Photoionization Study of Atoms, Molecules and Clusters and their Ions in the VUV and Soft X-Ray Region

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Motivation

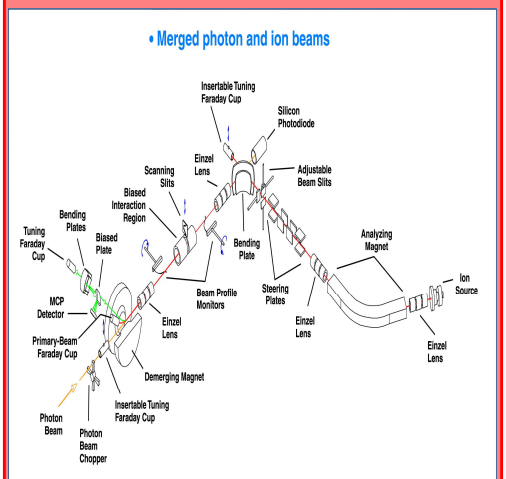
Fundamental:

- VUV photons are a highly selective probe of the internal electronic structure and dynamics of atoms, molecules and their ions.
- Systematic studies with ions permit probing and fine-tuning of electronic structure and interactions along
  - isoelectronic sequences *(different elements with the same number of electrons)*.
  - isonuclear sequences *(same element with different numbers of electrons)*.
- Photoionization theory is based on approximations but few experimental benchmarks exist for ions.

Applications:

- Most of the visible matter in the universe exists in the ionized plasma state.
- Photons carry most of the information we have about the distant universe and hot laboratory plasmas (e.g. fusion-energy research).
- Photon opacity databases used in modeling hot environments depend largely on untested theoretical calculations.
- Advanced extreme ultraviolet (EUV) lithograpy light sources that are being developed to produce the next generation of semiconductor chips are based on hot ionized plasma discharges.

Ion-Photon-Beam Endstation



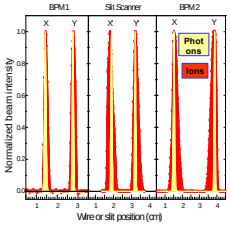
Merged-Beams Technique

The merged-beams technique has been widely used in successful experiments involving atomic or molecular species in electron-ion, ion-ion, ion-neutral, neutral-neutral as well as ion-photon collisions.
The absolute photoionization cross sections may be determined from measured experimental quantities by the equation:

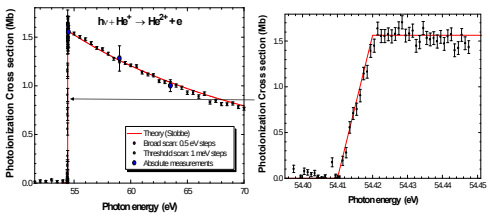
$$\sigma = \frac{R q e^2 v \eta}{I_+ I_\gamma \Omega L F_{av}}$$

R = product ion count rate
q = ion charge
v = ion velocity (cm/s)
e = electronic charge (C)
I_+ = ion current (A)
η = photodiode efficiency (electrons/photon)
Ω = ion detector efficiency
I_γ = photodiode current (A)
L = interaction length (29.4 cm)
F_{av} = average of form factors F(z) (cm^{-2})

$$F(Z) = \frac{\iint I_+(x,y)\, I_\gamma(x,y)\, dxdy}{\iint I_+(x,y)\, dxdy \iint I_\gamma(x,y)\, dxdy}$$



Absolute Photoionization Cross-Section Measurements for He+

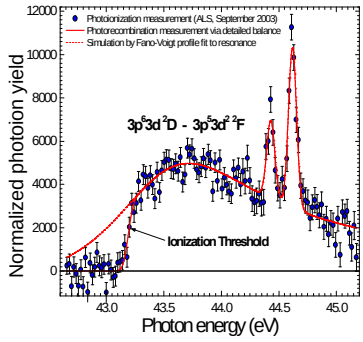


Threshold energy region

He^+ is a hydrogen-like ion with only one electron, for which the photoionization cross section can be exactly calculated. It therefore provides a critical test of absolute measurements using the IPB endstation.

Time-reversed processes: Ti^{3+} photoionization and e + Ti^{4+} photorecombination

Data points: S. Schippers et al J. Phys. B, 37: L209-L216, 2004

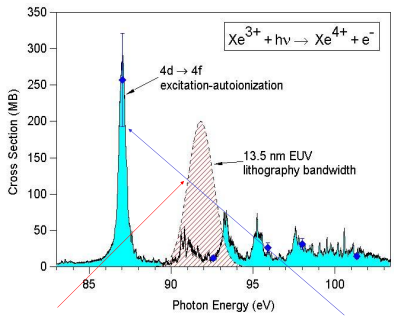


Curve: Photorecombination measurement of Schippers et al. [J. Phys. B **31**, 4873 (1998)] at theTSR storage ring in Heidelberg using principle of detailed balance.

An extremely broad resonance due to 3p – 3d excitation followed by a fast super-Coster-Kronig transition straddles and is truncated by the Ti^{3+} ionization threshold!

Photoionization of Xe $^{3+}$

E.D. Emmons *et al* Phys. Rev. A, **71**, 042704 (2005).

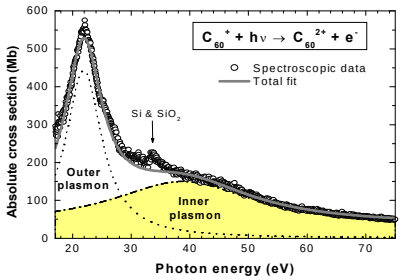


Photoionization measurements at ALS indicate that 4d – nl excitation of Xe^{3+} results in photoabsorption within the 13.5 nm EUV lithography light source window.

4d-4f excitation produces a doubly excited state of Xe^{3+} with all active electrons in the same shell. Because of the strong interaction between the electrons, this broad state autoionizes by a fast super-Coster-Kronig transition.

Photoionization of Fullerine Ions

S.W.J. Scully *et al* Phys. Rev. Lett. **94**, 065503 (2005).

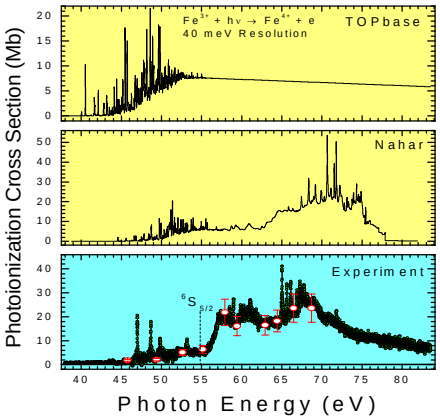


Recent absolute measurements at ALS show that photoionization of C_{60}^+, C_{60}^{2+} and C_{60}^{3+} ions are all dominated by a giant plasmon resonance, similar to that which was observed by Hertel *et al.* in photoionization of neutral C_{60}. This resonance is due to the collective motion of the 240 delocalized valence electrons.

Furthermore, photoionization measurements at higher photon energies show evidence for excitation of a second plasmon resonance. These two resonances are attributed to photoexcitation of surface and volume plasmons.

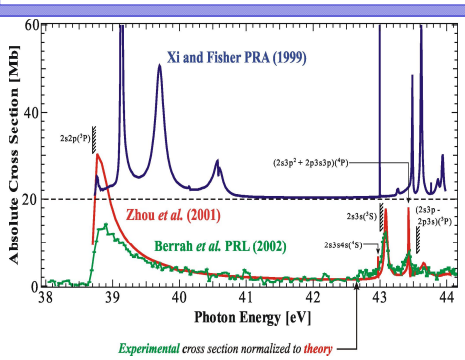
Comparison of Absolute Measurements with Theories for Photoionization of Fe^{3+}

$1s^22s^22p^63s^23p^63d^5$



- Theoretical R-matrix calculations predict resonances as well as broad features in the photoionization cross section.
- However, there are significant differences in the positions and strengths of the resonances, and of the broad features.

K-shell photoionization of He^-



- He^- is formed as long-lived metastable He^-:$1s^22s$.
- Two contradictory theoretical results for He^- photoionization preceded the experimentalm measurements.
- Experimental results clearly resolve the issue.

Selected Publications

1. Gharaibeh, M.F.; Aguilar, A.; Covington, A.M.; Emmons, E.D.; Scully, S.W.J.; Phaneuf, R.A.; Müller, A.; Bozek, J.D.; Kilcoyne, A.L.D.; Schlachter, A.S.; Álvarez, I.; Cisneros, C.; Hinojosa, G. Systematic Photoionization Study along the Iron Isonuclear Sequence: experiment and theory. (under process).
2. Scully, S.W.J.; Álvarez, I.; Cisneros, C.; Emmons, E.D.; Gharaibeh, M.F.; Leitner D.; Lubell M.S.; Müller A.; Phaneuf R.A.; Püttner R.; Schlachter A.S.; Schippers, S.; Balance C.P.; McLaughlin, B.M. Doubly Excited Resonances in the Photoionization Spectrum of Li+: experiment and theory. *J. Phys. B.* (under process).
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