# PROPOSALS FOR PUMP-PROBE EXPERIMENTS IN THE GAS PHASE USING THE TTF2-FEL

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*Abstract*

A selection of experiments is discussed, which will make use of the combination between VUV photon pulses produced by the TTF2-FEL in Hamburg (Germany) and pulses of an optical laser for studying atomic and molecular samples. By applying a two-photon pump-probe excitation scheme, the proposed investigations and the expected results will enable us, on the one hand, to characterize the FEL pulses and, on the other hand, to gain new insight in the dynamics of the electronic interaction in the electronic clouds of excited atoms as well as of the molecular dissociation processes.

## INTRODUCTION

Many processes induced by the interaction of XUV light with free atoms and molecules take place on a very short time scale. Photoexcitation of a core-electron to the ionization continuum or to a resonant state will, in general, be followed by fast electronic relaxation through autoionization or Auger process on a sub-picosecond time scale. The temporal width of the soft X-Ray pulses from the TTF2-FEL (100-300 fs) and of the synchronized, tunable optical laser (about 150 fs) will therefore be ideally suited to gain insight into the dynamics of these processes. By using time-resolved pump-probe techniques it will be possible to investigate the temporal evolution of a variety of processes, like electronic relaxation of autoionization states, coupling between two autoionization states, wave-packet formation of higher Rydberg states, coupling between electronic and nuclear motion in molecular systems, fast dissociation of molecules upon inner- and outer-shell photoexcitation, coherent population of fragment states etc.

First experiments have of course also to serve for characterizing the FEL pulses themselves, in particular the intensity, frequency and time structure of the individual pulses, since these values are only estimated on the basis of theoretical simulations. A powerful tool to obtain information about the shape of femtosecond pulses is given by cross correlation experiments [1-3]. This two-color method has been successfully applied to characterize the short X-Ray pulses generated by the generation of higher harmonics in an intense laser in a gas jet of rare gas atoms. It uses the measurement of the ionization signal induced by a XUV photon in the presence of a strong infrared beam (Figure 1). The latter induces sidebands on both sides of the main line in the photoelectron spectrum. The shape, intensity and position of the sidebands are directly correlated to the shape of the pulses and their time delay. At low photon energies, i.e. in the region between 15 and 50 eV, the ionization of the outer Xe 5p or Ar 3p shell can be studied. At higher energies more tightly bound electrons can be excited, e.g. Xe 4d at about 65 eV or Kr 3d at 90 eV.

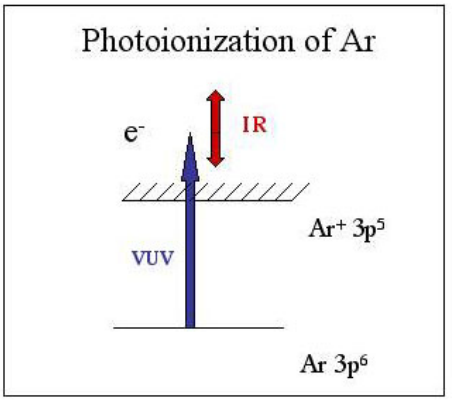


Figure 1: Schematic representation of the photoionization process of atomic Ar by a VUV photon in the presence of a strong IR laser field.

In addition, after excitation of a core electron it will also be possible to measure the corresponding Auger spectrum, where the dressing laser will give rise to the same type of side bands [4,5]. The spectra will be influenced by the interaction between different ionization continua (one-photon and two-photon ionization) and the resonantly excited state, which is characterized by an Auger lifetime of the same order as the exciting femtosecond photon pulses.

## CHARACTERIZATION OF FEL-PULSES

In order to select suitable systems for measuring and controlling the temporal delay between the FEL and the optical laser pulses and to find out the potentiality of the cross correlation method, we have performed a series of measurements [3] using the femtosecond laser system at the Laser Center (LLC) in Lund (Sweden). The XUV pulses are produced in Ar gas by generation of the higher harmonics (HHG) nof an intense (about 1 mJ at a 1 kHz repetition rate) 800 nm laser system giving 50 fs pulses [6]. The XUV pulses are used to ionize in the experimental chamber a sample of rare gas atoms and the photoelectron spectrum is recorded using a magnetic bottle spectrometer. The cross correlation measurements have been performed with part of the fundamental 800 nm beam, which is separated before entering the chamber for the production of the higher harmonics. In the presence of the strong IR laser pulse (dressing beam) the photoelectron spectra shows clearly some side-bands,

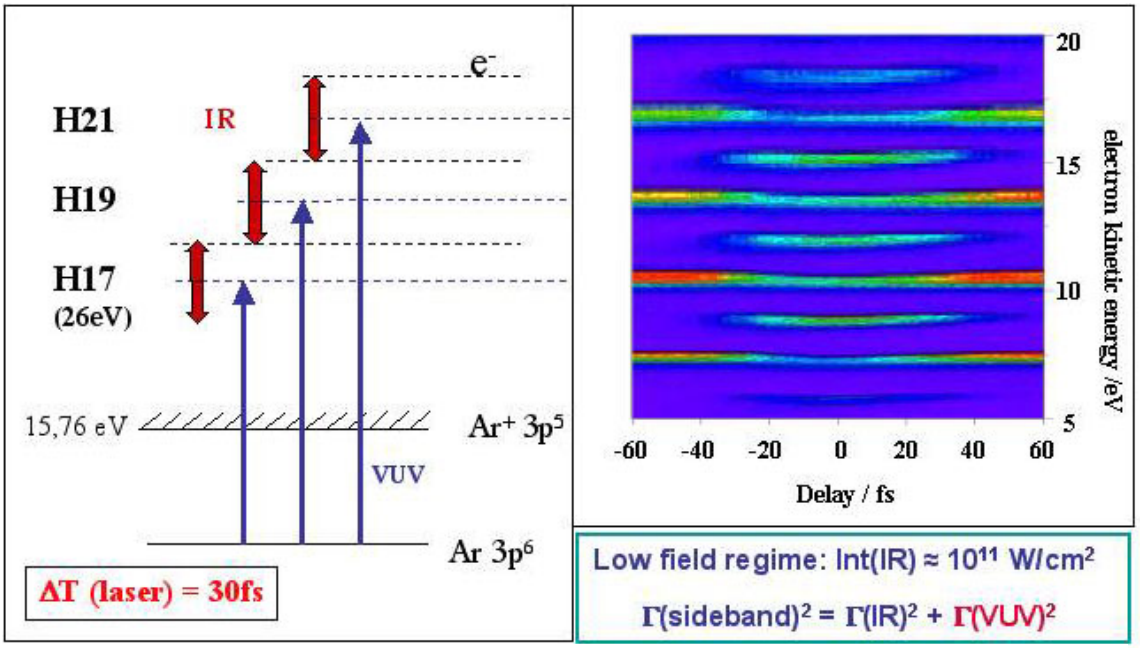


Figure 2: Schematic representation showing the photoionization of Ar atoms by VUV photons, which are produced through generation of higher harmonics of an intense IR laser, in the presence of the same IR laser in the interaction region (left-hand side). The resulting photoelectron spectra as a function of the time delay between the VUV and the IR pulses are given on the right-hand side of the figure.

which are due to an absorption or emission of an IR photon during the ionization process (Figure 2). The intensity, precise energy position and peak-shape of the electron lines strongly depend on the spatial and, more importantly, temporal overlap between the VUV and IR pulses as well as on the intensity of the dressing beam.

Due to the expected temporal jitter between the individual FEL pulses, the possibility to record single-shot spectra will be envisaged for the experiments at the TTF2-FEL. Simulations show (Figure 3) that that it is possible to determine the width and the relative position of the VUV pulses when the IR pulses are characterized providing a temporal resolution of better than 10fs. A precise analysis of the peak-shape allows in addition to determine the width of the VUV pulses. Since the IR pulses introduce a temporally non-uniform field the ionization conditions for different parts of the VUV pulses are also different, which results in an asymmetric, but characteristic lineshape. In this way the synchronization between fs-pulses can be analyzed and eventually also controlled.

In the experiments using the HHG source, spectra were recorded by averaging over 30 000 shots. This was mainly related to the intensity of the source providing about $10^6$-$10^7$ photons/pulse, which corresponds to about 3 electrons per pulse. With the projected FEL output of ~$10^{13}$ photons/pulse it can be estimated that cross correlation spectra are achievable with a single shot, if the IR laser can deliver ~0.5mJ. With the expected ~0.1mJ energy of the IR laser, the low field measurements yielding the phase shift will be possible at a 100 µm spot of the FEL. For measuring also the profile (10TW/cm$^2$) the experiment has to be performed with a 10µm diameter of the FEL in the interaction region. Problems might arise from space charge effect in the source volume when a huge amount of electrons are produce by the same FEL pulse. Simulations of the expected effects on the photoelectron spectra, in particular on the achievable kinetic energy resolution are under progress.

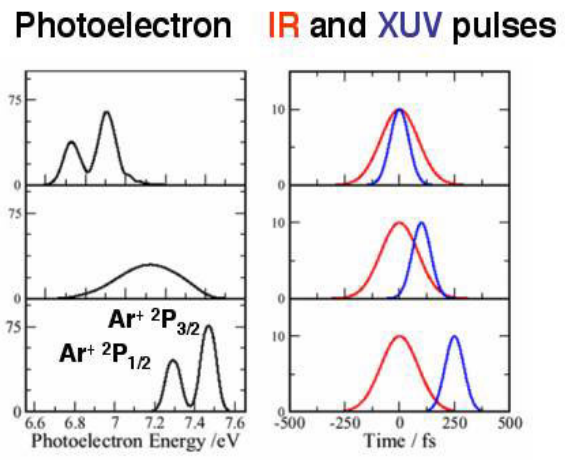


Figure 3: Simulation of the position and the shape of the Ar 3p photoelectron spectrum for different position of the VUV pulse with respect to the IR dressing pulse.

In general, the performed test experiments have demonstrated that the cross correlation measurements can be used to measure precisely the temporal parameters of a VUV pulse with respect to a well-characterized IR pulse.

## APPLICATIONS TO ATOMES

Depending on the final characteristics of the FEL with respect to photon energy range and tuneability, the pump-probe experiments will be extended to a large number of further experiments on atomic or molecular samples. To list just a few of them three examples are given:

(i) The investigations of resonances with same parity as the ground state, e.g. Xe* $4d^9 5s^2 5p^6 nd$, ms. Due to dipole selection rules, these states can only be excited either by a direct two-photon one-color excitation or by a two-photon two-color excitation via an intermediate resonant state, like the strong Xe* $4d^9$ 6p resonance ($h\nu$ (FEL) = 65.1eV). The short lifetime of the Xe* $4d^9$ 6p resonance ($\Gamma$=0.11eV, $\Delta$T=6fs) necessitates the combination and synchronization of fs-pulses (XUV + Visible). Resonant Auger spectroscopy will be used here to study the relaxation dynamics and additionally the influence of alignment of the intermediate state when the polarization of the two photon beams are changed in a controlled way. The results will extend and complete conventional studies using one-photon excitation and will provide a new and important basis for the theoretical treatment of electronic interaction and correlation in atomic multi-electron systems.

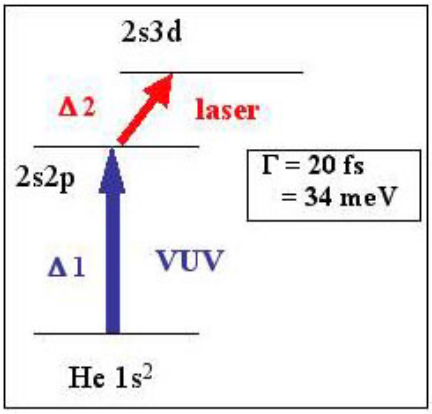


Figure 4: Excitation scheme for the coupling of autoionization resonances in He.

(ii) Coupling of autoionization states by a strong laser field. Similar to the above example, the FEL and a second synchronized fs-laser are used in a pump-probe arrangement to excite and to couple two autoionizing resonances, like He* 2s2p $^1$P and 2s3d $^1$D (at photon energies $h\nu$ (FEL) of about 60eV) (Figure 4). The profile of the resonances, being strongly asymmetric for He* 2s2p $^1$P [7], will strongly depend on the intensity of the two laser beams and on small changes in the relative time delay of the pulses and the photon energy of the second laser (detuning). Theoretical treatments [8,9] predict a pronounced and characteristic splitting in the resonance profile for laser intensities of 1GW/cm$^2$, which will be accessible in the planned experimental configuration.

## APPLICATIONS TO MOLECULES

(iii) Coherent population of excited states by fast dissociation: The photodissociation of $H_2$ or HCl in the photon energy region $h\nu$ (FEL)>17eV leads to the production of excited hydrogen atoms, in particular to the excited states with principal quantum number n = 2 or 3. Observation of the radiative decay of these states can separate the contributions from 2s and 2p as well as 3s, 3p and 3d levels by their different lifetimes (several nanoseconds). In a pump-probe experiment the FEL can be used to induce the dissociation process and the external laser to ionize the excited hydrogen atoms. The use of femtosecond pulses will allow the time resolution to be increased significantly and to observe quantum beats on the decay curve. The coherent population of the different fine-and hyperfine levels, which are energetically separated by 1 - 10 GHz, will induce a beat structure with a period of about 10 - 100ps (e.g. [10, 11]). For different resonant excitations different dissociation mechanisms are expected, which would lead to different, but characteristic quantum beats patterns of the decay curve.

## CONCLUSION

The given examples for experiments using the FEL pulses in combination with a synchronized optical fs-laser are part of working program, which has been proposed by a European collaboration including experimental groups at HASYLAB (Germany), Lund Laser Center and MAX-Lab (Sweden), Dublin City University (Ireland), Max-Born Institute for Nonlinear Optics and Short-Pulse Spectroscopy (Germany), and LURE (France). First experiments are schedules for spring 2005.

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