Nonlinear Attosecond Spectroscopy

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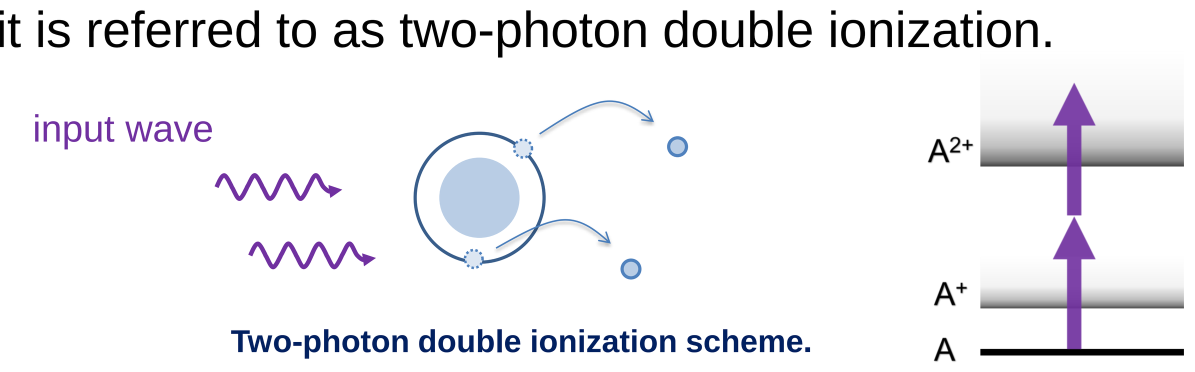
Abstract

In the project NONLINEAR-ATTO we are developing a system for the investigation of correlated electronic dynamics by nonlinear attosecond extreme ultraviolet (XUV) spectroscopy.
Simple quantum systems will be investigated in the frame of two-photon double ionization scheme giving insight into electron-electron correlation by performing XUV-pump XUV-probe measurements with attosecond time resolution.

Nonlinear XUV Optics

When we irradiate an atomic/molecular system with intense light pulses the medium's response becomes nonlinear. Thus, it may absorb two photons and emit a new photon with double frequency. When this absorption leads to ionization it is referred to as two-photon double ionization.



Two-photon double ionization scheme.

Multiphoton ionization *was first proposed by Maria Göppert Mayer, Nobel laureate in 1963, with her dissertation in the University of Göttingen in 1931, focused on 'two quanta jumps'.*

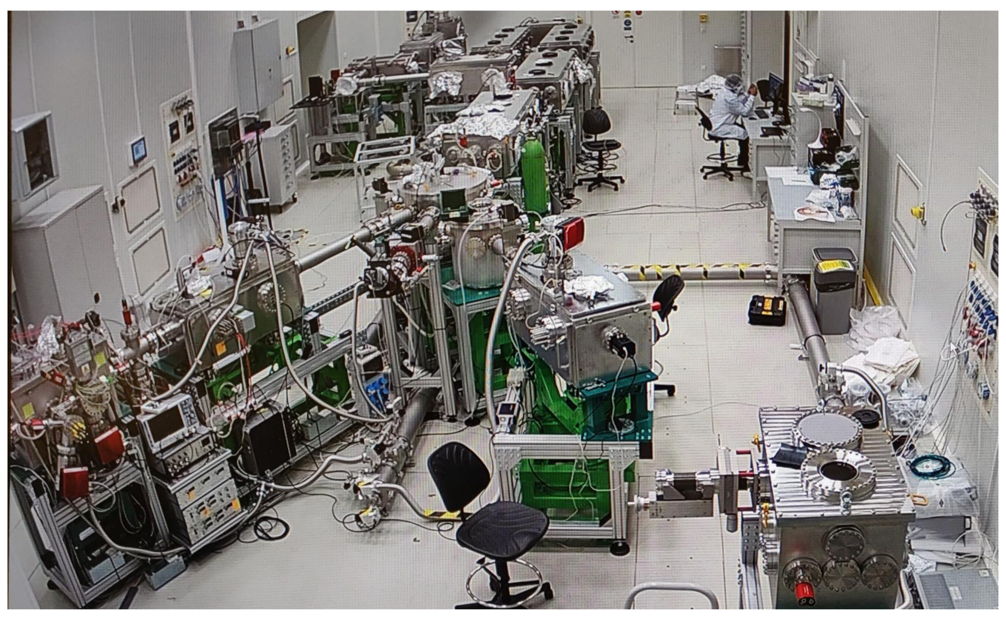
Ann. Phys., Lpz. B 9 273 (1931) *"Über Elementarakten mit zwei Quantensprünge ".*

Experimental Requirements

- Non-linear effects → induced by high intensities
- Timescale of electronic dynamics → attosecond pulses (1 as = 10^{-18} s)
- Coincidence spectroscopy → requires high repetition rates

Experiments will be performed in **ELI ALPS, Hungary** [3] in the beamline driven by the Single Cycle (SYLOS) Laser system.

- Intensities: 10^{15} -10^{16} W/cm^2
- Pulse duration: < 500 as
- Repetition rate: 1 kHz



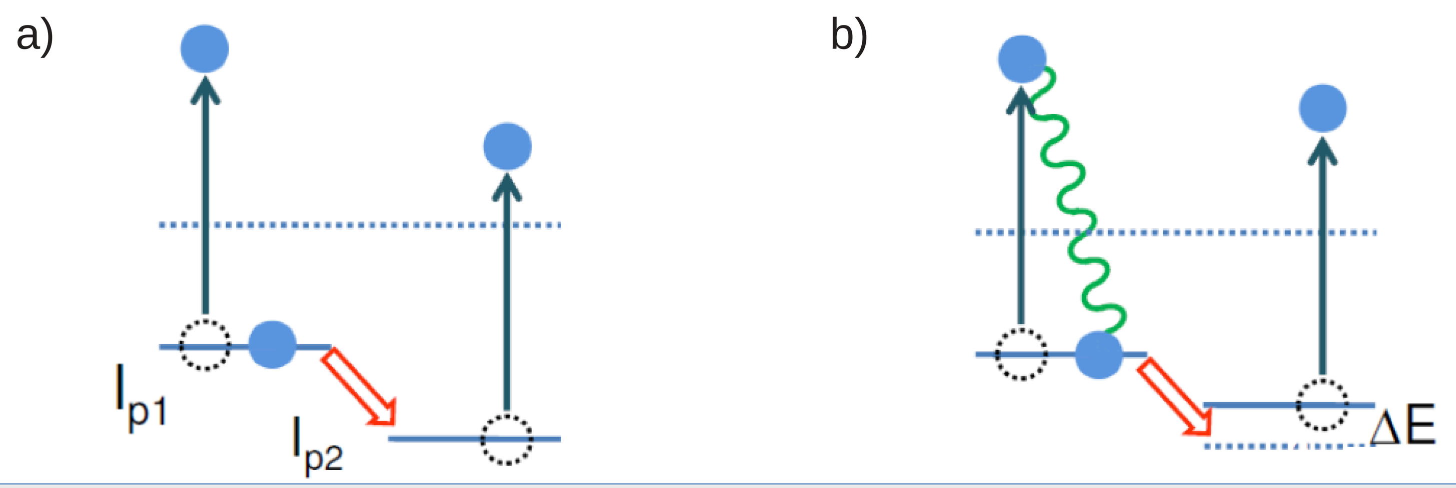
SYLOS-driven ELI ALPS beamline.

Investigating Electronic Dynamics in He

We choose a simple atomic system like **He** and we irradiate it with **XUV** light of higher photon energy than both the 1st and the 2nd ionization potential. Two possible ionization pathways would be the following [1]:

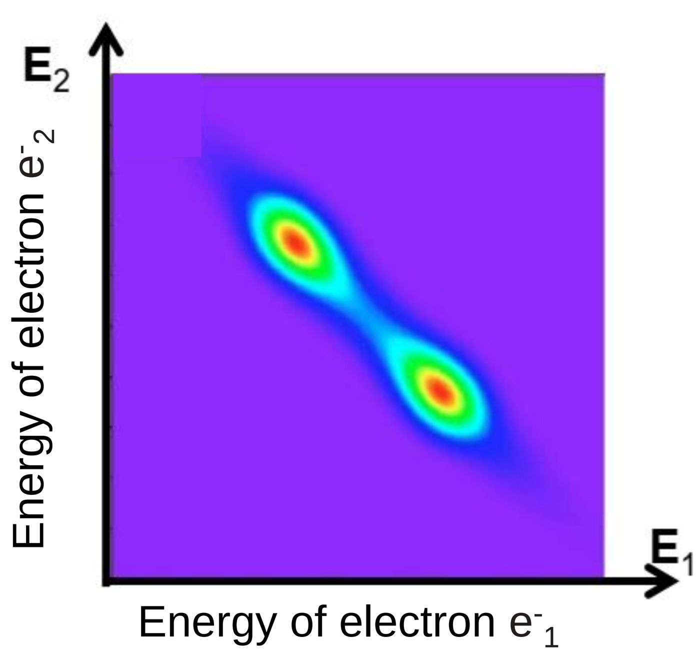
$$\mathrm{He} + \hbar\omega \rightarrow \mathrm{He}^{+}(1s) + e_1^- \,, \quad \mathrm{He}^{+}(1s) + \hbar\omega \rightarrow \mathrm{He}^{2+} + e_2^- \,,$$

$$\mathrm{He} + \hbar\omega \rightarrow \mathrm{He}^{+}(nl) + e_1^- \,, \quad \mathrm{He}^{+}(nl) + \hbar\omega \rightarrow \mathrm{He}^{2+} + e_2^- \,.$$



a) A 1st photon ejects one electron (e^-_1), leaving the ion in the ground state **He⁺**. At a later time, a 2nd photon is absorbed by **He⁺** leading to the emission of a second electron (e^-_2).
b) Here, electron correlation plays a role, as the first electron leaving the atom exchanges energy promoting the second electron to an excited state, which is subsequently ionized by a 2nd photon. [1]

Electron energy distribution.[2]

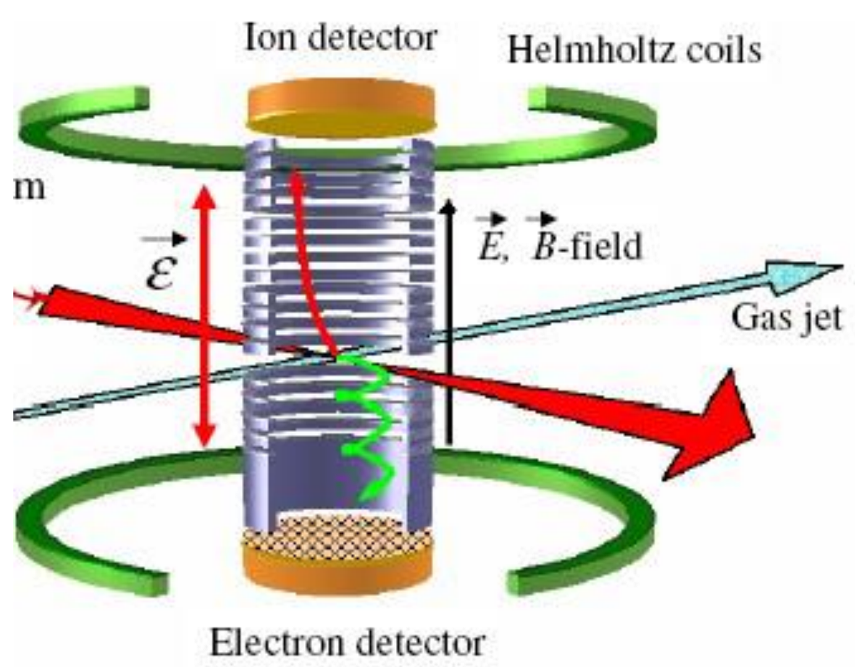


Distribution of the acquired energy for the two emitted electrons:

- When both electrons present similar kinetic energy, electron correlation has to be taken into account since it cannot be explained in the framework of an independent particle approach.
- Heisenberg uncertainty → this correlation is oscillating on an attosecond timescale:

$$\Delta t = \frac{\hbar}{I_{p2} - I_{p1}} = 22\ as$$

Reaction Microscope (REMI)



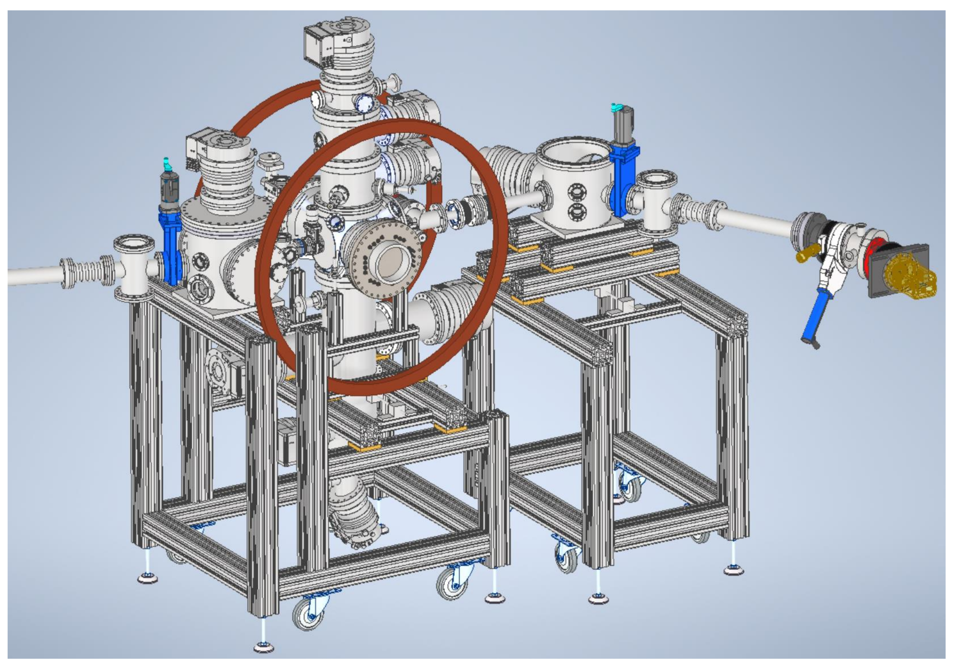
Reaction Microscope.

REMI is a momentum spectrometer [4]
→ 3D momentum distributions of ions and electrons in coincidence

- Ionization of atoms/molecules by intense XUV pulses
- Acceleration of electrons/ions by Electric field
- Confine electrons to spectrometer axis by Magnetic field
- Time of flight & impact position → **p**(x,y,z)

Upcoming Tasks

- Mounting/installing the setup
- Characterization of the split & delay unit for pump-probe experiments
- Setting up of an interlock system
- Acquisition software
- Testing the setup in Freiburg
- Experimental measurements in ELI ALPS, Szeged
- Data analysis



Sketch of the End station.

Acknowledgments

Funding from the Bundesministerium für Bildung und Forschung (Project 05K19VF1) is gratefully acknowledged.

Bundesministerium für Bildung und Forschung

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