few nm in depth. We foresee major impact on nano scale surface imaging of interest for industry (semi-conductor, photonics, aeronautics, biology, ...).

References:

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https://smr.u-bordeaux.fr/programmes-de-recherche/gpr-light

Information on HXUV CELIA team https://celia-bordeaux.cnrs.fr/recherche/hxuv/

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