LASERLAB-EUROPE

The Integrated Initiative of European Laser Research Infrastructures III

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WP32 – Innovative radiation sources at the extremes (INREX)

Deliverable D32.15
Interim reports on Deliverables 32.1 - 32.14

Lead Beneficiary: 15 MPQ

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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

1 Introduction

Over the last few years, the rapid evolution of femtosecond lasers towards ultra-high intensities and single optical cycles has revolutionized the generation of high intensity radiation over the whole electromagnetic spectrum from THz to hard x-rays. Thanks to the pulse durations produced using laser-like sources, which has been extended below 100 attoseconds, and the large number of photons per pulse (up to 10^{14}) over an extremely broad spectrum, these secondary sources are now regularly producing new scientific opportunities for Laserlab users. Compared with free-electron lasers, laser-driven secondary sources are intrinsically synchronised with lasers. This offers very rich insights into matter dynamic on unprecedented time and space scales, by allowing jitter-free pump-probe experiments (THz + X-rays; laser + as pulses; X-ray + attosecond, etc). The JRA “Innovative Radiation Sources at the EXtremes” (INREX) will further develop and apply these sources from the terahertz domain (0.3 to 20 THz, 10 – 600 cm^{-1}, 1 mm – 15 μm) to mid-infrared laser-like, single-cycle sources, emitting from 3 to 10 μm, from laser-based soft to hard x-rays ranging from 60 nm to 0.1 nm to plasma amplification (soft x-ray lasers) and emission of relativistic electrons in plasmas or in vacuum (betatron, Thomson scattering, laser-based wiggler and the FEL). Together with these sources, their associated diagnostics will be developed in a structured and collaborative programme that will coordinate the theoretical, numerical, and experimental efforts.

Co-ordinators of WP32: Reinhard Kienberger, MPQ; Philippe Zeitoun, CNRS-LOA; Stelios Tzortzakis, FORTH.

2 Interim progress report

2.1 Objective 1: Few-cycle to single cycle mid-IR sources

Several techniques have been used in the past to produce tuneable mid-IR pulses, but they are generally limited to relatively narrowband, multi-cycle pulses of low energy. The objective here is to develop high energy nearly single cycle pulses in the mid-IR, which will allow strong field studies in this spectral region.

Responsible Partners: POLIMI, ICFO, LENS

- Summary of task objectives:

Exploit the technological advances in OPAs and OPCPAs to develop powerful nearly single-cycle mid-IR sources. Study novel nonlinear media and phase matching schemes to further optimize the sources. Ultrafast spectroscopy, and strong field physics in the mid-IR. The mid-IR pulses coupled with other secondary sources will allow the investigation of a wide range of problems e.g. molecular spectroscopy, coherent control of vibrational wave packet motion, mass spectrometry and electron mobility in semiconductors etc. Femtosecond pulse shaping for the generation of tailored pulse sequences will be employed in time resolved four-wave mixing (photon echo) experiments.

- Task progress report and results

Deliverable 32.1

ICFO has developed a mid-IR OPCPA, currently providing 3100 nm, 5.5 cycle pulses at 160 kHz and 3.6W average power. ICFO has demonstrated nonlinear propagation in dielectrics for the first time in the anomalous dispersion regime and achieved clean 2.9-cycle pulses with 60% energy efficiency. The stability of the source was measured to 0.6% rms and 0.9% PV over 21 hours, corresponding to 140 days at 1 kHz. POLIMI has implemented a pulse shaper operating in the MIR spectral region. The system is a grating-based 4f-shaper equipped with a large-stroke deformable mirror in its Fourier plane; the shaper configuration and the deformable mirror were specially designed to operate at wavelengths from 12 to 20 micron. The capabilities of the shaper were tested on few-cycle MIR tunable from 15 to 23 THz, obtained by difference frequency generation between two near-infrared pulses generated by optical parametric amplifiers (OPAs). The shaper proved its capability to compress the pulses, to impart quadratic and cubic spectral phases and to split the spectral content of the light into double pulses. The electric field of such pulses was directly measured by electro-optic sampling. LENS has developed new mid-IR sources based on femtosecond frequency-combs. A quantum cascade laser has been actively phase-locked to a difference-frequency-generated (DFG) non-linear source referenced to a near-IR optical frequency comb, achieving a sub-kilohertz line-width and Cs-traceable mid-IR source delivering tens of mW. Moreover, a mid-IR frequency comb has been generated with more than 400 GHz span, 1 GHz rep. rate and sub-kilohertz-line-width teeth by an intra-cavity DFG process in a Ti:sapphire laser.

Publications:

1. M. Hemmer, A. Thai, M. Baudisch, H. Ishizuki, T. Taira, J. Biegert, “18 μJ energy, 160 kHz repetition rate, 250 MW peak power mid-IR OPCPA”, invited paper Chin. Opt. Lett. **11**(1), 013202 (2013). DOI:10.3788/COL201311.013202

2. M. Hemmer, M. Baudisch, A. Thai, F. Silva, J. Biegert, “Self-Compression to Sub-3-Cycle Duration of Mid-Infrared Optical Pulses in Bulk”, Opt. Exp. **21**, 028095 (2013). DOI:10.1364/OE.21.028095

3. I. Galli, M. Siciliani de Cumis, F. Cappelli, S. Bartalini, D. Mazzotti, S. Borri, A. Montori, N. Akikusa, M. Yamanishi, G. Giusfredi, P. Cancio, P. De Natale, Appl. Phys. Lett. **102**, 121117 (2013). DOI: 10.1063/1.4799284

4. I. Galli, F. Cappelli, P. Cancio, G. Giusfredi, D. Mazzotti, S. Bartalini, and P. De Natale, “High-coherence mid-infrared frequency comb,” Opt. Exp. (2013), in press

Deliverable 32.2

POLIMI has explored the possibility to steer, control or switch electron wave packets with light [1]. Such electron pulses were generated by strong-field-induced tunneling and acceleration of electrons in the near-field of a sharp gold tip, illuminated by an IR laser field with controllable CEP. The effect of the CEP on the generation and motion of electrons was observed together with clear variations in the width of plateau-like photoelectron spectra characteristic of the subcycle regime.

ICFO carried out several investigations with the uniquely developed few-cycle and CEP stable 160 kHz OPCPA. The wavelength scaling of strong field interaction of mid-IR pulses for HHG was investigated [2] together with the first measurement of strong field ionization momentum distributions in simple gas atoms and molecules in the tunneling regime [3]. Unexpected features were found which point at trapping at high lying states - theoretical investigations are ongoing.

CEA-SLIC has installed a mid-IR OPA (commercial system pumped by Ti:S, energy > 1 mJ in the 1.3-2 μm range, 1 kHz rep. rate) for driving HHG in gases. Ionization dynamics of molecules in strong field is currently studied using HHG spectroscopy (RABBIT).

LENS has implemented the phase-lock of a quantum cascade laser to a difference-frequency-generated (DFG) non-linear source referenced to a near-IR optical frequency comb, to perform sub-Doppler spectroscopy of CO2 transitions at 4.3 μm, obtaining absolute frequency measurements of several lines with few kHz uncertainties [4,5].

Publications:

[1] Björn Piglosiewicz, Slawa Schmidt, Doo Jae Park, Jan Vogelsang, Petra Groß, Cristian Manzoni, Paolo Farinello, Giulio Cerullo & Christoph Lienau: Nature Photonics, Published: 10 November 2013, doi: 10.1038/nphoton.2013.288

[2] D. R. Austin, J. Biegert: Strong-Field Approximation for the Wavelength Scaling of High-Harmonic Generation, Phys. Rev. A 86, 023813, 2012, DOI: 10.1103/PhysRevA.86.023813

[3] J. Dura, N. Camus, A. Thai, A. Britz, M. Hemmer, M. Baudisch, A. Senftleben, C.D. Schröter, J. Ullrich, R. Moshammer, J. Biegert, Sci. Rep. 3, 2675, 2013. doi:10.1038/srep02675

[4] I. Galli, S. Bartalini, P. Cancio, F. Cappelli, G. Giusfredi, D. Mazzotti, N. Akikusa, M. Yamanishi, and P. De Natale, Molecular Physics 111, 2041-2045 (2013). DOI: 10.1080/00268976.2013.782436

[5] I. Galli, S. Bartalini, P. Cancio, P. De Natale, D. Mazzotti, G. Giusfredi, M. E. Fedi, and P. A. Mandò, Radiocarbon 55, 213-223 (2013). DOI: 10.2458/azu_js_rc.55.16189

2.2 Objective 2: Ultra-short and intense THz sources

Here LASERLAB-EUROPE targets the implementation and/or the improvement of tabletop ultra-short and intense THz sources. The development will be based on different approaches including large area photoconductive antennas and nonlinear crystals, phase matched generation using pulse front tilt, and generation schemes based on laser filamentation in gases.

Responsible Partners: INFLPR, LENS, FORTH, CNRS-LOA, VULRC, CLPU, STRATH

- Summary of task objectives:

Development and optimization of intense ultrashort broadband THz pulse emission from gases excited by 2-color femtosecond laser pulses. The power will be optimized and the spectrum and polarization of the THz radiation will be controlled. Other schemes using large photoconductive antennas and nonlinear crystals or phase matching by pulse front tilt will be explored. THz spectroscopy and imaging set-ups will be developed to study excitation mechanisms and nonlinear optics. Examples of such applications include: THz pumping of nanostructures and semiconductors, metals, insulators, and correlated electron materials, coherent band-gap distortions and phase transitions, THz 2nd and 3rd order nonlinear effects, THz-optical nonlinear mixing.

- Task progress report and results

Deliverable 32.3

An advantage of THz sources from femtosecond laser induced plasma strings in air is the possibility to produce a THz source close to a distant target for remote applications. Terahertz wave generation from air excited by tightly focused fundamental and second harmonic pulses of femtosecond Ti:Sapphire laser have been investigated both experimentally and theoretically using the plasma microcurrent model and analyzing phase difference between the bichromatic pump components. At VULCR it has been shown that by varying optical pulse polarization parameters and optical path it is possible to control the yield and polarization state of generated ultrashort (picosecond) THz pulses. The experimental setup of THz source based on two-color laser plasma interaction was implemented at INFLPR. Numerical analysis of THz generation based on four wave mixing and free electron drifting model was performed. THz improved intensity yield has been studied at LOA by using coherent synthesis of terahertz radiation pulses from laser filaments organized in an array. THz comb generation and phase-locking of a 2.5 THz quantum cascade laser to a free-space comb, covering the 0.1--6 THz frequency range [1], has been demonstrated at LENS.

Publications:

[1] Phase-locking to a free-space terahertz comb for metrological-grade terahertz lasers, L. Consolino, A. Taschin, P. Bartolini, S. Bartalini, P. Cancio Pastor, A. Tredicucci, H. E. Beere, D. A. Ritchie, R.

Torre, M. S. Vitiello, and P. De Natale, Nature Communications 3, 1040 (2012).doi:10.1038/ncomms2048

2.3 Objective 3: Improving as pulses photon energy, flux and repetition rate

Conversion efficiencies for the generation of isolated ***as*** pulses in gases will be optimised in different wavelength regions by increasing the interaction volume, developing quasi-phase matching techniques, and using multi-colour fields. The generation of higher-energy photons will be pursued with IR and multi-colour laser fields, and from laser produced plasma on surfaces. The repetition rate will be increased to several 100 kHz or MHz.

Responsible Partners: LLAMS, MPQ, STFC-CLF, ICFO, LENS, LLAMS, CLPU, FVB-MBI, CEA-SLIC, CNRS-LOA

- Summary of task objectives:

Generation of high flux ***as*** pulses using QPM schemes for HHG will be developed. Optimisation of the interaction geometry and generating methods for efficient production of high-energy isolated ***as*** pulses by using multi-mJ driving pulses will be pursued. Intense ***as*** lasers pulses will be developed making use of double-optical gating with TW-class driver lasers, as well as by using nano-structures. Mid-IR driving lasers will improve the efficiency of harmonic generation in the 100-300 eV range will demonstrate conversion efficiencies exceeding 10^{-3} in the VUV with extremely high source brightness using multi-Joule fs laser systems. HHG in surface plasma in both the CWE and relativistic regimes will be investigated. A high repetition rate harmonic source using plasmonic structures will be developed.

- Task progress report and results

Deliverable 32.4

The ICFO group has been working on the development of high average power strong field relevant laser drivers in the long wavelength regime. They recently achieved 38.3 fs pulses at the kHz pulse repetition range, at 2000 nm and 0.9 mJ per pulse. Using these pulses, they have succeeded in approaching the water window range of the HHG cutoff. CLF is using the long wavelengths of an infrared OPA (1200 – 1600 nm) to extend HHG spectroscopy to photon energies up to 120 eV. LOA studied high-harmonic generation in gases in a two-field scheme by mixing the fundamental and the second harmonic of a Ti:Sa laser. The use of a hollow fiber for HHG is pursued. This work, performed in collaboration with PSI in Switzerland, shows strong emission around 200 eV. LENS has studied the enhancement of the conversion efficiency and the control of the spectral features of HHG in a region of interest for spectroscopy. Different interaction geometries and the use of gas regions with modulated density profiles were investigated to increase HHG yield. At LLAMS a new pump laser based on diode-pumped Nd:YAG modules was completed. With this pump laser a TW-class 800 nm / 300 Hz NOPCPA is constructed for high average power HHG [3]. This system will be used for diffractive imaging down to 10 nm, and an extension to the water-window soft-X-ray region is pursued with longer fundamental wavelengths of 1100-2100 nm based on the idler of the OPA. CEA-SLIC has optimized spatial properties and conversion efficiency of an HHG source using an XUV wavefront sensor as a sensitive diagnostic. Up to 10^{10} and 10^9 photons per laser pulse is routinely produced at 32nm (H25) and 20nm (H39), respectively, enabling applications such as single-shot coherent diffractive imaging of nanoscale objects. In collaboration with IST, CEA-SLIC also performed a first experimental demonstration of HHG in a cell with ionized gas by an electronic discharge creating a plasma channel, and HHG from singly-ionized argon gas was recorded. CLPU has carried out numerical simulations of HHG with TDSE. Using a temporally synthesized and spatially nonhomogeneous laser field, a dramatic extension of photon energy from HHG in helium, beyond the carbon K edge, has been demonstrated [1,2]. Also the generation of sub-as keV waveforms in the emission of HHG with mid-infrared lasers in gases was demonstrated theoretically. MPQ has imporved the stability and conversion efficiency of a few-cycle / 2.1µm / > 1 mJ / 1 kHz OPA system for HHG [4,5]. A 4 kHz booster system was used to amplify a CPA system and achieve ~1mJ/ 5 fs pulses, which were used for HHG [6].

Publications:

[1] J.A. Pérez-Hernández, M.F. Ciappina, M. Lewenstein, L. Roso and A. Zaïr, Physical Review Letters 110, 053001 (2013), doi:10.1103/PhysRevLett.110.053001

[2] C. Hernández-García; J.A. Pérez-Hernández; T. Popmintchev; M.M. Murnane; H.C. Kapteyn; A. Jaron-Becker and L. Plaja, Phys. Rev. Lett. 111, 033002 (2013), doi:10.1103/PhysRevLett.111.033002

[3] D.W.E. Noom, S. Witte, J. Morgenweg, R.K. Altmann, K.S.E. Eikema, Optics Letters 38, 3021-3023 (2013), DOI: 10.1364/OL.38.003021

[4] Alexander Schwarz, Moritz Ueffing, Yunpei Deng, Xun Gu, Hanieh Fattahi, Thomas Metzger, Marcus Ossiander, Ferenc Krausz, and Reinhard Kienberger, Optics Express, Vol. 20, Issue 5, pp. 5557-5565 (2012).

[5] Yunpei Deng, Alexander Schwarz, Hanieh Fattahi, Moritz Ueffing, Xun Gu, Marcus Ossiander, Thomas Metzger, Volodymyr Pervak, Hideki Ishizuki, Takunori Taira, Takayoshi Kobayashi, Gilad Marcus, Ferenc Krausz, Reinhard Kienberger, and Nicholas Karpowicz, Optics Letters 37, Iss. 23, pp. 4973–4975 (2012)

[6] W. Schweinberger, A. Sommer, E. Bothschafter, J. Li, F. Krausz, R. Kienberger, M. Schultze, Opt. Lett. 37 (17) 3573-3575 (2012).

Deliverable 32.5

ICFO has investigated the influence of spatial inhomogeneities on the strong field recollision mechanisms leading to HHG [1] as well as above threshold ionisation [2]. We find that the spatial field patterns can be tailored to enhance the harmonic cutoff significantly [3]. The Lewenstein model was extended to take the strong inhomogeneities, occurring in plasmonic nanostructures, into account. CLPU has performed theoretical investigations of HHG with few-cycle laser pulses in metal nanoparticles. It has been demonstrated that the strong nonhomogeneity of the laser field plays an important role in the HHG process and leads to a significant increase of the harmonic-cutoff energy. Theoretical simulations also predict the generation of high-energy electrons in metal nanotips, which can lead to very high photon energies in HHG [4,5].

Publications:

[1] M. Ciappina, J. Biegert, R. Quidant, M. Lewenstein, “High-order harmonic generation tailored using non-homogenous fields”, Phys. Rev. A. 85, 033828, 2012; also Virt. J. ultr. Sci., 11(4), 2012

[2] M. F. Ciappina, Srdjan, S. Aćimović, T. Shaaran, J. Biegert, R. Quidant, M. Lewenstein, “Enhancement of HHG by confining electron motion in plasmonic nanostrutures”, Opt. Exp. 20, 26261, 2012

[3] M. F. Ciappina, J. A. Perez-Hernandez, T. Shaaran, J. Biegert, R. Quidant, M. Lewenstein , “Above threshold ionization by few-cycle spatially inhomogeneous fields”, Phys. Rev. A, 86, 023413, 2012

[4] T. Shaaran, M.F. Ciappina, R. Guichard, J.A. Pérez-Hernández, L. Roso, M. Arnold, T. Siegel, A. Zaïr and M. Lewenstein, Physical Review A 87, 041402 (R) (2013), doi:10.1103/PhysRevA.87.041402

[5] M.F. Ciappina, T. Shaaran, R. Guichard, J.A. Pérez-Hernández, L. Roso, M. Arnold, T. Siegel, A. Zaïr and M. Lewenstein, Laser Physics Letters 10, 105302 (2013), doi:10.1088/1612-2011/10/10/105302

2.4 Objective 4: XUV optics, detection and diagnostic techniques

LASERLAB-EUROPE participants will develop *ultra broadband* ($\Delta\lambda/\lambda \sim 1/3$) XUV optics, which preserves or even corrects the *temporal* and *spatial phases* of the HHG beam. Sub-micrometer focusing of HHG pulses preserving the *as* pulse duration will be used to achieve high intensities e.g. for non-linear X-ray experiments. Spectral and temporal shaping of ***as*** pulses with versatile arrangements of broadband multilayer optics will be achieved. Detectors for 3D many-particle detection, co-variance imaging, (slice) imaging detectors and coincidence detection of ions and electrons will be improved. Second-order autocorrelation based techniques will be extended to isolated ***as*** pulses, while XUV surface SHG will be explored for ***as*** pulse diagnosis.

Responsible Partners: FORTH, MPQ, CEA-SLIC, CNRS-CELIA. FVB-MBI, LLAMS, LENS, CNRS-LOA, STFC-CLF

- Summary of task objectives:

MPQ will develop theoretical concepts and experimental realization of nano-focusing of HHG pulses that preserve the ***as*** pulse duration. Design, production and testing of ***as*** XUV optics in order to shape spectrally/temporally ***as*** pulses will be carried. Autocorrelation-like techniques will be developed and compared with frog-crab or streaking measurements. A velocity map imaging technique suitable for XUV/x-ray experiments will be developed and a reaction microscope setup will be optimised. Laser imaging of atomic inner-shell processes and molecular fragmentation reactions with full characterisation of 3D-recoil of correlated electrons and ions will be advanced. New schemes alternative to frequency combs to provide high-spectral resolution in the XUV by Ramsey or Fourier Transform spectroscopic techniques with HHG will be developed. Temporal characterisation techniques to solid target harmonics will be applied. Single shot XUV characterization techniques will be developed like interferometric techniques as well as non-linear processes, 2nd order autocorrelation based on split-mirror and ion imaging, and 2-XUV-photon FROG.

- Task progress report and results

Deliverable 32.6

CELIA implemented an imaging system providing both spectral and spatial resolution on a single shot basis. They observed many spatio-spectral structures in the XUV harmonic beam that are the signatures of spatio-temporal coupling during the generation process. Towards developing FORTH has modelled a single shot XUV non-linear autocorrelator based on spatially resolved two-XUV-photon ionization, induced by two intersected XUV beams. Utilizing a new ion-microscope, providing 1μm spatial resolution spatially resolved two-XUV-photon ionization of He has been demonstrated. MPQ has developed an angle-resolved time-of flight detector for *as* photoelectron spectroscopy. The detector is about to be implemented in an *as* beamline and to be tested.

2.5 Objective 5: Ultrafast XUV metrology/spectroscopy in atomic, molecular and solid state physics

Development of user-oriented beamlines is necessary for applications of XUV femtosecond and ***as*** pulses to atomic, molecular and solid state physics. The objective is 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, pump-probe experiments at surfaces will 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 will be probed by XUV radiation. Nonlinear optics with isolated ***as*** pulses will be implemented.

Responsible Partners: STFC-CLF, IST, CNRS-LOA, CEA-SLIC, POLIMI, CNRS-CELIA, FVB-MBI, FORTH, CLPU, CEA-SLIC, LLAMS, LU-LLC

- Summary of task objectives:

Several techniques (coincidence measurements, Two-photon spectroscopy, ***as*** XUV pump/***as*** XUV probe spectroscopy, UV-pump/XUV-probe techniques, HHG-based time- and angle-resolved photoemission, selection of two different VUV/XUV spectral intervals for pump/probe studies) will be used to investigate multi-electron dynamics in intermolecular processes, excitation/relaxation in solids excited by XUV, photoionisation in small molecules, atomic ionisation, molecular ionisation and dissociation, dynamics of polyatomic molecules and solids, as well as electron and nuclear dynamics in molecular systems, clusters, and condensed matter. Beamlines for the application of isolated ***as*** pulses based on ellipsoidal mirrors will be set up. Based on HHG in small aligned molecules, tomography imaging of valence molecular orbitals in asymmetrical systems will be developed. Techniques with ***as*** resolution based on a combination of XUV and IR wavelengths and the application of shaping, integrated with photo-electron and photo-ion coincidence imaging will be developed. Versatility, stability and control over ***as*** pulses will be improved.

- Task progress report and results

Deliverable 32.7

POLIMI has developed an optical system for micro-focusing as pulses, based on grazing incidence toroidal mirrors and featuring negligible aberrations, and the ability to insert a plane split-mirror for the generation of two XUV pulse replicas. LU-LLC has implemented a plane split-mirror at grazing incidence followed by two toroidal mirrors in Wolter configuration, to focus the XUV radiation of the Lund high energy harmonic beam line to a spot size of 3 μm diameter and an XUV intensity of 10^{14} Wcm^{-2}. FORTH has developed and operates an XUV-pump-XUV-probe facility, which has been used in studying 1 fs-scale dynamics in atoms and molecules. The dynamics of an atomic coherence induced above the ionization threshold of Xe atoms has been measured, and vibrational, electronic and ionization dynamics of H_2 have been tracked. STFC-CLF is operating an open-access facility for spectroscopy with 30 fs XUV probe pulses from 10-80 eV, with pump pulses tuneable from the UV to the mid-infrared. HHG-based time- and angle-resolved photoemission spectroscopy has been used to directly track electron dynamics in graphene and other novel materials. Ultrafast dynamics in water has been revealed through HHG photoelectron spectroscopy in a liquid jet. CNRS-LOA in collaboration with IST have developed a pump-probe technique for active spectroscopy of laser-produced plasma. The plasma is created by the interaction of 1 mJ, 30 fs pulse with a thin metallic filter and then probed by a high harmonic beam containing all the harmonic spectral components. CEA-SLIC has performed single-shot characterization of the magnetic nano-domain structure of a thin ferromagnetic Co/Pd multilayer film, using an optimized table-top high harmonic source. The scattering efficiency of the sample and average size of the magnetic domains are measured in a single 20 fs shot at photon energies of 45-70 eV. CEA-SLIC (in collaboration with Institut des Sciences Moléculaires d'Orsay, France) has investigated multi-color XUV/IR photoionization in rare gas atoms, where the relative phases of interfering 3-photon channels are varied on the *a*s timescale to modulate the photolectron angular distribution. CELIA has designed a new XUV-IR interferometer based on split mirrors approach for high temporal resolution and compatibility with high energy sources, and observed passive stability better than 100 asec. CLPU has commissioned a new laboratory hosting a 1kHz, 2 mJ, CEP-stabilized few-cycle Ti:Sapphire laser, XUV spectrometer and time-of-flight mass spectrometer for XUV-IR measurements in molecular gases. At LLAMS the new method of Ramsey-comb spectroscopy has been developed, based on multiple recordings with pairs of amplified frequency comb laser pulses. A first demonstration on the 5S-7S Rubidium transition near 760 nm has shown kHz accuracy with mJ-level amplified laser pulses. As a next step towards XUV spectroscopy with this system, up-conversion to deep-UV will be investigated for precision spectroscopic measurements on molecular hydrogen. MPQ has improved it´s UHV as beamline and performed several experiments on ultrafast electron dynamics in solids [3-7].

Publications:

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Deliverable 32.8

FORTH has demonstrated direct two-XUV-photon double ionization of Xe atoms induced by $1.5^{+0.2}_{-0.7}$ fs long pulses produced from HHG in gas medium, utilizing interferometric polarization gating of high peak power many-cycle lasers pulses [1]. FORTH and MPQ have achieved two-photon above-threshold ionization in Ar atoms, using intense XUV harmonic emission from relativistic laser-plasma interaction [2]. CELIA has used shaping of high energy driving laser pulses to optimize the energy and temporal/spatial profile of XUV pulses produced from HHG in gas medium [3]. The XUV pulses will now be used to observe nonlinear processes induced by XUV, such as two-photon double ionization of atoms. LOA has developed an IR beamline at kHz repetition rate dedicated to the production of intense *as* pulses from laser-solid interaction at relativistic intensity (>10^{19} W/cm^2). In parallel, LOA has upgraded the "Salle Noire" laser system into a double-stage CPA featuring a nonlinear filter to enhance the contrast of the pulses delivered on target. At the FERMI@ELETTRA FEL facility, POLIMI has investigated nonlinear two-photon excitation of the interatomic coulombic decay (ICD) in neon dimers. Exploiting the possibility for fine tuning of XUV line in steps of 20 meV, POLIMI has excited ICD mechanism through a resonant 2p2s$\leftarrow$2s^2 transition. Singly ionized neon monomers and neon dimers were measured as a function of the XUV photon energy. POLIMI performed as pump-probe transient absorption in N_2 combining an isolated as pulse and a CEP-stabilized few-cycle IR pulse, with sub-cycle synchronization [4]. Isolated *as* pulses were synthesized in two different spectral regions, corresponding to the excitation of N_2 bound states and of a series of Fano autoionizing resonances, using different metallic filters. It demonstrated that the IR pulse can influence the time-dependent polarization of the nitrogen ensemble, i.e., can steer the complex electronic molecular wave-packet excited by the broadband as pulse, on the sub-cycle time. CLPU has experimentally studied HHG in aligned molecules in the gas phase, using single-color and two-color mid-IR laser fields up to 2 µm wavelength at CLF. A two-color field experiment proved to support significantly higher

HHG efficiency than single-color one, but mainly at low harmonic orders. In collaboration with CELIA, CEA-SLIC has exploited XUV/IR 2-photon ionization of molecules (e.g., N_2, N_2O, CO_2) in the RABBIT technique to access quantum phase properties of the continuum states (theory in collaboration with LCPMR). In collaboration with the Institut des Sciences Moléculaires d'Orsay (Orsay, France), CEA-SLIC has studied XUV/nIR multi-photon ionization of atoms (Ar, He) involving interfering channels and subsequent modulation of the photoelectron angular distribution as a function of XUV/IR delay [6]. At LLAMS preparations have been made to extend the newly developed Ramsey-comb excitation technique with HHG. To this end the pump laser for the OPA used in the Ramsey-comb technique is upgraded to reach multi-mJ output. A toroidal XUV and NIR refocusing stage was designed for combined NIR-XUV multi-photon excitation schemes.

Publications:

[1] P. Tzallas et al. *J. Phys. B* **45**, 074007 (2012)

[2] P. Heissler et al. *New Journal of Physics* **14**, 043025 (2012)

[3] E Constant et al, J. Phys. B **45**, 074018 (2012)

[4] M. Lucchini et al, Phys Rev A **86**, 043404 (2012)

[5] J. Caillat et al, Phys Rev Lett **106**, 093002 (2011)

[6] Y. Picard et al, submitted to Phys Rev Lett (2013)

2.6 Objective 6: Synchrotron sources and free-electron lasers based on laser plasma wakefield accelerators

Laser-plasma wakefield accelerators (LFWA) have been developed to the point where they can now be considered as a driver of next generation compact radiation sources. The most challenging electron beam driven source is the free-electron laser because of its unprecedented brightness and coherence. The main objective of this task will be to pool experimental and theoretical efforts across the various groups to realise a LPWFA driven FEL. This will involve developing both LWFA and beam transport systems and investigating the short pulse regime in the FEL.

Responsible Partners: STRATH, CNRS-LOA, IST

- Summary of task objectives:

Synchrotron and free-electron laser developments depend on an understanding and matching of the accelerator and beam transport system. A programme to optimise the laser-plasma wakefield accelerator and beam transport will be undertaken to make it suitable for driving a free-electron laser or compact x-ray synchrotron source. This will involve both experimental and theoretical studies of the FEL driven by ultra-short electron bunches and the build up of coherence, including the effects of beam pipe wakefields. The injection of HHG source to control the FEL properties will be investigated.

- Task progress report and results

Deliverable 32.9

IST has focused on performing simulations, with the goal of designing the best electron acceleration regimes for the production of secondary sources, FEL or synchrotron. The control and means of manipulation of the injected beam was investigated, using the PIC Code OSIRIS. Acceleration was optimized with the goal of increasing the brilliance, through monochromatization of the source or increase of charge injection. The effect of frequency chirp and of plasma inhomogeneities on the laser propagation and on wakefield acceleration, and also of external magnetic fields have been explored. In collaboration with LOA and CEA, IST completed a series of XUV experiments, where a wavefront sensor was used to characterize the focus of an XFEL beam at the LCLS in Stanford. STRATH has carried out both experimental and theoretical investigations of injection, characterization of the electron beam to establish their suitability for driving a free-electron laser or as a compact synchrotron source. These studies have been augmented by investigating radiation due to betatron motion of electrons in the LWFA accelerating structure. All experiments have been carried out using preformed plasma capillary waveguides or gas jets and have been focused on single laser beam LWFAs, where injection occurs solely due to the nonlinear behaviour of the wake. Excellent beam stability has been observed. Good comparisons between a reduced model showing injection and PIC code simulations using OSIRIS and WAKE have been obtained. STRATH has also undertaken a comprehensive set of measurements of electron beam properties to characterize the energy spread, emittance, charge and bunch structure. [2-8]. LOA worked on the control of HHG source (HHG) for optimal seeding of FEL. We thus realize different setup to control the HHG polarization and to measure it. A high degree of circular polarization have been obtained of interest for FEL. Experiment at FERMI@ELETTRA has been done to measure the polarization of HHG seeded FEL.

Publications:

[1] V. B. Pathak, J. Vieira, R. A. Fonseca, and L.O. Silva New J. Phys, **14**, 023057 (2012).

[2] S. Cipiccia, S. M. Wiggins, D. Maneuski, E. Brunetti, G. Vieux, X. Yang, R. C. Issac, G. H. Welsh, M. Anania, M. R. Islam, B. Ersfeld, R. Montgomery, G. Smith, M. Hoek, D. J. Hamilton, N. R. C. Lemos, D. R. Symes, P. P. Rajeev, V. O Shea, J. M. Dias and D. A. Jaroszynski, Review of Scientific Instruments **84**, 113302 (2013)

[3Yevgen Kravets, Adam Noble and Dino Jaroszynski, Physical Review E **88**, 011201(R) (2013)

[4] G. Vieux B. Ersfeld, J. P. Farmer, M. S. Hur, R. C. Issac, and D. A. Jaroszynski, Appl. Phys. Lett. **103**, 121106 (2013).

[5] C. Ciocarlan, S. M. Wiggins, M. R. Islam, B. Ersfeld, S. Abuazoum, R. Wilson, C. Aniculaesei, G. H. Welsh, G. Vieux, and D. A. Jaroszynski, Phys. Plasmas **20**, 093108 (2013).

[6] C. Ciocarlan, M. Wiggins, S. Abu-Azoum, and D. Jaroszynski, Proc. SPIE 8779, 877918 (2013).

[7] Y. Kravets, A. Noble, and D. A. Jaroszynski, Proc. SPIE 8779, 87791X (2013).

[8] Silvia Cipiccia, Mohammad R. Islam, Bernhard Ersfeld, Richard P. Shanks, Enrico Brunetti, Gregory Vieux, Xue Yang, Riju C. Issac, Samuel. M. Wiggins, Gregor H. Welsh, Maria-Pia Anania, Dzmitry Maneuski, Rachel Montgomery, Gary Smith, Matthias Hoek, David J. Hamilton, Nuno R. C. Lemos, Rodolfo A. Bendoyro, Dan Symes, Pattathil P. Rajeev, Val O Shea, João M. Dias, Dino A. Jaroszynski, Nature Phys. **7**, 867 (2011)

Deliverable 32.10

Beam transport and FEL operation in the short pulse regime has been studied numerically. At IST, the study of the radiation emission in a plasma-wakefield accelerator scenario where a magnetic field is applied to assist the injection into the ion cavity was modeled with the particle in cell code OSIRIS [1,2]. Two- and three-dimensional simulations, supported by an analytical model, show that external static magnetic fields with suitable spatial profiles and amplitudes can relax the self-trapping thresholds in plasma based accelerators [3]. It is found that magnetic-field assisted self-injection can lead to the emission of betatron radiation at well defined frequencies [4]. This controlled injection technique could be explored using state-of-the-art magnetic fields in current/next generation plasma/laser wakefield accelerator experiments. The effect of radiation damping on the motion of particles in the ultrarelativistic regime was investigated [5]. Up to now such effects were not considered in PIC codes, but their impact needed to be taken into account. Several models were compared for the calculation of radiation damping force, with the goal of developing an algorithm for radiation damping in Osiris. A radiation damping algorithm has been recently implemented in OSIRIS, and its consequences for beam transport and FEL operation will be explored in the next reporting period.

STRATH has carried out both experimental and theoretical investigations of injection, characterization of the electron beam, LWFA driven undulator radiation emission in the VUV and also betatron radiation emission. The injection work has included understanding the evolution of the bubble structure in injection. We have now obtained good comparisons with a reduced model showing injection and PIC code simulations using OSIRIS and WAKE. The work has been submitted for publication. We have also carried out direct measurements of the bunch structure by measuring coherent transition radiation, which confirm the substructure predicted by the injection model that we have developed. This work has also been submitted for publication. An experimental paper has been published on the role of the plume in the LWFA based on a plasma capillary [9,10]. A new diagnostic technique for measuring the plasma density in a plasma channel has been developed [8]. Experiments have been carried out to measure VUV radiation from a LWFA driven undulator. This shows that narrow spectral width synchrotron spectra can be produced using a quasi-monoenergetic LWFA beam. Undulator radiation in the wavelength range 150 – 260 nm has been produced by 1.5 fs electron bunches from a 2 mm long laser plasma wakefield accelerator. The number of photons measured is up to 9×10^6 per shot for a 100 period undulator, giving a peak brilliance of > 3×10^{18} photons/s/mrad^2/mm^2/0.1% bandwidth. The radiation pulse duration is as short as 3 fs for 120 – 130 MeV electron beams.

The theoretical investigations of betatron radiation sources involve developing a unified theoretical model of betatron radiation, which has been submitted for publication. Experiments to characterize the betatron emission in a plasma channel have been carried out and published [6,12]. Investigations have been carried out on radiation reaction of particles in high fields [7,11].

Publications:

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[3] J. Vieira, S.F. Martins, V.B. Pathak, R.A. Fonseca, WB. Mori, L.O. Silva, Phys. Rev. Lett. 106 225001 (2011).

[4] J. Vieira, J.L. Martins, V.B. Pathak, R.A. Fonseca, W.B. Mori, L.O. Silva, Plasma Phys. Control. Fusion 54 124044 (2012).

[5] Marija Vranic, Joana L. Martins, Luis O. Silva, American Physical Society, 51st Annual Meeting of the APS Division of Plasma Physics, November 2-6, 2009, abstract #CP8.071.

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[7] Yevgen Kravets, Adam Noble and Dino Jaroszynski, Physical Review E 88, 011201(R) (2013)

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[11] Y. Kravets, A. Noble, and D. A. Jaroszynski, Proc. SPIE 8779, 87791X (2013).

[12] Silvia Cipiccia, Mohammad R. Islam, Bernhard Ersfeld, Richard P. Shanks, Enrico Brunetti, Gregory Vieux, Xue Yang, Riju C. Issac, Samuel. M. Wiggins, Gregor H. Welsh, Maria-Pia Anania, Dzmitry Maneuski, Rachel Montgomery, Gary Smith, Matthias Hoek, David J. Hamilton, Nuno R. C.

Lemos, Rodolfo A. Bendoyro, Dan Symes, Pattathil P. Rajeev, Val O Shea, João M. Dias, Dino A. Jaroszynski, Nature Phys. 7, 867 (2011)

2.7 Objective 7: Plasma and electromagnetic wigglers based on laser-plasma interactions

The very large transverse forces of a laser driven plasma density wake can act as a strong plasma wiggler. Relativistic electrons inside the "bubble" shaped wake will oscillate in its electrostatic potential and radiate very strongly. This so called "betatron" radiation is emitted as a very large spectrum of harmonics extending to hard X-ray photon energies. Significant radiation flux can be produced with high peak brilliance in the keV to 100's keV range. The micron size source results in an excellent transverse coherence. This, with the high photon energy, makes them useful for phase-contrast imaging and probing dense matter in spectral ranges where no brilliant sources exist. Control of the bubble shape and evolution will be studied as a way to control the spectral and/or the spatial properties of the radiation. Several possibilities will be explored to obtain shorter wavelengths with modest laser pulse energies.

Responsible Partners: CNRS-LOA, POLIMI, CNRS-CELIA, FVB-MBI, STFC-CLF, FORTH, CLPU, CEA-SLIC, LLAMS, LU-LLC

- Summary of task objectives:

Plasma (betatron) and electromagnetic (Thomson scattering) wigglers based on laser-plasma interactions will be investigated. The spectral range will be extended to 100's of keV, and the transverse and longitudinal coherent properties will be investigated. This will be supported by a theoretical study of betatron emission in an ion channel and include a study of the role of emittance, energy spread and charge in determining the X-ray pulse duration and brightness at the 100 keV level.

- Task progress report and results

Deliverable 32.11

IST has implemented of a post-processing radiation diagnostic in the OSIRIS code This work was then used for the study of magnetically assisted self-injection and radiation generation for plasma-based acceleration [1], a method allowing the generation of betatron radiation at well-defined frequencies. The betatron radiation model at IST was further developed with the extension of the diagnostic to the quantum regime. Betatron sources can now be characterized with a newer version of OSIRIS that takes into account radiation damping. LOA worked on a different version of hard X-ray emission induced by accelerated electrons. Indeed, Betatron is currently limited to few 10s keV with a slow scaling of photon energy with pump intensity. We thus studied the Compton effect that is the scattering of an infrared laser on an electron beam. In that case, the infrared laser while propagation in gas accelerates the free electrons. Since these particles are slightly slower than the speed of light in gas and in vacuum the infrared laser goes ahead of the electron bunch. By placing a mirror at the end of the gas jet, the infrared laser is back reflected, enabling efficient and naturally synchronize Compton scattering. Emission has been observed up to 200 keV with 100 TW laser [2].while betatron on the same laser is limited to about 10s keV

Publications:

[1] J Vieira et al, Plasma Phys. Control. Fusion 54, 124044 (2012)

[2] K. Ta Phuoc et al, Nat. Phot. 2012

2.8 Objective 8: Realize ultra-intense lasers at nanometre wavelengths

Short wavelength lasers will be developed using either the so-called “collisional excitation” or “recombination” schemes. Although widely studied for more than a decade, collisional schemes are the most promising as compact soft x-ray lasers and for reaching ultra-high intensities by seeding using an HHG source), while enlarging the spectral bandwidth of the plasma amplifier. The wavelength scaling with Z ($\lambda \propto [Z\text{-}1]^{-2}$ or $[Z\text{-}3]^{-2}$), has more potential than those based on collisional excitation scheme ($\lambda \propto [Z\text{-}10]^{-1}$ or $[Z\text{-}28]^{-1}$). Moreover, numerical simulations predict that the efficiency of recombination pumping is enhanced when the duration of the driving laser pulse is shortened. Numerical and experimental studies will be undertaken to achieve nm-scale soft x-ray lasers.

Responsible Partners: GSI, CNRS-LOA, CNRS-ISMO, CNRS-LUMAT, IST, PALS, MUT

- Summary of task objectives:

Optimization of plasma XRL towards higher output energy and beam quality. For this i) HHG as a seeding source with relatively narrow bandwidth, adapted to the gain profile of the XRL will be developed, and ii) an XRL set-up with two subsequent gain zones with different, increasing, beam diameter will be realized iii) strategies to enlarge the spectral bandwidth so as to support sub-picosecond amplification will be explored. Recombination scheme will be further studied on high Z elements both experimentally and theoretically.

- Task progress report and results

Deliverable 32.12

At PALS the possibility of creating an x-ray laser chain using three-stages was studied: HHG seed amplified in transient XRL preamplifier and boosted by QSS main amplifier. The advanced schemes of HHG employing quasi-phasematching were studied in collaboration with MUT. The transient XRL pumped under grazing incidence has been successfully implemented with Ti:S laser pumping. CNRS-ISMO and PALS have carried out an experiment aimed at a refined measurement of the spatial and temporal coherence properties of the Ne-like Zn plasma amplifier. The measured coherence time is shorter than ever measured with other types of XUV lasers, reaching 500 fs for a 2 cm amplifier. CNRS-LOA and CNRS-ISMO have collaborated with CNRS-LUMAT for the detailed characterization of a seeded XUV laser in Ne-like Ti, which was implemented at the LASERIX facility. At GSI-PHELIX, in collaboration with CNRS-LOA and CNRS-LUMAT a two-stage x-ray laser was demonstrated using a compact set-up with two Ti-like Mo stages. An improvement of more than 2 orders of magnitude in brilliance was observed. IST collaborated with LOA in the theoretical design and analysis of second generation X-ray lasers. Experimentally an XUV refraction diagnostic using a wavefront sensor was developed and tested [2]. These measurements help benchmark the refraction to better model the case of plasma amplifiers above the critical density. The multi-jet gas puff target developed at MUT-IOE has been used in joint experiments on high-order harmonics generation performed at PALS. The targets in a form of a 9-mm-long gas sheet with modulated gas density produced using 5-, 7-, and 9-orifice nozzles were irradiated with 40fs/1mJ laser pulses form a Ti:Sapphire laser. Dominating emission of 27th, 29th and 31st harmonics was observed. The experimental results have been confirmed by the numerical simulations performed at Cluj, Romania. The improvement of HHG seed sources was also reached in experiments of GSI-PHELIX team in collaboration with FSU-Jena using a scheme with two independent gas regions allowing for an enhancement and the variation of the bandwidth of different harmonics. At the IST facility, production of high harmonics with a ps driving beam has been studied, in view of a better coupling between the seed and the long-pulse amplifier.

Publications:
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2.9 Objective 9: Demonstrated high-average power HHG and soft x-ray lasers

: Several 100 kHz to MHz rep-rate driver sources (fibre lasers) will be used for HHG. The XUV yield will be improved to make single-event detection possible. On the other hand, the development of diode-pumped lasers is progressing very quickly and will have a very positive impact on soft x-ray lasers. However, to take full advantage of these high rep-rate lasers, several technical and fundamental issues still have to be solved. LASERLAB-EUROPE participants will develop high average power (> 0.1 mW) coherent soft x-ray lasers operating at 13.9 nm and 18.9 nm, which will then deliver photons to users.

Responsible Partners: FVB-MBI, CNRS-CELIA

- Summary of task objectives:

Thanks to the use of fiber lasers LASERLAB-EUROPE will increase the repetition-rate of HHG up to MHz or using thin-disk technology we will realize (> 0.1 mW) coherent soft x-ray lasers.

- Task progress report and results

Deliverable 32.13

CELIA has developed a new system specifically devoted to the collection of highly diverging harmonics emitted at high repetition rate in the tight focussing regime. After optimization, harmonics have been generated in Xenon, Krypton and Argon and characterized at a repetition rate of 100 kHz. At MBI a 400 kHz OPCPA producing CEP-stable 5.2 fs laser pulse has been developed. An upgrade boosting the average power from 2.2 W to (expected) 5 W is currently underway, and will form the basis for HHG experiments.

2.10 Objective 10: Develop ultrafast imaging with techniques adapted to soft x-ray lasers

LASERLAB-EUROPE participants will perform coherent diffraction experiments using soft x-ray lasers. The narrow bandwidth of the source should allow a better resolution than other sources to be achieved. Furthermore, the high number of photons per shot will be essential for single-shot imaging in the dynamic study of non-reproducible phenomena. Previous work has shown that using x-ray laser transmission and interferometry one can determine the opacity of hot dense matter and thus determine the rate of ablation of solid targets. It will be possible to undertake interferometric studies of warm dense target material and to perform coherent diffractive imaging using K-alpha radiation and x-rays produced by fast electrons in laser-plasmas.

Responsible Partners: CNRS-CELIA, CNRS-LOA

- Summary of task objectives:

LASERLAB-EUROPE will further develop ultra-fast imaging technique like XUV diffraction with HHG or new holographic or diffraction schemes adapted to the narrow spectral width of soft x-ray lasers.

- Task progress report and results

Deliverable 32.14

CEA-SLIC, in collaboration with Leibniz Universität *Hannover*, has extended 2D coherent imaging techniques in the soft X-ray range to 3D imaging. They have either exploited the large diffracted or twin beams in stereo imaging configuration. LOA, in collaboration with IST and CEA, worked on the development of *as* coherent imaging. They achieved holograms as well as diffraction images of different samples and using both a single wavelength HH pulse or multiple wavelength beam (up to 5 harmonics). On holography with reference, they demonstrated that using a dedicated target with a reference hole far enough from the sample, it is possible to separate the images corresponding to the different wavelengths during image retrieval treatment. CELIA has developed a new imaging system that provides both spectral and spatial resolution for XUV beam characterization on a single shot basis. Observing ultrashort XUV pulses generated at high energy with few cycle TW pulses, allowed them to observe many complex structures in the XUV beam. These structures are the signature of self-diffracting coherent XUV beams and contain rich information about the dynamics of the XUV pulse emission process and the spatio temporal coupling.