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Deliverable D32.8
Report on study of multiphoton processes in the XUV from gas and solid targets

Lead Beneficiary: 2 CEA

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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 studies of multiphoton processes in the XUV, in gas and solid targets, include three related topics: 1) Studies of multiphoton processes involving only XUV photons, 2) Studies of multiphoton processes involving at least one XUV photon, in combination with laser photons in the IR-vis-UV spectral range, 3) Development of new schemes for efficiently producing and controlling XUV photons, and implementing them in nonlinear studies. The nine partners have produced a variety of original demonstrations within the three topics.

B. Deliverable Report

1 Introduction

The studies of multiphoton processes in the XUV, in gas and solid targets, include three related topics:

1. ***Studies of multiphoton processes involving only XUV photons***, i.e., photons of energy between 10 and 1000 eV.
2. ***Studies of multiphoton processes involving at least one XUV photon***, in combination with laser photons in the IR-vis-UV spectral range.
3. ***Development of new schemes for efficiently producing and controlling XUV photons***, and implementing them in nonlinear studies.

In ***topic 1***, in the general case where they are non resonant, nonlinear processes have low cross sections (e.g., $\sigma < 10^{-52}$ cm^4s in two-photon ionization of valence shell atom). They become dominant over linear processes only at high XUV flux (number of photons on target > 10^{10} phot/s, intensity > 10^{12} W/cm^2). This favors choice of high density gas – as it is the case in contributions to D32.8-1 - or solid target. Many fundamental processes may be investigated in XUV/XUV pump-probe experiment, e.g., valence and core shelle photoemission, multiple ionization, electronic transfer and migration, ultrafast electronic and nuclear non adiabatic (non Born-Oppenheimer) dynamics.

In ***topic 2***, time-resolved multi-frequency studies, of the type [XUV/IR-vis-UV + target → products], have considered electronic and nuclear ultrafast dynamics in molecules in the gas phase. Since they involve temporal/spectral properties, e.g., coherence, duration, multi-frequency studies also serve to characterize the XUV field. Finally, besides the above case where XUV in the input channel is scattered on target, XUV can also be produced in the output channel by scattering [IR-vis-UV + target → XUV, products]. This is the case of HHG in gas itself, and the valuable applications of nonlinear harmonic spectroscopy to ultrafast dynamics in strong field.

In topics 1 and 2, besides they discrimination in specific detection channels (energy, charge and mass of product particles, absorbed/emitted light properties), measurement of nonlinear processes requires optimization of the XUV flux and special experimental arrangement which enhances their relative contribution. This is performed in ***topic 3***.

The nine partners in D32.8 have provided balanced contributions between the three above topics, i.e., a total of 4-6 contributions to each of them – see table. This results into 15 publications (with LASERLAB acknowledgements) and 3 articles under publication.

N°	partner	contribution	D32.8 topic
1	MBI	*Rare-Gas clusters in intense extreme-ultraviolet pulses from a high-order harmonic source*	1
6	FORTH-IESL	*Disclosing intrinsic molecular dynamics on the 1-fs scale through extreme-ultraviolet pump-probe measurements*	1
6	FORTH-IESL	*studies of direct (non sequential) two-XUV-photon double ionization in xenon*	1
6	FORTH-IESL	*studies of two-photon ATI in rare gas induced by a comb of harmonics*	1
6	FORTH-IESL	*studies of spatially resolved two-XUV-photon ionization of He*	1
15	Lund LLC	*Two-photon double ionization of neon using an attosecond pulse train*	1
3	CEA-SLIC	*Attosecond light and electronic vortices*	2
5	CNRS-CELIA	*Nonlinear cross correlation for attosecond pulse measurements in the temporal domain*	2
19	POLIMI	*Electron dynamics in molecules*	2
4	CLPU	*High-order-harmonic generation driven by metal nanotip photoemission*	3
5	CNRS-LUMAT	*Enhanced high harmonic generation driven by high-intensity laser in argon gas-filled hollow core waveguide*	3
16	LLAMS	*OPCPA system for driving HHG*	3
19	POLIMI	*Non-collinear high-order harmonic generation by three interfering laser beams*	3

Table 1 : contributions to the three topics of D32.8

2 Objectives

2.1 Multiphoton processes involving only XUV photons

Objective was to demonstrate NL processes involving only XUV photons. It has been attained in the studies by MBI, IESL and LLC in the gas phase.

2.2 Multiphoton processes involving combined XUV and IR-vis-UV photons

Objective was to demonstrate multi-frequency NL processes combining XUV and IR-vis-UV photons, in either time-resolved pump-probe studies of electronic and nuclear dynamics in target systems, or in cross-correlation techniques for characterizing the XUV fields. It has been attained in the studies by POLIMI, CEA-SLIC and CELIA in the gas phase.

2.3 New schemes for generation and control of XUV pulses and nonlinear studies

Objective was to demonstrate new schemes for efficient production of ultrashort XUV pulses, control of their properties and conditions of their interaction with the target. It has been attained in the studies by POLIMI, CNRS-LUMAT, CLPU and LLAMS.

3 Work performed / results / description

3.1 Multiphoton processes involving only XUV photons

MBI has implemented a HHG source using a loose focusing configuration to efficiently generate XUV pulses of energy >1 µJ in the 17-35 eV photon energy range. Using a spherical multilayer mirror with 7.5 cm focal distance, XUV intensity close to 1013 W/cm2 was achieved at the focus. With this source, MBI investigated sequential multiphoton ionization in Ar atoms and rare gas clusters using XUV-IR pump probe spectroscopy [Schütte2014a, Schütte2014b]. A THz source synchronized with HHG was as well developed and used to characterize the XUV pulses using the THz streaking technique, showing pulse duration of 20 fs or below.

IESL has completed previously reported studies of direct (non sequential) two-XUV-photon double ionization in xenon [Tzallas2012]. In a joint program using the XUV harmonic source from plasma on solid target at MPQ, IESL has reported two-photon ATI in rare gas induced by a comb of harmonics [Heissler2012]. In a time-resolved XUV-pump-XUV-probe study, IESL has investigated coupled electronic and nuclear non Born-Oppenheimer dynamics at 1-fs scale in molecular hydrogen [Carpeggiani2014]. IESL has reported of spatially resolved two-XUV-photon ionization of He to be used in the future for single shot 2^{nd} order autocorrelation measurements of XUV pulses (work under review).
In 2012-13, FORTH has implemented a major upgrade of its attosecond Science & Technology lab, reducing temporarily its experimental activities.

LU-LLC has significantly increased the pulse energy of the Lund high-intensity attosecond beam line by implementing a loose focusing geometry (focal length 9 m) together with long (length 6 cm) generation gas cell. The XUV source and beamline now deliver attosecond pulse trains in the photon energy range 17-50 eV, with total pulse energy at the µJ level. LU-LLC has worked out formal scaling laws for scaling of HHG to both low and high laser pulse energies. More generally, this formal scaling of energy and geometrical parameters applies to many nonlinear optics processes in gases.
The XUV beam is focused by two toroidal mirrors in Wolter configuration, to high intensity $>10^{12}$ W/cm^2, sufficient to induce two-photon double ionization in Ne and Kr. Currently, a double-sided velocity map imaging spectrometer is implemented to record simultaneously the momentum distribution of ions and photoelectrons. Future studies of electron correlations in nonlinear multiple ionization of atoms and molecules in the XUV range are envisioned.

3.2 Multiphoton processes involving combined XUV and IR-vis-UV photons

The nonlinear processes under study involve at least one XUV photon combined with IR-vis-UV photons from perfectly synchronized lasers. Time-resolved pump-probe studies investigate ultrafast electronic and nuclear dynamics in gas or solid state systems. Conversely, multi-frequency processes serve to characterize specific properties of the XUV pulses, such as temporal structure or orbital angular momentum.

CEA-SLIC has studied electronic dynamics in SF_6 under strong field irradiation, using high harmonic amplitude and phase spectroscopy. It gives evidence of multiple orbital dynamics in a multi-center molecular system [Manschwteus2015a]. CEA-SLIC has studied nonlinear HHG in the gas phase driven by IR field carrying orbital angular momentum (OAM). After the first works reported, the question of the OAM transfer in a highly nonlinear process such as HHG was still debated. CEA-SLIC used XUV/IR cross-correlation in the FROG-type RABBIT technique to characterize OAM of the XUV field. Measurement confirms that OAM is conserved in the nonlinear process [Géneaux2015].

Nonlinear cross correlation for attosecond pulse measurements in the temporal domain
CELIA has designed an original method to track the temporal profile of XUV atto pulses. It exploits the cross correlation between IR and XUV fields in the double ionization of a gas target. Tunnel ionization by the IR field produces a temporal sequence of bursts of singly charged ions, each of ~200 as duration. Ions are sequentially ionized by the atto pulse in the dication species. Form the nonlinear cross correlation between the ion bursts and atto pulse, one can directly extract the atto pulse duration. Experimental tests are currently performed in collaboration with the ILM-Lyon.

3.2.1 POLIMI

POLIMI has investigated charge migration in amino acid phenylalanine initiated by isolated attosecond pulses [Belshaw2012, Calegari2014, Calegari2015]. Electron transfer in molecular complexes is of crucial importance since it triggers the first steps in a number of biochemical processes. Efficient charge dynamics can be driven by purely electronic effects, which evolve on a few femtoseconds down to tens of attosecond timescale. In the pump-probe study on phenylalanine, an isolated XUV atto pulse triggers charge dynamics in the phenylalanine cation, whereas probe laser pulse produces double ionization and fragmentation, e.g., into immonium dication and COOH group. On the basis of theoretical simulations, coherent oscillation of the immonium dication yield is attributed to the ultrafast electron dynamics, prior to any nuclear motion.

3.3 New schemes for generation and control of XUV pulses and nonlinear studies

CLPU, in collaboration with ICFO and Imperial College, has theoretically explore new routes in HHG at high laser intensity, i.e., beyond the saturation intensity for which atoms are efficiently ionized [Pérez2014]. Detailed study of the electron dynamic outlines the contribution of “non adiabatic trajectories” that take place in the steep turn-on of the driving laser field. In parallel, CLPU has explored theoretically the possibility of generating ultrashort XUV pulses in HHG from metal nanotips; this alternative way of producing coherent XUV light seems promising [Ciappina2014a,b].

CNRS-LUMAT and CELIA have jointly worked on the optimization of the XUV yield in HHG. They demonstrated that 70-80 eV photons can be efficiently generated from guided propagation in long Ar medium at high laser intensity (above saturation intensity), in a regime where several nonlinear processes interplay to determine pressure-dependent phase matching [Cassou2014]. In collaboration with the General Physics Institute of the Russian Academy of Sciences (V. Strelkov), CELIA proposed a new approach to characterize the CEP of intense ultrashort pulse which is highly relevant for HHG optimization [Strelkov2014].

LLAMS has developed a double-pulse ultrafast amplifier based on OPCPA and a Nd:YVO4/Nd:YAG diode-pumped pump laser system. It can stably and equally (energy within 1%) amplify two ultrafast pulses from a frequency comb laser independently of each other to 1-5 mJ (depending on conditions) with pulse delays from 8 ns to well into microsecond time scales. With this system harmonics can be produced at XUV wavelengths <50 nm for multi-photon precision spectroscopy and pump-probe experiments. A XUV/NIR refocusing system based on toroidal grazing incidence mirrors is under construction for spectroscopy on ions.

POLIMI has demonstrated HHG driven in gas media by three synchronized, intense laser pulses organized in a non-planar geometry, i.e., the three beams converging from the three vertices of a square [Negro2014]. All harmonic orders are generated in the fourth vertex direction, without angular dispersion. This outcome allows study of HHG in non collinear geometry with a simple, background-free detection scheme, providing a powerful tool to high harmonic spectroscopy of excited samples.

4 Conclusion

The objective of demonstrating multiphoton processes in the XUV range has been attained in several types of experiments. Though requirement of high XUV flux were drastic, the studies could use the ultra-short harmonic source and moderate density gas targets. These conditions offer a variety of multi-frequency schemes for time-resolved pump-probe studies, in particular in the gas phase, which could be now implemented by all the LASERLAB partners. They prepare possibly “easier” experiments of multiphoton scattering on denser solid state targets, to the expense of a larger number of scattering channels to be resolved. The remarkable creativity of nonlinear studies on the harmonic source also prepares the studies on FEL at much higher XUV flux, which certainly constitutes one of the very promising perspectives of deliverable D32.8.

5 Publications

Note : submitted / published on ArXiv (no DOI)

1. [Belshaw2012] L. Belshaw, F. Calegari, M.J. Duffy, A. Trabattoni, L. Poletto, M. Nisoli and J.B. Greenwood, "Observation of Ultrafast Charge Migration in an Amino Acid," J. Phys. Chem. Lett. 3, 3751-3754 (2012). DOI: 10.1021/jz3016028
2. [Calegari2014] F. Calegari, D. Ayuso, A. Trabattoni, L. Belshaw, S. De Camillis, S. Anumula, F. Frassetto, L. Poletto, A. Palacios, P. Decleva, J. Greenwood, F. Martín, M. Nisoli, "Ultrafast Electron Dynamics in Phenylalanine Initiated by Attosecond Pulses," Science 346, 336-339 (2014). DOI: 10.1126/science.1254061
3. [Calegari2015] F. Calegari, D. Ayuso, A. Trabattoni, L. Belshaw, S. De Camillis, F. Frassetto, L. Poletto, A. Palacios, P. Decleva, J.B. Greenwood, F. Martín, M. Nisoli, "Ultrafast Charge Dynamics in an Amino Acid Induced by Attosecond Pulses," IEEE J. Selec. Topics in Quantum Electr. 21, 8700512-1-12 (2015). DOI: 10.1109/JSTQE.2015.2419218
4. [Carpeggiani2014] “Disclosing intrinsic molecular dynamics on the 1-fs scale through extreme-ultraviolet pump-probe measurements” P. A. Carpeggiani, P. Tzallas, A. Palacios, D. Gray, F. Martín and D. Charalambidis, *Phys. Rev.* **A89**, 023420 (2014), DOI: http://dx.doi.org/10.1103/PhysRevA.89.023420
5. [Cassou2014] “Enhanced high harmonic generation driven by high-intensity laser in argon gas-filled hollow core waveguide”, K. Cassou, S. Daboussi, O. Hort, O. Guilbaud, D. Descamps, S. Petit, E. Mével, E. Constant, and S. Kazamias, Optics Letters **39**, 3770 (2014). http://dx.doi.org/10.1364/OL.39.003770
6. [Ciappina2014a] “Coherent XUV generation driven by sharp metal tips photoemission”, M. F. Ciappina, J. A. Pérez-Hernández, T. Shaaran and M. Lewenstein, Eur. Phys. J. D **68**, 172 (2014), DOI: 10.1140/epjd/e2014-50060-4
7. [Ciappina2014b] High-order-harmonic generation driven by metal nanotip photoemission: Theory and simulations, M. F. Ciappina, J. A. Pérez-Hernández, T. Shaaran, M. Lewenstein, M. Krüger, and P. Hommelhoff, Phys. Rev. A **89**, 013409 (2014) DOI: 10.1103/PhysRevA.89.013409
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10. [Heyl2015] "Scale-invariant nonlinear optics in gases", C.M. Heyl, H. Coudert-Alteirac, M. Miranda, M. Louisy, K. Kovacs, V. Tosa, E. Balogh, K. Varjú, A. L'Huillier, A. Couairon, and C.L. Arnold, submitted 2015.
11. [Manschwetus2015a] "Self-probing spectroscopy of the SF6 molecule, a study of the spectral amplitude and phase of the high harmonic emission", B. Manschwetus, N. Lin, J. Rothhardt, R. Guichard, T. Auguste, A. Camper, P. Breger, J. Caillat, M. Géléoc, T. Ruchon, R. Taïeb, B. Carré, et P. Salières, J. Phys. Chem. A 119, 6111 (2015), DOI: 10.1021/acs.jpca.5b00446.
12. [Manschwetus2015b] "*Two-photon double ionization of neon using an attosecond pulse train",* B. Manschwetus, L. Rading, F. Campi. S. Maclot, H. Coudert-Alteirac, J. Lahl, P. Rudawski, C.M. Heyl, B. Farkas, T. Mohamed, A. L'Huillier, and P. Johnsson, submitted 2015
13. [Negro2014] "Non-collinear high-order harmonic generation by three interfering laser beams", M. Negro, M. Devetta, D. Faccialà, A.G. Ciriolo, F. Calegari, F. Frassetto, L. Poletto, V. Tosa, C. Vozzi, and S. Stagira, Optics Express **22**, 29778 (2014), doi: 10.1364/OE.22.029778
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6 Persons involved :

6.1 MBI

B. Schuette, F. Campi, M. Arbeiter, Th. Fennel, M.J.J. Vrakking, O. Kornilov, A. Rouzée

6.2 CEA-SLIC

Romain Geneaux (PhD), Thierry Ruchon, A. Camper, T. Auguste, O. Gobert

6.3 CLPU

J.-A. Pérez Hernández, Luis Roso, collaboration ICFO (M. F. Ciappina, T. Shaaran, M. Lewenstein) and MPQ (M. Krüger, P. Hommelhoff)

6.4 CNRS-CELIA

L. Quintard, C. Ballage, A. Dubrouil, D. Descamps, F. Catoire, E. Constant, E. Mével collaboration with Institut Lumière Matière-Lyon (F. Lépine, V. Loriot, Ch. Bordas, A. Marciniak)

6.5 CNRS-LUMAT

K. Cassou, S. Daboussi, O. Guilbaud, S. Kazamias POLIMI

6.6 IESL

P. Tzallas, P.A. Carpeggianni, D. Gray, G. Kolliopoulos, E. Skantzakis, D. Charalambidis

6.7 LLC

B. Manschwetus, L. Rading, P. Rudawski, B. Farkas, F. Campi, C.M. Heyl, A. L'Huillier, P. Johnsson

6.8 LLAMS

J. Morgenweg, R. Altmann, L.S. Dreissen, S. Galtier, K.S.E. Eikema

6.9 POLIMI

M. Negro, M. Devetta, D. Faccialà, A.G. Ciriolo, F. Calegari, F. Frassetto, L. Poletto, V. Tosa, C. Vozzi, S. Stagira