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D9.2
HHG Optical setup for XUV spectroscopy

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DELIVERABLE DETAILS

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D9.2	HHG Optical setup for XUV spectroscopy

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Development of a high-harmonic generation (HHG) optical setup for XUV spectroscopy that will be readily available to NFFA-Europe users

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☐ PP - Restricted to other programme participants & EC: (Specify)
☐ RE - Restricted to a group (Specify)
☐ CO - Confidential, only for members of the consortium

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Contents

Executive Summary

Ultrashort pulsed laser techniques such as harmonic generation and multi-photon excitation, have recently found their application in nanomaterials research, as non-invasive tools for imaging and manipulation of nanosystems, and spectroscopy with unprecedented time resolution. The Attosecond Science and Technology (AST) activity at FORTH-IESL focuses on the generation, characterization and applications of intense Extreme Ultraviolet (EUV) radiation emitted in the form of pulses of duration less than 1fs (attosecond pulses). It targets the development, upgrades and running of a state of the art, table-top, attosecond beam lines dedicated to the investigation of ultrafast dynamics in all states of matter, as well as of non-linear and strong field phenomena induced solely by the XUV radiation. Deliverable 9.2 refers to the development of a high-harmonic generation (HHG) optical setup for XUV spectroscopy that will be readily available to NFFA-Europe users.

1. Attosecond beam lines

The recently upgraded AST laboratory, is equipped with two gas phase high-order harmonic generation (HHG) beam lines which can deliver ultra short pulses in the spectral range of 15 eV to 35 eV with energy in the GWatt and MWatt range. Both lines can be used for pump-probe studies in the XUV spectral range. The driving laser is a commercial Ti:sapphire system emitting pulses of maximum 400 mJ pulse energy, ≤ 18fs minimum pulse duration at 10Hz repetition rate and central wavelength 800 nm. About one fifth of the total energy is used in driving the HHG source.

The XUV beam lines are divided in four units as is shown in the block diagram of Figure 1. The 1st unit, named "Laser Beam Delivery (LBD)", which is used for the manipulation of the driving field towards the generation of the XUV radiation, contains the laser beam steering, polarization shaping, beam shaping, pulse manipulation and focusing optics. The 2nd unit, named "XUV Generation (XUV-G)", is used for the generation of the XUV radiation and contains the XUV generation medium. The 3rd unit, named "XUV Separation (XUV-S)", contains the optical elements which are used for the isolation of the generated XUV radiation from the driving laser field and the steering (and XUV wave front splitting in case that is needed) of the XUV beam. The 4th unit, named "XUV Manipulation (XUV-M)", contains optical elements used for spatial and spectral selection of the XUV radiation. In this unit, XUV diagnostics can also be used. The 5th unit, named "End Station (ES)", which is used for the temporal characterization and the applications of the asec pulses, contains the XUV focusing elements, the XUV diagnostics and the detectors required for performing experiments using the XUV radiation.

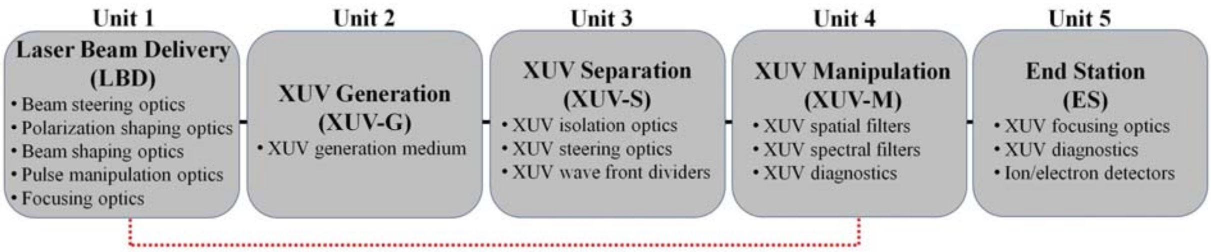

Figure 1. Block diagram of XUV beam lines. From Ref. [1]

In the 1st unit, the "beam steering optics" are broadband high reflectivity plane mirrors and beam splitters (or drilled holey mirrors) used for alignment and splitting of the driving laser field. The beam splitters are used to steer part of the driving field away from the XUV generation area (red dashed line in Figure 1). This auxiliary pulse, is used for the characterization of the IR beam used for the generation of High- harmonics. The "polarization shaping optics" are zero-order half- and/or quarter-wave plates used to control the polarization of the driving field. "Beam shaping optics" concern optical arrangements used for the optimization/manipulation of the XUV generation process through the control of the spatial intensity distribution of the driving field. Apertures and/or beam stops are "Beam shaping optics" which are commonly used for the formation of annular shaped beams and are very useful for the optimization and the spatial isolation (from the driving laser beam) of the XUV radiation generated in gas phase media. Also, they can be used for the creation of beams where an inner beam of small diameter is surrounded by an annular one. Such beams can be used for the manipulation of the harmonic generation process and for the temporal characterization of asec pulses via IR/XUV cross-correlation approaches (see for example Refs. [2, 3]). The "pulse manipulation optics" concern the optical arrangements used for the manipulation of the XUV generation process through the control of the driving field waveform. Such arrangements are used for the generation of isolated asec pulses by multi-cycle laser fields, utilizing the PG [4, 5-7] approaches. "Focusing optics" concern the optical elements/configurations (lens, focusing mirrors, deformable mirrors) used for the optimization of the XUV emission through the focusing conditions.

In the 2nd unit, the "XUV generation medium" concerns the type and the properties of the medium used for the generation of the XUV radiation. Gas phase media can be introduced in the unit by means of pulsed nozzles (pulsed gas targets) or leaking valves (static gas cell targets).

In the 3rd unit, the "XUV isolation optics" are used in order to reflect the XUV beam towards ES and significantly reduce the laser driving field which can damage the XUV elements existing in the units 4 and 5. The "XUV steering optics" are used for the alignment of the XUV beam towards the ES. The XUV optics in this unit are specially designed multilayer mirrors with high reflectivity in a specific broadband XUV energy region or plane mirrors placed at grazing incidence angle for high reflectivity in a broadband XUV range. Silicon plates, placed at the Brewster angle of the fundamental laser frequency and grazing incidence BK7 mirrors with antireflection coating the fundamental laser frequency have been used in order to fulfill these requirements. For example, a single split Silicon plate placed on translation/tilting stages can serve as an XUV isolator, steering element and wave front beam splitter. Additionally, due to the strong dependence of their reflectivity on the polarization direction of the driving field, these plates (in combination with the half wave plates of unit 1), can provide a precise control of the energy of the reflected driving field. This is a crucial point in case that the driving field is used as auxiliary pulse (in this case the XUV filters have to be removed from the beam path) for IR/XUV pump-probe experiments at the ES. This can be achieved using in unit 1 the beam shaping optics used for the temporal characterization of asec pulses via IR/XUV cross-correlation approaches.

In the 4th unit, the "XUV spatial filters" (apertures and/or beam stops) are used in order to spatially select part of the XUV beam and block the outer part of the XUV beam which may contain part of the driving field. This is crucial for the selection of XUV radiation generated by different electron trajectories in gas phase media and for the isolation of single asec pulses. The "XUV spectral filters" are metal (Al, Sn, In, etc.) foils of ~150 nm thickness used as band pass filters for the spectral selection of the XUV radiation. Also, these filters eliminate any residual part of the driving field which is reflected by the "XUV isolation optics". Both, spatial and spectral filters, are placed on motorized translation stages for the precise control of their position with respect to the position of the XUV beam. For the measurement of the XUV energy, and the spatial/spectral characterization of the XUV beam which enters the ES, calibrated XUV detectors (like photodiodes, PMT, MCP etc.), XUV diagnostics (like XUV beam profilers and XUV spectrometers) are also placed at the end of this unit.

In the 5th unit, the "XUV focusing optics" concern grazing- or normal-incidence XUV focusing mirrors. Grazing incidence focusing optics (toroidal mirrors) with high reflectivity (40%–50%) in a broadband (~ 20 eV bandwidth at ~ 40 eV photon energy range) XUV spectral range, while normal-incidence focusing optics are unprotected metal coated or multilayer spherical mirrors with high reflectivity (10%–20%) in a more confined (~ 6 eV bandwidth) (compared the toroidal mirrors) XUV spectral range. Although, both configurations can deliver asec structure in the XUV interaction region, the advantages and disadvantages of each of them is a matter under investigation. Normal incidence spherical mirrors with reflectivity 10%–20% have been used for focusing an XUV beam of ~ 20 eV photon energy in the sub-4 μm level reaching intensities $>10^{13}$ W/cm^2 [8]. This XUV focusing geometry has been extensively used for the temporal characterization of asec pulses via IR/XUV cross-correlation [2, 3, 9], 2nd-order autocorrelation approaches (using spit spherical mirror (Fig. 2) in unit 5 or split silicon plates in unit 3) [2, 10, 11], and for imaging the ion distribution produced by linear and non-linear processes at the focus of the XUV beam [8, 12, 13]. In order to characterize the XUV beam after the interaction with the system under investigation, XUV diagnostics as those described in unit 4 can also be placed at the output of the 5th unit.

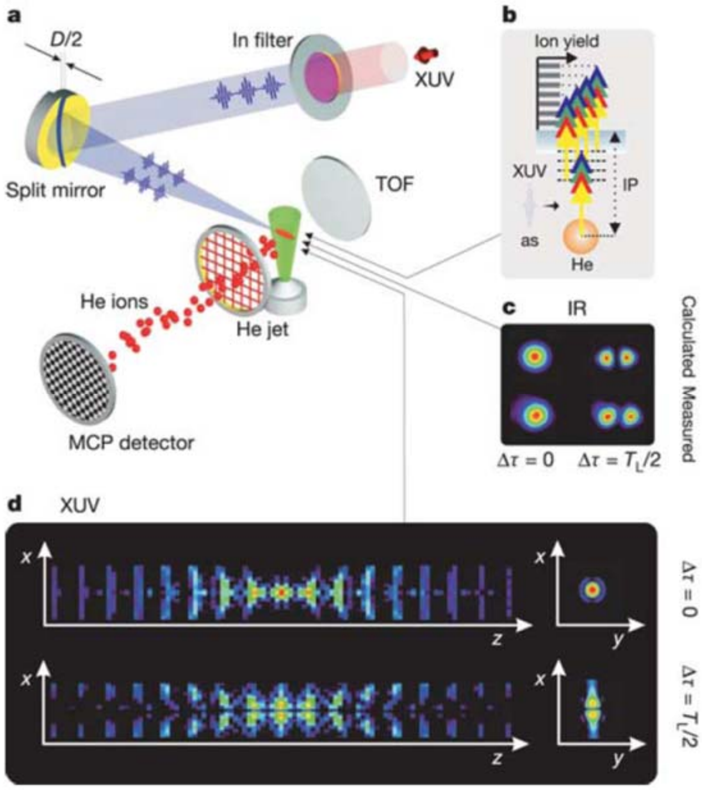

Figure 2. (a) The second-order XUV autocorrelator consists of a spherical mirror, split into two halves, serving as a focusing wavefront divider. As a nonlinear detector, a two-photon ionized He gas (b) is used and the He ion yield, recorded by a time-of-flight (TOF) mass spectrometer, provides the autocorrelation signal. (c) and (d) show the principle of the volume autocorrelator. (c) For zero and $\Delta\tau=T_L/2$ delay, the calculated and measured transverse intensity distribution for the laser frequency (IR) changes from an Airy spot to a double maximum distribution. The three-dimensional colour contours in (d) depict, in logarithmic scale over three decades and for a displacement of 0 and D= $\lambda/2$ ($\Delta\tau=T_L/2$ delay), the simulated XUV intensity distribution at the ionization region. Although the total energy remains the same, this variation in the spatial distribution allows for second-order autocorrelation measurements. From. ref. [10].

1.1 MWatt XUV beam line

The beam line can deliver attosecond pulse trains (having an overall XUV pulse duration < 10 fs) and isolated attosecond pulses in the 15-30 eV spectral range and energy in the target area (placed into the end station) in range of hundred-nJ/pulse. The XUV peak intensity at the XUV focus can reach the level of 10^{14} W/cm^2. One branch of the beam line is equipped with a wave front splitting arrangement (split spherical mirror) and is dedicated used for XUV-pump-XUV-probe studies in the attosecond time scale [1, 10, 11]. The second branch is equipped with an ion imaging detection (Ion Microscope) used for the spatial characterization of the XUV beam and for quantitative measurements in the linear and the non-linear XUV regime [8, 13].

Figure 3 shows a picture of the ≈ 10 m long MWatt attosecond XUV beam line. The beam line has been used for the generation, characterization and applications of intense asec pulses generated in gas-phase media [1].

The IR beam is focused with an f = 3 m focal length lens into a pulsed gas jet, where the harmonic radiation is generated. When Xe gas is used the energy of the emitted XUV radiation at the output of the medium is in the order of 1μJ. The XUV beams after from the XUV-S and XUV-M untis is focused into the target area (placed into the End Station) with a spherical gold mirror of 5 cm focal length. With this configuration XUV peak intensities up to 10^{14} W/cm^2 have been achieved at the focus of the XUV beam [8, 11, 13]. The energy of the XUV radiation in the interaction region (tens-μJ/pulse) is obtained from the measured pulse energy using an XUV calibrated photodiode (placed in the XUV-M unit) taking into account the reflectivity of the gold spherical mirror. The harmonic spectrum can be measured either by recording energy resolved photoelectron spectra resulting from the single-photon photoionization of Ar by the harmonic comb, or by means of an XUV spectrometer.

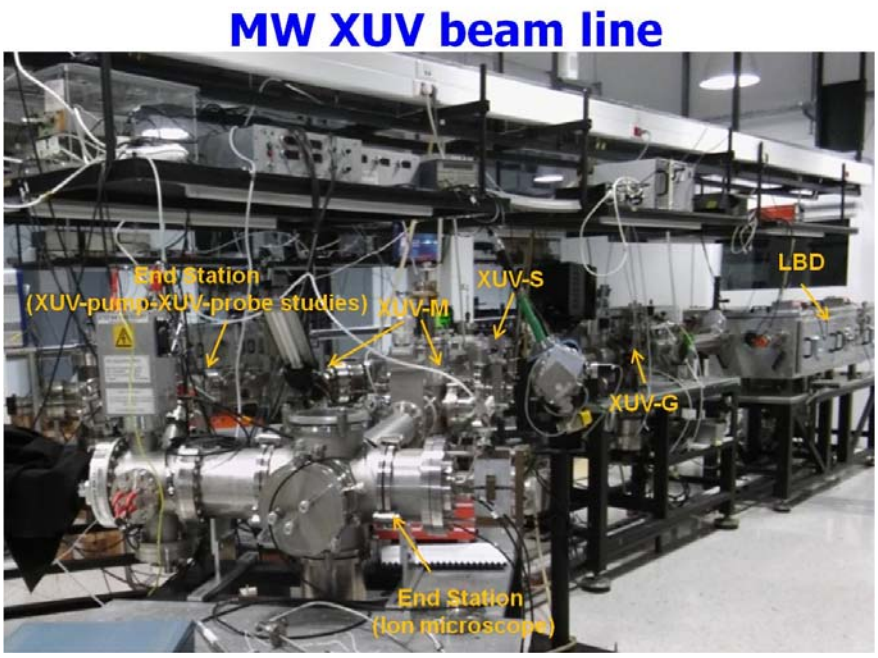

Figure 3. Picture of the ≈ 10 m long MWatt XUV beam line. LBD: "Laser Beam Delivery", which is used for the manipulation of the driving field towards the generation of the XUV radiation, contains the laser beam steering, polarization shaping, beam shaping, pulse manipulation and focusing optics. XUV-G: "XUV Generation" is used for the generation of the XUV radiation and contains the XUV generation medium. XUV-S: "XUV Separation", contains the optical elements which are used for the isolation of the generated XUV radiation from the driving laser field and the steering of the XUV beam. XUV-M: "XUV Manipulation", contains

optical elements used for spatial and spectral selection of the XUV radiation. End Station (XUV-pump-XUV-probe): Used for the temporal characterization and the applications of the asec pulses, contains the XUV focusing elements, the XUV diagnostics and the detectors required for performing XUV-pump-XUV-probe experiments in the attosecond time scale.

The electron spectra were recorded using a μ-metal shielded time-of-flight (TOF) ion/electron spectrometer, attached to the 1st XUV beam-line branch. The TOF can be set to record either the photoelectron energy distribution or ion-mass spectrum. Using a split spherical mirror of 5 cm focal length (Fig.2) in the TOF branch the duration of the asec pulses in a train was obtained by means of 2nd order volume autocorrelation (2-IVAC) measurements [1, 2, 10]. When a PG optical arrangement [7, 14] is introduced in LBD unit the XUV spectrum switches form a harmonic comb to continuum which reflects the generation of isolated asec pulses. It has been found that the intensities of the XUV pulses in the interaction area can be up to 10^{14} W/cm^2 [11], while their durations can be obtained by means of 2-IVAC measurements in case the CEP of the driving field is stabilized or measured and tagging approaches are applied [15]. These pulses were used for the observation of two-XUV-photon double ionization in Xenon gas [11], for time-resolved XUV spectroscopy studies [16] and XUV-XUV pump-probe measurements of ~1 fs scale dynamics in atoms [11] and molecules [17]. The temporal characterization of asec pulses generated in gas-phase media will be described in Section 4.

The second branch of the beam line is equipped with an Ion imaging detector (Ion Microscope) [13, 18] (with spatial resolution of ≈ 1 μm) and used for the spatial characterization of the XUV beam (Fig. 4) and for quantitative measurements in the linear and the non-linear XUV regime [8, 13].

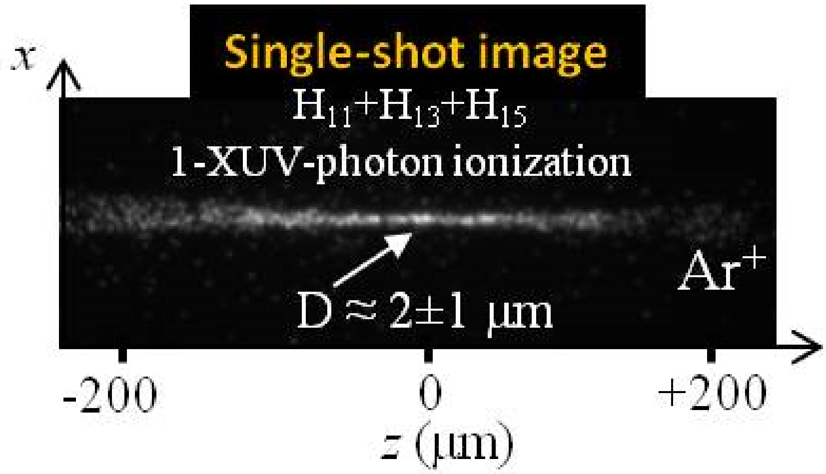

Figure 4. Single-shot ion distribution recorded at the focus of an XUV beam which contains the harmonics from 11th to 15th. The ion distribution was produced through 1-XUV-photon ionization of argon. From. ref. [13].

1.2 GWatt XUV beam line

The beam line can deliver XUV pulses (in the form of attosecond trains with overall duration < 10 fs) in the 15-35 eV spectral range with energy in the range of hundreds of μJ/pulse at the exit of the harmonic generation medium [19]. The energy of the XUV in the target area placed into the end stations is in the range of tens-μJ/pulse and the peak intensity (in case of using a spherical mirror of f=5 cm) is in the range of 10^{16} W/cm^2.

Figure 5 shows a picture of the ≈ 20 m long GWatt attosecond XUV beam line. The enchancment of the XUV photon flux was achieved by utilizing loose focusing geometries and the precise control of the phase matching conditions achieved by means of single-/multi gas jet configurations [19].

The laser beam is focused into the generation area through a f=9m spherical mirror. For the generation two to four gas jets, with variable mutual distance, can be used utilizing what is called quasi phase matching. Rare gases are used for the generation process. The separation of the IR is by reflection on a Si plate and absorption of the residual IR by an band pass thin metal filter which allow the transmission of harmonics with order ≥ 11th . The harmonic spectrum is measured either by measuring the energy resolved photoelectron spectra of the single photon ionization of Ar or by a flat field XUV spectrometer. The total energy of the comp of the harmonics is measured using a calibrated XUV photodiode. We have measure 100-200 μJ scale pulse energies when Xenon or Argon gas is used for the generation, which translates to 10^{13} photons/pulse scale XUV photon fluxes. The generation is maximized when two Xenon jets are used in a phase matched arrangement. The duration of the XUV pulse train has not been measured.

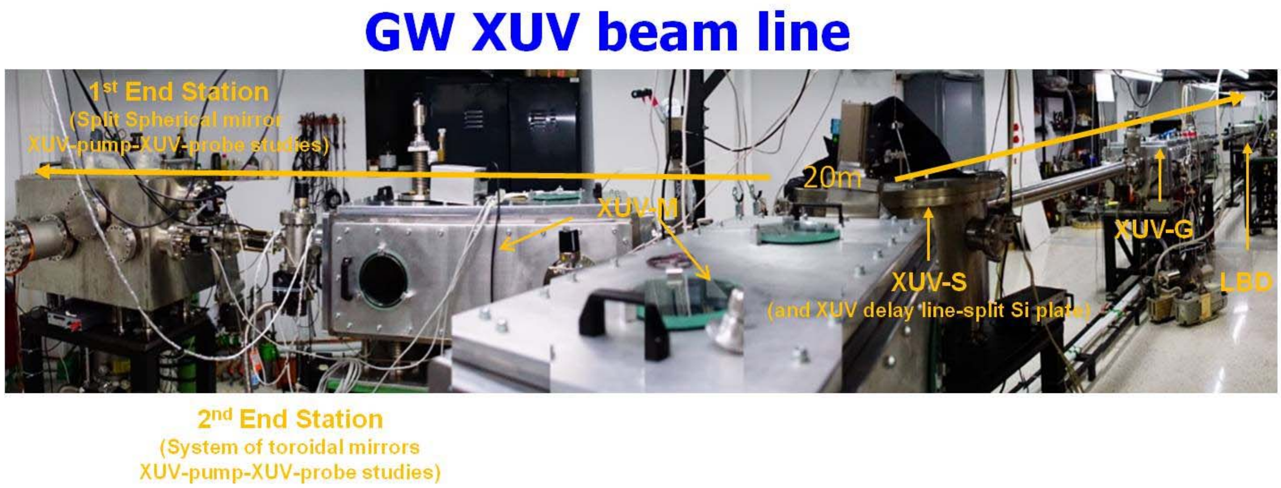

Figure 5. Picture of the ≈ 20 m long GWatt XUV beam line. LBD: "Laser Beam Delivery", which is used for the manipulation of the driving field towards the generation of the XUV radiation, contains the laser beam steering, polarization shaping, beam shaping, pulse manipulation and focusing optics. XUV-G: "XUV Generation" is used for the generation of the XUV radiation and contains the XUV generation medium. XUV-S: "XUV Separation", contains the optical elements which are used for the isolation of the generated XUV radiation from the driving laser field and the steering (and XUV wave front splitting in case that is needed) of the XUV beam. XUV-M: "XUV Manipulation", contains optical elements used for spatial and spectral selection of the XUV radiation. 1st End Station: Dedicated for the temporal characterization and the applications of the asec pulses. It contains the XUV focusing elements (Split spherical mirror of 5 cm focal length), the XUV diagnostics (Flat field XUV spectrometer) and the detectors (TOF electron/ion spectrometers) required for performing XUV-pump-XUV-probe studies in the attosecond time scale. 2nd End Station: Dedicated for the temporal characterization and the applications of the asec pulses. It contains the XUV focusing elements (system of toroidal mirrors of 50 cm focal length), the XUV diagnostics/detectors (TOF electron/ion spectrometers and Ion microscope) required for performing XUV-pump-XUV-probe studies in the attosecond time scale.

The generated high XUV photon flux has been verified by measuring multiple ionization of Ar. The XUV beam was focused by a gold coated spherical f=5 cm mirror. The results have been published in ref. [18].

2. Conclusions and Perspectives

Two high power XUV beam lines are currently operational in the upgraded Attosecond Science and Technology Lab. of FORTH-IESL for performing time resolved studies in the XUV spectral range.

The first beam line can deliver attosecond pulse trains (having an overall XUV pulse duration < 10 fs) and isolated attosecond pulses in the 15-30 eV spectral range and energy in the target area (placed into the end station) in range of hundred-nJ/pulse. The XUV peak intensity at the XUV focus can reach the level of 10^{14} W/cm^2 [1].

The second beam line can deliver XUV pulses (in the form of attosecond trains with overall duration < 10 fs) in the 15-35 eV spectral range with energy in the range of hundreds of μJ/pulse at the exit of the harmonic generation medium [19]. The energy of the XUV in the target area placed into the end stations is in the range of tens-μJ/pulse and the peak intensity (in case of using a spherical mirror of f=5 cm) is in the range of 10^{16} W/cm2.

Each beam line consist two end stations equipped with XUV focusing elements (Split spherical mirror of 5 cm focal length), XUV diagnostics (Flat field XUV spectrometer) and detectors (TOF electron/ion spectrometers, Ion microscope) required for performing XUV-pump-XUV-probe studies in the attosecond time scale and quantitative studies in the linear and non-linear XUV regime.

The HHG workstations developed in the current Deliverable have been employed in two Research articles [18,19] acknowledging the funding from NFFA-Europe.

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