Probing XFEL Radiation Induced Many Electron Dynamics

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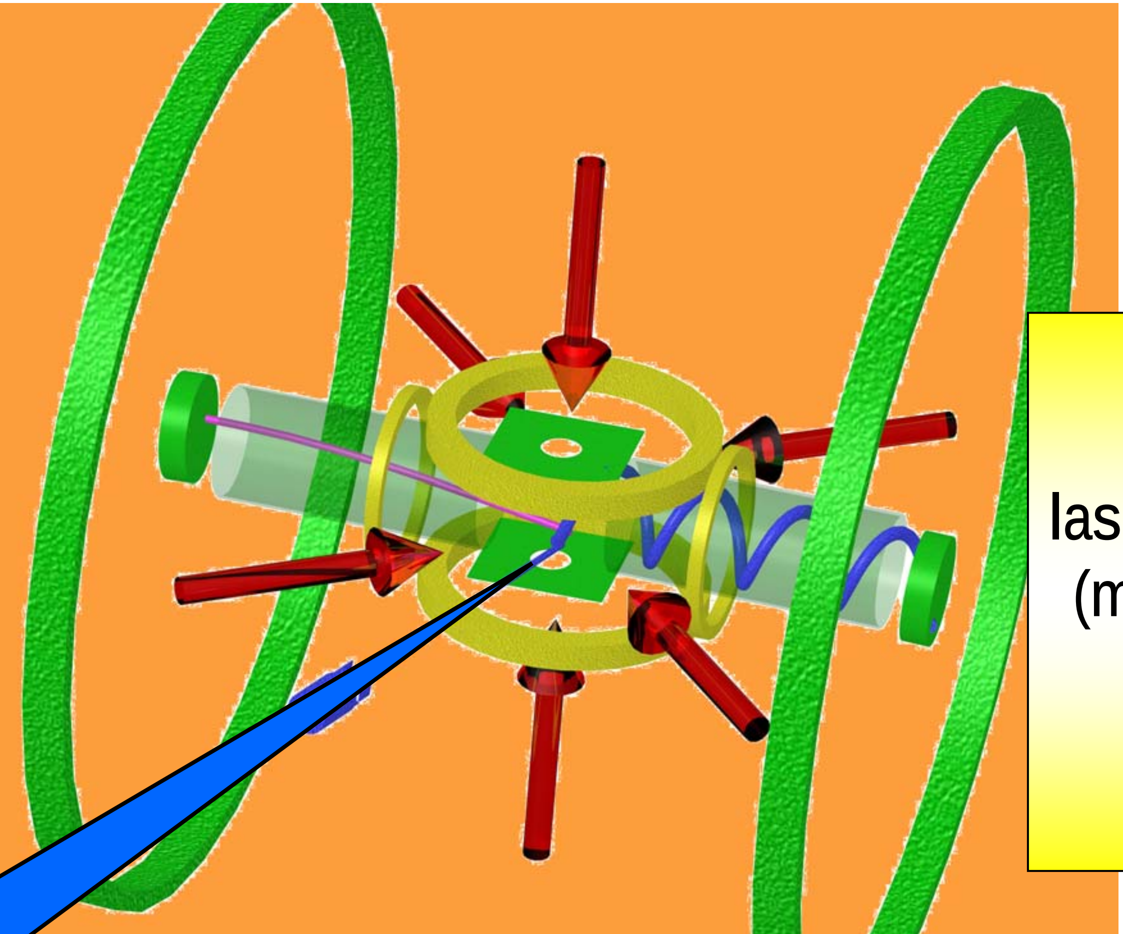

XFEL radiation
+
laser target cooling and trapping
(magneto-optical + dipole trap)
+
multi-particle imaging
(reaction microscope)

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- Motivation for our FEL studies

  Single-photon multiple ionization

- Experimental approach MOTRIMS

- Results of a pilot experiment at FLASH (lithium)

- Possible Future Studies using XFEL radiation (sodium)

- Summary

Fundamental Few-Body Dynamics

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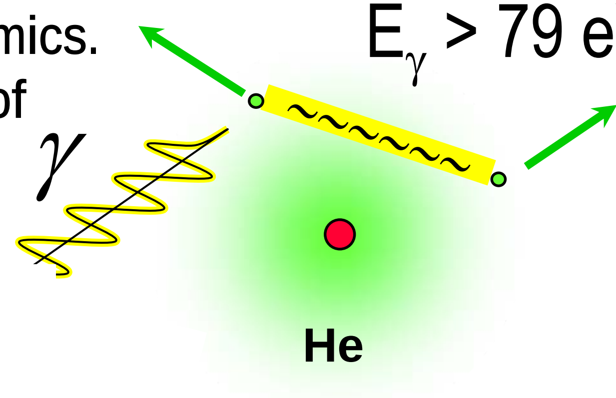
Our Initial Motivation

- Quantitative description of n-body Coulomb dynamics. Presently only n = 3, e.g. photo double ionization of helium is studied in detail.

$\sigma^{2+} \leq 10^{-20}$ cm^2

$E_\gamma > 79$ eV



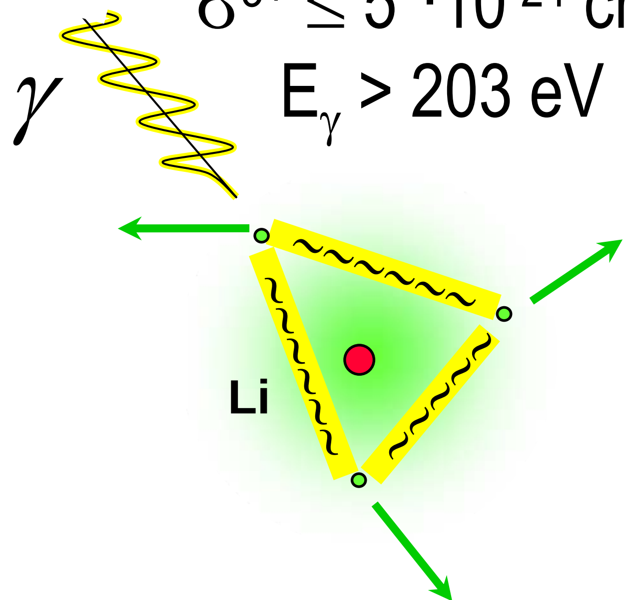
Goals

- Triple ionization: investigation of the three electron dynamics, the complete fragmentation of lithium.

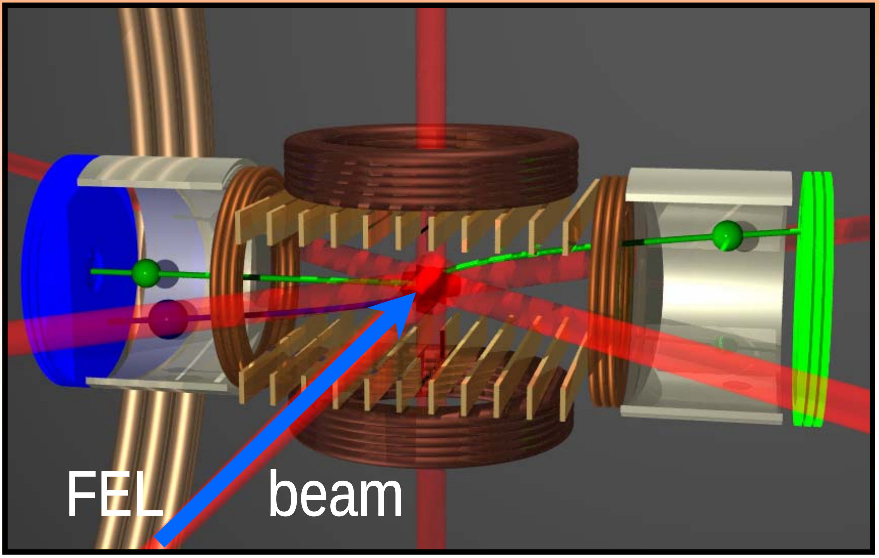
- Double ionization: variation/control of the correlation by optical pumping of the target.

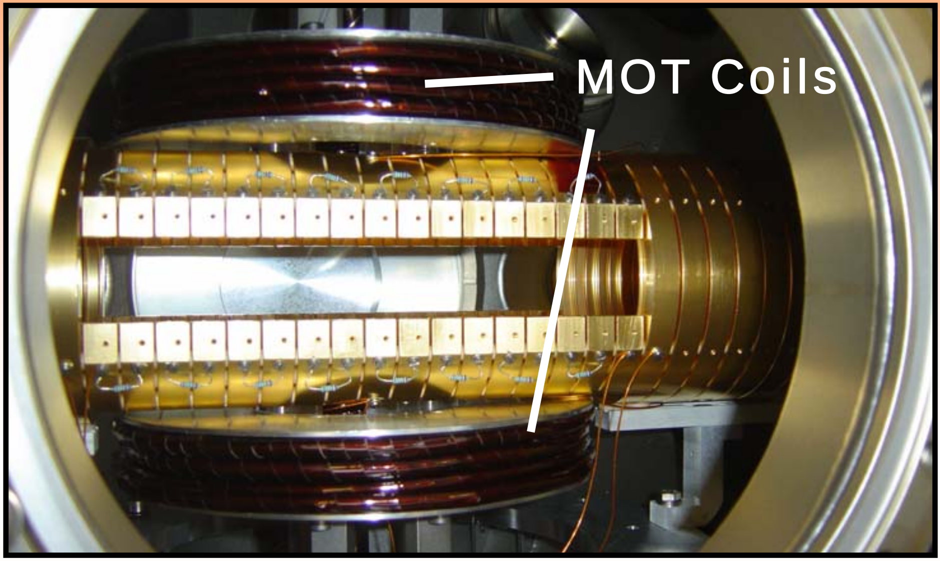
$\sigma^{3+} \leq 5 \cdot 10^{-24}$ cm^2

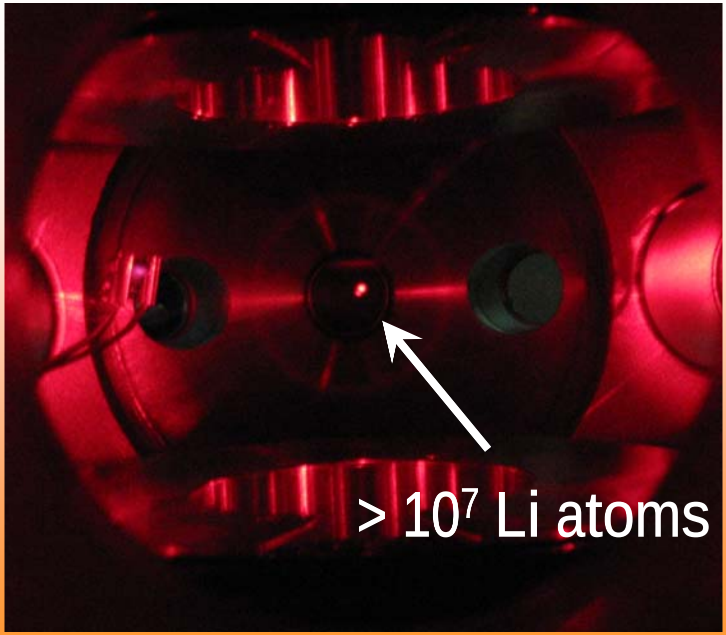
$E_\gamma > 203$ eV



Experimental Set-up







Specifications:

Target temperature: T_{MOT} = 300 μK (13 neV)

Momentum resolution for ions: $\Delta p < 0.05$ a.u.

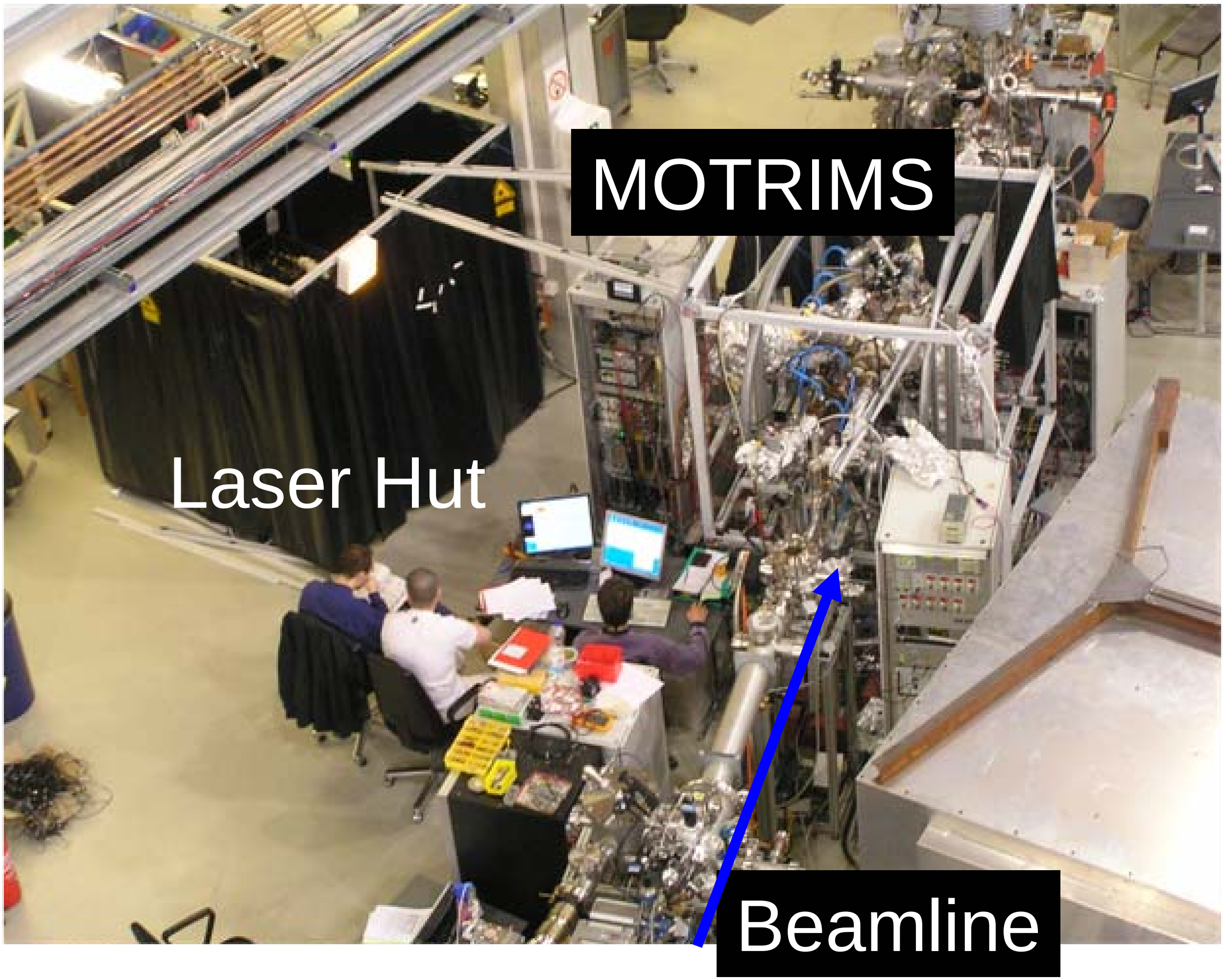
Resolution for electrons: $\Delta p = 0.05$ a.u.

Acceptance: 4π solid angle for all target fragments

Density: $\rho = 10^{11}$ cm^{-3}

UHV: $P = 10^{-11}$ mbar

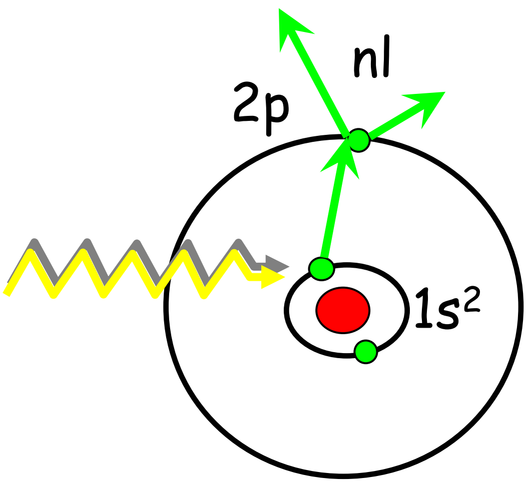
Pilot Experiment at FLASH



Double Ionization of Laser Prepared Quantum States

- Control of the electronic correlation.
- Selection rules depend on the symmetry.
- Ionization of an aligned / oriented valence orbital.

$Li^*(1s^2\, nl\, SLM_L)$

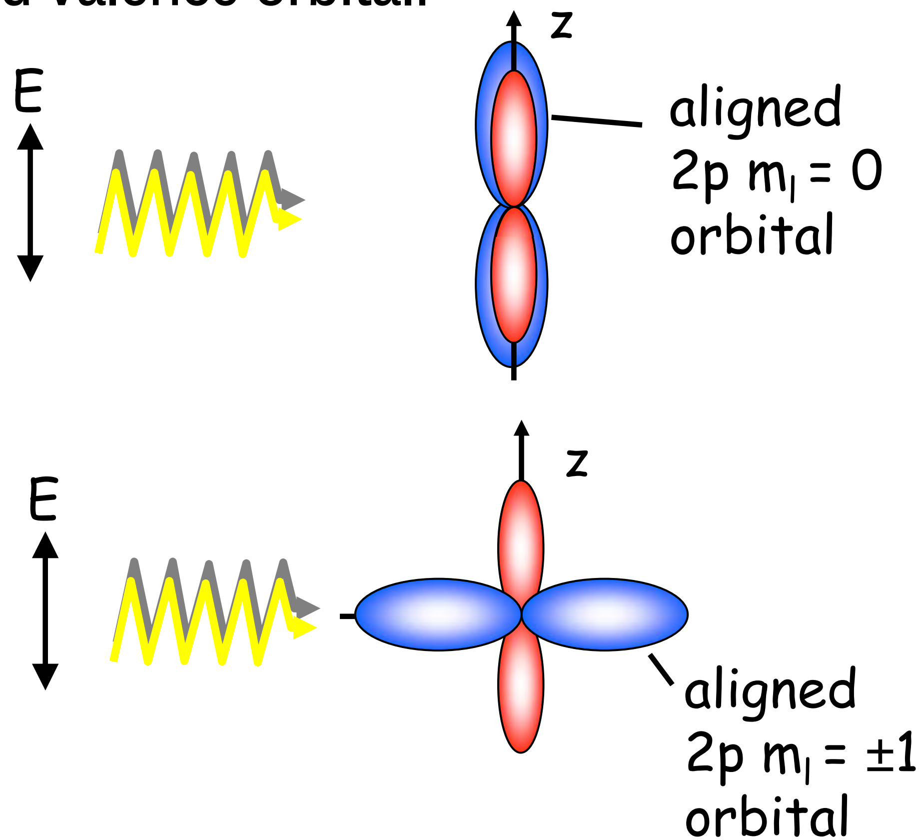


$\sigma^{2+} \leq 10^{-20}$ cm^2

$E_\gamma > 81$ eV,

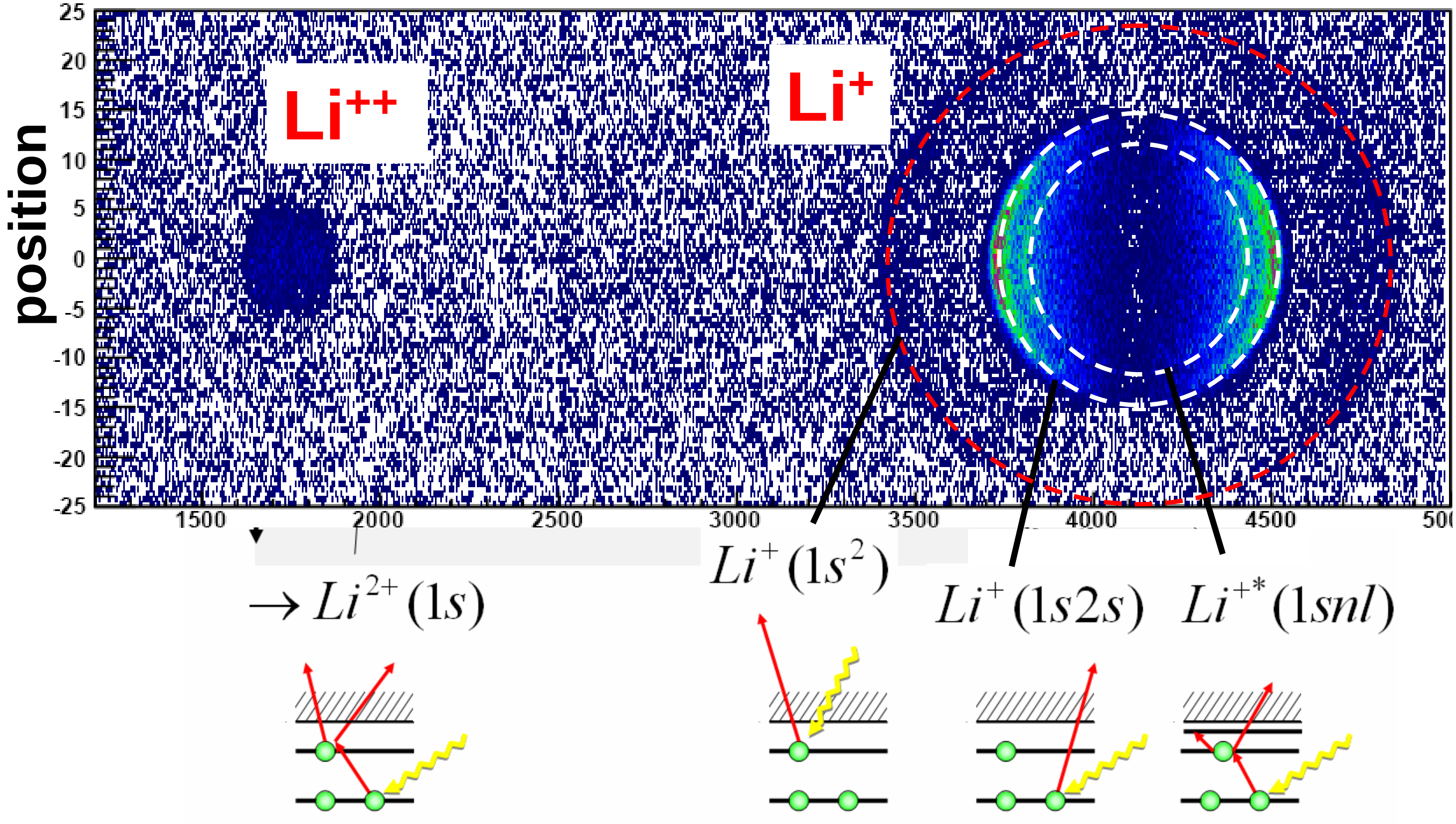
$\lambda < 15$ nm

2 μJ @ 20 Hz → 1 Hz signal

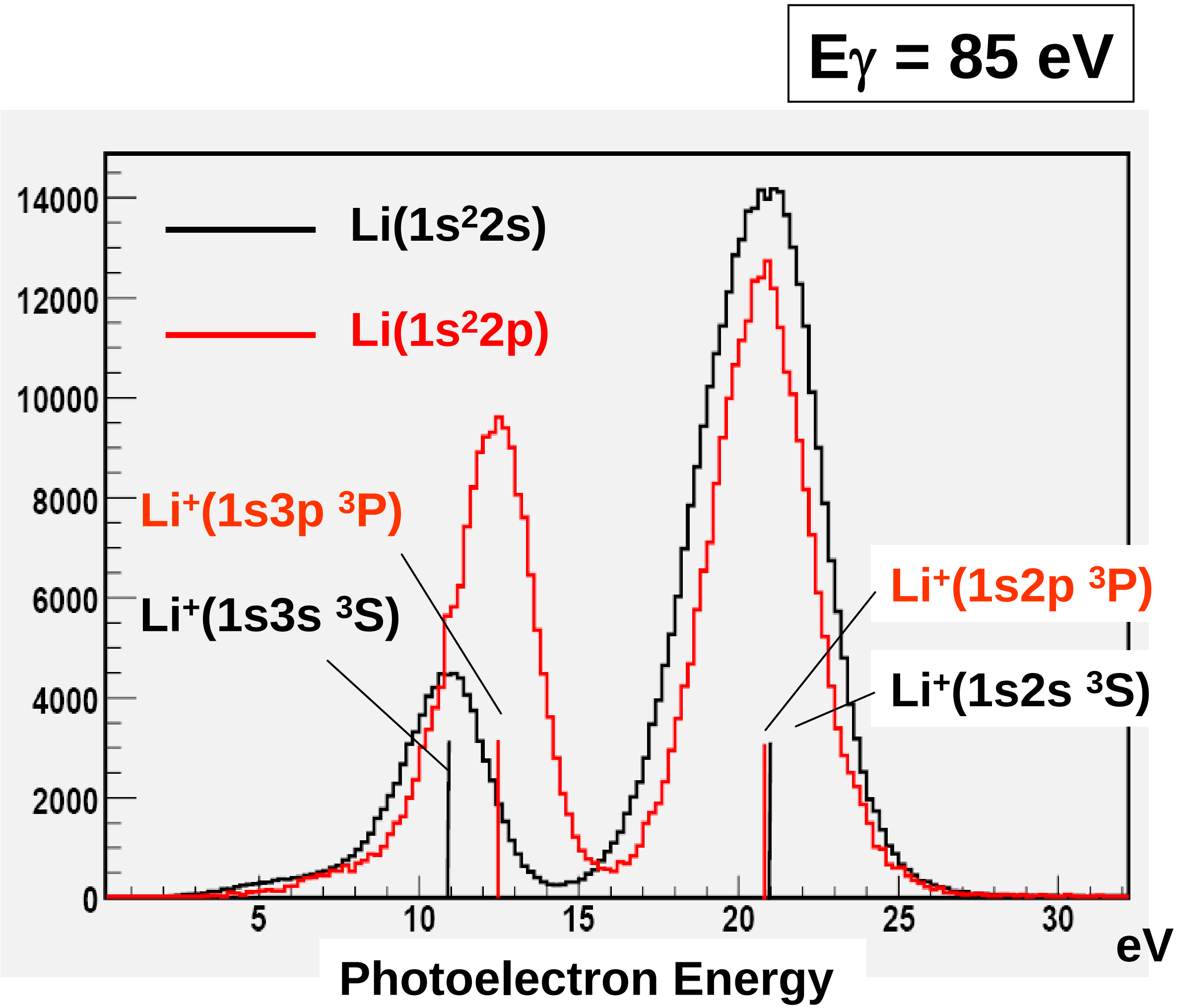


Raw Spectrum

2D (TOF & Position) Recoil Ion Spectrum

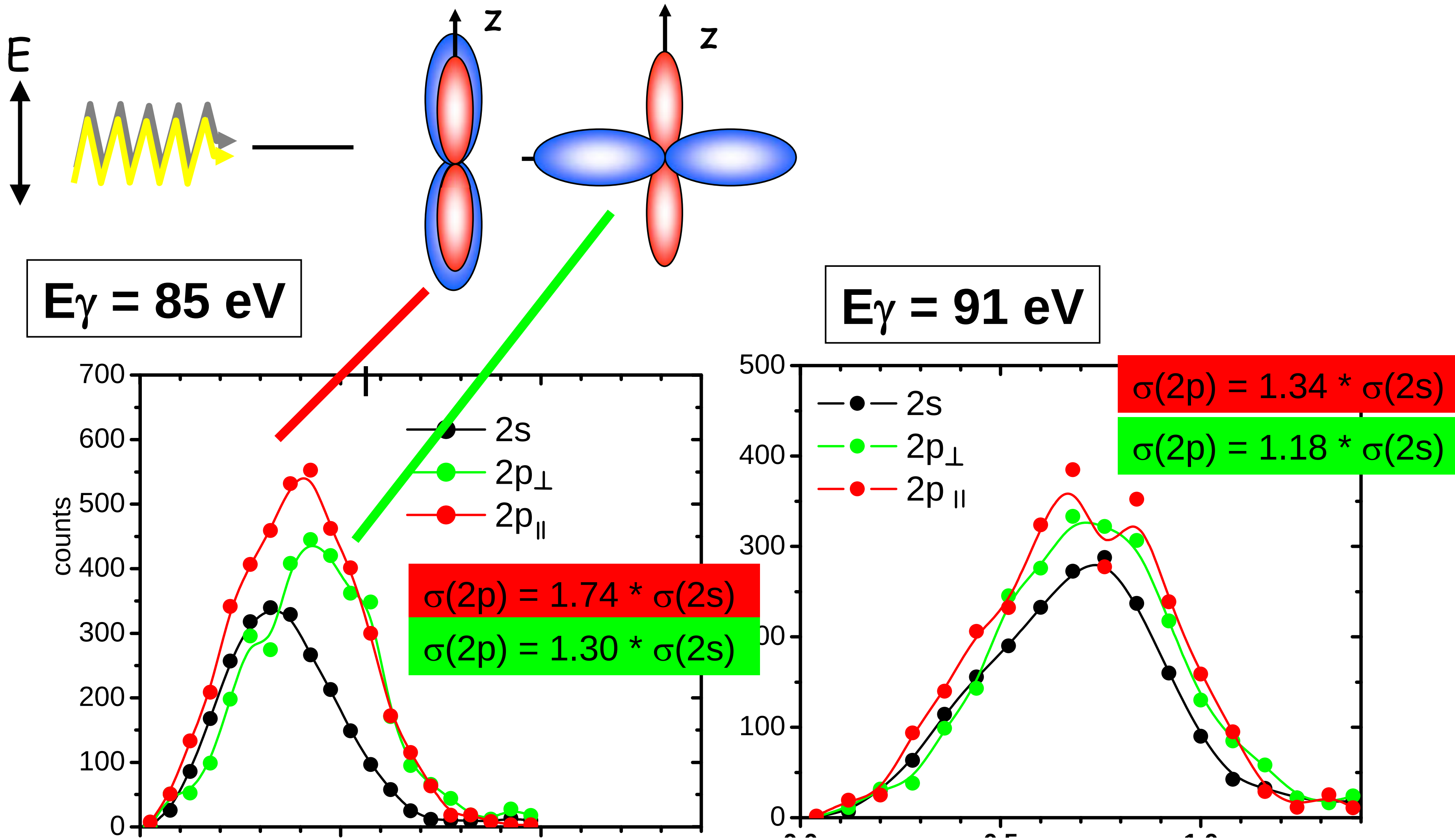


Simultaneous Ionisation-Excitation



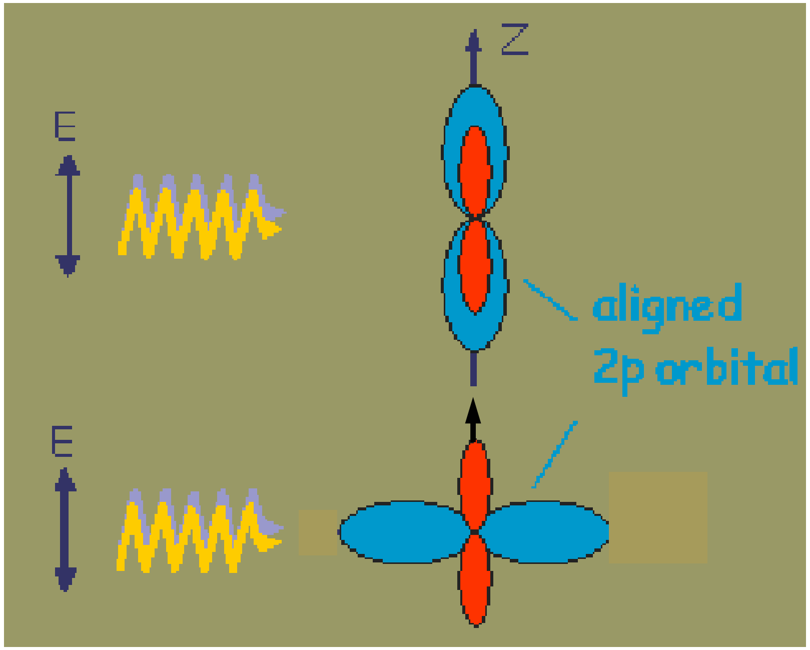
No Dependence on Initial State Alignment! (2007)

Alignment dependent Ion Momentum

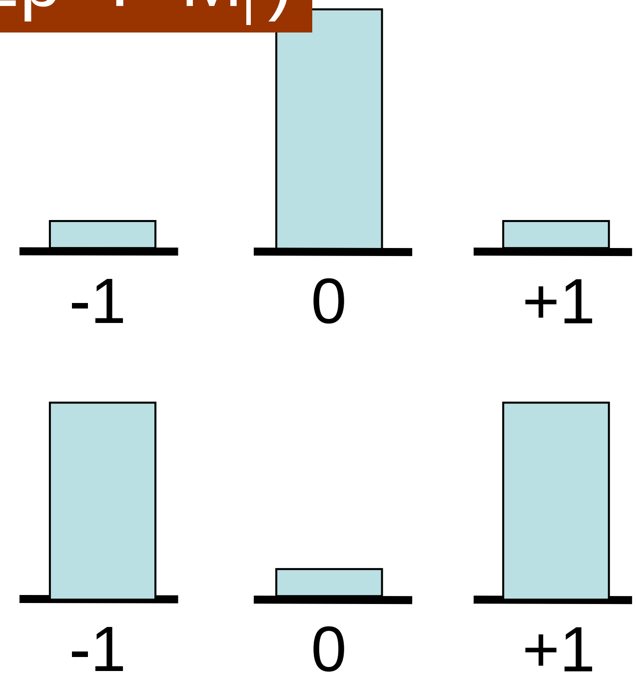


Strong Dependence on Initial State Alignment!

"Proper" Formal Treatment



$Li^*(2p\ ^2P\ M_l)$



$Li^*(2p\ ^2P,\ M_P=0) + \gamma \rightarrow Li^{2+}(1s\ \ ^2S) + 2e^-\ (\varepsilon s\ m=0\ ,\ \varepsilon d\ m=0)$

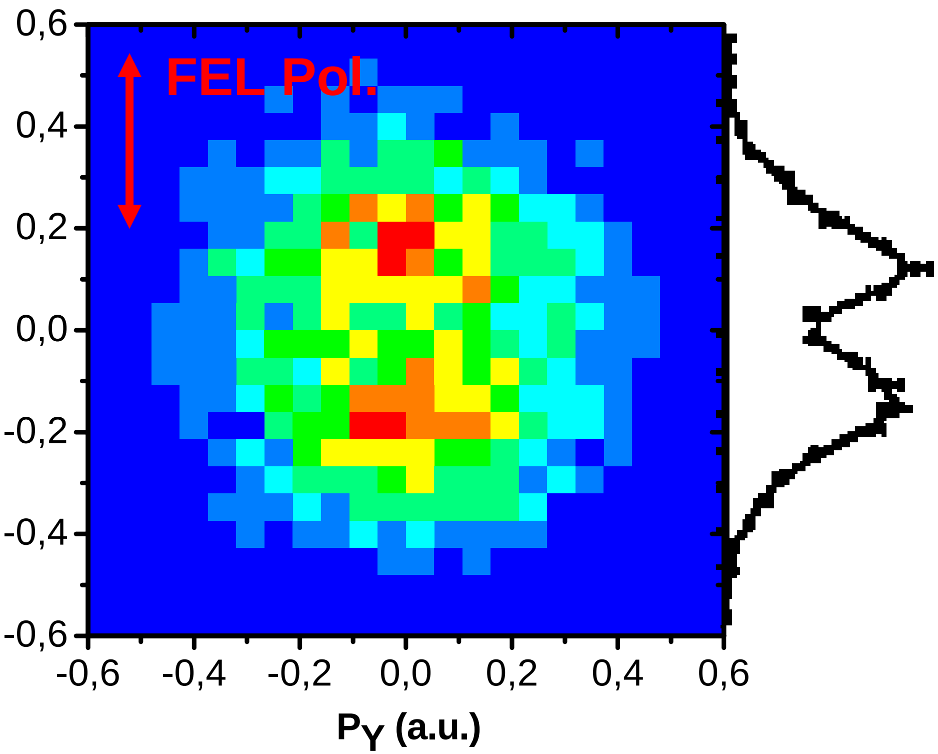
$Li^*(2p\ ^2P,\ M_P=\pm 1) + \gamma \rightarrow Li^{2+}(1s\ \ ^2S) + 2e^-\ (\varepsilon d\ m=+1,\ \varepsilon d\ m=-1)$

$Li(2s\ ^2S,\ \ M_S=0) + \gamma \rightarrow Li^{2+}(1s\ \ ^2S) + 2e^-\ (\varepsilon p)$

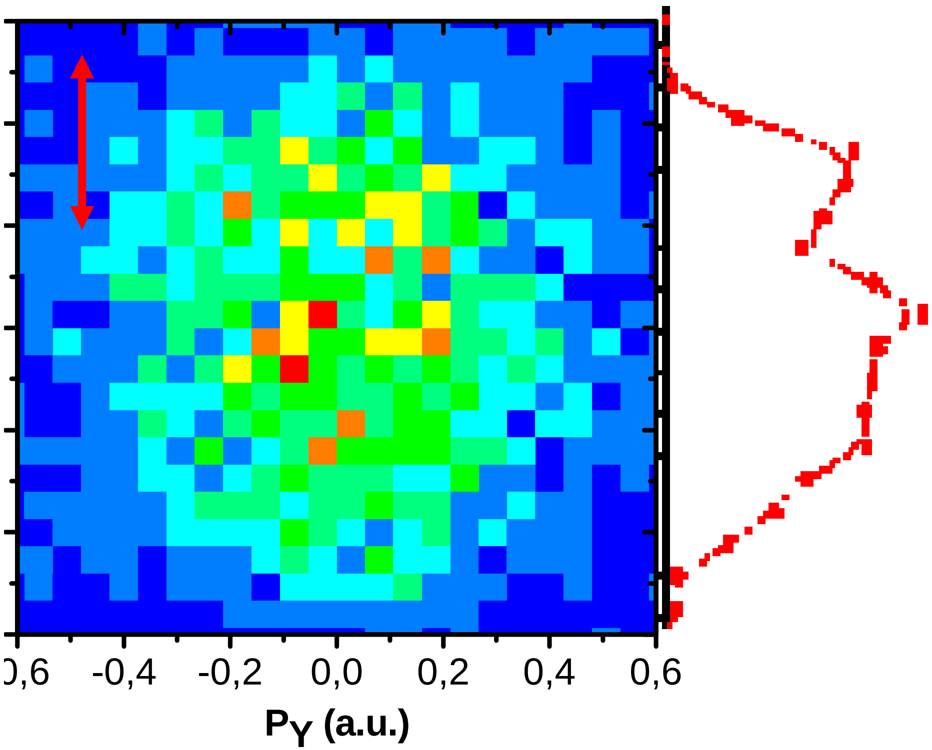
Influence of Selection Rules

85 eV (γ ,2e)

Li ($1s^22s$)

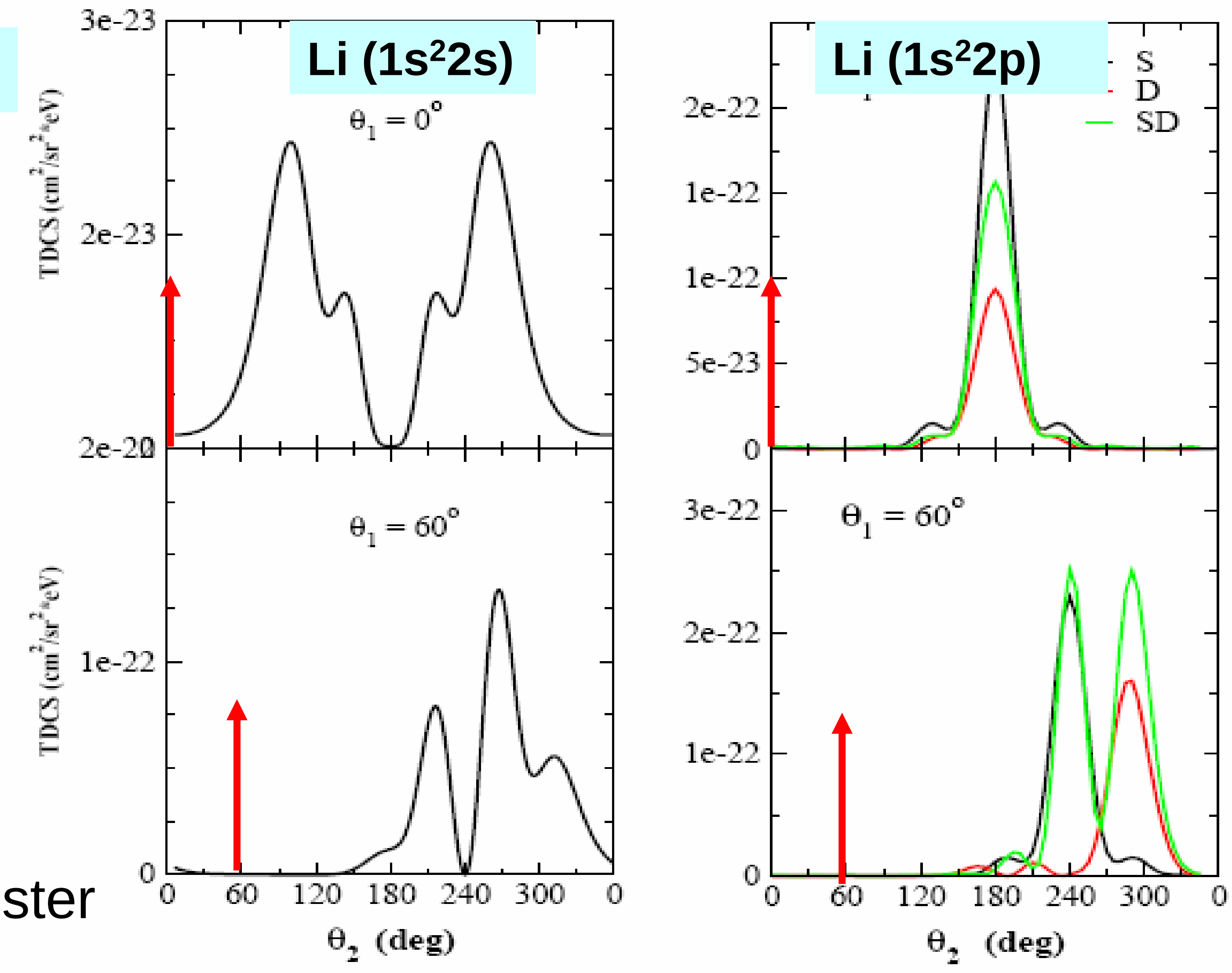


Li ($1s^22p$, ||)



Future: Fully Differential CS

90 eV (γ ,2e)



Time dependent close-coupling calculation.

J. Colgan, M. Foster

Next Steps

- Implementation of a dipole trap $\rightarrow$ field free trapping, coincident electron spectroscopy will be possible.
- One-photon triple ionization (10^{15} γ/s, >10 kHz rep. rate)

Future Studies Using XFEL Radiation

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Scientific advisory committee for LCLS (2000)

- Atomic Physics Experiments
- Plasma and Warm Dense Matter Studies
- Structural Studies on Single Particles and Biomolecules
- Femtochemistry
- Nanoscale Dynamics in Condensed Matter Physics

Atomic physics:
R.R. Freemann, P.H. Bucksbaum, K. Kulander,
L. Young, R. Falcone

Multi-Photon Multi-Electron Dynamics

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- Production of multiple vacancies in a specified inner shell

- Inner-shell ionization dominates, Photoionization occurs „inside-out“

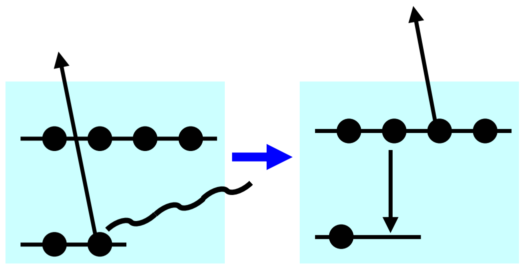
- Ionization rate easily exceeds inverse pulse length → multiple vacancies

- Cascading to high charge states.

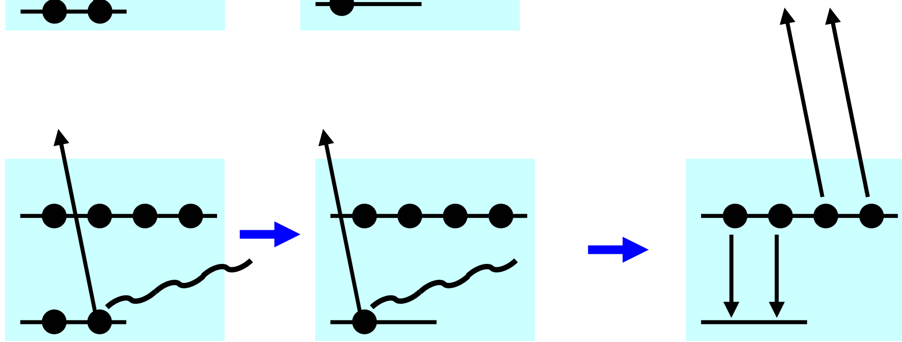
- Coherent multiphoton effects. Nonlinear processes in inner cores.
→ Diagnostics of temporal beam properties. E.g. two-photon photoionization.
Signature: Photons or electrons with energies greater than the XFEL

- Ions produced in excited states:
Study of x-ray laser mechanisms in many materials.

New Ionization Processes:
Multi Core Hole Formation + K-Shell Multiphoton Ionization

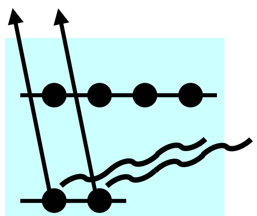


Photon ionization

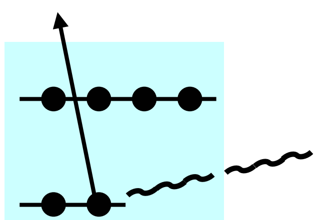


Sequential multiphoton ionization
First direct multiple core hole formation
→ „Hypersatellite“ Auger electrons

New



Direct multiphoton ionization



2-Photon absorption (also resonant)

The Sodium Target

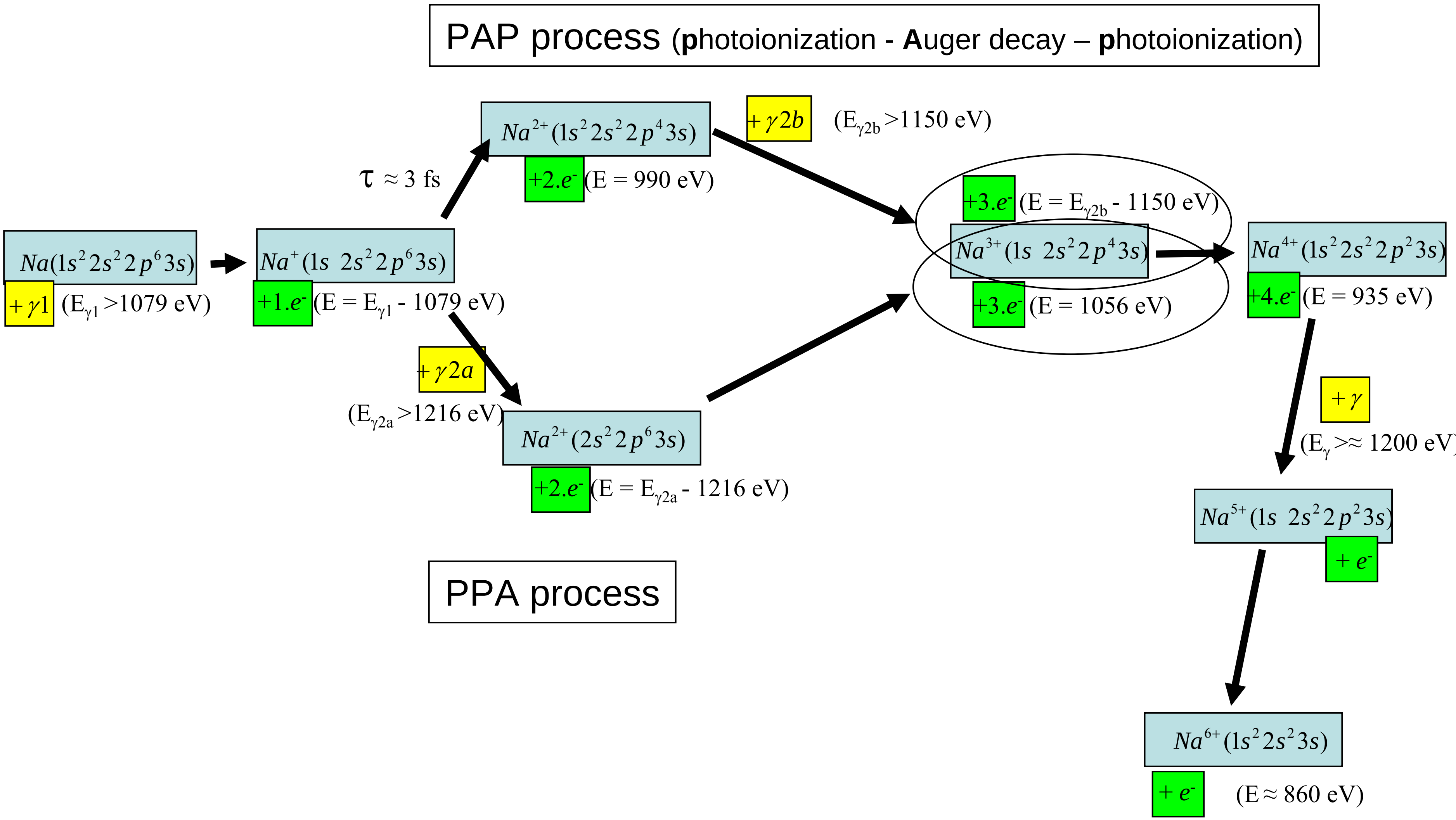
- Neon electronic structure + valence electron.

- Much better ion momentum resolution (0.05 a.u.) than for Ne.

- Target can be localized to 10 μm in the dipole trap.

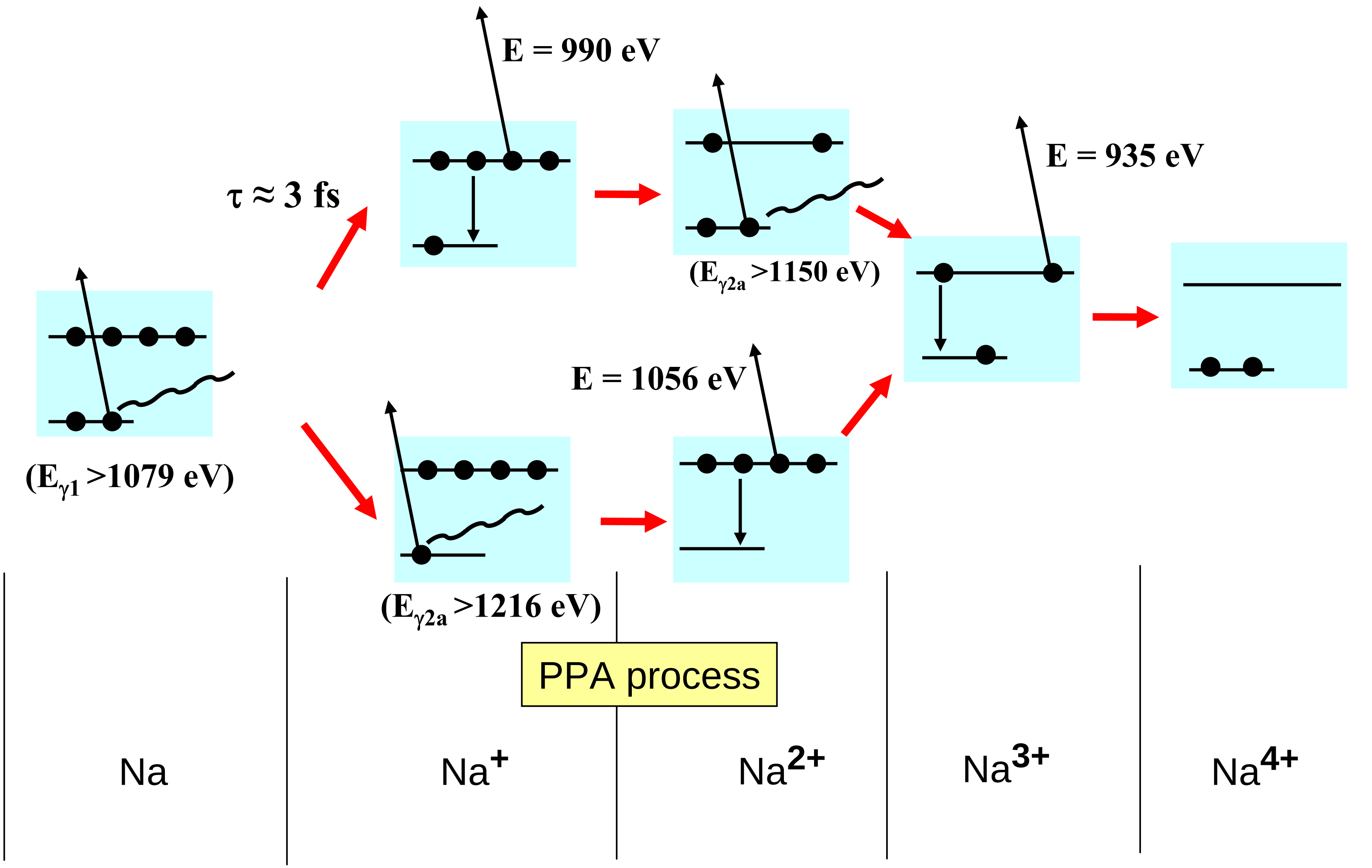
- Choice of laser excited initial state (e.g. K-edge shifting).

Sodium: Sequential Multiple K-shell Ionization



Sodium: Sequential Multiple K-shell Ionization

PAP process (**P**hotoionization - **A**uger decay – **P**hotoionization)



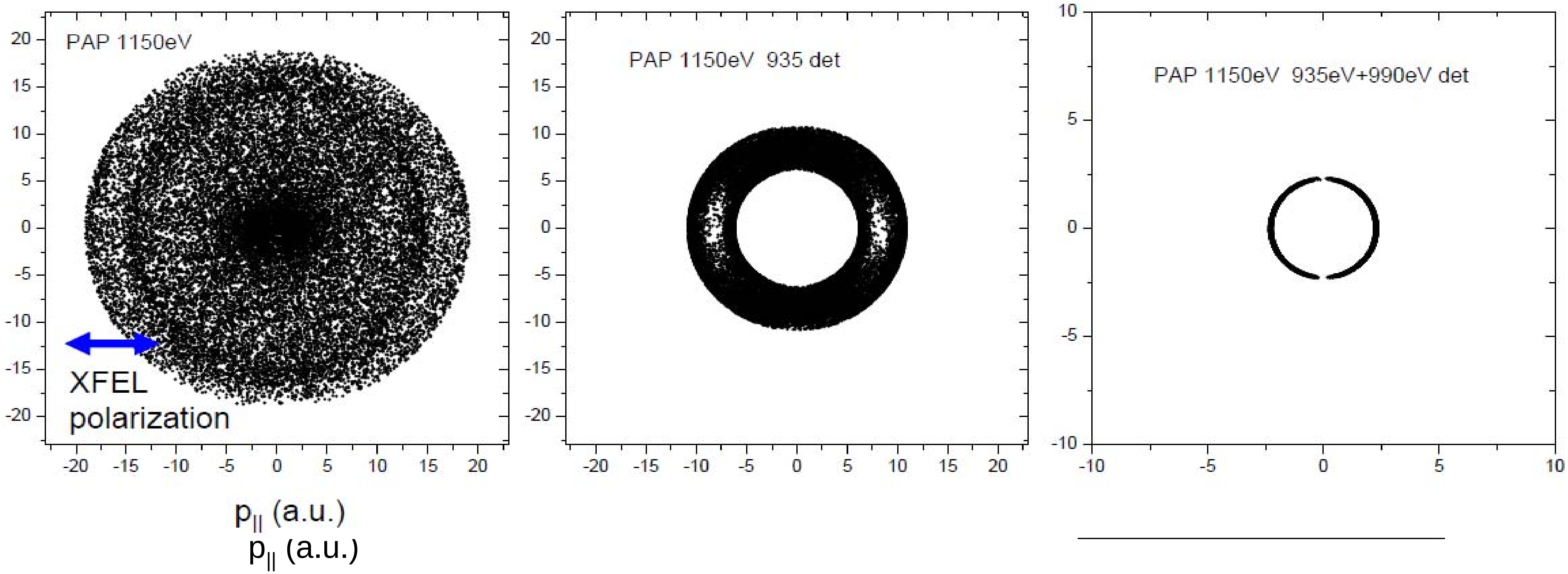
1. Example E_γ = 1150 eV, only the PAP process is possible:

PAP

$Na + \gamma$ (1150 eV) $\rightarrow Na^{4+}$

1.e^-)	71 eV	*pe*$^-$	(1. photoelectron)
2.e^-)	990 eV	*Ae*$^-$	(1. KLL Auger electron)
3.e^-)	0 eV	*pe*$^-$	(2. photoelectron)
4.e^-)	935 eV	*Ae*$^-$	(2. KLL Auger electron)

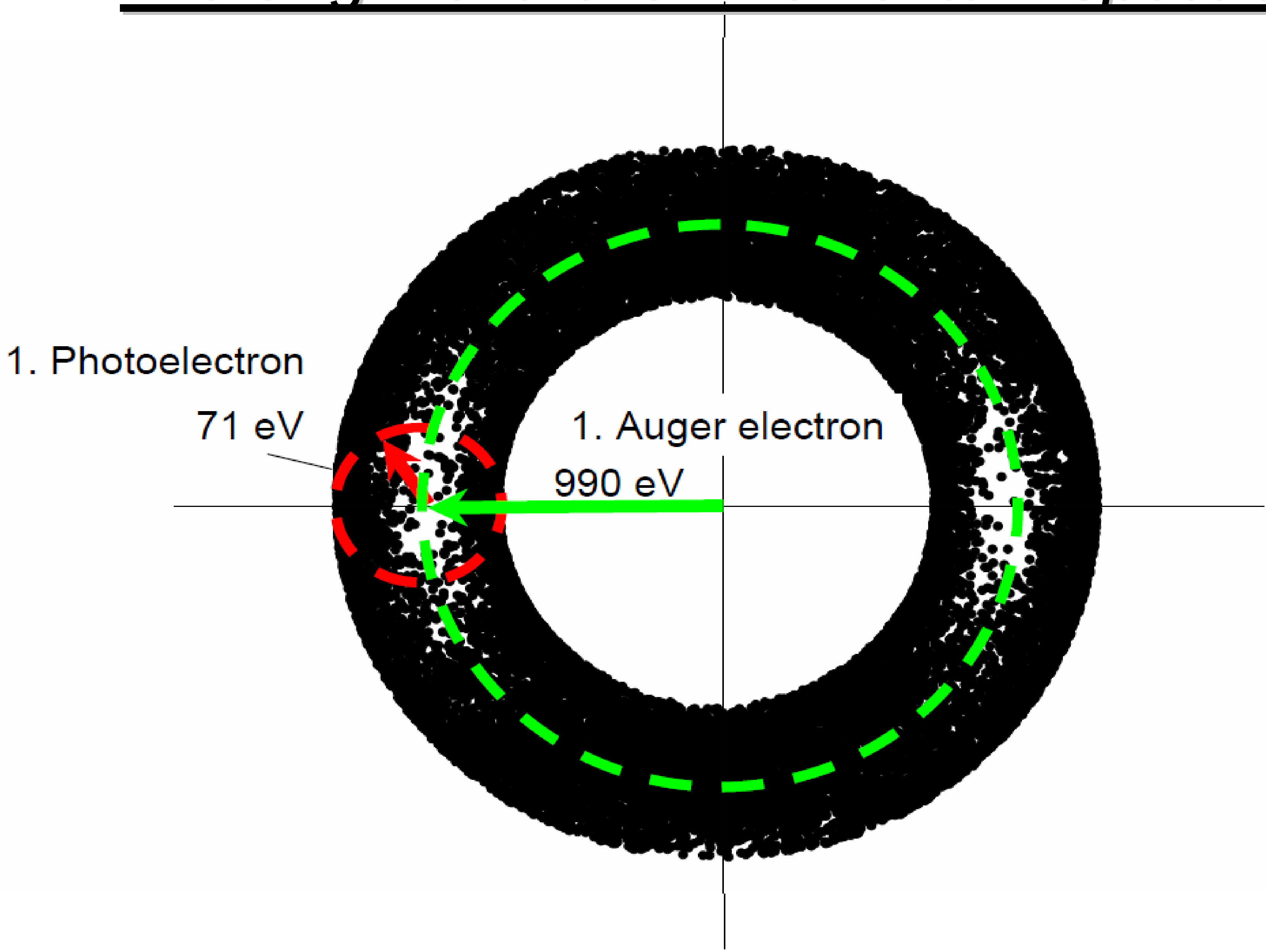
Na^{4+} recoil ion momentum
Cut in $P_{||}$, P_x plane



1 Auger electron (935 eV) in coincidence

2 Auger electrons (935 eV + 990 eV)

The Origin of the Ion Momentum Spectra



2. Example E_γ = 1216 eV (threshold for 2.e^- in PPA):

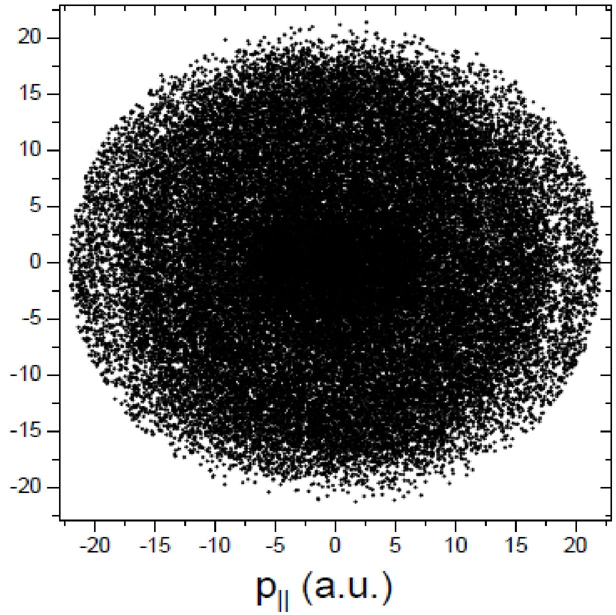
$Na + \gamma\ (1216\ eV) \rightarrow Na^{4+}$

	PAP		PPA
1.e^-)	137 eV pe^-	↔	137 eV pe^-
2.e^-)	990 eV Ae^-	↘ / ↙	≈ 0 eV pe^-
3.e^-)	66 eV pe^-	↗ / ↖	1056 eV Ae^-
4.e^-)	935 eV Ae^-	↔	935 eV Ae^-

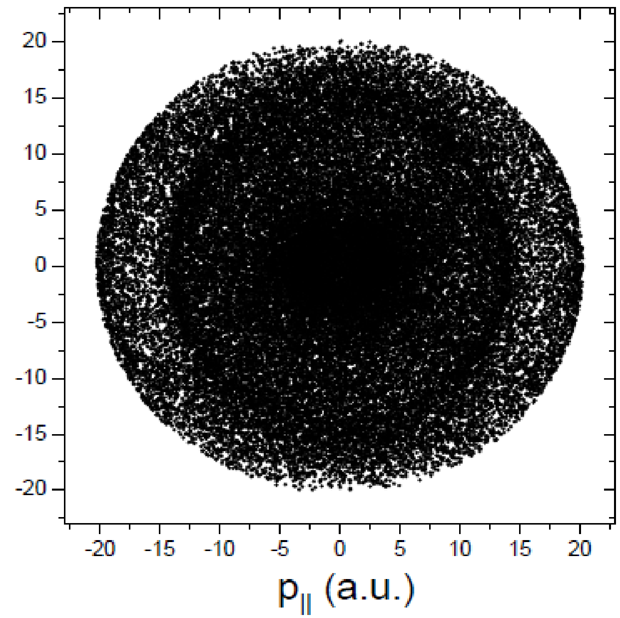
Na^{4+} recoil ion momentum
Cut in $P_{||}$, P_x plane

FEL polarization

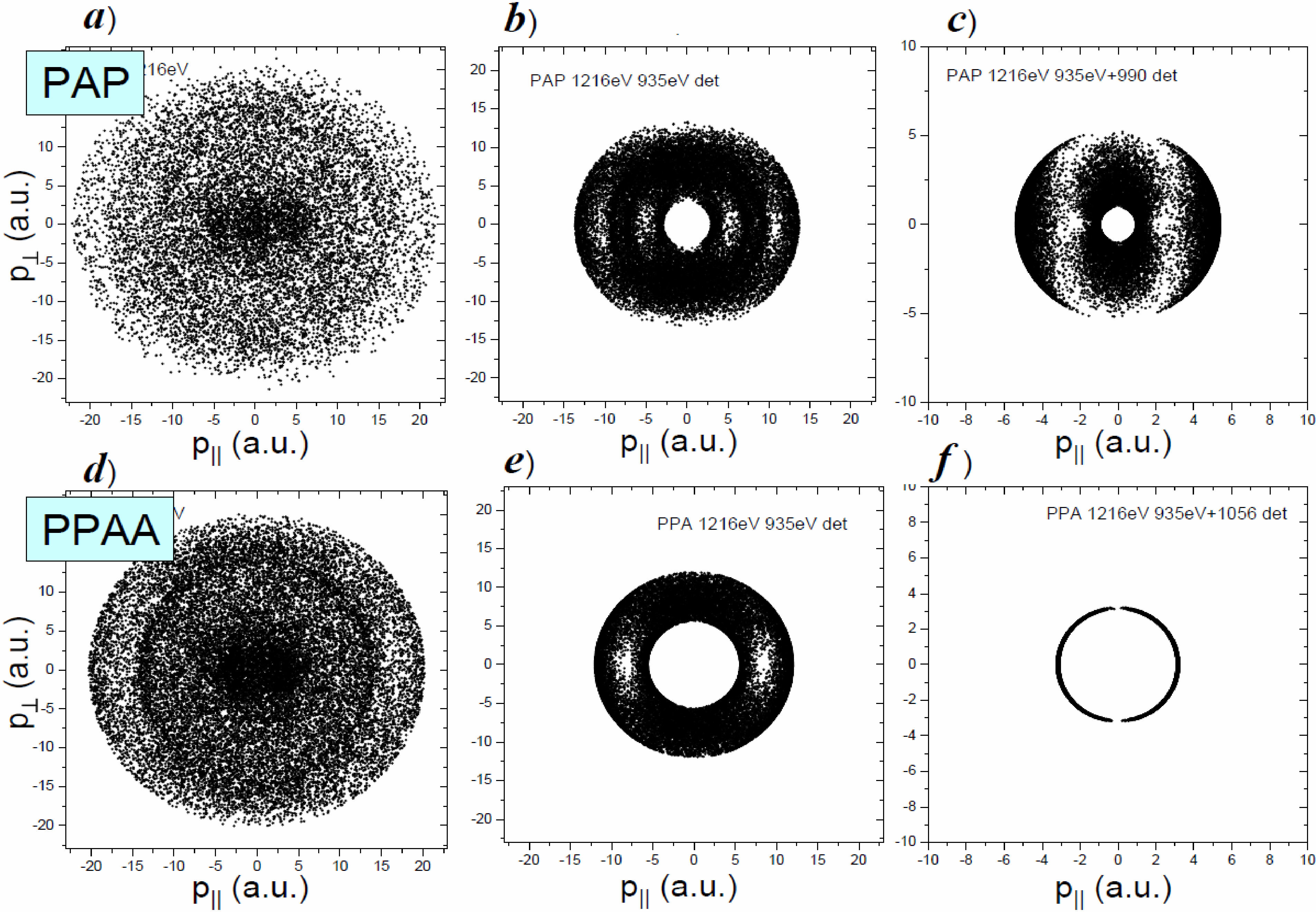
PAP



PPA



pe^-: photoelectron (dipolar)
Ae^-: Auger electron (isotropic)



a)
PAP
216eV
b)
PAP 1216eV 935eV det
c)
PAP 1216eV 935eV+990 det
d)
PPAA
V
e)
PPA 1216eV 935eV det
f)
PPA 1216eV 935eV+1056 det
p⊥ (a.u.)
p|| (a.u.)

K-edge Shifting by Laser Excitation

$Na^{+}(1s\ \ 2s^{2}2p^{6}3s\ \ {}^{3}S)$ (E_{γ} = 1079.5 eV)

$Na^{+}(1s\ \ 2s^{2}2p^{6}3p\ \ {}^{3}P)$ (E_{γ} =1083.6 eV)

- Should become possible for 0.3 % XFEL bandwidth and below.

- Allows to extract pure double K-hole contribution.

Rate Estimations

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Photons/pulse	10^{13}	
Pulse width	100 fs	
Cross section	$3 \cdot 10^{-19}$ cm^{-2}	
KLL Auger rate	$4 \cdot 10^{14}$ 1/s	

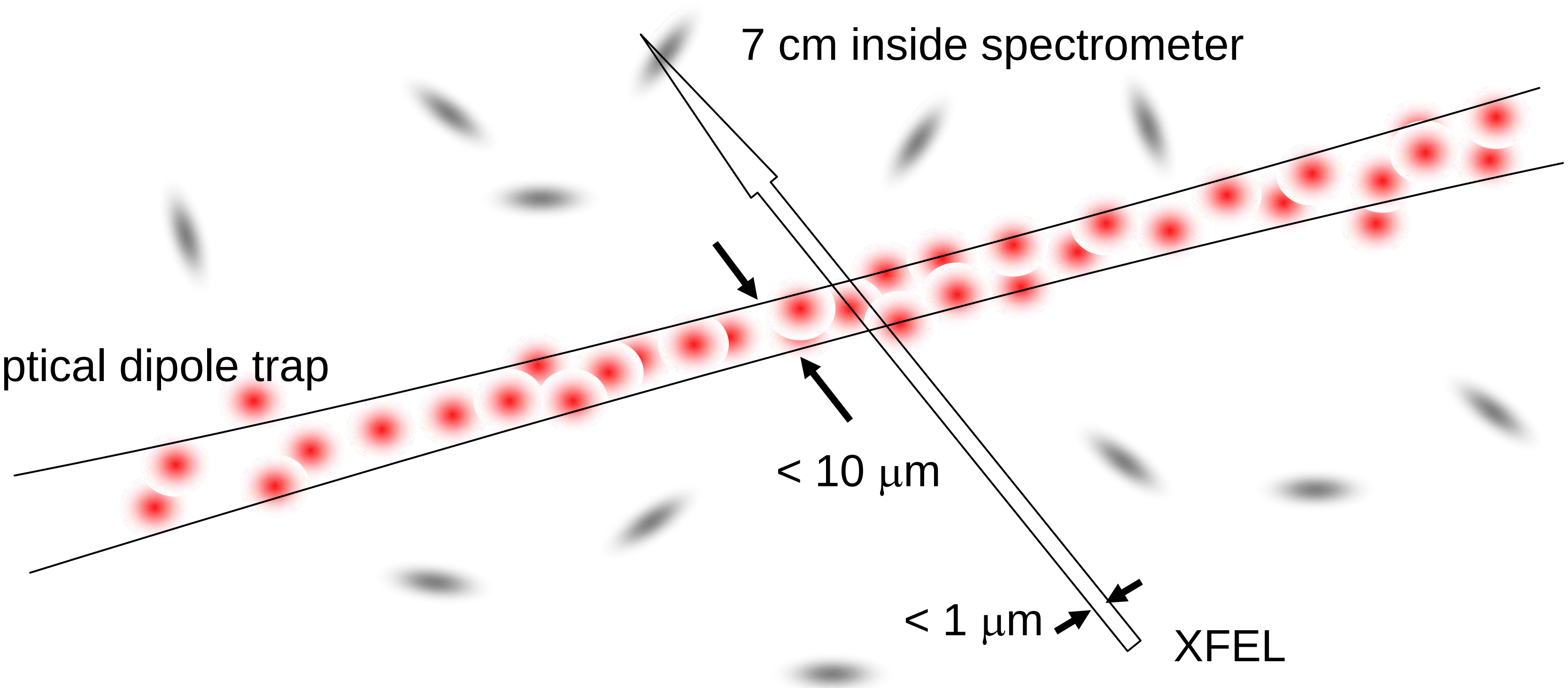
XFEL focus	20 μm	1 μm
Ionization rate	10^{13} 1/s	$4 \cdot 10^{15}$ 1/s
Probability for Multiple ionization	$2.5 \cdot 10^{-2}$	1

Count Rate Estimation

Target density $\rho = 1 \cdot 10^{11}\ 1/cm^3 = 1/\ 10\mu m^3$ → 1 target atom in the XFEL beam

Background ($1 \cdot 10^{-11}$ mbar) $\rho = 3 \cdot 10^{5}\ 1/cm^3$ → 0.02 background atom in the beam

→ **XFEL beam diameter decisive**



Summary

- Successful commissioning of Motrims at FLASH, electron – ion coincidences are envisaged.

- Manipulation/control of electron correlation in photo-double ionization.

- Photo-triple ionization requires $>10^{15}$ photons/sec. @ >> 10 kHz rep. rate.

- XFEL proposal aims at sodium for first studies.

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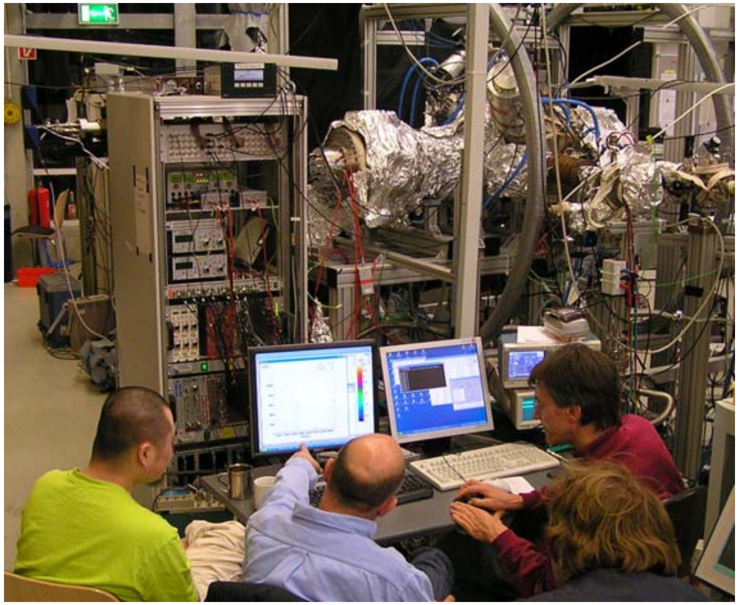

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