Pulse length and photon energy dependence of multi-photon processes

Michael Martins
Universität Hamburg
Institut für Experimentalphysik

31.10.2011

Overview

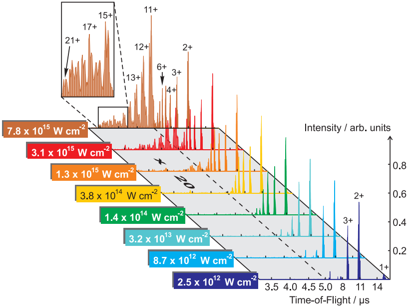
- The 4d giant resonance
  - Photon Energy dependence of ion spectra in Xenon
  - Xenon and Iodine
  - A molecular effect ?
- The 3p giant resonance in Manganese
  - Ion spectra in and off resonant
  - A two-photon giant resonance ?
- Pulse length dependent effects

Thanks to

- Universität Hamburg
  Nils Gerken
  Stephan Klumpp
  Florian Sorgenfrei
  Vera Linsenmann
  Karolin Mertens
  Ricarda Laasch
- Kai Tiedtke
- The FLASH Team
  Vladimir Rybnikov and Tim Wilkens for the DAQ System

Funding: DFG MA2561/4-1 , SFB925 A3

Resonances and Nonlinear processes



- Xe 4d $\rightarrow$ (4ϵ)f giant resonance
- High charge states up to Xe^{21+}
- What is the influence of the giant resonance ?

A.Sorokin et al., PRL **99**, 213002 (2007)

- Discussions on the process of the production of these charge states in the short pulse (10-20 fs) of FEL radiation
- Resonance position of the atom might be important only for the first ionization steps

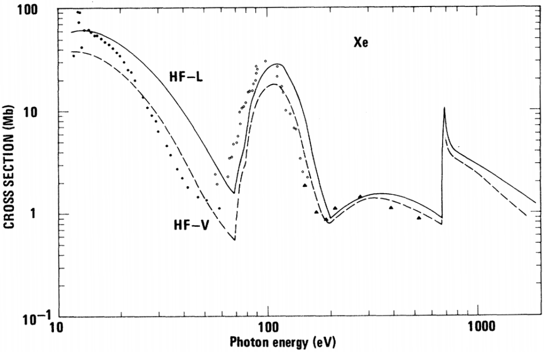
Resonances and Nonlinear processes

- Choose other systems with giant resonance of different types
  Shape resonance vs. discrete/autoionizing resonances
- Shape resonance:
  Iodine, barium, ... $(4d \rightarrow (4\epsilon)f)$
- Discrete resonance:
  Europium $(4d \rightarrow 4f)$,
  Manganese $(3p \rightarrow 3d)$
- What is the difference in resonant and non-resonant spectra ?
- What is the influence of the pulse length/structure ?

Experiments

- Simple setup with ion TOF and electron TOF spectrometers
- Experiments are performed at the FLASH BL2 beamline in the standard focus of $\approx 20 - 30\mu m$
- MCP Detectors are operated in the linear regime recording single shot spectra with each FLASH pulse
- FLASH was operating the multibunch mode with 30-200 bunches and 250 kHz repetition rate

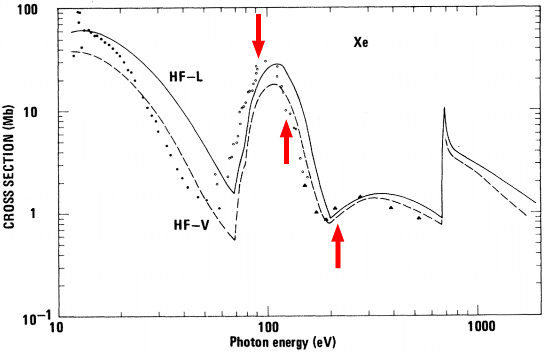
Xe $4d \rightarrow (4\epsilon)f$ – Photoabsorption



Kennedy and Manson, Phys. Rev. A **5**, 227 (1972)

- $\cong$ 93 eV: Maximum of the shape resonance
  $\cong$ 123 eV: Slightly above the shape resonance
  $\cong$ 217 eV: Above the shape resonance – Cooper minimum

Xe $4d \to (4\epsilon)f$ – Photoabsorption



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Xe $4d \rightarrow (4\epsilon)f$ – Ion spectra

- Pulsenergy $\approx 110\mu J$ and $\cong 30\mu m$ spot size
- Maximum charge state observed is Xe^{11+} $4d^7$

Xe $4d \rightarrow (4\epsilon)f$ – Summary

- For mid charge states sequential processes are dominating
- Xe resonances shift the mean charge state to 7+
- Resonances have to be included in calculations
  Many of the resonances are not well known

Iodine – Ion spectra

- CH_2I_2 and CH_3I molecular samples
- Similar behavior as for Xe
  Cut off around at I^{10+} – $4d^7$ @ 93 eV and 110 μJ

Iodine – Nonlinear molecular effect ?

$$CH_3I \longrightarrow I^{n+} + CH_x^{k+}$$

$$CH_2I_2 \longrightarrow I^{n+} + I^{m+} + CH_x^{k+}$$

- Strong Coulomb interaction in the CH_2I_2 system due to the interaction of I^{m+} and I^{n+} ion fragments
- Is there an influence of the molecular environment on the multi-photon process ?

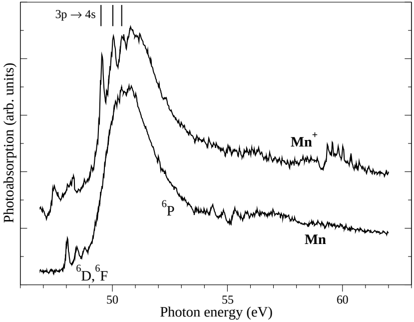
Iodine – Nonlinear molecular effect ?

Integrated charge distribution for $\mathbf{I}^{n+}$

$\cong 6\mu J$ $\cong 110\mu J$

- Modified fragment charge distribution for CH_2I_2 and CH_3I
- Comparison with I_2 and other I molecules !

Manganese Giant Resonance

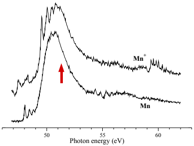


J.T. Costello et al, Phys.Rev.A **43**, 1441 (1991)

- Mn $3s^2 3p^6 3d^5 4s^2$
- Different Type of Resonance
- Fano type giant resonance $3p \rightarrow 3d$
- Ion spectra in the resonance and above
- Which charge states can be reached in comparision to xenon or iodine ?

- Double core hole ionization might be used as a tool for the dynamics of processes in 3d metal compounds

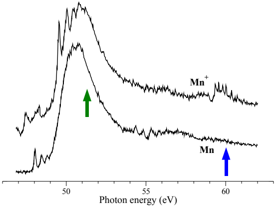
Mn – Resonant ion spectra



- Excitation at ≅52 eV in the giant resonance
  $IP(4+) = 51.2\ eV \qquad IP(5+) = 72.4\ eV$
- Lower charge states are observed in comparison to Xe and Iodine

Mn – Ion spectra

How does the 3p-3d resonance influences the charge distribution ?



Experiments at 20.3 nm were runnung at low irradiance levels of $\approx 5\mu J$

Mn – Resonances

Influence of the 3p-3d resonance seems to be rather weak

Generation of Mn^{1+} to Mn^{4+} mainly due to sequential processes

Mn – Nonlinear Giant Resonance

- Might there be a nonlinear giant resonance ?
- One photon: $3p \rightarrow 3d$
  Two photons: $3s \rightarrow 3d$
- First, principle calculation within a HF and CI approximation
- Two, 2-photon resonances:
  (1) $3s \rightarrow 3d$
  (2) $3p^2 \rightarrow 3d^2$
- $3s \rightarrow 3d$ is well seperated and might be observed
- Experiments at different photon energies are necessary

Pulse dependent effects

“old” and “new” ion spectra for Xe^{4+} to Xe^{9+}

Comparable number of photons $\cong 2 \times 10^{11}$ photons

Sorokin et al., PRL

Comparable irradiance $\cong 10^{14} W/cm^2$

- The charge state distibution varies quite strongly
- Newer data shows higher medium charge states
- What is the difference $\rightarrow$ pulse length
  Old data $\cong 10 - 30\ fs$ New data $100 - 300\ fs$ (?)

Pulse length

- Evidence for change in the pulse length after the FLASH upgrade
- LOLA shows longer electron bunches
- SASE spectra taken e.g. with the PG2 monochromator [1] shows a strongly increased number of modes
- Direct autocorrelator experiments show bunch length of 100-300 fs depending on the bunch charge
- Comparison to Ne ion data from the LCLS [2]
  Bunch length influences the ion charge distribution

[1] M.Martins et al., RSI **77**, 115108 (2006)

[2] L.Young et al., Nature **466**, 56 (2010)

Pulse length

Okt 2010 ≈250 fs – April 2011 ≈100 fs

- Charge state distribution can be explained by the longer pulses in the newer data
- Theory more complicated as compared to Ne at the LCLS due to the complex resonance structure in the Xe ions

Pulse length – Bunch trains

- Sort the Xe ion spectra according to the pulse intensity (100μJ) and bunch number in a bunch train
- Charge distribution is changing with the bunch number

$\rightarrow$ Bunch length/structure is changing in a bunch train

Summary

- Strong influence of 4d resonances in different initial charge states of Xenon and Iodine on the sequential multi-photon processes
- Influence of the 3p giant resonance in Manganese is rather weak
- Molecular environment modifies the charge state distribution in Iodine molecules
- The charge state distribution is effect by the pulse length of the FEL radiation
- The pulse length of FLASH varies with the bunch number

Thank You