Nonlinear Attosecond Spectroscopy

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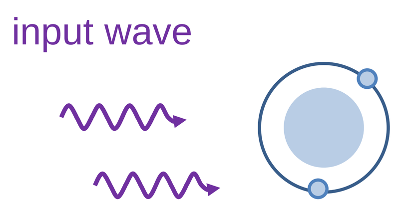
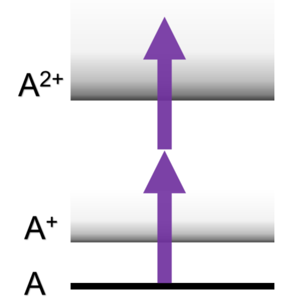
Abstract

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 by two-photon double ionization giving insight to electron-electron correlation and by performing XUV-XUV pump-probe experiments 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.





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'.*

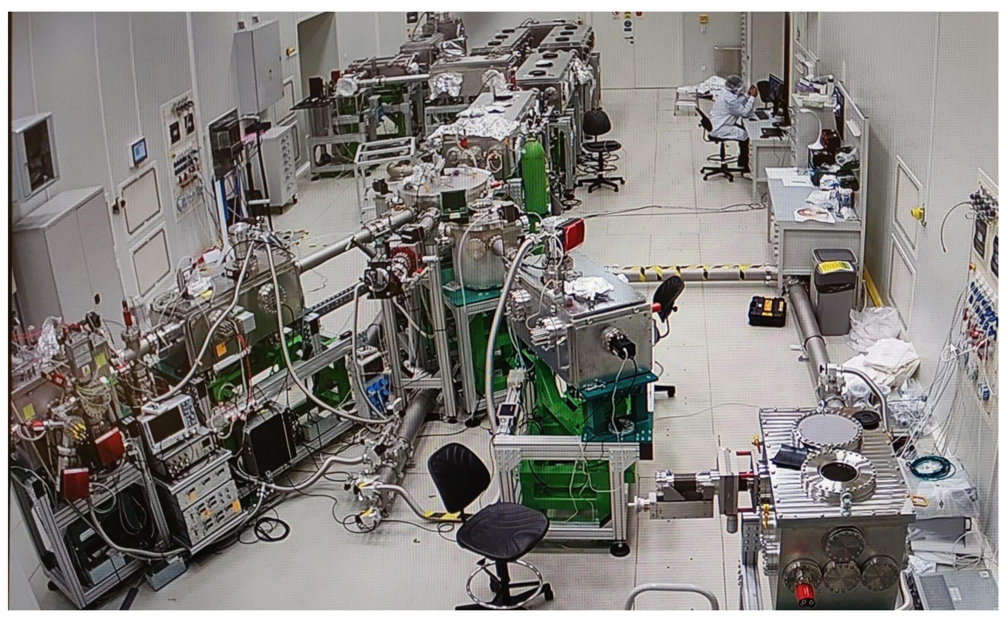
Ann. Phys., Lpz. B 9 273 (1931) "Über Elementarakten mit zwei Quantensprünge ".

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 held in **ELI ALPS, Hungary** [3] utilizing SYLOS laser system

- Intensities: ≤ 10^{17} W/cm^2
- Pulse duration: < 500 as
- Repetition rate: 1 kHz



ELI- ALPS beamline

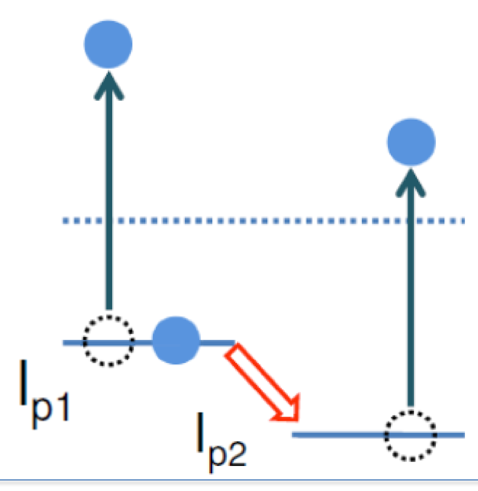
Investigating Electronic Dynamics

We choose a simple atomic system like He and we irradiate with **XUV** photons which acquire higher energy than the ionization potential. Let us have a closer look to the ionization pathways [1]:

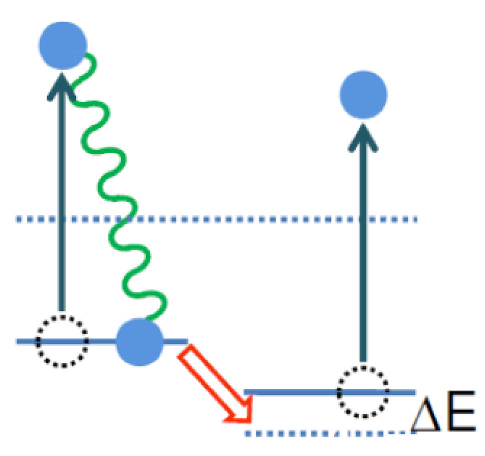
$$\begin{aligned} \mathrm{He} + \hbar\omega &\rightarrow \mathrm{He}^+(1s) + e_1^- , \\ \mathrm{He}^+(1s) + \hbar\omega &\rightarrow \mathrm{He}^{2+} + e_2^- , \end{aligned}$$

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

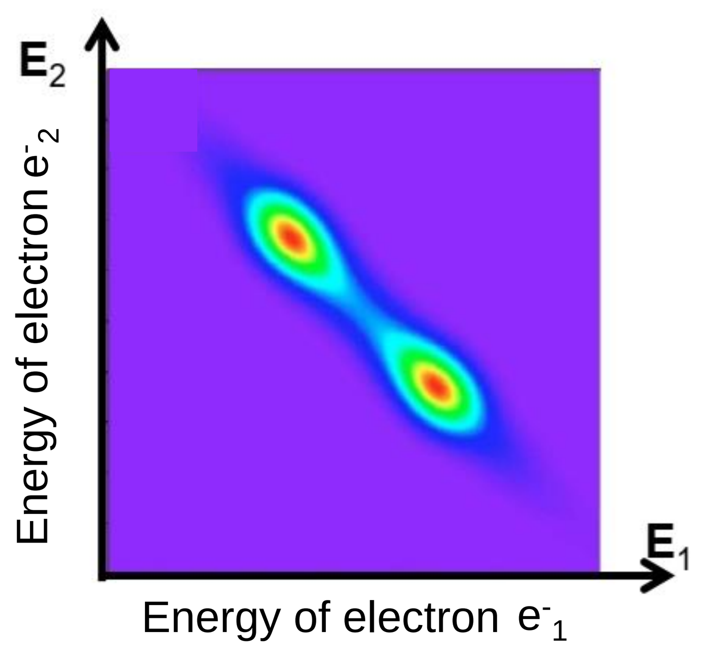
a)



b)



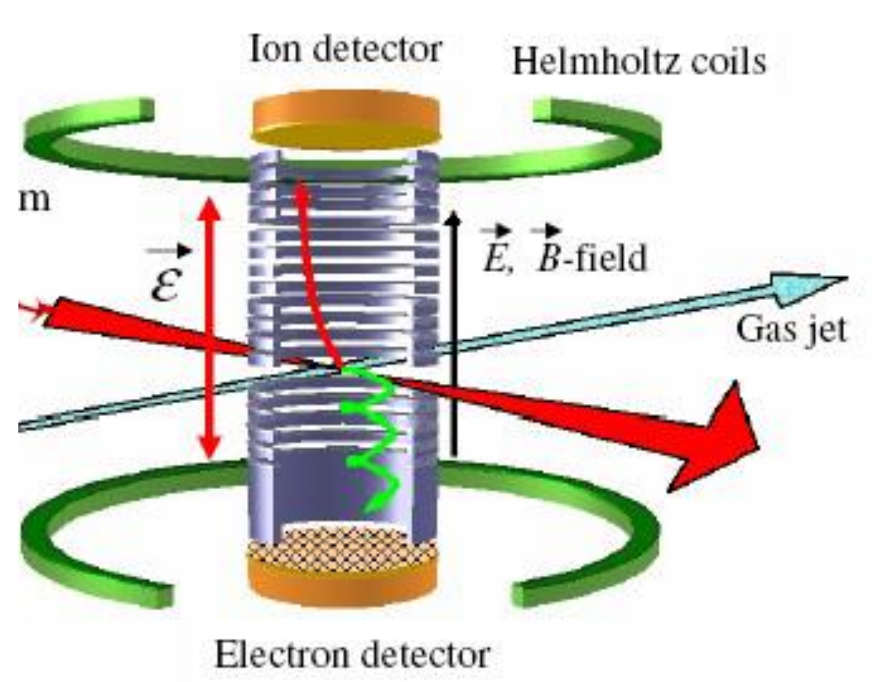
a) A 1st photon ejects one electron (e^-_1), leaving the ion in the ground state **He⁺**. At a later time, a 2nd photon is absorbed by **He⁺** leading to a the emission of a second electron (e^-_2).
b) Here, electron correlation plays a role as the first electron leaving the atom can exchange energy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



The plot depicts the acquired energy for the two emitted electrons [2]. When both electrons present similar energy electron correlation has also to be taken into account.
Considering the Heisenberg uncertainty, this correlation is oscillating on an attosecond timescale:

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

Reaction Microscope



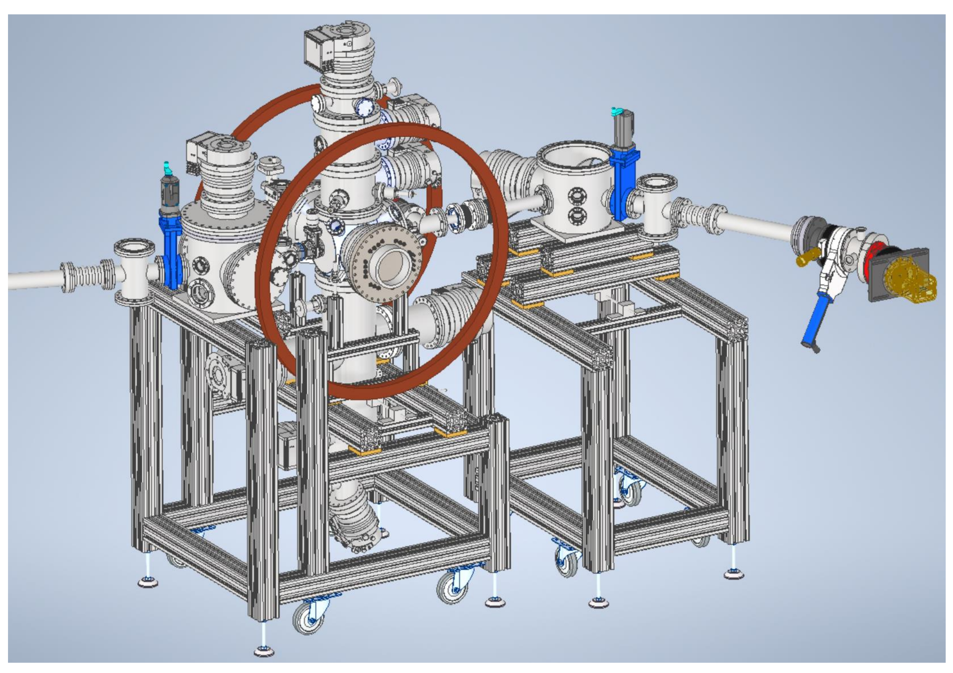
Reaction Microscope

3D coincidence spectroscopy performed by means of the reaction microscope [4]

- Ionization of atoms/molecules by intense XUV pulses
- Acceleration of electrons/ions by E-field
- Guidance of electrons by B-field
- Impact position → $p_{x/y}$
- Time of flight → p_z

Upcoming Tasks

- Mounting the setup
- Split-delay unit for pump-probe experiments → characterization
- Setting up of an interlock system
- Acquisition software
- Measurements (testing the setup in Freiburg, Experimental measurements in ELI ALPS, Szeged)
- Data analysis



Sketch of the end station

Acknowledgments

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