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32.7
Report on various facilities for pump/probe spectroscopic studies in XUV

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Deliverable Nature	
R = Report, P = Prototype, D = Demonstrator, O = Other	R
Dissemination Level	
PU = Public PP = Restricted to other programme participants (incl. the Commission Services) RE = Restricted to a group specified by the consortium (incl. the Commission Services) CO = Confidential, only for members of the consortium (incl. the Commission Services)	PU

A. Abstract / Executive Summary

The development of user-oriented beamlines is required for applications of XUV femtosecond and attosecond pulses to atomic, molecular and solid-state physics. Many **INREX** participants have worked to develop these beamlines, which have enabled access experiments to be performed in gas-phase molecules and novel materials. Beamlines are also being developed to deliver high-energy XUV pulses to target, enabling XUV-pump XUV-probe experiments. In parallel, **INREX** partners have been developing new techniques for generating and doing spectroscopic experiments in the UV, which will enable novel spectroscopic XUV experiments in the near future.

B. Deliverable Report

1 Introduction

The goal of this deliverable is to make available a wide range of user-oriented beamlines and spectroscopic techniques enabling the application of ultrashort pulses of XUV to a variety of application areas.

2 Objectives

The objective of this deliverable was to develop techniques to apply ultrafast (phase-controlled) laser sources to pump-probe studies in atomic and molecular systems, ranging from pump-probe experiments involving photons and electrons as probes and pumps and vice-versa, with arbitrary wavelengths.

In the condensed phase, the goal was to enable pump-probe experiments at surfaces to be executed with few-fs pulses or with single ***as*** pulses in streak-camera measurements. Time- and angle-resolved photo-emission, Fermi-surface dynamics and optical-pulse excited gratings on metals/semiconductor surfaces can then be probed by XUV radiation.

A further goal is to enable nonlinear optics with isolated ***as*** pulses.

3 Work performed / results / description

Several **INREX** participant laboratories have further developed user-oriented beamlines for applications of XUV pulses.

POLIMI have developed an optical system for micro-focusing attosecond pulses using grazing-incidence toroidal mirrors, with negligible aberrations; long exit arm and a plane split-mirror to generate pump and probe pulses [1]. The optical scheme is based on two sections to first de-magnify the source and then correct for coma. An XUV spot with no evidence of coma and FWHM diameter of 8 μm has been measured. Trains of attosecond pulses have been generated with a measured peak intensity of about 3×10^{11} W/cm^2.

CELIA has developed a new split-mirror arrangement with large aperture and high stability that can be used at 45° incidence. This split mirror has a passive control only and allowed 50 attosecond stability on the IR-XUV delay control to be achieved. A large aperture (15 mm diameter) was necessary to use this system with a high energy XUV line developed on a 10 Hz TW laser system (Figure 1). A time-of-flight ion spectrometer has also been developed and implemented on this line. The high energy XUV source, split mirror and time of flight apparatus are now all coupled together and the ensemble is being used to perform characterization of attosecond pulses in the temporal domain (see D32.8).

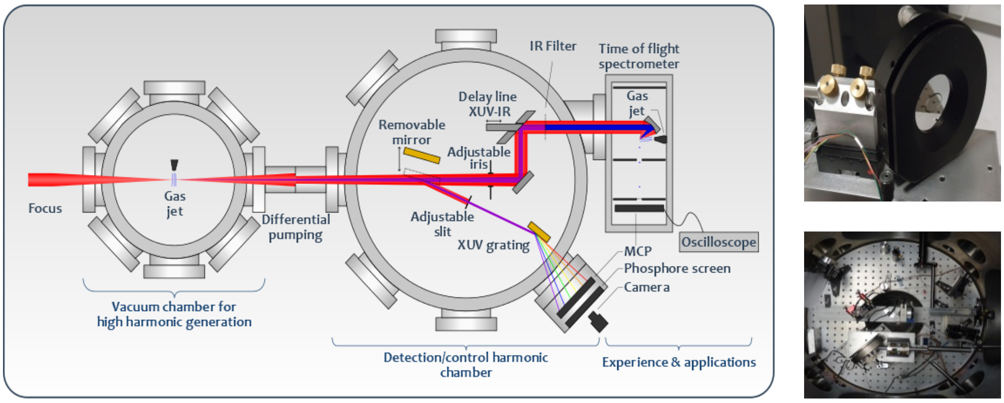

Figure 1. *XUV beamline designed for TW laser system at CELIA (left), showing the split mirror (top right) and detection/control harmonic chamber (bottom right).*

The **CLF** has upgraded the performance of the Artemis open-access facility for time- and angle-resolved photoelectron spectroscopy with HHG pulses by increasing the pump energy delivered to target in the infrared (up to 20 uJ at 11 micron); increasing the pump photon energy range to the UV; installing an additional high resolution grating to provide energy resolution <100 meV and demonstrating <10 fs time-resolution with <500 meV energy resolution on the beamline.

CEA-LIDyL-SLIC and **CNRS-LOA** are investigating the potential of the atto lighthouse configuration of HHG in gases and plasmas for new types of time-resolved studies. **CEA-LIDyL-SLIC** has completed a versatile attosecond beamline installed on the PLFA 1 kHz laser [2]. It combines a very complete set of diagnostics (harmonic amplitude, phase and polarization) and advanced electron spectrometers (MBES and COLTRIMS) enabling high harmonic spectroscopy and photoionization experiments. Passive and active stabilization allow long-term stability for experiments requiring long acquisition times.

At **CLPU**, a new XUV user facility based on a CEP-stabilized ultrafast laser system is in development.

FVB-MBI commissioned a series of new pump-probe beamlines [3], including a setup with a time-compensating XUV monochromator and a setup for femtosecond time-resolution XUV pump-XUV probe experiments and a setup for two-color IR/UV-XUV experiments where high energy XUV is generated by means of two-color HHG.

These XUV beamlines have been used for a wide-variety of pump-probe studies in the XUV.

The **CLF** beamline at Artemis has been used for studies of electron dynamics in novel materials including graphene, liquid jets and gases, several of which were supported by Laserlab access [4-8]. **FORTH** has performed the ever first XUV-pump-XUV-probe study of molecular vibrational, electronic and ionization dynamics in the 1 fs temporal regime using broad band coherent XUV continua generated through polarization gating assisted HHG in atomic Xenon. 1 fs ionization dynamics have been measured in molecular Hydrogen. **CEA-SLIC** have collaborated with **CNRS-LOA** to carry out pump-probe studies using Coherent Diffractive Imaging (CDI) in the XUV spectral range. They have completed a study of the field-induced demagnetization dynamics of magnetic nano-domains in a thin ferromagnetic Co/Pd film [9]. The fast demagnetization (~100 fs) is attributed to direct spin transfer between neighboring domains. In a similar pump-probe scheme using CDI, time-resolved studies of plasma expansion in nanostructures are being performed. **IST** studied the feasibility of probing solid density plasmas with High Harmonic Generation in collaboration with **CNRS-LOA** and **CEA-SLIC**. The wavefront stability of the HHG probe was measured, showing that wavefront deflections above lambda/3 could be detected, which validated the design of a density measurement of warm dense plasmas using a wavefront sensor [10].

In parallel, techniques for generating ultrashort UV pulses, and for performing spectroscopy in the UV are being developed.

POLIMI has generated ultra-broadband UV pulses by sum frequency generation of a narrowband near-infrared pulse and a broadband visible pulse from a non-collinear optical parametric amplifier (NOPA). A novel device for the characterization of the spectral phase of the ultraviolet pulses has been developed, which is a variant of two-dimensional spectral interferometry (2DSI) based on difference frequency generation. UV pulses with µJ-level energy ranging from 320 to 380 nm as short as 8.5 fs have been generated and fully characterized.

LLAMS has developed a new method of Ramsey-comb spectroscopy, and the special ultrafast double-pulse amplifier required for it [11-12]. The phase information extracted from the Ramsey signals enables kHz accuracy spectroscopy with ultrafast high power laser pulses, as shown on the 5S-7S two-photon transition at 760 nm in Rb. Because of the high peak power, easy frequency conversion is enabled for precision spectroscopy in the deep-UV and the extreme ultraviolet. Doppler-free two-photon spectroscopy in krypton was demonstrated at 212 nm with an accuracy of 100 kHz, which is 35 times better than achieved before. Efforts to extend Ramsey-comb spectroscopy with HHG to the XUV wavelength domain are in progress.

4 Conclusions

Many **INREX** participants have worked to develop beamlines to deliver femto- and attosecond XUV pulses for applications in atmic/molecular and solid-state physics. These have enabled access experiments to be performed in gas-phase molecules and novel materials. Beamlines are also being developed to deliver high-energy XUV pulses to target, enabling XUV-pump XUV-probe experiments. In parallel, **INREX** partners have been developing new techniques for generating and doing spectroscopic experiments in the UV, which will enable novel spectroscopic XUV experiments in the near future.

5 Publications

[1] F. Frassetto, A. Trabattoni, S. Anumula, G. Sansone, F. Calegari, M. Nisoli, L. Poletto, *"High-throughput beamline for attosecond pulses based on toroidal mirrors with microfocusing capabilities",* Rev. Sci. Instrum. **85**, 103115 (2014). doi:10.1063/1.4898671

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[9] M Ducousso, X Ge, W Boutu, D Gauthier, B Barbrel, F Wang, A Borta, A-I Gonzalez, M Billon, B Vodungbo, J Gautier, R Hawaldar, B Tudu, R Delaunay, M Tortarolo, P Zeitoun, J Lüning and H Merdji, *"Single shot studies of a Co/Pd thin film's magnetic nanodomain structure using ultrafast xray scattering"*, Laser Physics **24**, 025301 (2014). doi:10.1088/1054-660X/24/2/025301

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6 Persons Involved

CEA-SLIC: S. Weber, B. Manschwetus, V. Gruson, T. Ruchon, M. Géléoc, Willem Boutu, B. Carré;

CNRS-LOA: Ph. Zeitoun,

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CLPU: J A Perez Hernandez;

CNRS-CELIA: Ludovic Quintard, Ondrej Hort, Antoine Dubrouil, Dominique Descamps, F. Catoire, Eric Mével and Eric Constant;

FORTH: D. Charalambidis, P. Tzallas, P.A. Carpeggiani, D. Gray, G. Kolliopoulos; **LLAMS:** J. Morgenweg, I. Barmes, R. Altmann, L.S. Dreissen, S. Galtier, K.S.E. Eikema; **POLIMI:** A. Trabattoni, G. Sansone, F. Calegari, M. Nisoli, A. Oriana, G. Cerullo, C. Manzoni.

IST: S. Künzel, G. O. Williams, M. Fajardo