A UV-XUV attosecond beamline with few-femtosecond tunable ultraviolet pump pulses

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Abstract. Ultrafast charge transfer processes in organic materials which occur in organic materials are fundamental for advancing solar energy conversion technologies. Understanding these phenomena on a short time scale induced by visible and ultraviolet (UV) light is crucial for future control and engineering of these molecules. Here, we present a novel attosecond beamline featuring Resonant Dispersive Wave emission for generating sub-3 fs tunable pump pulses in the UV region and High Harmonic Generation (HHG) in a semi-infinite gas cell for isolated attosecond pulse generation in the Extreme ultraviolet range.

1 Introduction

Photoinduced electron transfer (ET) and charge transfer (CT) processes occurring in organic materials are the cornerstones of technologies aiming at the conversion of solar energy into electrical energy and at its efficient transport. Understanding these processes when induced by visible (VIS) and ultraviolet (UV) light is crucial for advancing the development of more effective organic opto-electronic materials, which are typically characterized by absorption spectra within the 200-400 nm range [1]. By using femtosecond pulses, it is possible to study processes such as isomerization, nuclear vibrations, hydrogen migration, etc., which certainly affect ET and CT at "longer" time scales. However, to fully understand and control the early stages of ET and CT, attosecond (as, 10^{-18} s) pulses have proven indispensable for measuring ultrafast charge dynamics in organic molecules [2]. The pump-probe scheme which aims at resolving such processes in the very first tens of femtoseconds need to include: (i) a tunable ultrashort pump in the UV/VIS spectral range which initiates the dynamics, short enough not to perturb the subsequent evolution; (ii) an attosecond pulse which probes the dynamics by projecting the neutral evolution into the cationic states.

Here, we present the realization of a novel attosecond beamline in which pump pulses with sub-3-fs duration and tunable in the UV/VIS spectral range, generated through Resonant Dispersive Wave (RDW) emission are combined with Isolated Attosecond pulses (IAP) in the 20-50 eV range. Pulse durations for both pump and probe are confirmed by *in-situ* characterization.

2 Beamline overview

The attosecond beamline is seeded with a commercial Ti:Sapphire CEP stable laser (Coherent Legend HE-Elite CEP) which delivers 800 nm at 25 fs pulses with energy per pulse of 6 mJ and 1-kHz repetition rate. The output pulses undergo pulse post-compression in a Hollow Core Fiber (HCF) setup [3]: using a pressure gradient configuration with 1.8 bar of Helium in a 3 m long fiber and a set of chirped mirrors (Ultrafast Innovations) the pulses are shortened down to 4 fs, with an energy per pulse of 2 mJ. The duration of the compressed pulses is measured using a home-built D-scan [4].

The pump-probe scheme is realized through a Mach-Zender interferometer, where the pulses are split using an 80/20 broadband beamsplitter into the HHG and RDW arms. The first is focused into a semi-infinite gas cell (SIGC) for the generation of XUV attosecond pulses, which are used as probe pulses. The remaining part of the beam traverses a delay line before being directed into a capillary for the generation of pump pulses in the UV spectral region. After removal of the driving infrared pulses in both arms, the UV pump and XUV probe pulses are independently focused and then recombined non-collinearly into the interaction region onto the gas target with a recombination angle of $< 0.3°$. A Time-Of-Flight (TOF) spectrometer is used to detect either the ions or electrons produced by the interaction of both pulses with the gas phase target.

2.1 UV generation in hollow capillaries

RDW emission in gas-filled capillary is an innovative and efficient technique based on soliton dynamics, which enables the generation of few-femtosecond pulses in the deep UV (DUV) and UV spectral regions, at microjoule

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energy levels, with tunability achieved through gas pressure adjustments [5]. A complete temporal characterization of these pulses has been recently demonstrated [6]. In our setup we implemented a rigid hollow capillary directly attached to the vacuum chamber of the attosecond beamline. The capillary is 60-cm-long, with a core diameter of 160 µm and it was filled with neon in a pressure gradient configuration. After generation, the driving field is removed by using a pair of silicon mirrors at Brewster angle. In Fig. 2 we reported the different spectra measured after the separation, by varying the neon pressure inside the fiber.

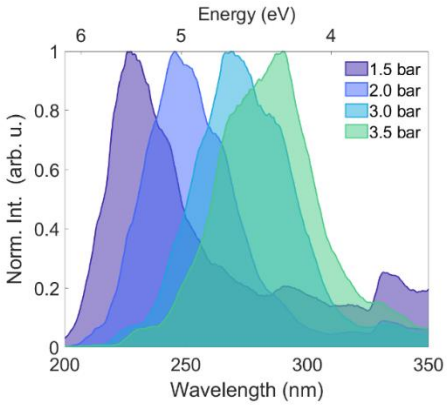

Fig. 1. Normalized experimental RDW spectra as a function of the gas pressure at fiber input.

The carrier wavelength of the pump pulses can be easily adjusted by changing the pressure at the fiber input. For each pressure, the transform-limit duration associated is below 3 fs. Afterwards the pump pulses are focused on the interaction region by using a spherical Al mirror, with an energy per pulse of 0.5 µJ on target and reaching intensities of the order of 10^{11} - 10^{12} W/cm^2. The duration of the pulses has been characterized *in-situ* by cross-correlation photoionization of Krypton.

2.2 XUV generation in a semi-infinite cell

Isolated attosecond pulses are produced using a long medium geometry, realized through a SIGC [7] in combination with ionization gating technique [8] This allows us to exploit the generally high conversion efficiency of long-medium geometries with the temporal properties of isolated attosecond pulses. The cell consists of 3 main regions separated by small orifices. In the first region, filled with around 5.8-mbar of argon, the driving field interacts with the gas and generates the harmonics. Focus position is just a few millimeters before the first orifice (200 µm), which opens to a 7 mm channel and creates differential pumping and assures that most of the radiation is not reabsorbed. The two following regions create additional differential pumping before reaching the 10^{-6} mbar level. The XUV pulses are filtered by the fundamental using metallic filters and refocused by using a gold coated toroidal mirror. An XUV spectrometer is used for diagnostic of XUV radiation. A typical spectrum of isolated attosecond pulse is reported in Fig. 2. It spans in a photon energy region in between 20 and 50 eV. These pulses have been characterized using XUV – NIR Attosecond Streaking [9], confirming a duration of 180 as.

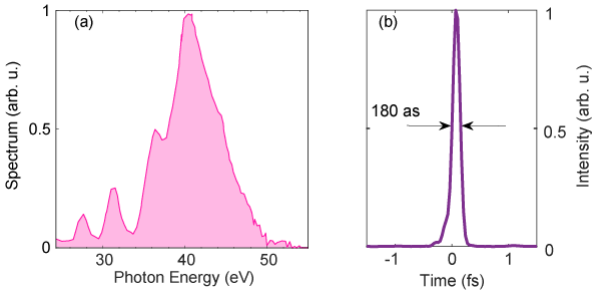

Fig. 2. (a) Normalized spectrum of isolated attosecond pulses obtained in Argon at 5.8 mbar using ionization gating in a SIGC. (b) Normalized intensity temporal profile of the IAP.

Conclusions

We have demonstrated the realization of a novel attosecond beamline which combines sub-3 fs pump pulses, tunable in the 200-350 nm and reaching 0.5 µJ energy per pulse on target. These pulses are combined with IAPS in the 20-50 eV and 180-as duration, maximizing the temporal resolution.

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