- **High field atomic physics**
  Interaction of high-intensity, short-duration laser pulses with atoms at and beyond the limit of perturbation theory.

Determination of multiphoton ionization cross sections

This activity relates to the quantitative investigation of multophoton processes. Quantification of MPI reefers to the determination of ionization saturation intensities and generalized multiphoton ionization cross sections, as defined below.

In the multiphoton regime the temporal evolution of the ionization probability $P(t)$ according to the lowest order perturbation theory (LOPT) obeys the relation

$$\frac{dP(t)}{dt} = (1 - P(t))\sigma^{(n)} \left(\frac{I(t)}{\hbar\omega} \right)^n$$

$I(t)=I_0F(t)$ being the ionizing laser intensity (I_0 the intensity maximum and $F(t)$ the pulse temporal profile with unity maximum) n the number of absorbed photons and $\sigma^{(n)}$ the n-photon generalized ionization cross section. Thus the probability to find the atom ionized when the laser pulse is gone reads

$$P(t \to \infty) = 1 - \exp\left[-\sigma^{(n)} \int_{-\infty}^{\infty} \left(\frac{I(t')}{\hbar\omega} \right)^n dt' \right]$$

Far from saturation of ionization (exponent<<1) expansion of the above gives

$$\ln P(t \to \infty) = \ln \sigma^{(n)} + \ln\left[-\int_{-\infty}^{\infty} \left(\frac{I(t')}{\hbar\omega} \right)^n dt' \right]$$

Upon saturation of ionization the probability becomes unity and thus

$$\sigma^{(n)} = \frac{\hbar\omega}{I_{SAT}} \left[\int_{-\infty}^{\infty} \left(F(t') \right)^n dt' \right]^{-1}$$

where I_{SAT} is the so called saturation intensity and $I(t)=I_{\mathrm{SAT}}F(t)$.

Multihpoton processes have been for many years investigated qualitatively. These studies have lead to the discovery of a number of new effects. Quantification though becomes important in order these processes to be utilized in applications such as in elemental analysis or other types of diagnostics. The work on quantitative MPI studies at FORTH-IESL, conducted with 0.5ps pulses at 248nm (KrF amplified DFDL laser system), includes the following two approaches.

- Defeating volume effects in MPI

One of the major problems in High field physics originates from the need of using focused laser beams in order to achieve the required high intensities. As a consequence there is a 3D laser intensity distribution at focus, which due to the non-linear laser intensity dependence of processes observed, does not allow quantification of the results unless the laser intensity distribution is stable and very well known.

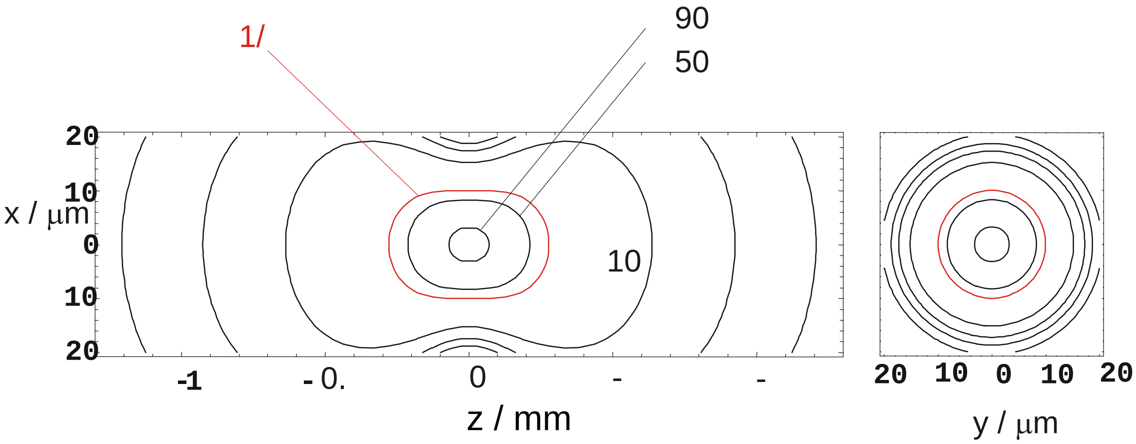

Fig. 1 Spatial intensity distribution of a Gaussian laser beam at focus

Experimentally the volume effect problem can be defeated by the technique of a four (or multiple) grid reflectron TOF mass spectrometer. The technique has been developed in the group of Dr. H. Schröder at MPQ, who in collaboration with our group and Dr. B. Witzel (now at Univ. of Laval) has installed such an apparatus in the laser and applications lab of FORTH-IESL to be used with the 500fs KrF laser beam. The principle of the apparatus is that it selects ions from a strongly confined volume of the interaction region. Within this volume the laser intensity is practically spatially constant and thus free of volume effects. The confinement occurs in two dimensions through a small spectrometer aperture and in the third dimension through a specific electrostatic ion mirror (four grid reflectron).
Using this apparatus saturation intensities and absorption cross-sections at 248nm and 500fs have been determined for a number of elements of the periodic system, as well as effects related to quantitative elemental analysis have been investigated [see e.g. B. Witzel et al. *Phys. Rev.* A **58**, 3836 (1998); B. Witzel et al. *Phys. Rev.* A **60**, 3311 (1999), E. Varoucha et al. *Phys. Rev.* A **65**, 012901 (2002) and references therein].

- *Determination of MPI generalized cross sections from ponteromotive shifts*

During the process of photoionization the absorbed photon energy is shared between the processes of a) the ejection of the electron in the continuum and b) the quiver motion of the free electron in the field. While at low intensities the energy needed for the second process is negligible (it is proportional to the laser intensity and inversely proportional to the square of the photon energy), at high intensity ionization may become substantial. This effect causes an effective shift of the ionization potential towards higher energy and thus a shift of the photoelectron energy towards smaller values. This shift is known as ponteromotive shift given by

$$U_P = \frac{(eE_0)^2}{4m\omega^2}$$

We have developed a quantitative method through which the generalized cross section of a non resonant MPI process can be determined from the detailed shape of the photoelectron spectra (ATI). This shape is caused by the varying ponderomotive shift due to the temporal intensity distribution of the laser pulse $I(t)=I_0F(t)$. The method is described in C.J.G.J. Uiterwaal et al. *Phys. Rev.* A **57**, 392 (1998).

Testing of models though correlated electron-ion measurements.

The project targeting the study of correlated systems in strong fields was focused on the study of multiple ionization of many electron atomic systems. The problem of multiple ionization has been a central problem in multiphoton processes almost since the field was born. The question raises was whether a double (or multiple) ionization occurs by sequentially ionizing the system (i.e. the atom is first singly ionized and the produced ion is further ionized) or two electrons can be simultaneously ejected leading to as so called "direct" double ionization. For a while sequential double ionization seemed to be the dominant double ionization channel. The observation of the "direct" double ionization became a challenge and this is because a) from the physical point the problem is a three body problem under strong field conditions and b) from the practical point of view direct double or multiple electron excitation high up in the continuum is identical with putting a large amount of energy, which was hoped to be gained back in the form of short wavelength coherent radiation and eventually as x-ray laser radiation.

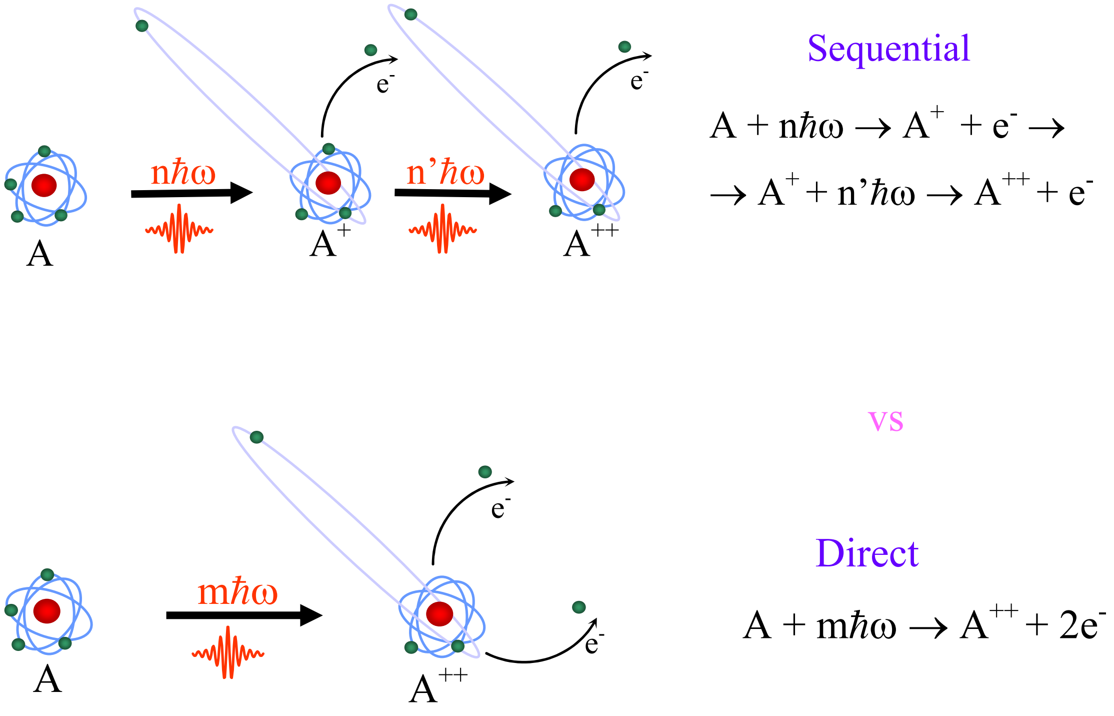

Fig. 3 Schematic illustration of the sequential and direct double ionization processes.

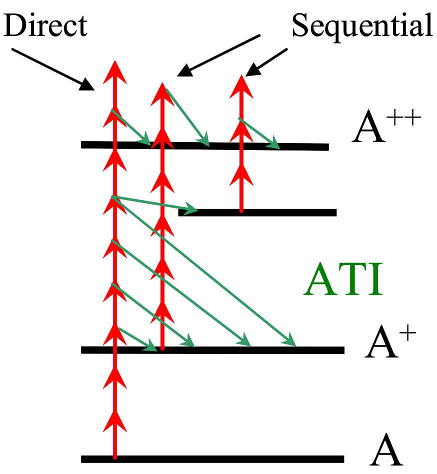

Fig. 4 Sequential and direct double ionization processes in the Multiphoton Ionization regime

For many years the investigations in this field were targeting simply trends because of the large variety of new effects that could be discovered. However, the field has reached some years ago a high degree of maturity so that sophisticated measurement techniques became necessary for a detailed investigation of the processes under consideration. In year 1994 a non perturbative laser intensity dependence of the double ionization yield of He atoms when interacting with *fs* laser pulses of a Ti:Sapph laser system (780 nm) has been observed by Walker et al. [PRL **73**, 1227 (1994)]. This enhanced double ionization, attributed to a non sequential process, has attracted the attention and efforts of many scientist of the field. More general the correlated multiple electron excitation or ejected became a field of enhanced investigations. The information gained from experiments in this type of studies was conventionally based on ion yield vs laser intensity measurements or on energy (and/or angularly) resolved photoelectron spectra [see e.g. D. Xenakis, et al. *Phys. Rev.* A **60**, 3916 (1999)]. This type of experiments provide some insight about the ionization channels involved, however, more complete information can be gained through correlated ion charge state – energy/ angularly resolved electron measurements. This additional information becomes necessary in particular in order to test the validity of the different existing theoretical models (e.g. sequential, shake off, rescattering) of multiple ionization.

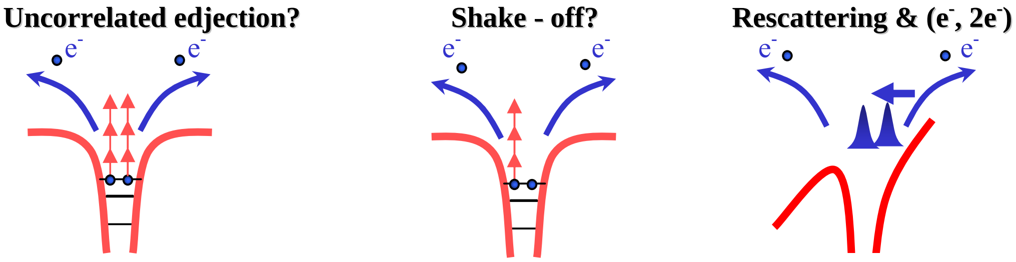

Fig. 5 Schematic illustration of double ionization models

In our group has been demonstrated the first high energy resolution electron-ion coincidence experiments in the high field regime [B. Witzel et al. *Phys. Rev. Lett* **85**, 2268 (2000)]. Improvement of the statistics of the experiment will allow the measurement of detailed structures in the multiple ionization spectra. Furthermore the application of the method for the investigation of correlated molecular photo-fragments is planed. The project was in collaboration with the Univ. of Freiburg (Dr. B. Witzel) were complementary studies of electron – ion correlations with angular information have been performed

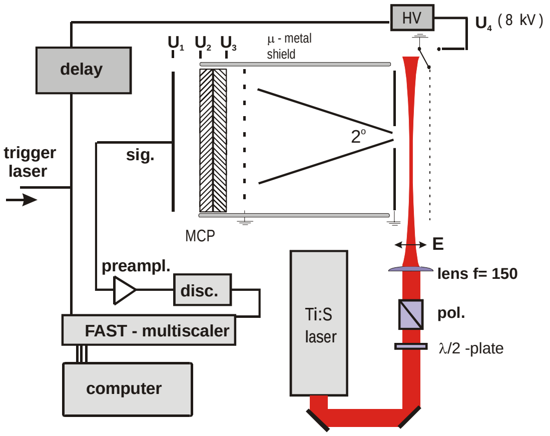

Fig. 6a Ion-electron coincidence apparatus. The same detector is used for both particles. After the electrons have reached the detector a pulsed extracting high voltage initiates the ion Time of Flight.

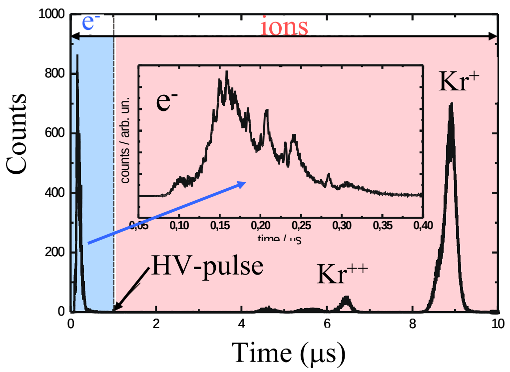

Fig. 6b The TOF spectra measured with this apparatus thus consist of an energy resolved electron spectrum appearing in the beginning of the spectrum followed by the ion spectrum