ELI Flagship Experiment Proposal

BRIEF INFORMATION

Title:
Intense, spectrally-tunable XUV pulses from High-Harmonic Generation to study collective dynamics at the nanoscale

Short Title/ ID of Project: XUV tuning

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ELI Facility: ELI-BL

Experimental Area: XUV science, ultrafast dynamics

Experimental chamber / station: MAC station, HHG source, Allegra laser

Phase: Flagship Experiments

Estimated Duration (days of beamtime & preparation): 60

Risk estimation & quantification: Moderate.

Preliminary work shows that the Allegra-HHG beamline has the ability to generate XUV pulses with a tunability across >1.5 eV in the XUV range (e.g. from below 21 to above 22.5 eV, covering the He absorption centered at 21.6 eV). Risks are mainly related to the speed and reliability that can be achieved for the tuning process. Furthermore, the development of methods for normalizing the experimental data is a challenge.

LASER SYSTEM PARAMETERS

Pulse duration (fs): <20 fs **Wavelength (nm):** Tunable, from about 840 to 770 nm (2nd harmonic 385-420nm)

Pulse energy (J): 30 to 70 mJ **Repetition rate (Hz):** kHz

Others: The ability to generate 2nd harmonic in the HHG beamline from the high power Allegra beam. Generating higher harmonics with the 2nd harmonic of the drive laser gives higher harmonics with a greater spacing which is preferable (when used together with multilayer XUV optics or the XUV monochromator) for truly monochromatic probing (or pumping) of the sample. Additionally, due to the phase matching conditions, which needs to be fulfilled for efficient SHG generation, natural shot-to-shot fluctuations of NIR spectrum are not transferred to fluctuations of central photon energy of HHGs.

The project requires stable operation of L1 Allegra laser and careful laser beam diagnostics through the full experimental chain, including the Allegra laser, Beam Transport, HHG beamline and the MAC end-station. Therefore, efficient communication and coordination of activities between 3 teams; L1 Allegra, HHG and MAC together with the external user group are essential for this project.

Diagnostics: Laser: Diagnostics for the spectral characteristics as a function of Dazzler settings, for wavelength dependent divergence, and the spatial chirp. Furthermore, standard laser diagnostics: pulse energy, pointing, spectrum. XUV: pulse energy, spectrum, pointing relative to drive laser and ideally wavefront. Experimental station: Development of suitable diagnostics in the interaction region of the experiment.

Special Technical Requirements for the beams: It is essential to find an optimal set of parameters for the Allegra laser within desired NIR spectral region while keeping operation of complex laser system stable.

OTHER TECHNICAL INFORMATION

EMP expected: N

Compatibility with vacuum: Y

Vacuum contamination risks: Certain sample delivery systems represent a vacuum challenge in terms of gas load, but not a vacuum contamination risk

Special technical works requested on-site: The experiment does not require significant technical works on site.

SCIENTIFIC PROPOSAL

Aims / Objectives:

As a result of OPCPA amplification scheme, the NIR spectrum of the Allegra laser is naturally very broad. Therefore, different parts of the NIR spectrum in the spectral region between 700 and 950 nm can be selectively amplified. This tunability of the fundamental laser provides an opportunity to also tune the HHG central photon energy within a certain spectral range and, therefore, optimize absorption of the sample by going into resonance with specific absorption edges in the VUV/XUV energy range of the HHG source (roughly 10 to 100 eV). When established, this tunability will be combined with the focusing geometries and pump-beam capabilities available at the multipurpose MAC station.

The central goal of the proposed development is to establish a reliable and flexible possibility for users to request VUV/XUV photon energies on demand. As a pilot, user-driven experiment utilizing this new ability we suggest the investigation of the collective autoionization (CAI) dynamics of multiply excited pure and doped helium nanodroplets. To resonantly excite the He nanodroplets, the Allegra laser will be tuned across the main He absorption resonance using the new scheme based on consecutive SHG in a BBO crystal followed by HHG in a gas cell. Systematic measurements of CAI yields and spectra will uncover new details of the CAI mechanism and potentially novel relaxation processes such as heterogeneous interatomic Coulombic decay (ICD) and CAI in helium nanodroplets doped with foreign atoms and molecules. Eventually, the back-focusing scheme will enable studies of highly nonlinear optical phenomena in unsupported molecules and nanosystems irradiated by wavelength-tunable intense XUV radiation.

Brief description of the scientific background and rationale of the project:
Just about all important functions are related to electronic excitations and the subsequent electron dynamics, including the relaxation pathways of the initial excitation. With an increasing ability to engineer materials, even on the nanoscale, the ability to determine site-specific contributions to such dynamics becomes critical. In this perspective, the ability to tune ultrashort XUV pulses to selected absorption edges gives unique abilities to study properties and functions of nano-engineered samples.

Although the suitable pilot experiments, and other immediate applications, are within the field of AMO science, other areas will benefit strongly from the development of the tuning capacity and can be relevant for flagship experiments. This includes also fields such as material science and resonant imaging. An example is in the resonant study of transition metal M edges of e.g. Fe and Ni as exemplified in the work in demagnification dynamics (e.g. 4792 PNAS|March 27, 2012|vol. 109|no. 13).

Proposed experimental method and working plan:
Q3/4 2021: Initial experiments on HHG tuning in resonance with the He absorption at 21.6 eV.
Verify the ability to tune across an absorption edge. In the case of the He pilot experiment between 20 to 22.5 eV.
Q1/2 2022: Follow up data analysis and optimization of the experimental set up.
Q2/3 2022: Further pilot experiments. Establish pump-probe experiments with the tunable XUV beam.
Q3/4 2022: Follow up data analysis and optimization of the experimental set up
Q4 2022: Main experimental run for the flagship experiment.
Studies of collective dynamics at the nanoscale using intense, tunable XUV pulses from High-Harmonic Generation
Q1/2 2023: Follow up data analysis and optimizations of the experimental set up.
Q2 2023: Outlook/contingency experiment. Beamtime to investigate the applicability of the established technique on a wider range of scientific questions. E.g. in AMO science in a different XUV energy range (different sample/absorption) or even on a different target system, e.g. a solid state target.
If necessary this beamtime can be used as a contingency beamtime to achieve the flagship experiment goal.
Q3/4 2023: Data analysis and reporting.

Description of the experimental arrangement including main optics, targetry, and diagnostics
The experimental arrangement is based on the experimental chain using the Allegra laser together with the ELI Beamlines HHG source and the multipurpose MAC scientific station.

The pulses from the Allegra laser are first used to generate 2nd harmonic in BBO, then HHG is done in Xe gas from the 2nd harmonic. In the example of the pilot experiment, tuning the L1 spectrum and the SHG stage allows tuning of the wavelength of the XUV pulses (HH7) to 21.6 eV photon energy (absorption resonance of He droplets). Initially a back-focusing mirror with a narrow reflectivity bandwidth is used to focus the XUV pulses to high intensity (potentially sufficient for non-linear XUV/matter interactions) and with high spectral purity (ensured by the wider harmonics spacing achieved when generating with the 2nd harmonic). Main detection techniques for the pilot experiment are ion and electron Time of Flight spectroscopy and Velocity Map Imaging (VMI). A challenge will be to superimpose the HHG and SHG beams to realize pump-probe experiments. New approaches for determining the spatial and temporal overlap will have to be developed.

Expected outcome / yield

Although outstanding tools for ultrafast science, HHG sources traditionally suffer from limitations due to the way that their XUV output is restricted to multiple harmonic orders of the HHG driving laser, as well as from the limited photon flux they offer. The proposed development will address both of these central aspects and will place the HHG beamline at ELI Beamlines in a unique parameter regime. We will develop the ability to provide requested photon energies for users in order to optimize photon absorption of a sample by tuning the XUV photon energies to specific resonances. In the process of establishing this ability we will also optimize the focusing conditions of the HHG pulses in the interaction region in order to achieve the highest possible demagnification of the XUV source in the experiment, and thus, significantly increase photon flux on the sample (as well as minimize photon losses by using fewer optics in the XUV beam paths). What we aim to realize will lead to publications based on the technical development:

- Demonstrate the wavelength tunability of XUV pulses using a HHG source.
- Establish the back-focusing geometry to reach high XUV intensities; characterize the spot size and intensity.
  Develop a scheme for HHG-SHG pump-probe spectroscopy in the back-focusing geometry.
- Investigate the potential for this tunable source development for applications beyond AMO science.
- Investigate the potential to combine the tuning capability with broadband in-line focusing (potentially with the use of the XUV monochromator available on the HHG beamline) to allow tunability in the entire XUV range of the HHG source for selected harmonics or the broad band emission.

Pilot experiments on “the investigation of the collective autoionization (CAI) dynamics of multiply excited pure and doped helium nanodroplets” will showcase the new abilities potentially generating publications in a high-ranking journals on a number of topics, e.g.:

- Characterization of the transition from ICD (double excitation of He droplets) to CAI (multiple excitation) in He nanodroplets and in Ne clusters.
- Measurement of the angular distribution of electrons created by ICD of Xe and Li attached to He nanodroplets upon resonant excitation of the droplets.
- Pump-probe measurement of the dynamics of heterogeneous ICD in the systems (He*+Xe and He*+Li).
- Pump-probe measurement of the dynamics of CAI of multiply excited He nanodroplets.

Relevant publications in the field by participating scientists, in reverse chronological order

No.		Year

1	A Multipurpose End-Station for Atomic, Molecular and Optical Sciences and Coherent Diffractive Imaging at ELI Beamlines E Klimešová, *et al*., The European Physical Journal Special Topics, 1-12	2021
2	Enhancement of Above Threshold Ionization in Resonantly Excited Helium Nanodroplets, R. Michiels, *et al*., Phys. Rev. Lett. 127, 093201	2021
3	Ultrafast resonant interatomic Coulombic decay induced by quantum fluid dynamics, A. C. LaForge, *et al*., Physical Review X 11, 021011	2021
4	Ultrafast relaxation of photoexcited superfluid He nanodroplets, M. Mudrich *et al*., Nat. Commun. 11, 112	2020
5	High-flux source of coherent XUV pulses for user applications, O Hort, *et al*., Optics express 27 (6), 8871-8883	2019