WORKPACKAGE

WP-3: HHG and ultrafast electron imaging

Coordinators: P. Salières (CEA), A. Assion (FEMTO, Spectra Physics Vienna)

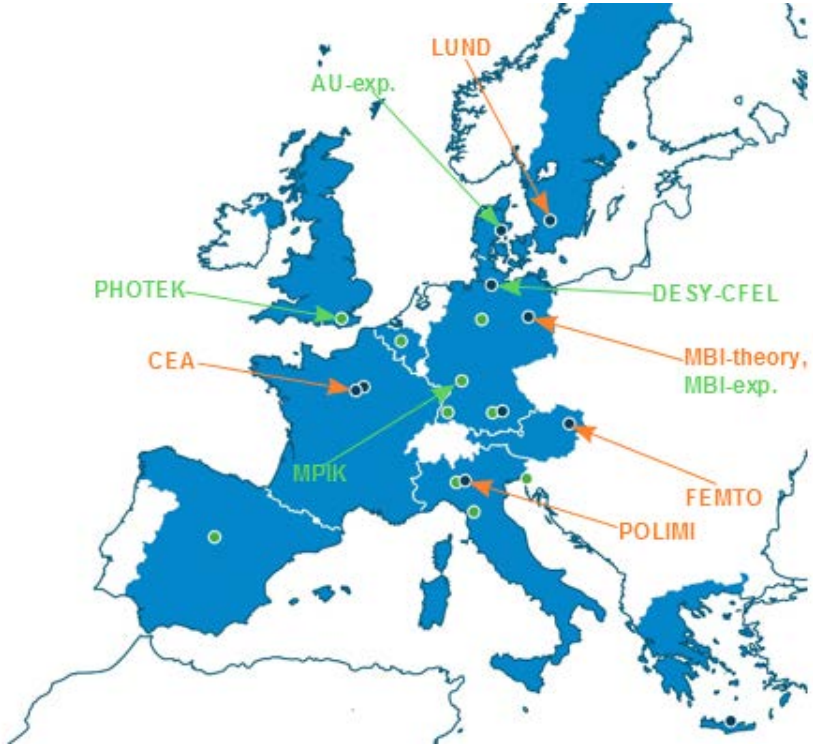
Period: Start Month 4 – End Month 48

- Leading Participants (Orange in the picture):

Commissariat à l'énergie atomique et aux énergies alternatives (**CEA**), Politecnico Milano (**POLIMI**), Max Born Institute (**MBI-theory**), Spectra Physics Vienna (prior Femtolasers) (**FEMTO**), University of Lund (**LUND**)

- Partner Organisations (Green in the picture):

Aarhus University(**AU-exp.**), **LUND**, Deutsches Elektronen Synchrotron (**DESY-CFEL**), Photek (**PHOTEK**), Max Planck Insitute for nuclear physics Heidelberg (**MPIK**)



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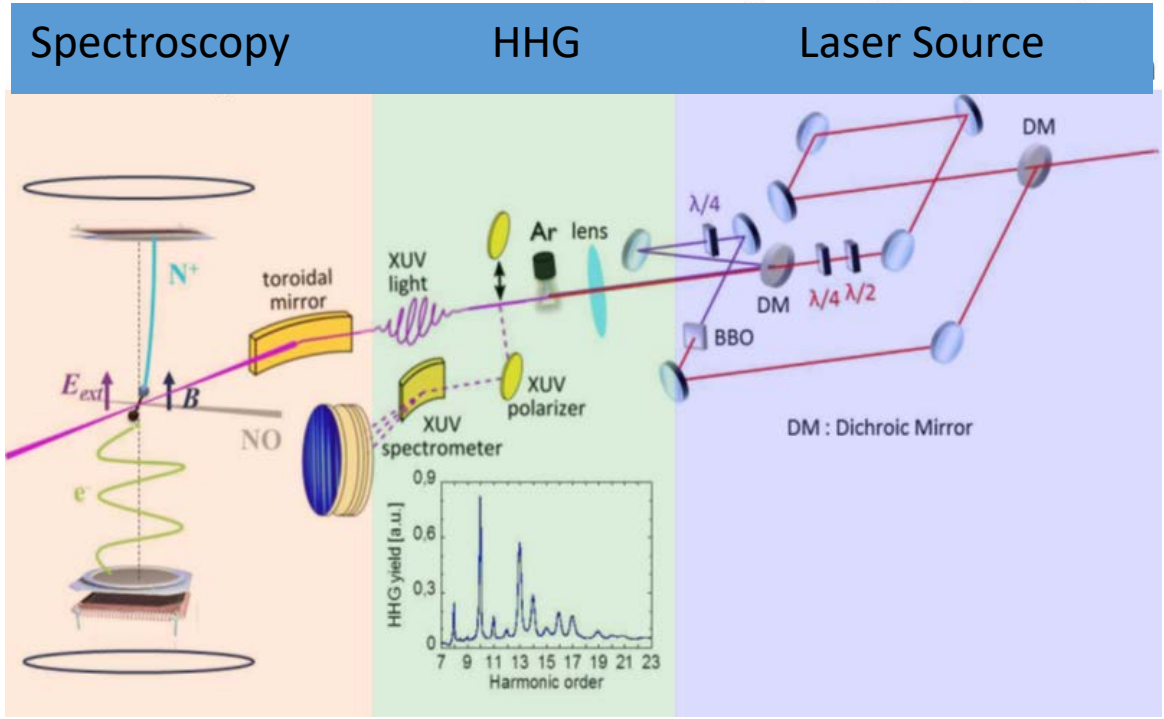
WP3 Overall



Make use of ultrashort (attosecond and femtosecond) electron wave-packets to image multi-electron and nuclear dynamics in molecules.

The wave packets will be created either by:

- Strong field IR photoionization – leading to HHG or LIED, both of which contain information on electron/nuclear dynamics after the ionization event.
- Single photon absorption of an XUV pulse – used for imaging of structural changes.

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SUB WORKPACKAGES

Sub-workpackages:

- *WP 3.1*
  - **CEA**, **AU-exp.**, **POLIMI**, performs experiments aimed at imaging the attosecond oscillation of the hole created by the tunnel ionization of multiple orbitals in different molecules.
  - **MBI-theory** supports these experiments
  - **MBI-theory** also investigates possibilities of calibrating ionization and recombination times in different molecular orbitals.
  - **PHOTEK** develops a low temperature valves in order to achieve high density cold molecular beams necessary for the alignment of molecular ensembles .

- *WP 3.2*
  - **FEMTO** develops new laser source which generates mid-IR CEP-stable light pulses
  - **MPIK**, **AU-exp.** and **DESY-CFEL** investigate the diffraction pattern generated by the core-shell photoelectrons emitted by specific atoms of aligned molecules using FEL XUV pulses.
  - **LUND** and **MBI-exp.**, with the help from **PHOTEK** collaborate on extending such experiments from FEL sources to HHG sources.

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EARLY STAGE RESEARCHERS' PROJECTS IN WP

- **ESR CEA** and *AU-exp.* performs experiments aimed at imaging the attosecond oscillation of the hole created by the tunnel ionization of multiple orbitals in triatomic molecules such as N_2O.
- **ESR POLIMI-2** and *AU-exp.* investigates more complex systems such as the small hydrocarbons C_2H_2, C_nH_{2n}, and allene C_3H_4 using a CEP-stabilized laser source at 1.5 µm.
- **ESR MBI-2** supports these experimental activities by extending the theoretical treatment of hole dynamics.
- **ESR FEMTO** develops a novel approach for achieving few-cycle CEP-stable pulses at 4 µm, ideal for laser-induced electron diffraction experiments.
- **ESR LUND-2** and *MBI-exp.* investigates the diffraction patterns generated by the core-shell photoelectrons emitted by specific atoms of aligned molecules via HHG sources.

WP-3 ESRs:

1. ESR-CEA 1 (1)
2. ESR-POLIMI-1 (2)
3. ESR-MBI-2 (2)
4. ESR-FEMTO 1 (1)
5. ESR-LUND-2 (2)

In total 7 ESR's

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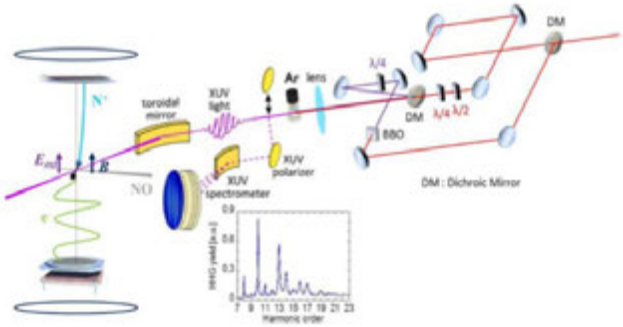
SCIENTIFIC ACTIVITIES AND GOALS IN PROGRESS

➢ **CEA**:

- Performed a complete characterization on the high harmonic polarization state, (helicity and degree of polarization) using molecular polarimetry
- Studied elliptical XUV emission (SF_6, N_2,Argon). Investigation reveals smaller ellipticity than found in previous optical polarimetry measurements, and significant depolarization.
- Performed theoretical studies on Harmonic emission from diatomic molecules. Findings questions the strict separation between continuum dynamics and recombination advocated, e.g., in the 3-step model, with strong implications for high harmonic spectroscopy.



- **CEA**: Complete characterization of the polarization state of the harmonic emission from bicircular counter-rotating ω and 2ω fields

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SCIENTIFIC ACTIVITIES AND GOALS IN PROGRESS

➢ **POLIMI**:

Interplay between structural and dynamical effects in (HHG) spectroscopy. Advanced alignment techniques will be investigated for HHG-based tomography of nonplanar molecules.

- Investigation of alignment of non planar molecules
- Reconstruction of the highest occupied molecular orbital for the case of ethylene was undertaken. Results: Inversion in the shape of the rotational revivals above 90 eV.
- Conflict: ionization limits the cutoff in HHG driven by Ti:Sapphire sources.
- Conclusion: need mid-IR ultrafast laser sources for orbital tomography in this kind of molecules

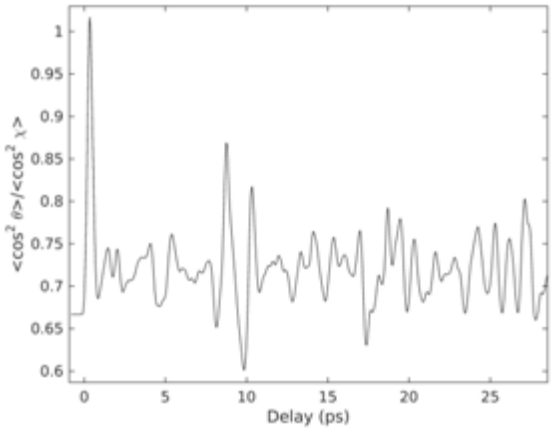
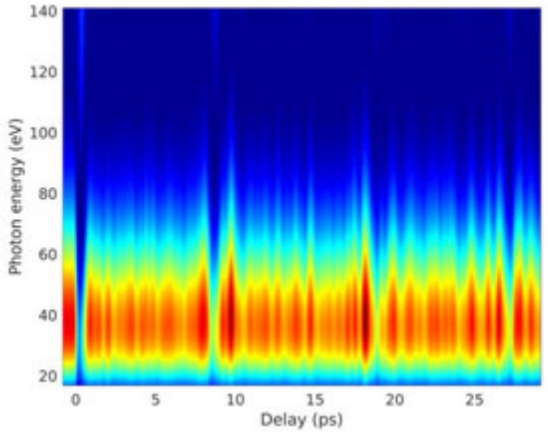


Figure 4 Temporal evolution of the ratio $\langle\cos^2\theta\rangle/\langle\cos^2\chi\rangle$ in ethylene after impulsive alignment; see text for details.



- **POLIMI**: Calculated macroscopic emission from ethylene as a function of the delay between aligning and HHG-driving pulses.

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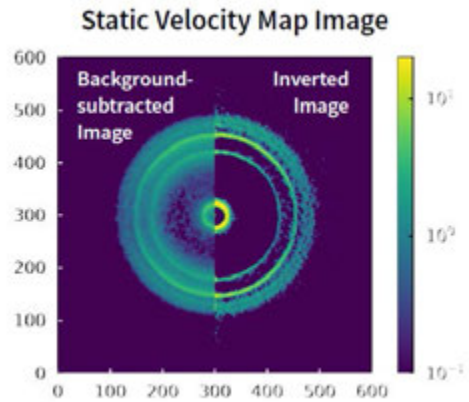
SCIENTIFIC ACTIVITIES AND GOALS IN PROGRESS

➢ MBI:

▪ Experimental

- Generation of an isolated harmonic using 400 nm high order harmonic generation
- Imaging the electronic structure of valence orbitals:

  XUV ionization of aligned CF_3I molecules and measurement of the photoelectron angular distribution
- Results: PAD is highly sensitive to symmetry and shape of the ionzed orbital

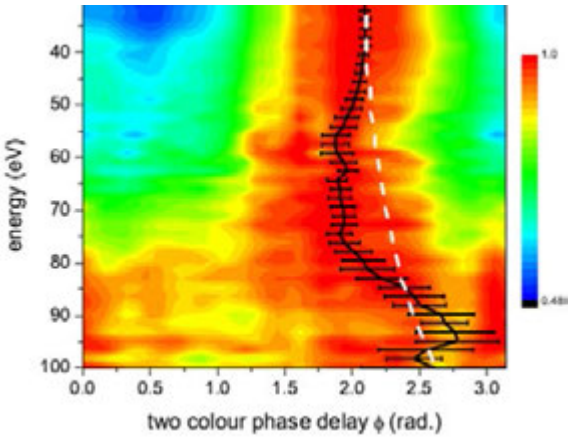


▪ Theoretical

- Development of a two-color HHG spectroscopy scheme fundamental pulse plus a weak second harmonic pulse (orthogonal polarized), with controlled delay. HHG signal recorded vs the two-color delay
- In order to establish a intramolecular interferometer concept to interpret the experimental results.

Comparison Theory experiments

- **First experimental evidence of correlation-assisted tunnelling.**
- **Direct experimental evidence of strong contribution of excited ionic states to HHG in mid-IR field.**



- **MBI**: Results of two-color HHG spectroscopy for the CO2 molecule, for the (laser intensity I=1.3 10^{14} W·cm^2)

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SCIENTIFIC ACTIVITIES AND GOALS IN PROGRESS

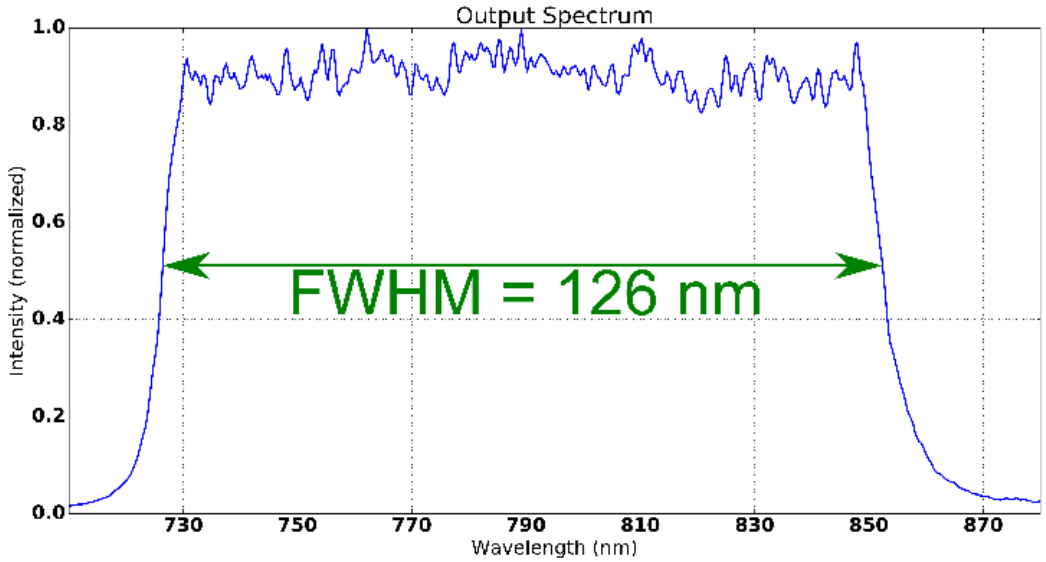
➢ **FEMTO**:

Seed and pump OPA

- Development of a sub-15 fs Ti-sapphire amplifier.
- Comparison simulation and experimental results to design Gauß filters for the small signal gain regime
- Define number of amplification passes
- Proof of principle: Ultra broadband amplification FWHM > 100 nm achieved corresponding pulse duration 15 fs (measured).

OPA

- Investigate (N)OPA design: pump/seed, crystal selection, phase matching type
- Investigate intrinsical CEP stability of Idler waves (1800 nm)



- **FEMTO**: Spectrum of the ultra broadband Ti-sapphire amplifier (pulse energy 1 mJ, repetition rate 1 kHz)

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SCIENTIFIC ACTIVITIES AND GOALS IN PROGRESS

- **LUND**: Make use intense XUV pulses produce through high-order harmonic generation for ultrafast imaging of molecular dynamics using e.g. photoelectron diffraction techniques.

  - Design of XUV pump-XUV probe experiments, offering a high XUV flux throughput
  - Development of a double velocity map imaging spectrometer optimized for experiments using high-intensity XUV sources
  - Integration of a high-density pulsed valve (Even-Lavie)with the photo ion/-electron spectrometer, enabling experiments on gaseous, liquid and solid samples.
  - **Two-photon double ionization of neon was demonstrated using an intense extreme ultraviolet (XUV) attosecond pulse train (APT) in a photon energy regime where both direct and sequential mechanisms are allowed.**

- **PHOTEK**:

  - Low temperature valves are developed through another project (called Puff) which is in collaboration with David Parker in Nijmegen. Expected finish date is later date in 2017.
  - In collaboration with LUND a coincidence detection VMI spectrometer will be finished in 2017.

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SCIENTIFIC ACTIVITIES AND GOALS IN PROGRESS

- **AU**:
  - AU, **DESY-CFEL** and other groups started a major experimental campaign at FLASH aiming to follow, in real time, the ring opening of thiophenone.
  - Time-dependent changes were observed and the data set is currently being analyzed.
- **DESY-CFEL**:
  - The collaboration with AU mentioned above.
  - Collaboration **MPIK**, **AU-exp.**, and **DESY**,

    study the diffraction patterns generated by photoemission from the iodine 4d orbital in various halogenated hydrocarbons.
  - The molecules are strongly aligned is space using adiabatic alignment and manipulation techniques developed by **AU-exp.** and **DESY**.
  - The data analysis is currently in progress.

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Summary

	Papers: Published	Papers: preparation/submitted	Conference talks
WP-3.1			
		2	1
	1		2
WP-3.2			
		1	
	4		
	2		3
		1	2
Total	7	4	8

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