Electron Spectroscopy on ion beams

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Outline

- Photoionization of ions
- Interest
- Difficulties
- Presentday experiments: examples
- What will be possible and how could it be done?

Photoionization of ions ($h\nu$ range)

Single or multiple photoionization of ions:

Singly charged positive Ions: $h\nu > \approx 20$eV ($Na^+ \approx 47$eV; $Ca^+ \approx 12$eV)

$X^+ + h\nu \rightarrow X^{++} + e^-$ or $X^{(m+1)+} + me^-$ (multiple ionisation: Auger decay…)

(detection of ions OR electrons)

Multiply charged ions :($Ne^{++} \approx 63$eV, $Sc^{++} \approx 25$eV, $Ca^{++} \approx 50$eV, $Be^{++} \approx 153$eV)

$X^{n+} + h\nu \rightarrow X^{(n+m)+} + me^-$

Need for VUV, XUV light sources (synchrotron,…VUV, XUV lasers)

Electron correlations become **smaller** when charge increases.

Negative ions: very strong electron correlations in ground and excited states.

Hartee-Fock approximation not good enough to give electron affinity.

Only **one bound state** but **resonances** (doubly excited states) exist (decay by autodetachment)

Photodetachement of atomic negative ions: $h\nu > 0$ to 3.6 eV (for Cl^-) ($H^- \approx 0.75$ eV) (IR or visible lasers)

Photon energy for resonance excitation: $h\nu \approx 10$eV; higher for inner-shell excitation

$A^- + h\nu \rightarrow A^{-**}$ (resonance) $\rightarrow A^0 + e^-$ (detection of electrons or neutrals)

$\rightarrow A^0 + e^-$ (direct detachment)

$\rightarrow A^+ + 2e^-$ (double detachment, detection of positive ions)

Interest

<u>Plasma modeling:</u>

OPACITY and IRON projects: need for absolute photionization cross-sections
Low density astrophysical plasmas and also fusion plasmas.
Need to compare with experimental absolute cross sections for singly (atomic and molecular) and multiply charged positive ions but more information is obtained from differential cross sections $\Rightarrow$ electron spectroscopy .

Negative ions:
Example: search for H^- 2s2p $^1P^o$ shape resonance in stellar absorption at 11eV but never observed (?)
General interest for the study of electron correlations, decay by electron emission $\Rightarrow$ electron spectroscopy

Electron spectroscopy is the only way to follow the evolution of excited states: **branching ratios, Auger decays, partial cross sections, contribution of different sub-shells…**

Why is it difficult?

Ion beam density: typically less than **10^6 ions/cm^3**
(Example: H^- : 1keV, 1nA, S=1mm^2 $\Rightarrow$ N $\approx$ 1.5x10^4 ions/cm^3)
corresponds to a neutral density in vacuum less than **10^{-10} torr** (ultra-high vacuum)
Increasing ion beam intensity induces space-charge effects that modify electron energies (I >> 1 μA)
Up to now only two electron spectroscopy experiments have done on positive ion beams (Bizau *et al*, PRL 67, 576 (1991); Al Moussalami *et al*, PRL 76,4496 (1996) on Ca^+ giant resonance $\sigma\approx$2200 Mb; Gottwald *et al*, PRL 82, 2068 (1999) on Xe^+)
Experiments on photoionization of ion beams have been mostly devoted to absolute cross section measurements performed by ion spectroscopy.
Photon flux is typically **10^{12-13} ph/s** (undulator) (i.e. μW power)
To increase the interaction region $\Rightarrow$ merged ion and photon (SR) beams (~50 cm), ultra-high vacuum (~10^{-10} torr), detection of ions.

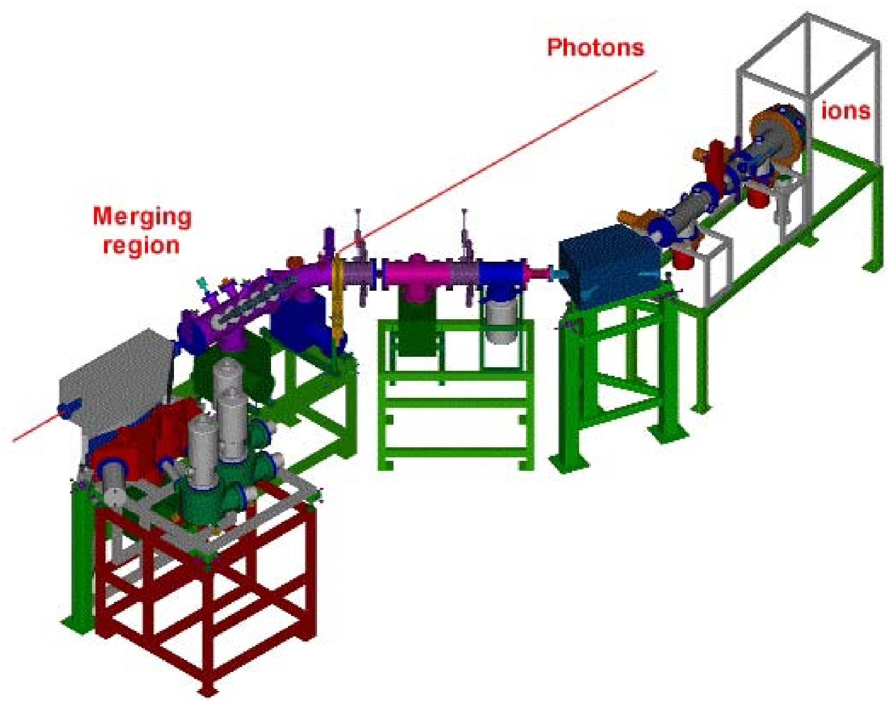
LURE (J-M Bizau); ASTRID (DK); ALS (USA); SPRING8 (JAPAN)….

- Others problems:
- Ionization of background gas gives electron signal (noise!) (not ion signal), collisions of ions on the background gas gives also electron signal (and ion signal: more critical for negative ions since collision detachment cross section are $\sim 10^{-15}$ cm^2 while photodetachment cross sections are $\sim 10^{-18}$ cm^2)
- $\Rightarrow$ extreme high vacuum **<10^{-12} torr** (cryogenic cooling for instance on ELISA electrostatic storage ring for ions Aarhus)

- With new photon sources:
- Gain in photon flux (>>10^{15}ph/s) will allow crossed beam experiments better adapted to electron spectroscopy specially with “complete” experiments.
- The pulsed structure is well adapted to time-of-flight electron spectroscopy

Merged beam experiments

- Exemple in ASTRID: measurement of absolute ionization cross-sections
- http://www.ifa.au.dk/amo/atomphys/atomphys.htm
- He^{+}, Li^{-}, C^{+}, N^{+}, O^{+}, Mg^{+}, Al^{+}, Al^{++}, S^{+}, K^{+}, Ca^{+}, Ca^{++}, Cr^{+}, Fe^{+}, CO^{+}, I^{-}, Cs^{+}, Ba^{+}, Ba^{++}, …etc



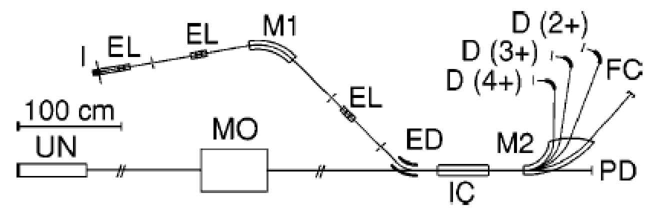


FIG. 1. Schematic drawing of the experimental setup. I, ion source; EI, Einzel lenses; M1 and M2, deflection magnets; ED, electrostatic deflector; IC, interaction chamber; D, detectors for 2+(3+), 3+(4+), and 4+ ions from photoionization of 1+(2+) ions; FC, Faraday cup for 1+(2+) ions; UN, undulator (not in scale); MC, Miyake monochromator (not in scale); PD, calibrated Al photodiode.

Experimental set-up

Some examples: Molecular ion: CO^+, Multicharged ions: Xe^{n+}

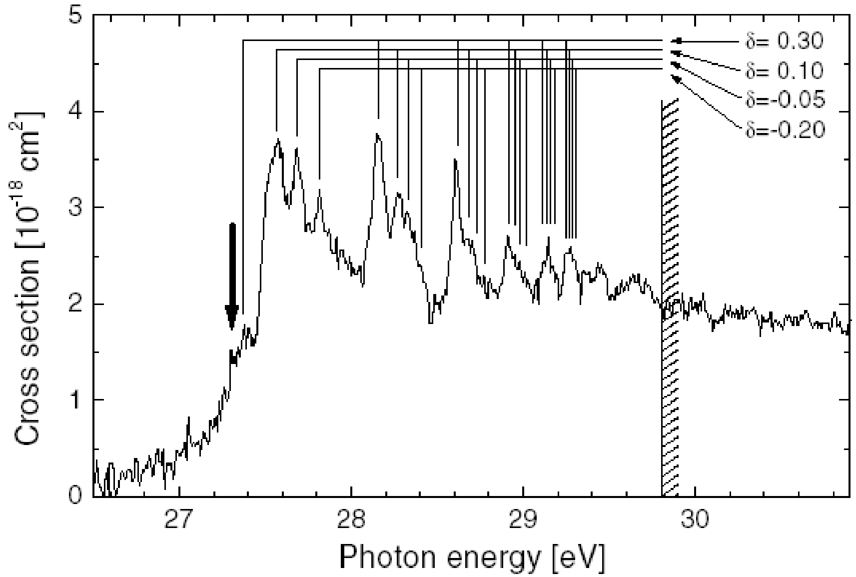


Figure 2. Details of the threshold region. The arrow points to the threshold for CO^{2+} ion production at 27.31 eV; the series limit for the Rydberg states is indicated at 29.80 eV.

T. Andersen, *et al*, 2001 J. Phys. B **34** L327

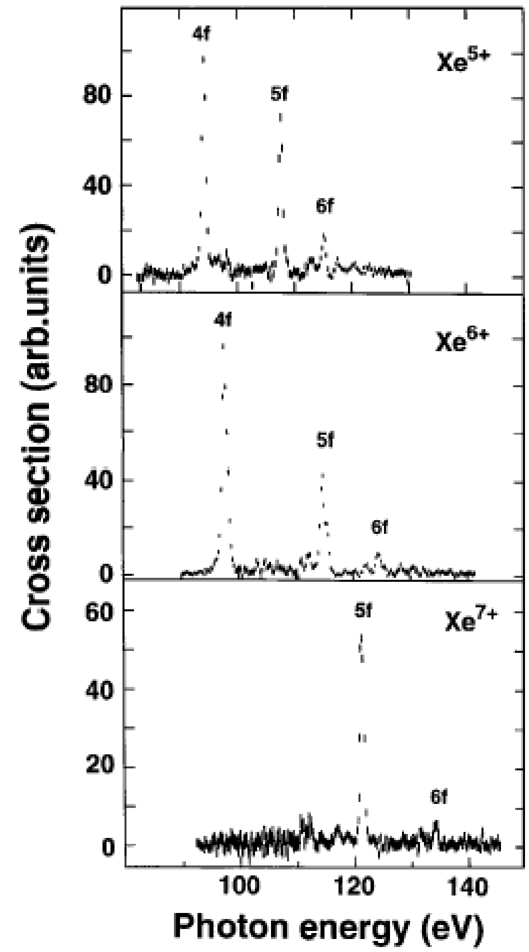


FIG. 2. Experimental single photoionization spectra of Xe^{5+}, Xe^{6+}, and Xe^{7+} measured below the 4d-ionization thresholds. Autoionization lines following $4d \rightarrow nf$ excitations dominate the spectrum. Weaker lines are due to the decay of excited states formed by $4d \rightarrow np$ $(n > 5)$ excitations.

Bizau et al, Phys. Rev.Lett. 84, 435 (2000)

Negative ions

Synchrotron experiments: example Li^- (ASTRID and ALS)

$Li^- + h\nu \rightarrow Li^+$ (signal to noise ~ 1/10)

H. Kjeldsen, et al 2001 J. Phys. B 34 L353

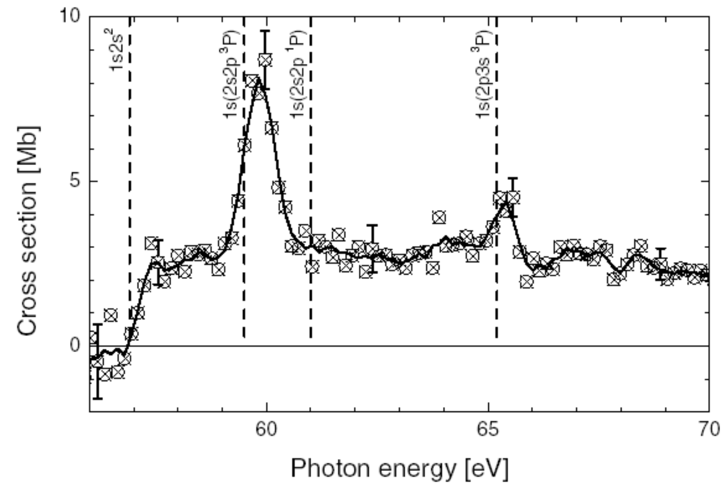


Figure 1. Absolute cross section for photodetachment of Li^- leading to Li^+ formation. Various core-excited states of neutral Li are indicated by vertical dashed lines. Circles: experimental data points; full curve: smoothed experimental spectrum (average of 3 adjacent points). Statistical error bars are shown for selected points. The systematic uncertainty is 20%.

Discrepancy between theory and experiment: explained by electron recapture by Li+ ⇒ neutral Li formed, need for electron spectroscopy

Gorczyca et al Phys. Rev. A **68**, 050703 (2003)

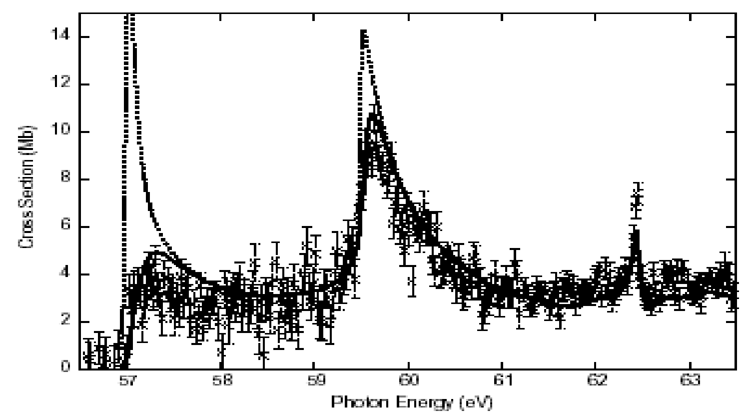


FIG. 2. Photodetachment of Li^- yielding Li^+ ions. The experimental results from Ref. [5] (crosses, shifted by −0.165 eV) are compared to the standard R-matrix results (dotted line) and the optical potential results (solid line).

Experiments on H^- in LAMPF

(800MeV beam, crossed with YAG laser, Doppler energy tuning)

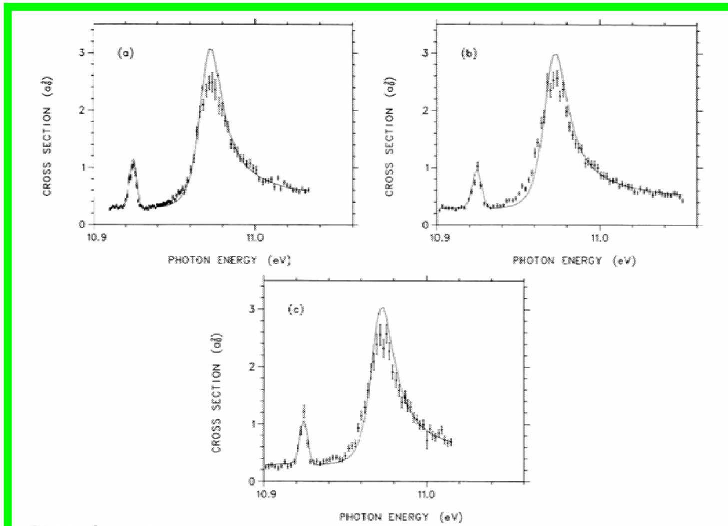


FIG. 5. Fits of Broad and Reinhardt's calculation (Ref. 30) to three runs over the zero-field resonance region. Experimental resolution has been folded into the theoretical curve (solid line). See Table III. Three independent runs are shown so that run to run variations in data and fitted parameters can be seen.

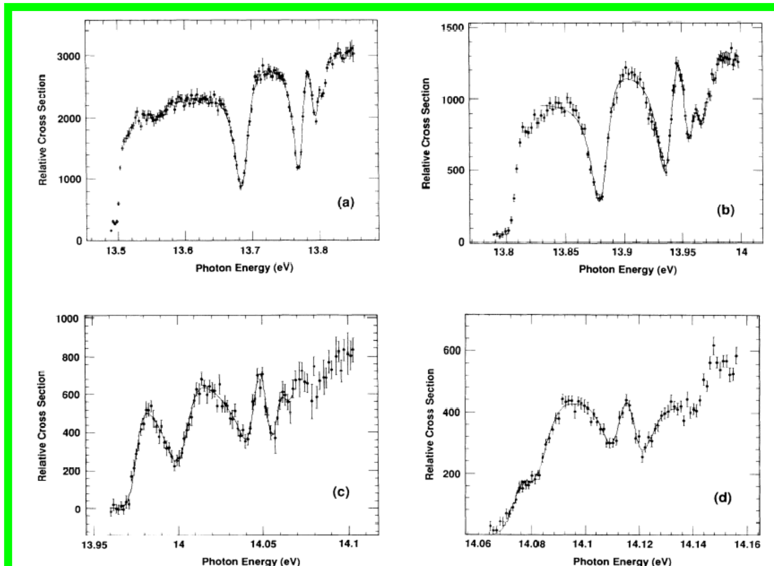


FIG. 8. Partial photodetachment cross sections of H^-, showing production of neutral hydrogen in (a) $n \geq 4$ ($\chi^2/\nu = 0.97$, CL is 55%); (b) $n \geq 5$ ($\chi^2/\nu = 1.85$, CL is 0.01%); (c) $n \geq 6$ ($\chi^2/\nu = 0.88$, CL is 73%); and (d) $n \geq 7$ ($\chi^2/\nu = 0.98$, CL is 50%). Instrumental resolution is 8.3 meV. Threshold energies as follows: $n = 4$, 13.5054 eV; $n = 5$, 13.8084 eV; $n = 6$, 13.9746 eV; $n = 7$, 14.0748 eV; $n = 8$, 14.1398 eV. The solid lines are fits to sets of Fano profiles (see text).

Observation of H^- 2s2p $^1P^o$ Feshbach and shape resonances around H(n=2)

Bryant et al, Phys. Rev. A 27, 2889–2912 (1983)

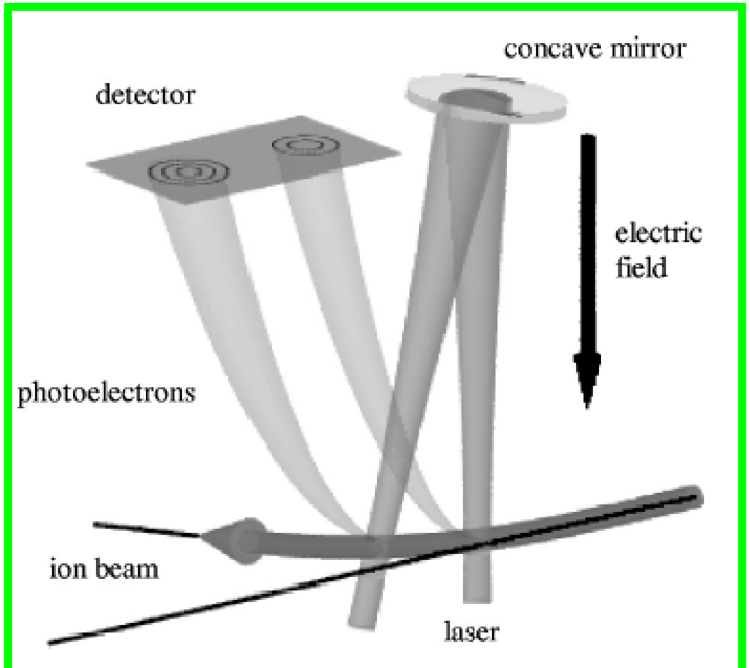
H^- resonances up to H(n=9)

P.G. Harris *et al.*, Phys. Rev. A **42**, 6443 (1990).

The branching ratio for the decay of resonances are mainly unknown: could be measured by electron spectroscopy

Photodetachment spectroscopy of negative ions

Photoelectron microscope



Blondel et al, Phys. Rev. A **64**, 052504 (2001)

Limited to the threshold region:
measurement of electron affinity

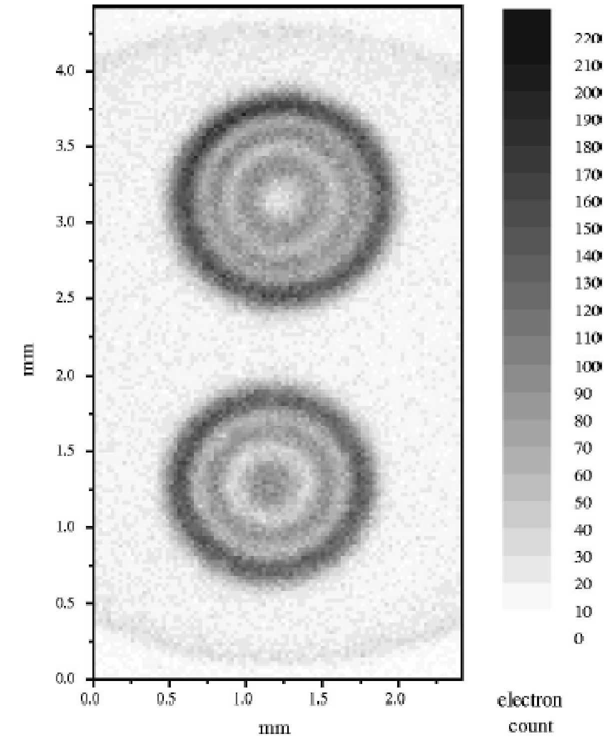


FIG. 2. A photodetachment double spot from $^{16}O^-$, in a field $F \simeq 536$ V m^{-1}. Detection takes place 0.514 m away from the laser-ion crossings. The ion kinetic energy is 500 eV. There is of course only one laser frequency, but the Doppler shift is larger for the incoming laser beam (top spot) than for the reflected laser beam (bottom spot), which results in initial electron kinetic energies of 0.980 and 0.863 cm^{-1}, respectively, and the observed differences in the ring patterns. The electron affinity deduced from this very pair of spots is 11 784.679(1) cm^{-1}, which shows the accuracy of the fitting procedure. Other sources of uncertainties have to be taken into account, which make the final error bar of the electron affinity larger (see text). Rainbows can be observed, at the top and bottom of the image, which are due to more energetic electrons detached from fine-structure excited $^{16}O^-$.

Example of negative ion resonances: He-

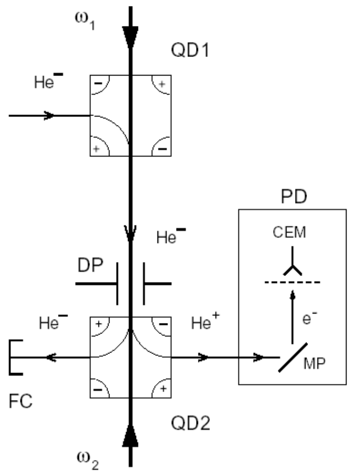
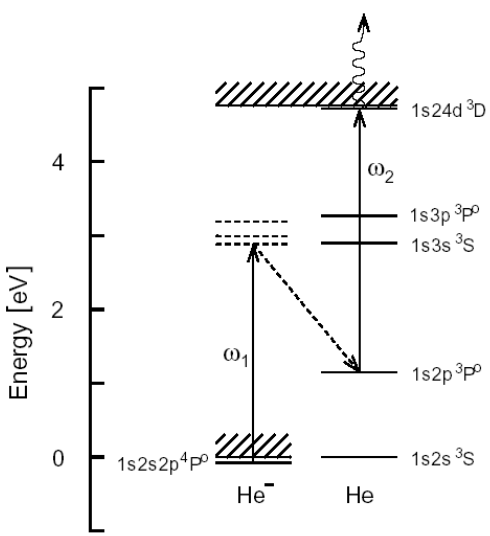


Figure 2. The interaction and detection regions: QD1, QD2, electrostatic quadrupole deflectors; CEM, channel electron multiplier; DP, deflection plates; PD, positive ion detector; FC, Faraday cup; MP, metal plate.



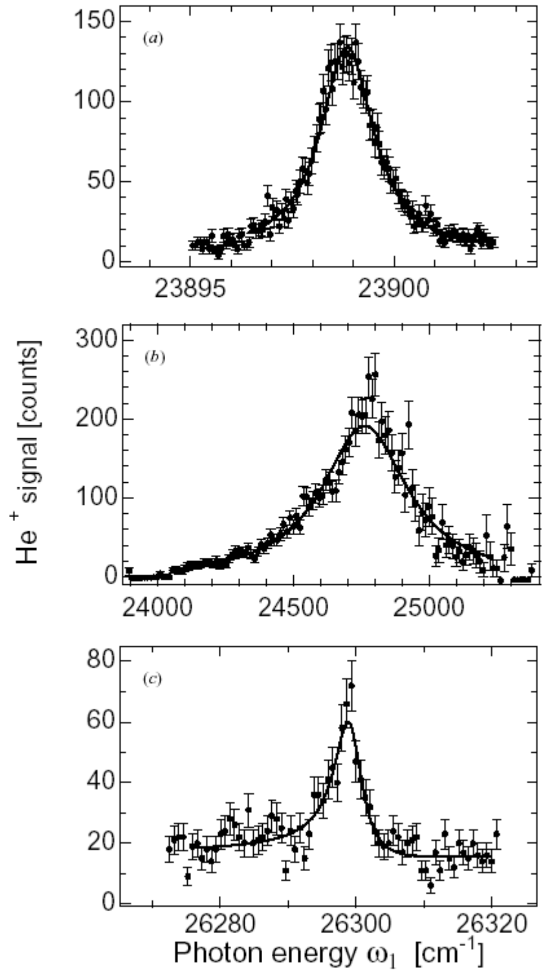


Figure 3. Yield of He^+ ions versus photon energy ω_1. The solid line is a fit to the experimental data (dots) using equation (4). The three curves show the (*a*) 1s3s4s ^{4}S; (*b*) 1s3p^2 ^{4}P and (*c*) 1s3p4p ^{4}P resonances. The figures show the measured data without corrections for the Doppler shift (see text). Note that the energy scale in (*a*), (*b*) and (*c*) is different.

Klinkmüller et al, J. Phys. B: At. Mol. Opt. Phys. **31** (14 June 1998) 2549-2557

He$^-$ and C$^-$ inner shell excitation

Berrah et al, Phys. Rev. Lett. **88**, 093001(2002)

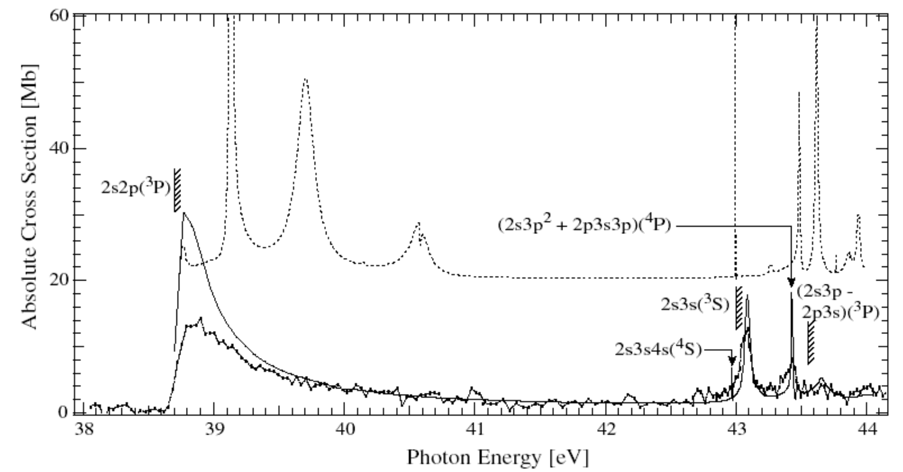


FIG. 1. Photodetachment cross section of the $1s2s2p$ ^{4}P state of He$^-$ in the $1s$ photoabsorption region producing He$^+$. The low-resolution experimental results are shown along with the theoretical results of Refs. [1] and [2] shown with dotted lines, respectively. The He thresholds and He$^-$ resonances are indicated by the hatched lines and arrows, respectively.

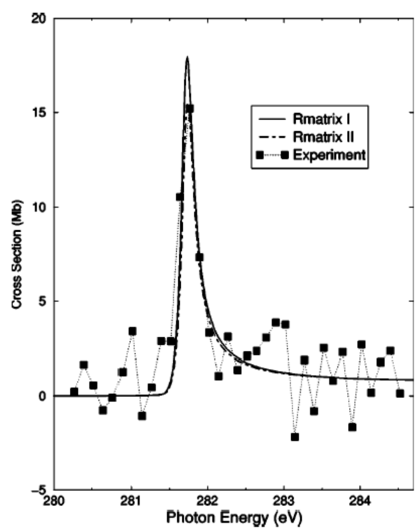


FIG. 2. Comparison between theoretical and experimental cross sections for photodetachment of C$^-$ leading to C$^+$. The solid line (R-matrix I) corresponds to the nonorthogonal B-spline results, whereas the dash-dot line (R-matrix II) corresponds to the standard, single orthogonal basis set results.

N. D. Gibson et al., Phys. Rev. A 67, 030703 (R) (2003)

Experiments in ALS: Detection of positive ions again

What can be done?

- With VUV lasers we can compensate the low density of the ion beam by increase of the photon flux compared to SR. Can operate with crossed beams which are better adapted to electron spectroscopy.

- In this case we can use 4π detection with magnetic field spectrometers. We can have similar count rates for electrons and ions.

Electron spectroscopy with merged beams?

- Need a longitudinal magnetic field to guide electron trajectories along the beam. Electrons from the whole interaction region can be detected and energy analyzed with an electrostatic spectrometer

- Advantage: for fast ion beam, translational spectroscopy can achieve very good resolution for very low energy electrons (particularly shape resonances of negative ions decaying to the closest level)

Magnetic bottle spectrometers

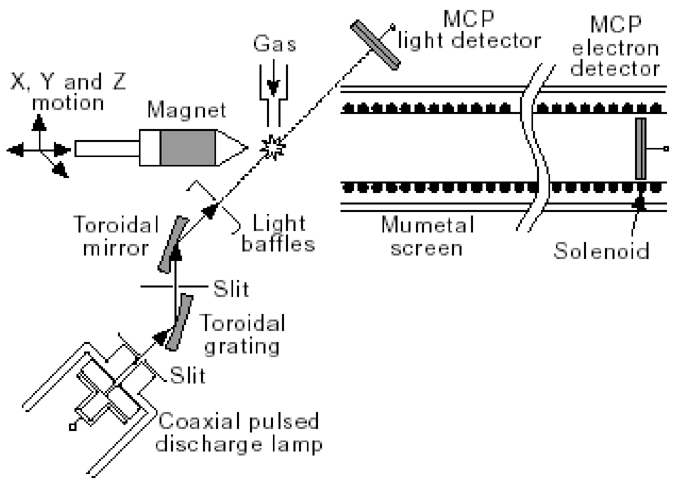
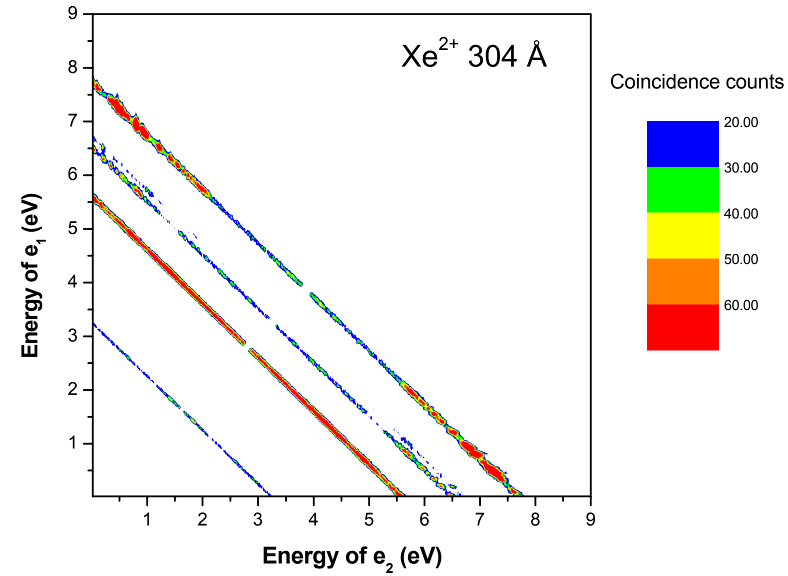


FIG. 1: Scheme of the apparatus. Pumping by turbo molecular pumps at both ends of the flight tube provides a base vacuum of 2 10^{-7} mbar.



Detection of **all** electrons on **4π** solid angle. **Transmission** independent of energy. Good **resolution**. Observation of **multiple** ionization in rare gases with very low photon flux. Xe double and triple ionization have been observed.

Well adapted to pulsed structure of the laser (μ seconds TOF) but need for high repetition rates to compensate the low target density and to avoid multiple ionization events in the same pulse. Possibility of multi-electron coincidences which can be a good way to eliminate background signal.

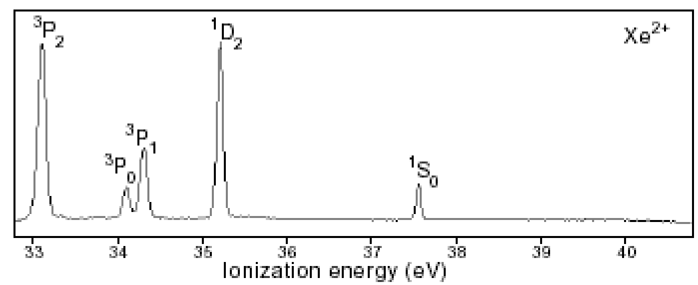


FIG. 3: The spectrum of Xe^{2+} states produced by photoionization at 40.8 eV.

Eland et al, Phys. Rev. Lett. **90**, 053003 (2003)

COLTRIMS type experiment

All particles can be detected in **coincidence**: electrons and fragment ions (molecular ions). Fully differential cross section.
Need extreme high vacuum (10^{-12} torr) to avoid multiple events in one laser-shot.

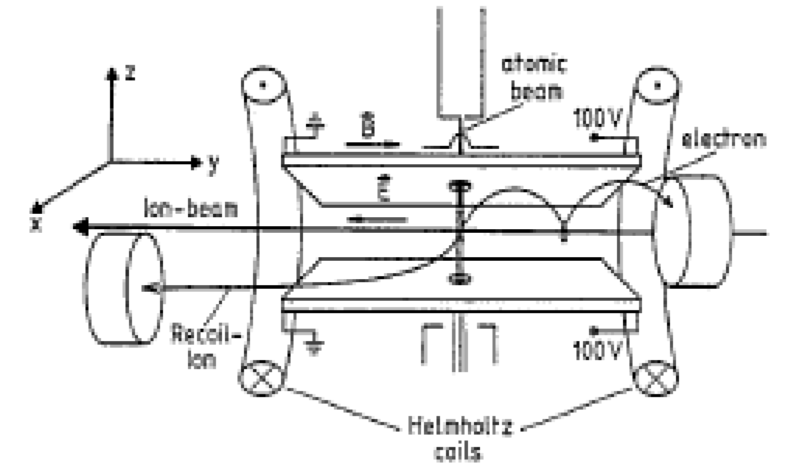


Fig. 1. Schematic drawing of the combined recoil-ion electron spectrometer.

Moshammer et al, NIMB 108, 425 (1996)
Dorner et al, Phys Rep 330, 95 (2000)

Conclusions

Electron spectroscopy on very dilute species like ions in an ion beam will become feasible with the VUV lasers due to orders of magnitude increase in the photon flux compared to SR.

The developments in vacuum technology will allow vacuum limits in the **10^{-12} torr** in order to avoid signal from the background gas.

Due to the **pulsed structure** of the laser with delays in the μsecond range **electron time-of-flight spectroscopy** is an easy task.

Use of magnetic spectrometers will allow detection of **all electrons in coincidence** giving electron spectroscopy in 1, 2 and 3-D. The electron signal should be comparable to the ion signal.

This technique could also be applied to **molecular ions** (positive or negative). Possibility to detect fragment ions. Macro-molecular ions. For negative ions it could be a way to study species that cannot be studied as neutrals (electro-spray ion sources).

- Collaborations:
- J-M Bizau… (LIXAM, Orsay)
- H.Kjeldsen… (ASTRID, Aarhus)
- P.Lablanquie…
- …