

XTRA

HUMAN RESOURCES AND MOBILITY (HRM)  
ACTIVITY

MARIE CURIE ACTIONS  
Research Training Networks (RTNs)

PART B

Ultrashort XUV Pulses for Time-Resolved and Non-Linear Applications

“XTRA”

## ***Research topic***

In the last few decades the development of ultrashort laser pulses has led to significant advances in physics, chemistry and biology. With femtosecond lasers the elementary motions of atoms and molecules can be observed, leading - for example - to a better understanding of complex biological systems. Though electron dynamics often plays a crucial role in these systems, experimental studies have thus far been mostly limited to investigating the ‘slow’ particles (i.e. the atoms) that move under the influence of a potential that results from a time-average over all interactions involving electrons. In order to investigate electron dynamics *in real-time*, attosecond laser pulses (1 attosecond =  $10^{-18}$  seconds) are needed.

Within the last two years spectacular progress has led to the first experimental realization and detection of attosecond laser pulses. This progress can be illustrated by the number of citations involving the word ‘atto’ in the research literature in the last few years, which has grown from 17 papers in refereed journals in 2001 to 32 in 2002. Attosecond pulses are ‘hot’ and justifiably so, because the development of attosecond pulses opens up the possibility to study a new class of physics problems – most notably concerning the role of electron dynamics – that in the past we could not even begin to consider studying. Therefore, the development of attosecond laser pulses represents both an accomplishment and a tremendous challenge for the future, where we have to learn to give meaning to the concept of ‘attoscience’. It is the ambition of this network to play an important role in this process. The network unites a number of groups that in the last few years have broken the attosecond barrier with teams in atomic physics, molecular physics and solid-state physics that are eager to explore the possibilities for application of ultrashort XUV pulses in their respective fields, with the atomic physics segment that was crucial in the development of the technique providing a vital link. The nature of the project is very much a dialogue between specialists in the development of optical techniques for the generation and characterization of unique new light sources and a community of researchers continuously looking for new tools and techniques to further the understanding in their field. Through the research carried out in this network a new generation of young research scientists will be trained that in the future can play an important role in the further development and spread of this emerging technology. The young researchers trained by the network will later in their career benefit from the fact that their education did not just consist of the development of a new technique or the use of existing technologies in their application field of interest. Rather, they will have witnessed and have been instrumental in the birth of an important new technology. Through their network training they will become people who do not develop technologies for their own sake, but who are very aware of the fact that a new technology has to serve a purpose (in science or industry). This is very challenging but ultimately one of the most rewarding learning experiences that a young researcher can have.

In the network the method for the generation of ultrashort XUV laser pulses is harmonic generation. For the foreseeable future this is the technique of choice, and all the applications that will be attempted in the network will make use of this technique. At the same time this proposal is very much relevant to the ongoing development of other techniques with the potential to generate attosecond and femtosecond XUV pulses. Within a few years the 2<sup>nd</sup> stage of the TESLA Free Electron Laser project will be completed, leading to the generation of ultra-high brightness pulses in a wavelength range that is covered by high harmonic generation. The network can and will play an important role in the dialogue between laser-based and synchrotron-based research teams pushing the capabilities and applications of ultrashort XUV pulses.

## ***Project objectives***

The main objective of this proposal is the development and exploitation of state-of-the-art techniques for the generation of attosecond and femtosecond extreme ultra-violet (XUV) laser pulses in important and challenging experiments in atomic, molecular and solid state physics. To make this possible the objectives of the project are organized into 4 tasks:

### **Task A: Development and characterization of state-of-the-art femtosecond and attosecond light sources**

In the 5<sup>th</sup> framework programme, the EU sponsored the formation of the Research Training network ATTO (*Generation and Characterisation of Attosecond Pulses in Strong Laser-Atom Interactions: A Step towards Attophysics*). The results of this network have been nothing short of spectacular. In ATTO the *first ever* attosecond pulses were created and characterized, and the *first ever* time-resolved measurement using an attosecond pulse was performed, representing a level of progress that far exceeded expectations and firmly established the position of Europe at the forefront of this emerging field. These results have generated enormous excitement and have already led to new initiatives in several countries (the United Kingdom, the Netherlands, Canada, Switzerland) for large-scale national programs to acquire and develop the capabilities in this field. Many aspects of the generation, characterization and implementation of attosecond pulses still have to be investigated, before attosecond pulses can become a routine tool that can be as widely used (and is as easy to use) as present day femtosecond lasers operating in the visible and IR part of the wavelength spectrum. Therefore in the present network the first task represents a continuation of the work that was successfully initiated during ATTO, where the teams that participated in ATTO join forces with a few new teams with complementary expertise, and will focus their efforts on the following aspects:

- 1) *The generation and characterization of ultra-high power carrier envelope phase-controlled light sources.*

High power phase-stable laser pulses are a vital key to the controlled production of single attosecond pulses, which in turn is an essential condition for the use of attosecond pulses in time-resolved experiments with attosecond time resolution. Combining techniques like hollow core fiber pulse compression, the use of an optical parametric amplifier for chirped pulse amplification (OPCPA) and multi-stage carrier envelope phase stabilization we will *develop a Terawatt peak power few-cycle (~5 fs) carrier envelope phase-stabilized laser*. Diagnostics for the carrier envelope phase will be developed based on angle-resolved photoelectron spectroscopy.

- 2) *The generation and characterisation of attosecond and femtosecond XUV pulses*

Within ATTO high harmonic generation was developed as a route to the generation of attosecond and femtosecond XUV pulses. In the present network we will generate *single attosecond pulses using carrier envelope phase-controlled high energy pulses* (Task A.1) Complete characterization of femtosecond and attosecond XUV pulses is absolutely crucial for the interpretation of experiments where these pulses are used (Tasks B-D). Therefore we will develop a series of tools for the *complete characterization of femtosecond and attosecond laser pulses*. These tools will include second-order autocorrelation measurements, angular-resolved photoelectron spectroscopy (AR-PES), cross-correlation frequency-resolved optical gating (XFROG) and spectral interferometry for direct electric field reconstruction (XSPIDER).

- 3) *New approaches for the generation of intense ultrashort XUV pulses.*

If attosecond and femtosecond XUV pulses are to have the impact on science that the development of femtosecond visible and IR lasers has had in the past decade, then it is

absolutely crucial that methods are developed that can not only be used in specialized laser laboratories but that can be *readily available to users in diverse areas of research*. We will explore several highly promising schemes that will increase the generation efficiency and availability of femtosecond and attosecond XUV pulses, while greatly reducing their cost.

### **Task B: Applications of attosecond and femtosecond XUV pulses in Atomic Physics**

High harmonic generation developed a little more than a decade ago out of studies of atomic ionization processes using the intense lasers that were coming available at the time, and were first observed in experiments on the strongly-related phenomenon of above-threshold ionization (ATI). In ATI and harmonic generation an electron is launched into the continuum via tunnel-ionization, the final fate either being the production of a free electron in ATI or the production of an XUV photon if recombination takes place. The development of attosecond optical pulses makes it possible for the first time to consider the possibility of studying the harmonic generation process that is responsible for the attosecond pulse generation *itself in real-time!* At the same time attosecond and femtosecond XUV pulses allow us to address several of the main questions occupying researchers in atomic laser physics. While one-photon double-electron ionization, many-photon one-electron ionization and many-photon many-electron ionization are all quite well understood, one of the main remaining challenges for theorists and experimentalists is understanding few-photon few-electron ionization processes. The availability of intense femtosecond and attosecond XUV pulses now allows experimental studies on these problems for the first time. At the same time, the spectral characteristics of attosecond and femtosecond XUV pulses allow us for the first time to dig deeper into the atom, and study dynamical processes not only for outer shell electrons but also for inner shell electrons. Along with important new possibilities in spectroscopy and metrology the network will study:

1) *Time-resolved photoionization processes*

We will study sub-fs dynamics of inner shell ionization processes, starting from single-state lifetime measurements that can be compared to available spectroscopic measurements, and proceeding to *inner shell relaxation processes involving quantum beats* (as a result of coupling between adjacent states) and *non-exponential decay* (as a result of competing and interfering autoionization pathways). Using atomic ionization as a benchmark system, a streak camera for ultrafast detection of XUV photoionization processes will be developed, that can also be applied in Tasks C and D.

2) *The dynamics of strongly-driven electrons*

The formation of attosecond XUV pulses results from recombination of strongly-driven electrons formed by tunnel ionization. Using an attosecond pulse train formed by high harmonic generation, we will *study the motion of the electron wavepacket within the cycle of the optical driving pulse*. And we will perform experiments with IR, THz and MHz driver pulses, where the electron dynamics can be inferred from *full characterization of the harmonics generated or directly visualized in the time-domain*. Extending the laser intensity to relativistic intensities we will explore the potential for strongly driven electrons to *excite and resolve nuclear processes*.

3) *Electron-electron correlation in few-photon few-electron ionization processes*

Few-photon few-electron ionization (including the preparation of inner-shell excited atoms) is an experimentally completely unexplored field that is at the center of attention of current theoretical work in atomic laser physics aimed at understanding electron-electron correlations. We will study the *competition between direct and sequential double ionization by two-photon XUV absorption*. In related work we will investigate electron-electron

correlation by *studying ionization (or excitation) of inner shell states as a function of the degree of excitation of an outer shell electron* that is resonantly excited to a bound state or a coherent superposition state.

4) *Applications in atomic spectroscopy and metrology*

High harmonic generation provides unique opportunities for spectroscopy in the otherwise ‘difficult’ XUV part of the spectrum. We will develop *Fourier-Transform spectroscopy and Ramsey-type spectroscopy to study (short-lived) excited atomic states*. Another application will be in femtosecond laser produced laser plasmas where the temporal and spectral properties of harmonic radiation allow to study the plasma at high electron densities and with high spatial and temporal resolution. Using a high repetition rate high power few-cycle laser we will generate a harmonic frequency comb extending from 40 nm to 800 nm, for use in ultra-high resolution spectroscopy.

### **Task C: Applications of attosecond and femtosecond XUV pulses in Molecular Physics**

Ultrashort XUV pulses produced by high harmonic generation (HHG) can be extremely useful in femtochemistry. XUV light can both trigger and probe electronic and nuclear dynamics in molecules, since *single-photon excitation* of both valence shells ( $h\nu < 50\text{eV}$ ) and inner-shells ( $h\nu < 300\text{eV}$ ) is possible. State-to-state studies of molecular reaction dynamics (e.g. dissociative ionization or reactive collisions) are thus feasible, in contrast with commonly used multiphoton excitation schemes in the UV-visible part of the wavelength spectrum, where secondary processes often mask the process of interest. Furthermore it has recently been demonstrated that the same strongly-driven electrons that form the basis of the harmonic generation process (task B2) can *themselves* be used for structural determination of the molecules from which they originate. Using strongly-driven electrons in the first femtoseconds of a molecular rearrangement process and an attosecond or femtosecond XUV pulse at ‘longer’ timescales, molecular structural changes can be studied on arbitrary timescales. Within the network we will study:

1) *Time-resolved electronic and nuclear wavepackets*

The fundamental timescale for molecular re-arrangement processes is the vibrational period, which limits the timescale for making and breaking of molecular bonds. We will reach the ultimate timescale for neutral chemistry by *observing the fundamental vibrational period of the  $H_2$  molecule*. At the same time, we will study *electron dynamics in strongly optically driven  $H_2$  molecules*, as a model system for laser-controlled chemistry where molecular bonds can selectively be weakened or strengthened.

2) *Time-resolved molecular structure determination*

We will use strongly-driven continuum electrons to *measure the structure of a simple molecule via electron diffraction* and measure electron re-collision in larger systems, as a step towards the use of time-resolved electron diffraction using continuum electrons for monitoring structural changes in molecules on attosecond and femtosecond timescales.

3) *Photodissociation dynamics of polyatomic molecules*

We will perform time-resolved XUV-UV and UV-XUV pump-probe experiments to elucidate *the role of conical intersections in the photodissociation of benchmark polyatomic molecules* like  $NH_3$  and  $C_2H_4$ , using time-resolved photoelectron and photofragment spectroscopy to monitor molecular re-arrangement and dissociation processes.

**Task D: Applications of attosecond and femtosecond XUV pulses in Solid State Physics**

One of the most exciting potential application areas of isolated XUV pulses generated by high harmonic generation is the elucidation of the real time dynamics of hot electrons at surfaces and within nanostructures. Experimentally ultrafast time-resolved photoemission experiments can probe the real time scattering of intermediate, excited electron states as a function of the energy of the state (i.e., in a state-specific fashion) and can be correlated with the band structure of the system. Or the relaxation dynamics can be probed by measuring *resonant* XUV reflectivity spectra of shallow core levels, providing *chemical* and *site specific* information. The interaction of XUV pulses with materials is also of interest itself, since the XUV light can excite core-level states that relax by forming high concentrations of electron-hole pairs through mechanisms that have yet to be elucidated and that can give rise to localizations and defects. Finally, the ability of nanoscale surface features to act as preferential binding sites for biological molecules, e.g., proteins and photoreceptors, raises the ultimate prospect of investigating electron dynamics in biological systems on (sub)-femtosecond timescales using time-resolved photoelectron spectroscopy. Within the network we will study:

1) *Time-resolved electron dynamics*

The application of femtosecond XUV pulses in surface science makes it possible to study electron dynamics in real time using time-resolved photoelectron emission. Using this technique we will study *magnetization changes and phase transitions in magnetic materials, as well as carrier mobility, scattering and relaxation phenomena inside quantum-confined structures*, such as “zero-dimensional” quantum-confined systems based on the use of nanostructured surfaces.

2) *Time-resolved structural dynamics*

Time-resolved photoemission spectroscopy can also be applied to monitor structural modifications of solids. We will study photo-induced melting of semiconductors by an intense UV laser as well as photo-induced nuclear dynamics of physisorbed molecules on surfaces.

3) *Non-linear processes on surfaces*

It has recently been demonstrated that the high peak power of harmonic pulses makes it possible to drive two-photon processes. We will perform experiments aimed at determining estimates of the XUV two-photon cross sections on solid surfaces, which are basically totally unknown. Processes like two-photon absorption and above-threshold ionization, or two-photon harmonic generation, can be considered.

<b>Team</b>	<b>Scientist-in-charge</b>	<b>Task A</b>	<b>Task B</b>	<b>Task C</b>	<b>Task D</b>
1. AMOLF	<i>Dr. M.J.J. Vrakking</i> (Network Coordinator & Sub-Coordinator Task C)	<b>X</b>	<b>X</b>	<b>X</b>	
2. Lund	<i>Prof. A. L'Huillier</i> (Sub-Coordinator Task B)	<b>X</b>	<b>X</b>	<b>X</b>	
3. CELIA-Bordeaux	<i>Dr. E. Constant</i>			<b>X</b>	<b>X</b>
4. Vienna	<i>Prof. F. Krausz</i> (Sub-Coordinator Task A)	<b>X</b>	<b>X</b>		
5. Milano	<i>Prof. M. Nisoli</i>	<b>X</b>			<b>X</b>
6. IESL Heraklion	<i>Prof. D. Charalambidis</i> (Sub-Coordinator young researcher training)	<b>X</b>	<b>X</b>		<b>X</b>
7. Saclay	<i>Dr. P. Salières</i>	<b>X</b>	<b>X</b>	<b>X</b>	
8. MPQ-Garching	<i>Dr. G. Tsakiris</i>	<b>X</b>	<b>X</b>	<b>X</b>	
9. LOA-Palaiseau	<i>Dr. Ph. Balcou</i>	<b>X</b>	<b>X</b>		<b>X</b>
10. Birmingham	<i>Prof. R.E. Palmer</i> (Sub-Coordinator Task D)			<b>X</b>	<b>X</b>
11. Oxford	<i>Prof. I.A. Walmsley</i>	<b>X</b>			
12. Szeged	<i>Prof. S. Szatmári</i>	<b>X</b>	<b>X</b>		
13. Zürich	<i>Prof. U. Keller</i>	<b>X</b>		<b>X</b>	
14. NRC-Ottawa	<i>Prof. P.B. Corkum</i>		<b>X</b>	<b>X</b>	
15. FOCUS-Michigan	<i>Prof. P.H. Bucksbaum</i>	<b>X</b>	<b>X</b>		

## ***Work plan***

### **Task A: Development and characterization of ultrashort XUV laser pulses (sub-coordinator Krausz, Vienna)**

#### **Sub-task A1:** *Generation and characterization of carrier phase-controlled light sources*

Research teams involved: Vienna, Milano, Oxford

A1.1 Compression and carrier-envelope phase (CEP) stabilization of Terawatt pulses. *Vienna, Milano, Oxford*

A1.2 Carrier-envelope phase metrology *Vienna, Milano, Oxford*

#### **Sub-task A2:** *Characterization of attosecond and femtosecond pulses*

Research teams involved: AMOLF, Lund, Vienna, IESL, Saclay, MPQ, LOA, Oxford, Zürich, FOCUS

A2.1 Generation and characterization of single attosecond pulses generated using carrier phase-controlled light sources *Vienna, Oxford*

A2.2 Energy-resolved second-order autocorrelation of attosecond pulse trains *MPQ, IESL*

A2.3 Detection of harmonic phase-locking by angle-resolved photoelectron spectroscopy *AMOLF*

A2.4 XUV Cross-correlation frequency-resolved gating (XFROG) *Lund, Saclay, LOA, Zürich*

A2.5 XUV Spectral interferometry for direct electric field reconstruction (XSPIDER) *Lund, Oxford, Saclay, FOCUS*

#### **Sub-task A3:** *New approaches for the generation of intense ultrashort XUV pulses*

Research teams involved: Vienna, Milano, Saclay, MPQ, LOA, Oxford, Szeged, Zürich

A3.1 Harmonic generation using a high-energy MHz repetition rate light source *Vienna, Zürich*

A3.2 Harmonic generation using a high-energy sub-ps excimer laser *Szeged*

A3.3 Development of a high flux-high quality harmonic source *Saclay, Oxford*

A3.4 Generation of intense attosecond pulse trains using surface harmonic generation *MPQ*

A3.5 Optimal control of ultrafast non-linear processes *Milano, LOA, Saclay*

### **Task B: Applications of ultrashort XUV pulses in Atomic Physics (sub-coordinator L'Huillier, Lund)**

#### **Sub-task B1:** *Time-resolved photoionization processes*

Research teams involved: AMOLF, Vienna, IESL, Szeged, FOCUS

B1.1 Measurement of sub-fs super-Coster-Kronig Auger decay *Vienna*

B1.2 Quantum beat spectroscopy of atomic inner-shell states *Vienna*

B1.3 Non-exponential decay in interfering Auger processes *Vienna*

B1.4 Excited state wavepacket dynamics *AMOLF, IESL, Szeged, FOCUS*

B1.5 Electron-electron correlation in inner shell-outer shell interactions *IESL*

#### **Sub-task B2:** *The dynamics of strongly-driven electrons*

Research teams involved: AMOLF, Lund, NRC, FOCUS

B2.1 Sub-cycle probing of multi-photon ionization at optical frequencies *Lund, NRC*

B2.2 Multi-photon ionization and harmonic generation at IR, THz and MHz frequencies *AMOLF, FOCUS*

B2.3 Attosecond nuclear physics *NRC*

#### **Sub-task B3:** *Electron-electron correlation in few-photon few-electron ionization processes*

Research teams involved: Lund, IESL, MPQ

- B3.1 Two-photon direct and sequential XUV double ionization *Lund, MPQ, IESL*
- B3.2 Multi-photon inner shell ionization *Lund*

**Sub-task B4: Applications in atomic spectroscopy and metrology**

Research teams involved: Vienna, Saclay, LOA

- B4.1 XUV Fourier-Transform spectroscopy and Ramsey-type spectroscopy *Saclay*
- B4.2 Time-resolved diagnostics of dense plasmas *Saclay, LOA*
- B4.3 Metrology using a harmonic frequency-comb extending from 40 to 800 nm *Vienna*

**Task C: Applications of ultrashort XUV pulses in Molecular Physics  
(sub-coordinator Vrakking, AMOLF)**

**Sub-task C1: Time-resolved electronic and nuclear wavepackets**

Research teams involved: CELIA, Birmingham

- C1.1 Time-resolved Coulomb explosion imaging of molecular vibrational wavepackets *CELIA*
- C1.2 Electron dynamics in strongly optically-driven H<sub>2</sub> molecules *Birmingham*
- C1.3 Dynamics of molecular Rydberg wavepackets *Birmingham*

**Sub-task C2: Time-resolved molecular structure determination**

Research teams involved: AMOLF, Zürich, NRC

- C2.1 Control of strongly driven electron wavepackets *NRC, AMOLF*
- C2.2 Attosecond and femtosecond electron diffraction imaging of molecular structures *NRC*
- C2.3 Photoionization and photodissociation of fixed-in-space molecules *AMOLF, Zürich*
- C2.4 Attosecond electron diffraction imaging of molecular dissociation processes *NRC*

**Sub-task C3: Photodissociation dynamics of polyatomic molecules**

Research teams involved: AMOLF, Lund, CELIA, Saclay, MPQ

- C3.1 Nuclear wavepackets probed using femtosecond time-resolved XUV photoelectron spectroscopy *AMOLF, Lund, CELIA*
- C3.2 The role of conical intersections in the dissociation of polyatomic molecules probed using femtosecond time-resolved fragment translational spectroscopy *AMOLF, Lund*
- C3.3 Time-resolved dissociation of double-bonded ( $\pi$ ) systems *MPQ, Saclay*

**Task D: Applications of ultrashort XUV pulses in Solid State Physics  
(sub-coordinator Palmer, Birmingham)**

**Sub-task D1: Time-resolved electron dynamics**

Research teams: IESL, CELIA, Milano, Birmingham

- D1.1 Ultrafast electron dynamics in magnetic materials *Milano*
- D1.2 Ultrafast electron dynamics in quantum-confined structures *IESL, Birmingham*
- D1.3 Electronic relaxation in solids via electron-electron and electron-phonon coupling *CELIA*

**Sub-task D2: Time-resolved structural dynamics**

Research teams: IESL, Birmingham

- D2.1 Probing of surface physiochemical processes via time-resolved UPS/soft XPS *IESL*
- D2.2 Dynamics of physisorbed molecules *Birmingham*

**Sub-task D3: Non-linear processes on surfaces**

Research teams involved: LOA

- D3.1 XUV two-photon photoemission on dielectrics and scintillators *LOA*