WORKPACKAGE

WP1: Attosecond XUV-IR spectroscopy

Coordinators: Anne L'Huillier (LUND); Thomas Metzger (TRUMPF)

Period: 4-48 Month

The aim of WP1 is to **characterize attosecond and femtosecond electronic processes and nuclear dynamics occurring in molecules and clusters**. To achieve this goal, **high-repetition rate table-top HHG sources** will be developed by the academic and industrial partners of the network exploiting established and novel laser technologies. Using these systems, we will conduct XUV-IR pump-probe experiments implementing spectroscopy techniques based on the measurement of charged particles (**photoelectron and photoion angular distributions**) and photons (**absorption spectroscopy**).

This project has received funding from the European Union's Horizon 2020 research and innovation programme under the Marie Skłodowska-Curie grant agreement No 641789

DETAILS AND SUB WORKPACKAGES

WP 1.1 Development of high-repetition rate lasers for electronic correlation and electron-nuclear coupling in small systems

(ESR LUND-1, ESR AMPL-1, ESR AU-1, ESR FREIB-1)

- *High-repetition rate laser systems (industrial development)*
- *Photoelectron and photoion spectroscopy in small molecular systems*

WP 1.2 Multi-electron dynamics in large molecules

(ESR DESY, ESR MPQ)

- *Generation of controlled beams of complex molecules*
- *Femtosecond and attosecond dynamics in complex molecules*

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EARLY STAGE RESEARCHERS' PROJECTS IN WP

1) **ESR LUND-1 High repetition rate attosecond source for experiments with full energy, angular and temporal resolution (Jan Lahl)** Supervisors: A. L'Huillier

2) **ESR AMPL-1 CEP-stabilized multi-kHz regenerative amplifiers for attosecond experiments (Anna Golinelli)** Supervisor: X. Chen

3) **ESR AU-1 Kinetic energy release spectra in dissociative ionization of diatomic molecules (Qingli Jing)** Supervisor: L. Madsen

4) **ESR FREIB-1 Coincidence spectroscopy in small molecules (M. Moioli)** Supervisor: G. Sansone

5) **ESR DESY Attosecond dynamics in conformer-selected amino acids (Melby Johny)** Supervisor: J. Küpper

6) **ESR MPQ Electron delocalization at the molecule-surface interface (Dionysios Potamianos)** Supervisor: R. Kienberger

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List of Deliverables WP1

Deliverable Number[14]	Deliverable Title	Lead beneficiary	Type[15]	Dissemination level[16]	Due Date (in months)[17]
D1.1	High-repetition rate photoelectron spectroscopy	3 - LUND	Report	Public	24
D1.2	Regenerative amplifier	7 - AMPL	Demonstrator	Confidential, only for members of the consortium (including the Commission Services)	48
D1.3	Time-resolved dynamics in conformer selected beams	4 - DESY	Report	Public	36
D1.4	Attosecond dynamics in small and large molecules	3 - LUND	Report	Public	48

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Schedule of relevant Milestones WP1

Milestone number	Milestone title	Lead beneficiary	Due Date (in months)	Means of verification
MS1	Demonstration of high repetition rate single attosecond pulse	3 - LUND	24	as pulses
MS2	XUV/IR cross correlation	3 - LUND	30	XUV/IR cross correl.
MS3	Novel design for regenerative amplifiers for the generation of CEP-stable pulses	7 - AMPL	24	Novel regen 0.1mJ
MS4	Montecarlo wavepacket technique	6 - AU	24	Extension of the Monte Carlo wavepacket technique
MS5	Combination of high-density valve with imaging detection	4 - DESY	22	high-density, high repetition valve optimized for generation of cold beams of aminoacids
MS6	Molecule/surface interface imaging	5 - MPQ	24	Characterization of the molecule/surface interface by spectroscopic techniques
MS29	High-repetition rate photoionization experiments	11 - FREIB	11	Photoionization experiments at high rep. rates

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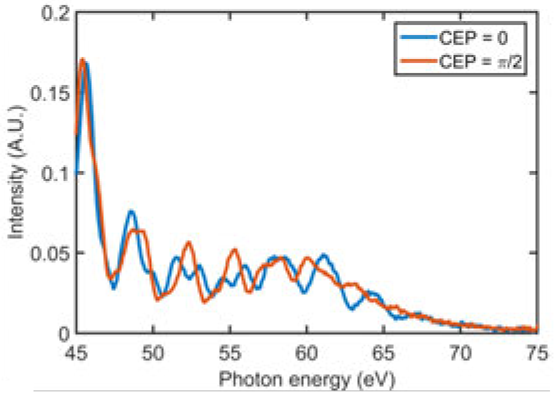
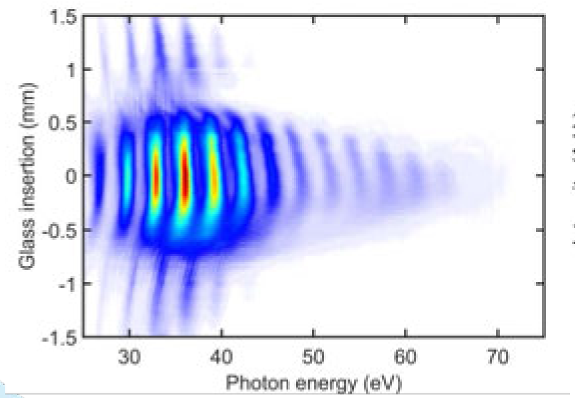
Highlights of WP1: *High repetition rate Optical-Parametric Chirped Pulse Amplification (OPCPA) laser and PEEM*

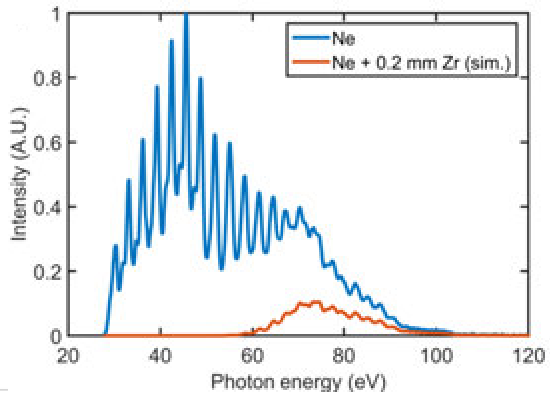
LUND and VENTEON

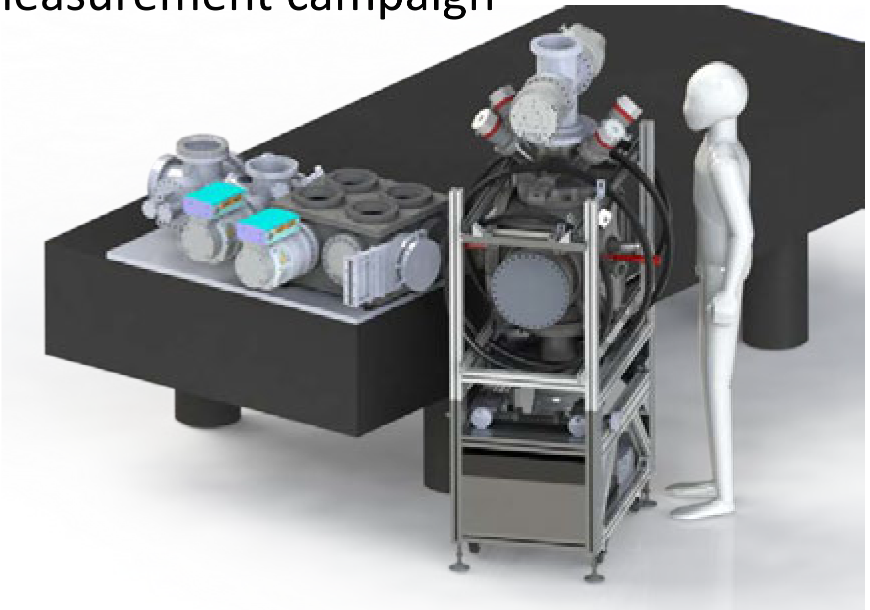
Developed system: 2 stage OPCPA: 5µJ (>10 µJ planned); <7fs; 200kHz; CEP stab. **(VENTEON)**

Experiment: HHG generation demonstrated, XUV-IR interferometer designed and realized, photo-emission electron microscopy (PEEM) was build up (for atomic and molecular experiments with full energy, angular and temporal resolution, a first measurement campaign will be started soon. **(LUND)**









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Highlights of WP1: Montecarlo wavepacket technique

AU

- Qingli Jing & Prof. Lars Bojer Madsen: formulating a stochastic Monte Carlo wavepacket approach to the problem of dissociative ionization (for single pulse processes & pump-probe schemes) .
- Because of increasing interest in mid-IR: It was suggested to study laser-induced dissociative double ionization of molecular hydrogen with wavelengths from the near-infrared and into the mid-infrared regime.
- Now: Work continues on dissociative ionization in various pump-probes schemes, including also attosecond XUV-IR pump-probe spectroscopy and possible elucidation of the effects of autoionizing states.

Qingli Jing and Lars Bojer Madsen, "Laser-induced dissociative ionization of H_2 from the near-infrared to the mid-infrared regime", Phys. Rev. A **94**, 063402 (pages 11) (2016).

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Highlights of WP1: High-Density, High Repetition Valve

DESY

- Theoretically investigated the separability of glycine conformers in a cold beam: computationally shown: different experimentally observed conformers can be separated by dispersion of a cold (~1 K) molecular beam.
- Performed detailed investigations of the possibility to thermally vaporize and supersonically expand glycine form an Even-Lavie valve, but could not observe a beam of intact glycine molecules but fragments.
- Performed different laser-desorption based experiments and have produced warm plumes of glycine as well as beams of cold glycine molecules entrained in supersonic jets of rare gases. We expect that these beams can be dispersed and at least relatively pure (>90 % purity) samples of conformers I and II, maybe also III (and possibly IV) can be produced in our separation setups.

- PhD thesis of Thomas Kierspel in spring 2017
- Soon: Teschmit, Horke, Küpper, et al.

MEDEA

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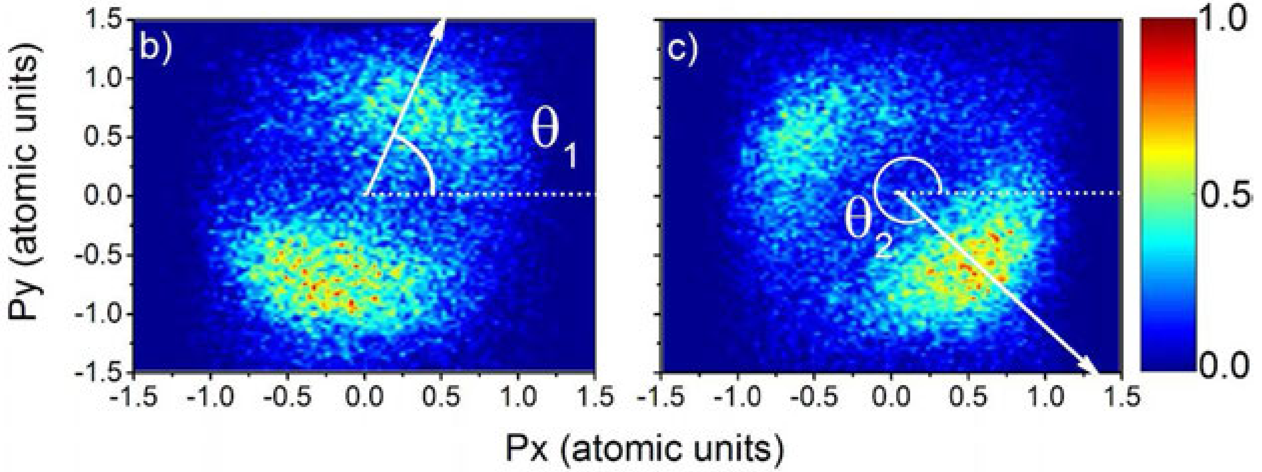
Highlights of WP1: *Determination of the polarization direction of isolated attosecond pulses at 10 kHz*

POLIMI, AMPL and MPIK

Fully characterize the vectorial electric field of few-cycle pulses (at POLIMI with MPIK) based on the photoelectron spectrometer installed in the Reaction Microscope, which gives access to the 3D angular distribution of the photoelectrons. Isolated as pulses were generated by modulating in time the polarization state of few-cycle driving pulses using two birefringent plates (cryst. quarz generating a delay and lambda/2). Polarization depends on delay.

By fitting the photoelectron angular distributions the polarization direction was determined.

Photoelectron angular distribution generated by an isolated attosecond pulse for (left) and (right). The angle of the polarization axes of the attosecond pulses is indicated by (left) and (right)



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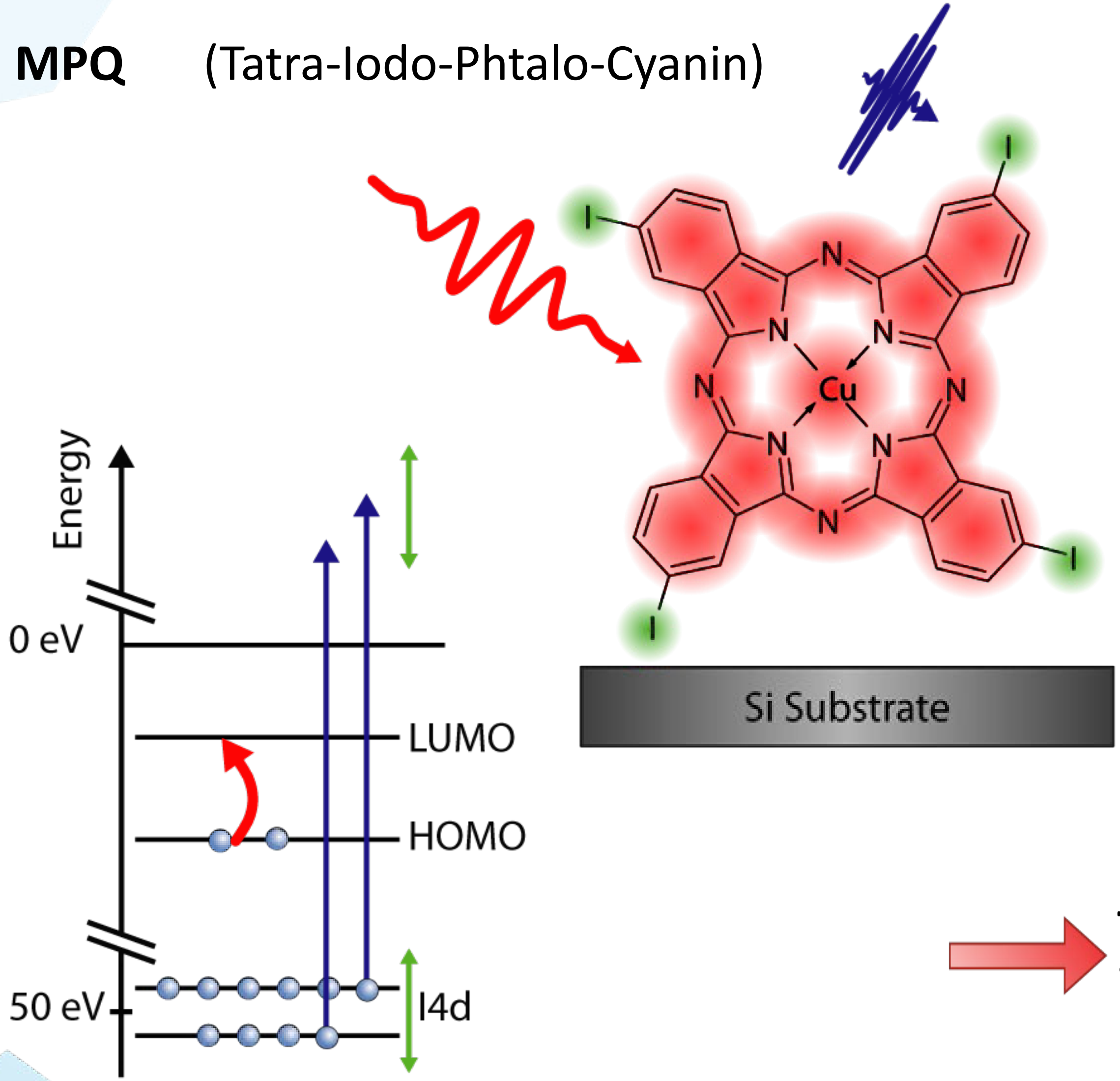
Highlights of WP1: Charge transport in chemical systems: light harvesting

MPQ (Tatra-Iodo-Phtalo-Cyanin)



- **Visible light absorption induces π-π^* transition**
- **Time dependent chemical shift via coulomb interaction**
- **Ionization of iodine 4d orbitals by XUV probe**

Tracking of electron motion in molecules with 1 fs resolution

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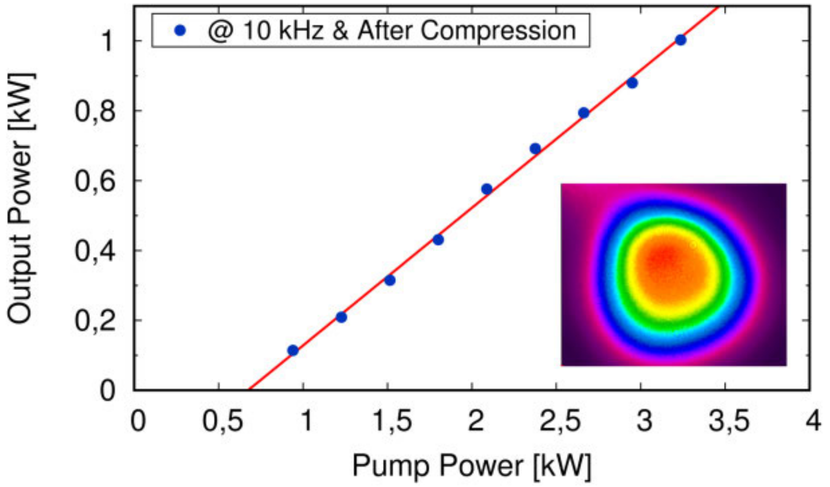
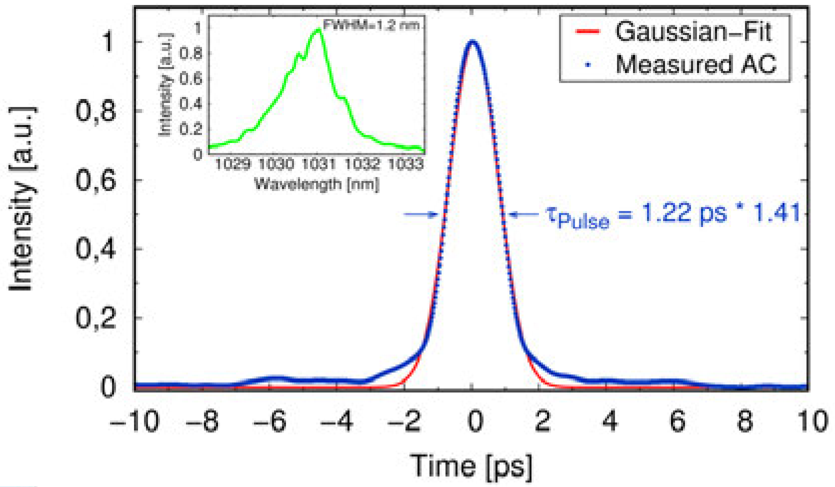
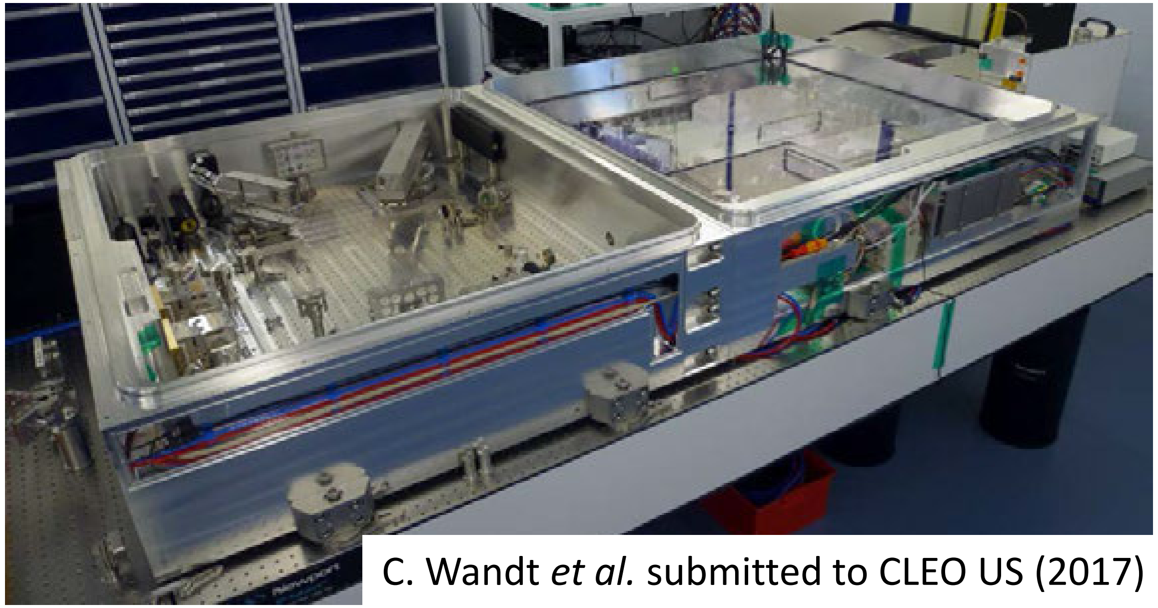
Highlights of WP1: *Development of high-repetition rate laser systems at* ***TRUMPF*** *(MPQ)*

Developed system: Pump laser for OPCPA: 10-100mJ; 1ps, 10-100kHz; 1kW average power





- improve pulse compression and beam profile
- 200mJ; 5kHz; 1ps nearly completed
- 1J/multi-kW development has started
- pump seed synchronization (2-20fs rms jitter) developed and commercialized (S. Prinz et al., Opt. Express **22**, 31050-31056 (2014)



C. Wandt *et al.* submitted to CLEO US (2017)

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