

***Atomic, molecular and cluster science  
using X-ray FELs  
with momentum imaging***

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Tohoku University, Japan***

*Matsushima*

# *X-FEL and EUV-FEL Facility in Japan*

X-FEL (will be in operation in 2010)

EUV-FEL  
(in operation)

SPring-8



# Outline

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## 1. Introduction to ion momentum imaging

Historical background

Towards zero dead-time detections

Apparatus for ion momentum imaging

## 2. EUVFEL experiments

3-1. Higher harmonics probed by He recoil momentum spectroscopy

3-2. Sequential double and triple ionization of atoms and diatoms

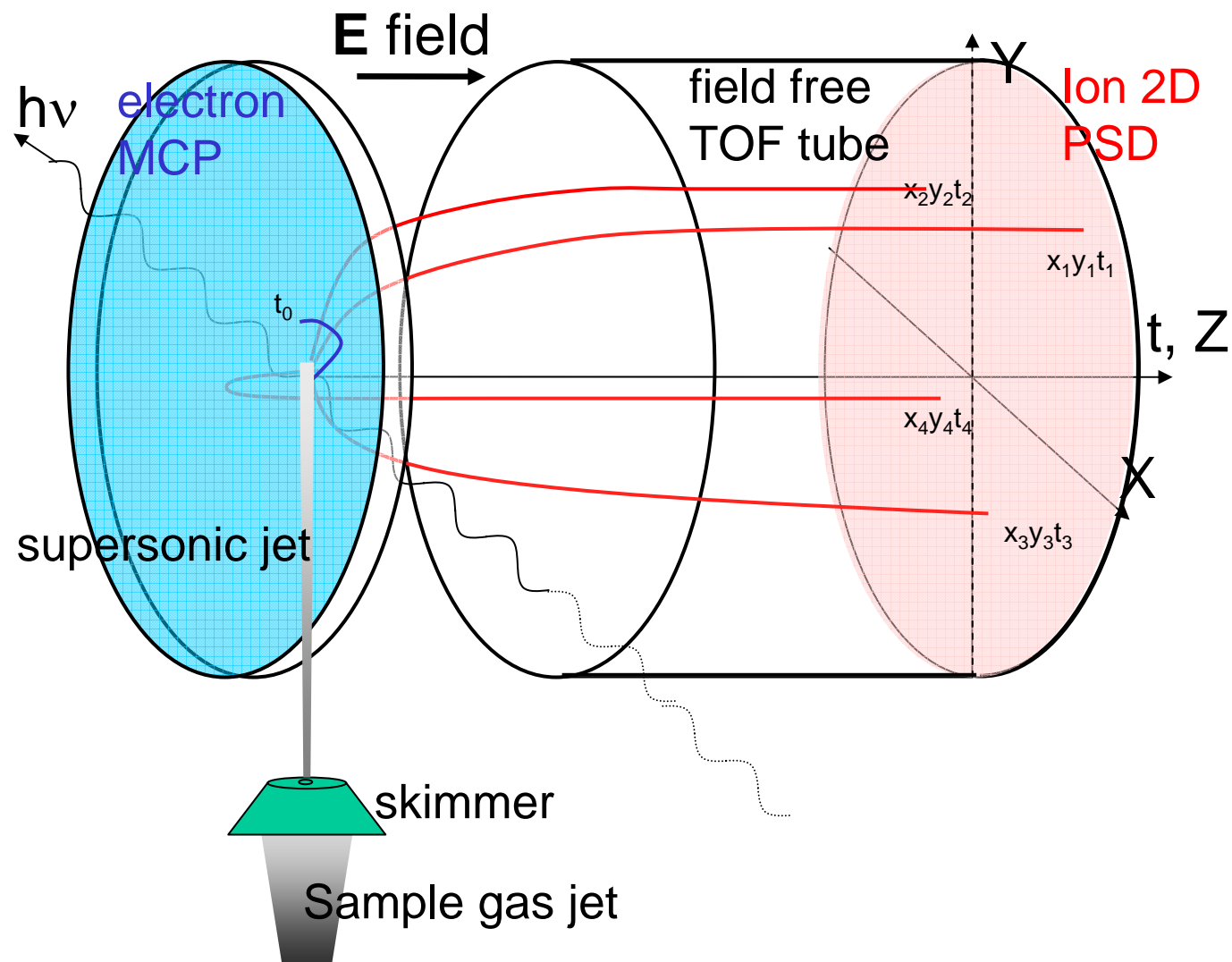
3-3. Step-wise multiple ionization of rare-gas clusters

## 3. Toward future, toward XFEL experiments

Apparatus for e and ion momentum imaging

Comments on single-shot X-ray diffraction microscope

# Multiple-ion coincidence imaging setup



*position & time of flight ( $x, y, t$ )* →

*3D momentum of each particle*

# ***History of developments for momentum imaging techniques***

Combining PSD with TOF technique → momentum imaging

Multi-anode PSD: multi-hit capability

F. Heiser, U. Becker (FHI, Germany), M. Lavollée (LURE, France)

Delay-line anode PSD:

J.H.D. Eland (Oxford, UK)

O. Jagutzki, H. Schmidt-Böcking (Frankfurt, Germany)

Dead-time problem – solved by the development of **HEX anode**

(resort routine: by Achim Czasch)

Electronics:

TDC (for SR and high rep. rate laser experiments)

Multi-channel (16ch) multi-hit (up to 8 or more), common stop capability

TDC-8 (Roentdek) developed by J. Eland

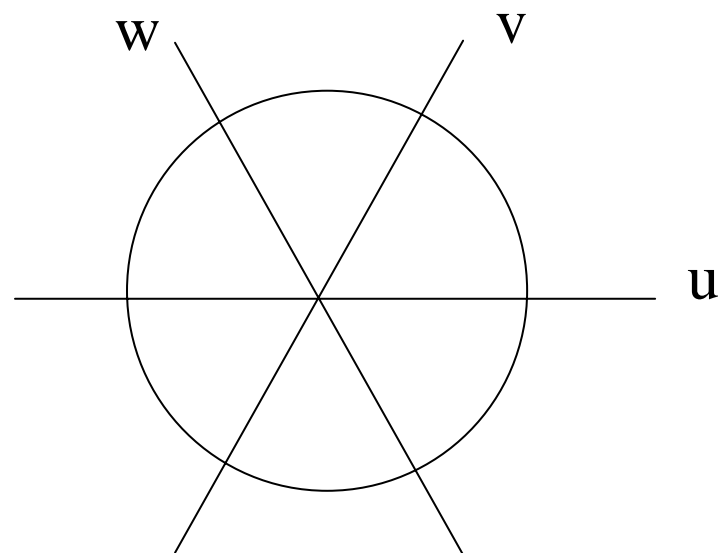
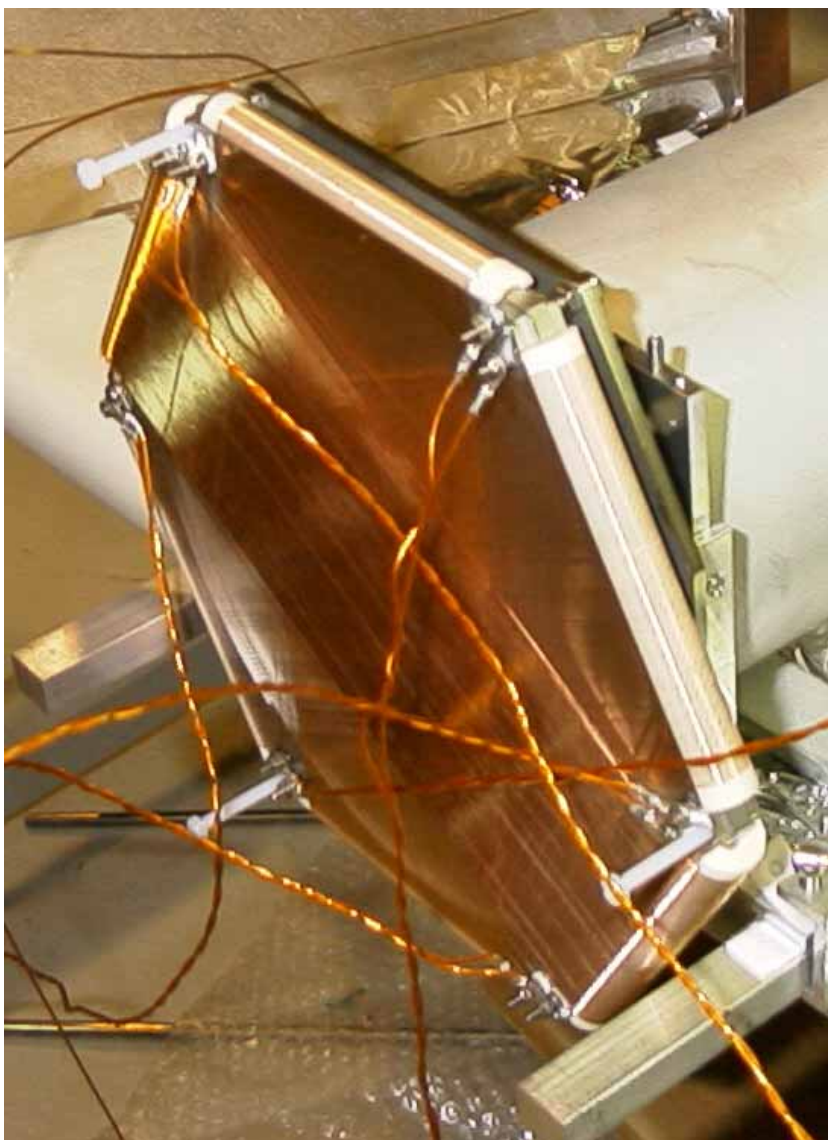
(Multi-channel TAC/TDC developed in Japan)

New TDC-8HP (by Till Jahnke)

**Digitizer** (for FEL)

**Soft-CFD** (by Lutz Faucher, Achim Czasch)

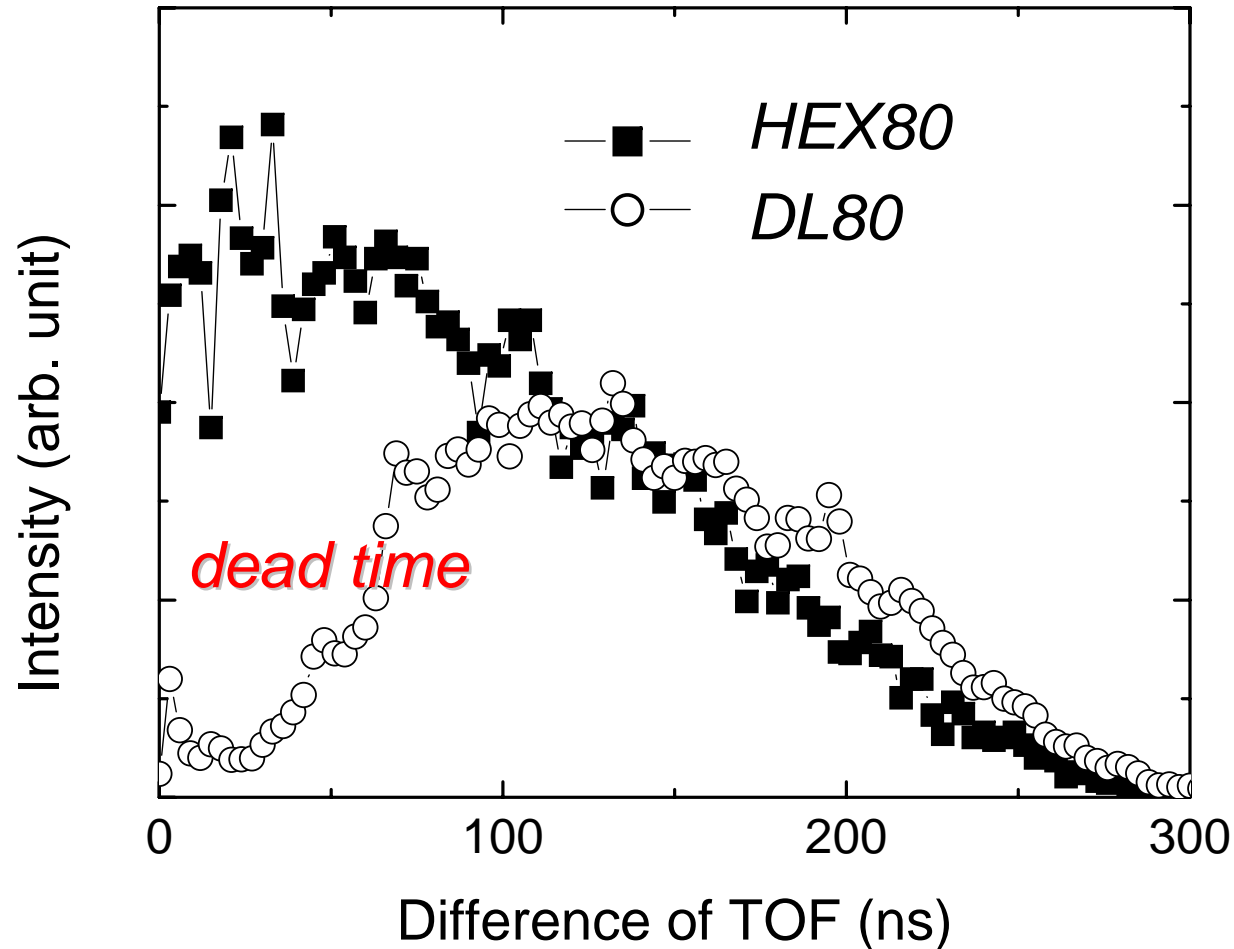
# Hexagonal delay-line anode



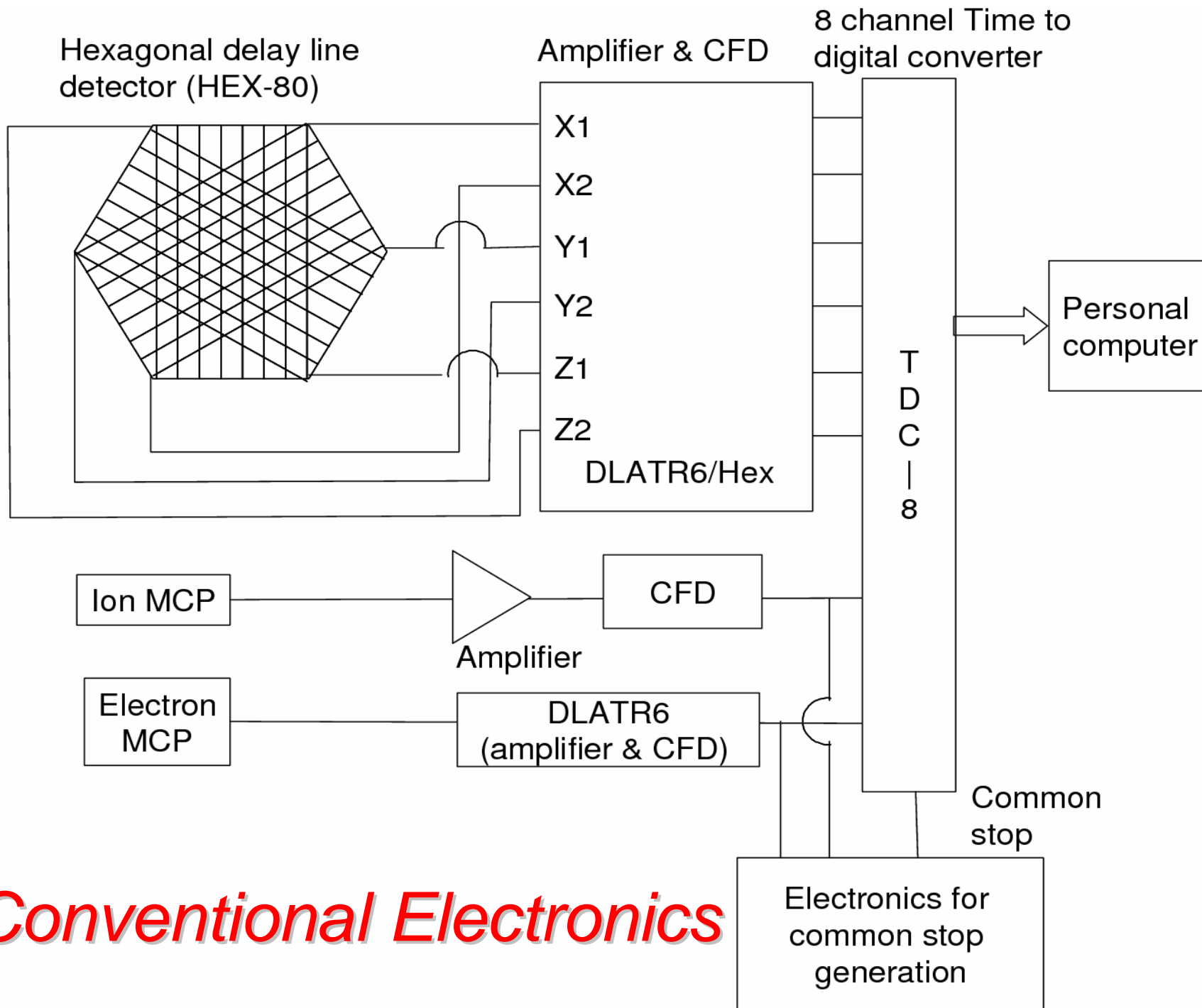
Redundant information from a three-layer delay line anode and a sophisticated logic is necessary to reconstruct time and positions for more than one “hit”s in 100 ns.

*Roentdek, A. Czasch*

# Towards zero dead time detection



Distribution of events as a function of the TOF difference of the two  $O^+$  ions produced from  $CO_2^{++}$

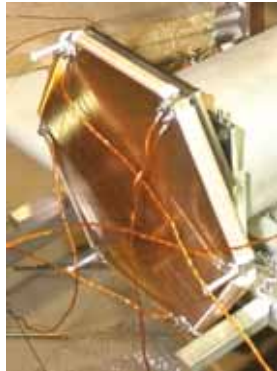


# *Conventional Electronics*

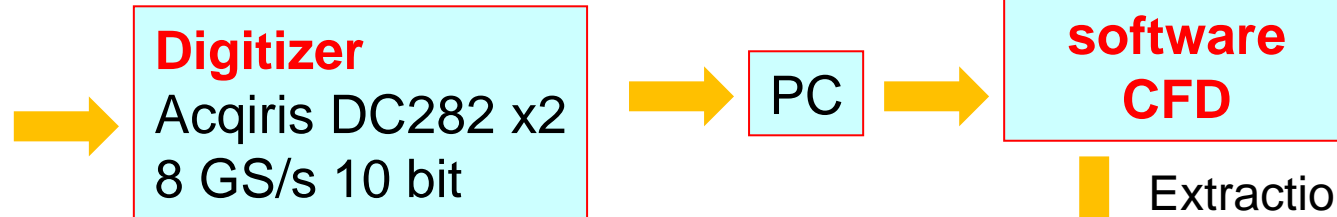
# Towards zero dead-time detection system

## Digitizer + software CFD

*L. Faucar*



HEX detector



Analog signals are fed to PC

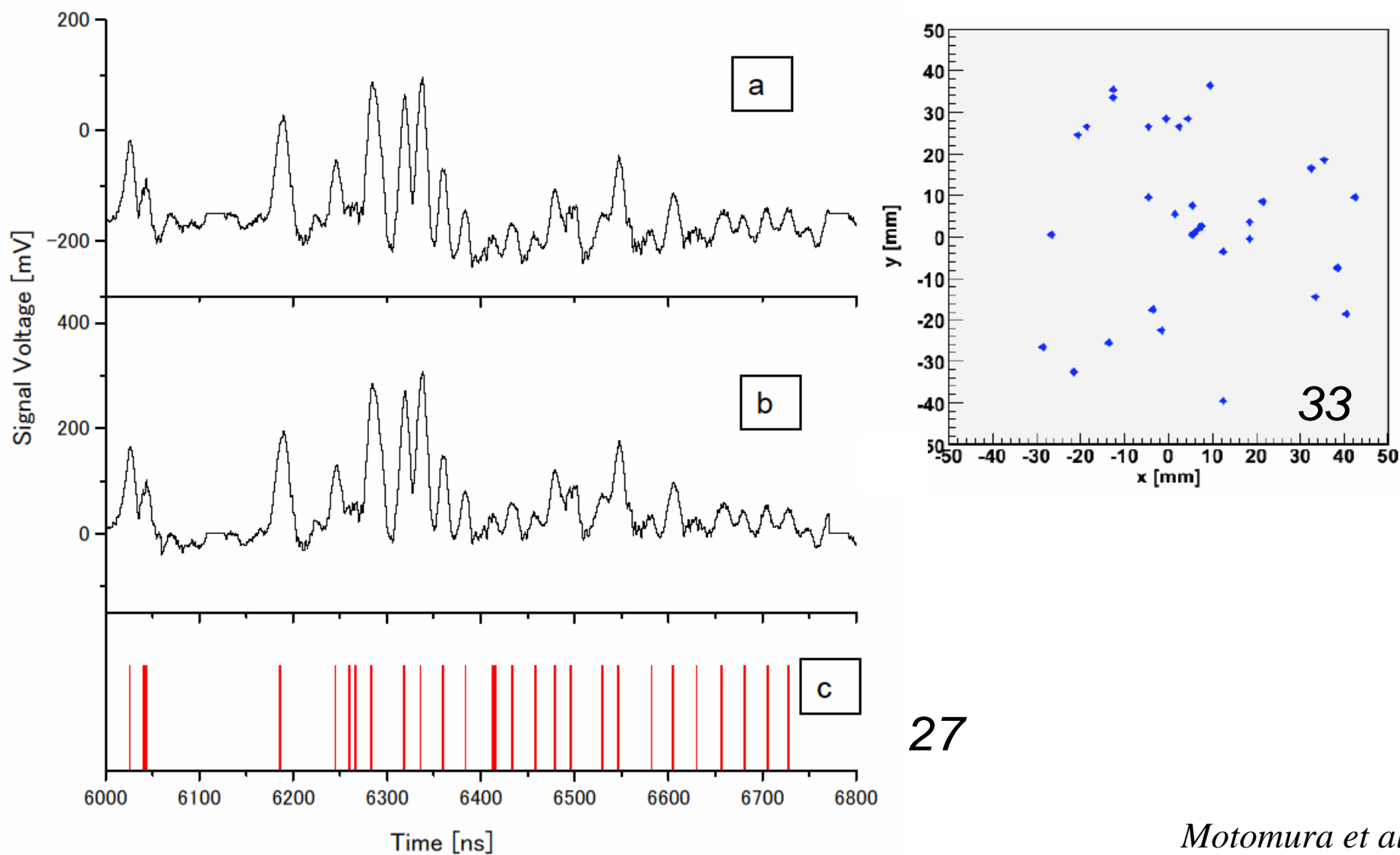


Extraction of the time information from the waveform

- TOF spectrum
- Position
- Momentum

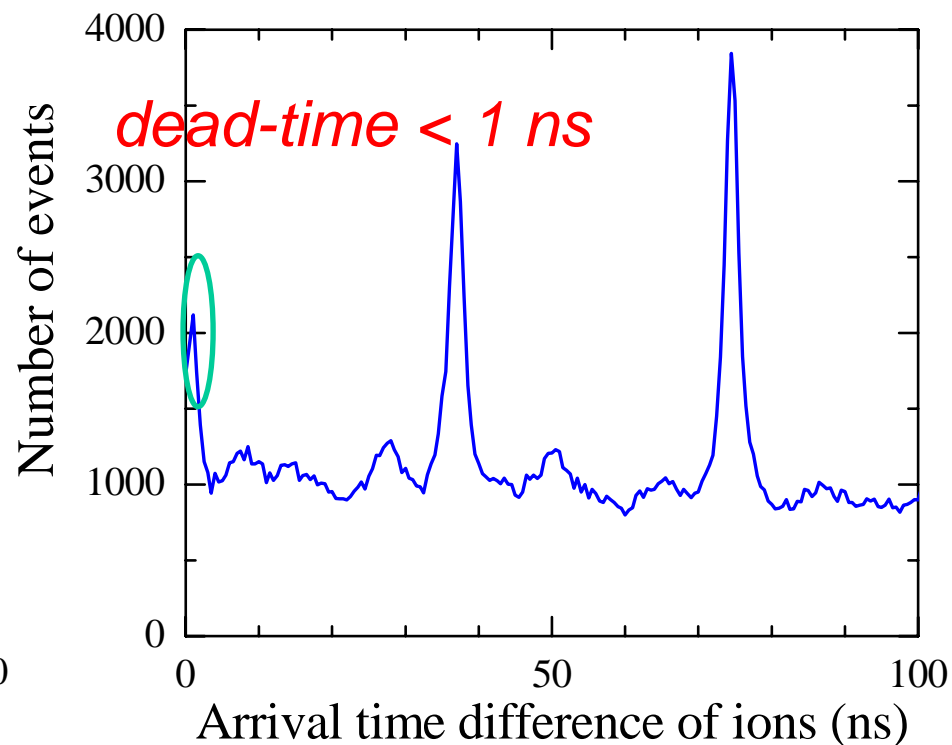
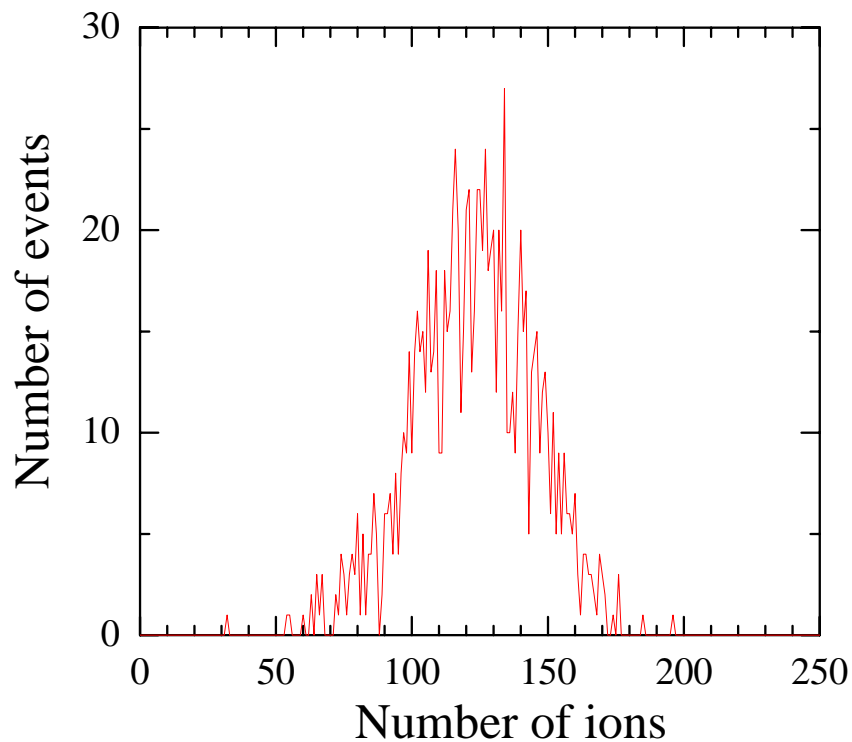
# Software CFD

*Kr<sup>+</sup> via Coulomb explosion of Kr clusters irradiated by FEL*



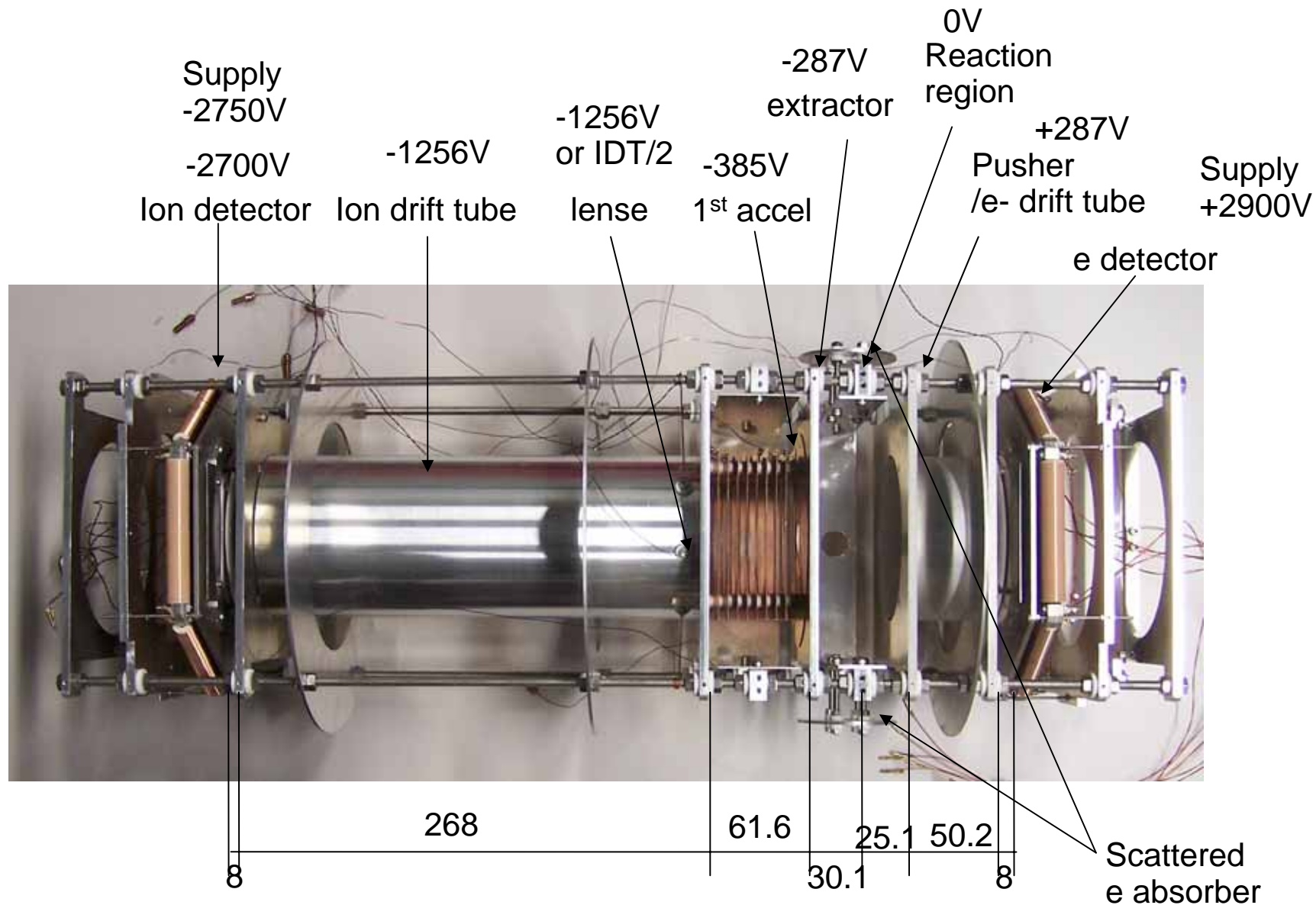
# Zero dead-time measurements

*Coulomb explosion experiment of Kr clusters by FEL*

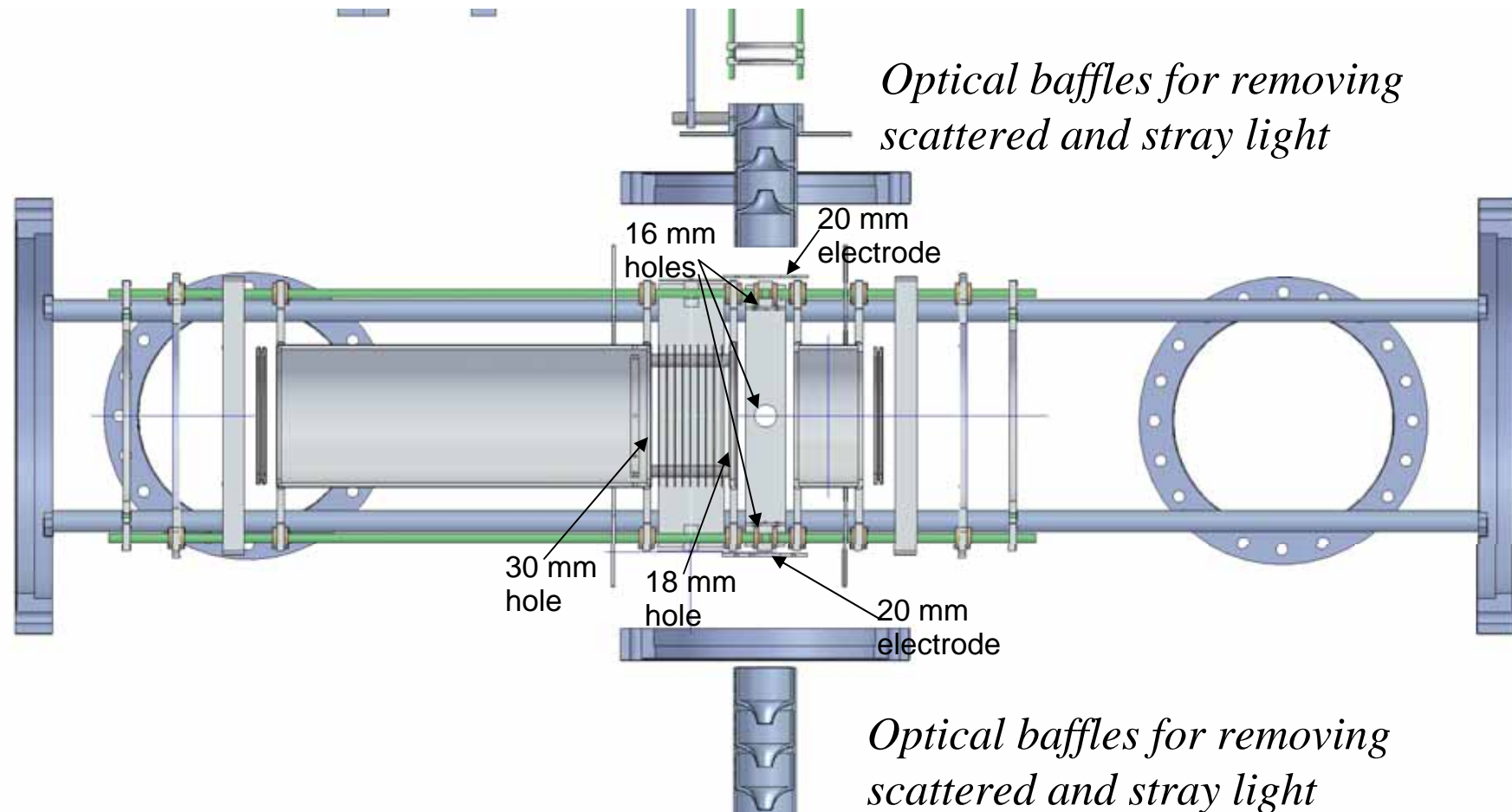


# Spectrometer and typical voltage conditions

Two-stage acceleration + velocity focusing optics



# *Spectrometer mounted in the chamber*



# ***EUV-FEL Experiments :***

*Mar 10 – Mar 21, 2008*

*Setup version 1.0*

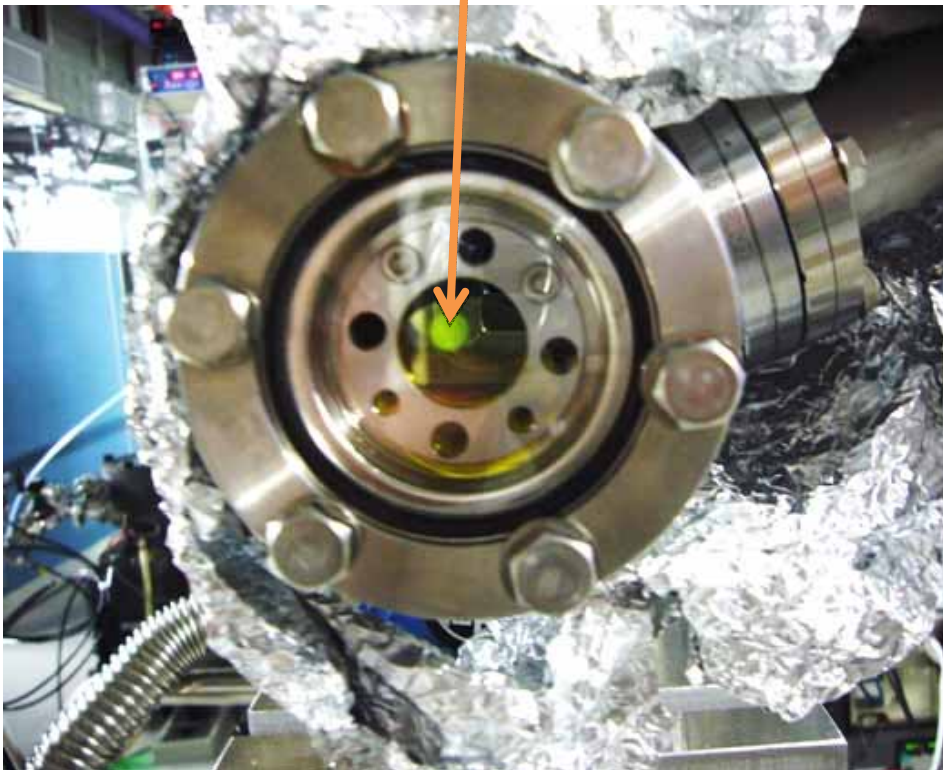
*May 26 – June 6, 2008*

**Setup version 1.1**

*Sep 29 – Oct 3, 2008*

*Setup version 2.0*

**EUV-FEL light**



# *Multiple ionization of rare gas clusters by intense EUV-FEL at SPring-8*

Tohoku U.

H. Fukuzawa

X.J. Liu

G. Pruemper

M. Okunishi

K. Shimada

K. Ueda

Apparatus &  
Measurements

Tohoku U.

T. Harada

M. Toyoda

M. Yanagihara

M. Yamamoto

Optics

Kyoto U.

H. Iwayama

K. Nagaya

M. Yao

Cluster source

AIST

K. Motomura

N. Saito

Analysis  
software

Special Thanks

RIKEN SCSS test accelerator operation group

IMRAM technical service section, Tohoku U.

A. Belkacem & LBL optics group

Budget support: MEXT, JSPS, MPG

MPI Heidelberg

A. Rudenko

J. Ullrich

Measurements

Frankfurt U.

L. Foucar

A. Czasch

R. Doerner

Acquisition  
software

RIKEN

M. Nagasono

A. Higashiya

M. Yabashi

T. Ishikawa

R. Tanaka

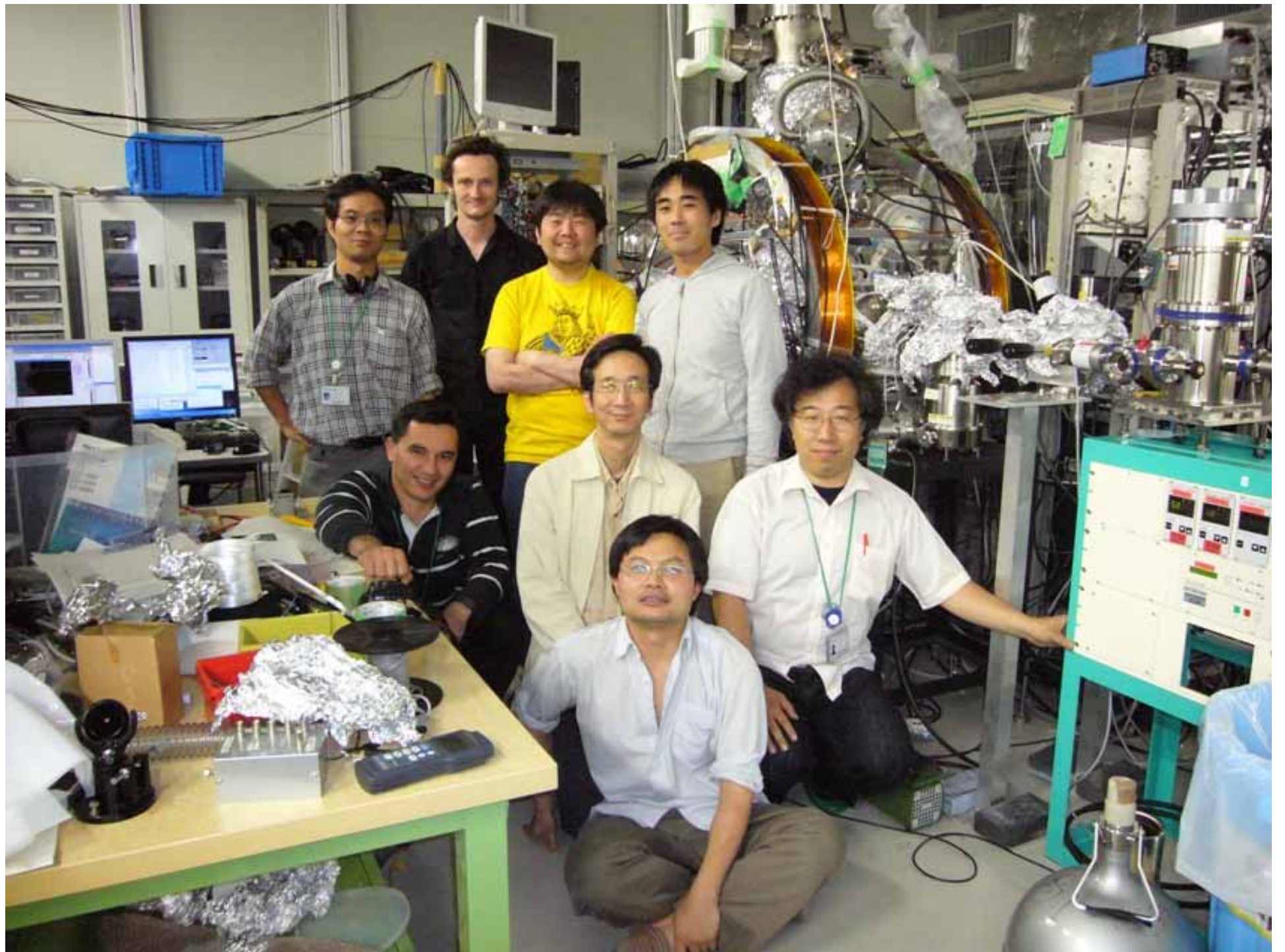
Light source

RIKEN/JASRI

H. Kimura

H. Ohashi

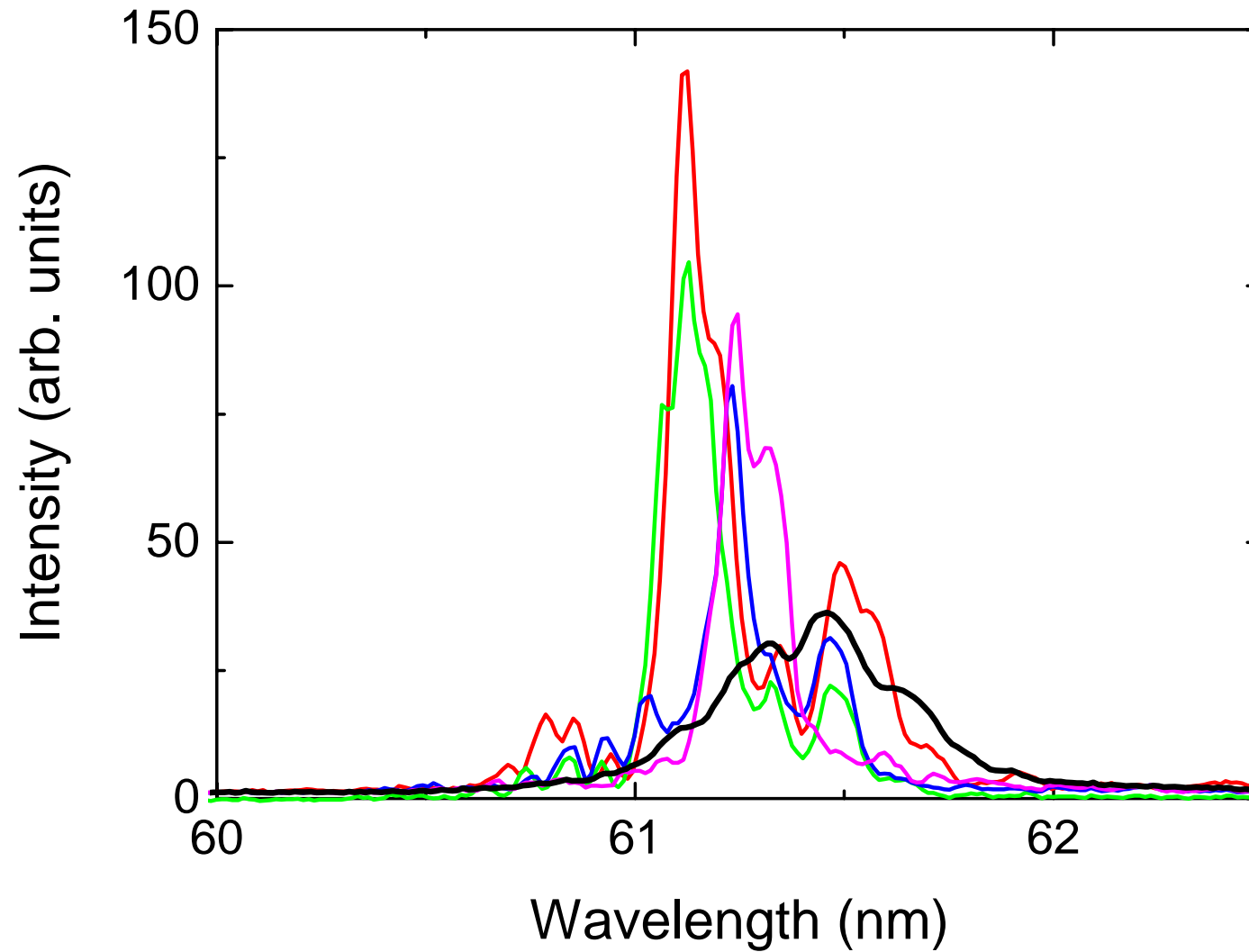
Beam line



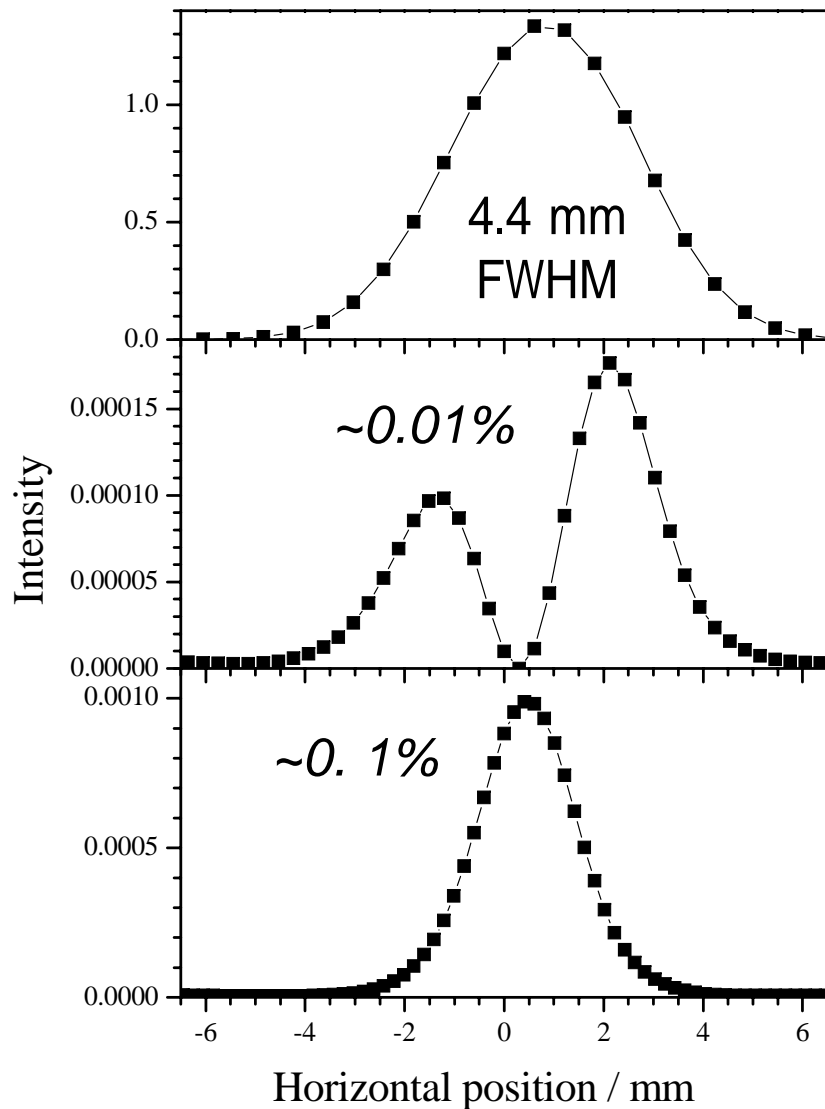
# *Characteristics of EUV-FEL*

Energy range	<b>51 - 61 nm (20 - 24 eV)</b>
Band width	< 1 %
Pulse Energy	> 10 $\mu$ J at 61 nm
Stability of pulse energy	< 20 %
Pulse width	~ 100 fs
Period of Bunch	10 Hz ( 60 Hz in future)
Polarization	Horizontal (> 99 %)
Higher harmonics at 54 nm	Second < 0.1 % Third < 1 %

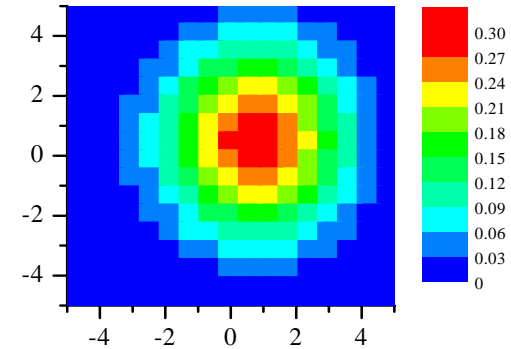
# *Example of EUV-FEL spectra*



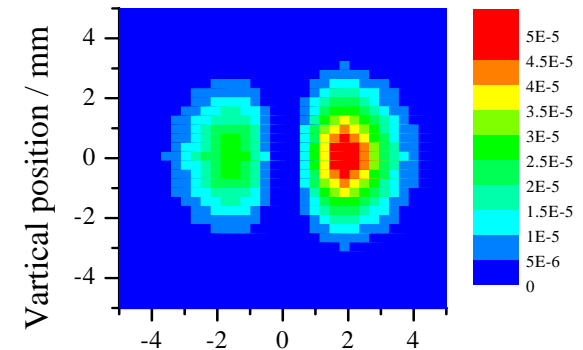
# Higher order harmonics



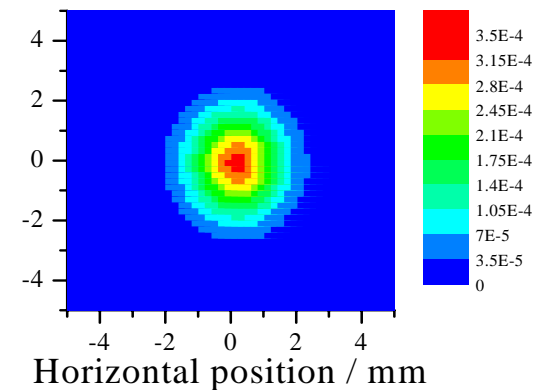
Simulation  
29 m from  
source point  
*first*



*second*



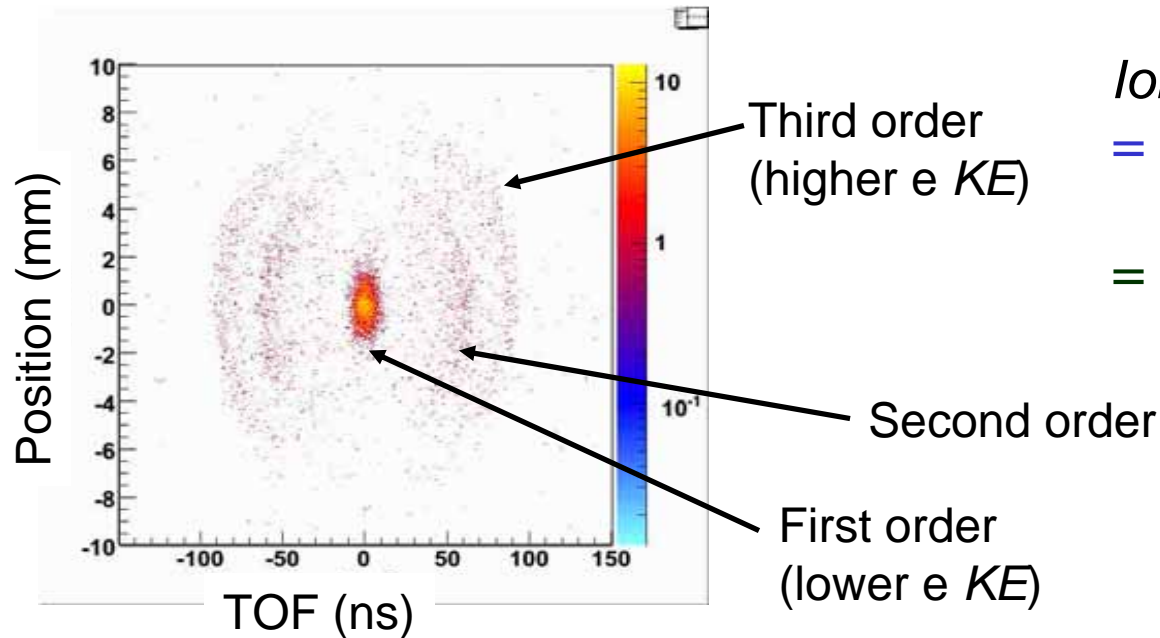
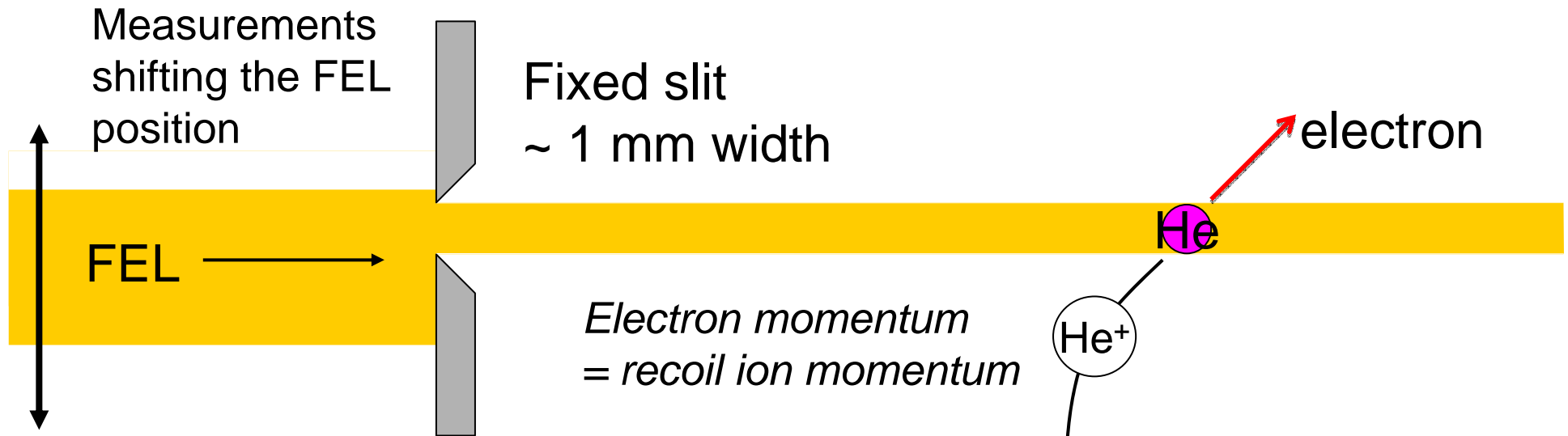
*third*



Tanaka

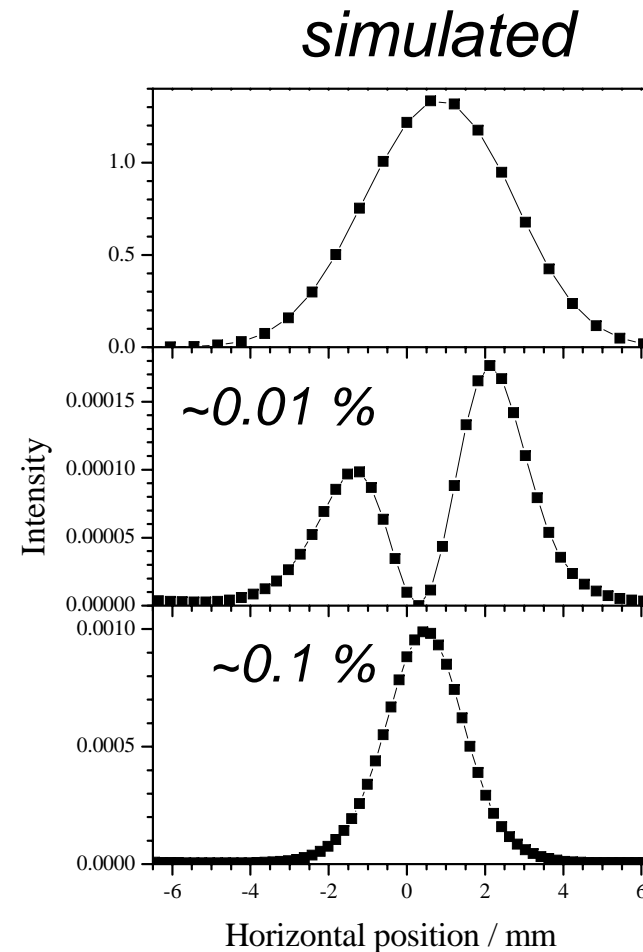
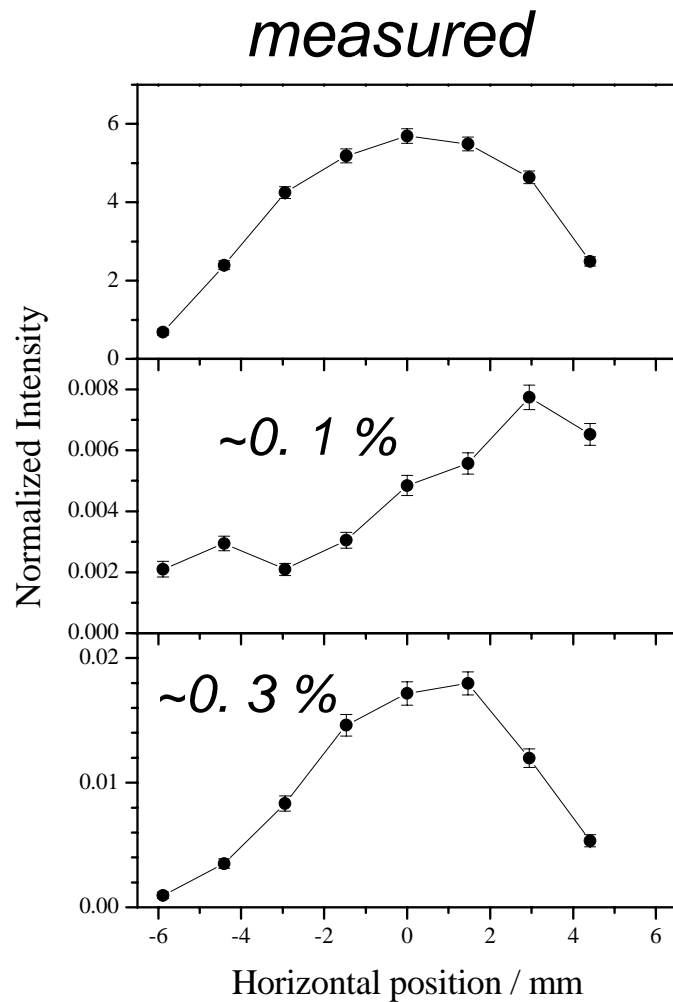
Task: measurements of relative intensities and spatial distributions

# Intensity distribution measurements for the higher order lights



Ion momentum imaging  
= Recoil-ion momentum spectroscopy  
= energy-resolved electron angular-distribution measurements

# Intensity distributions for the higher order lights

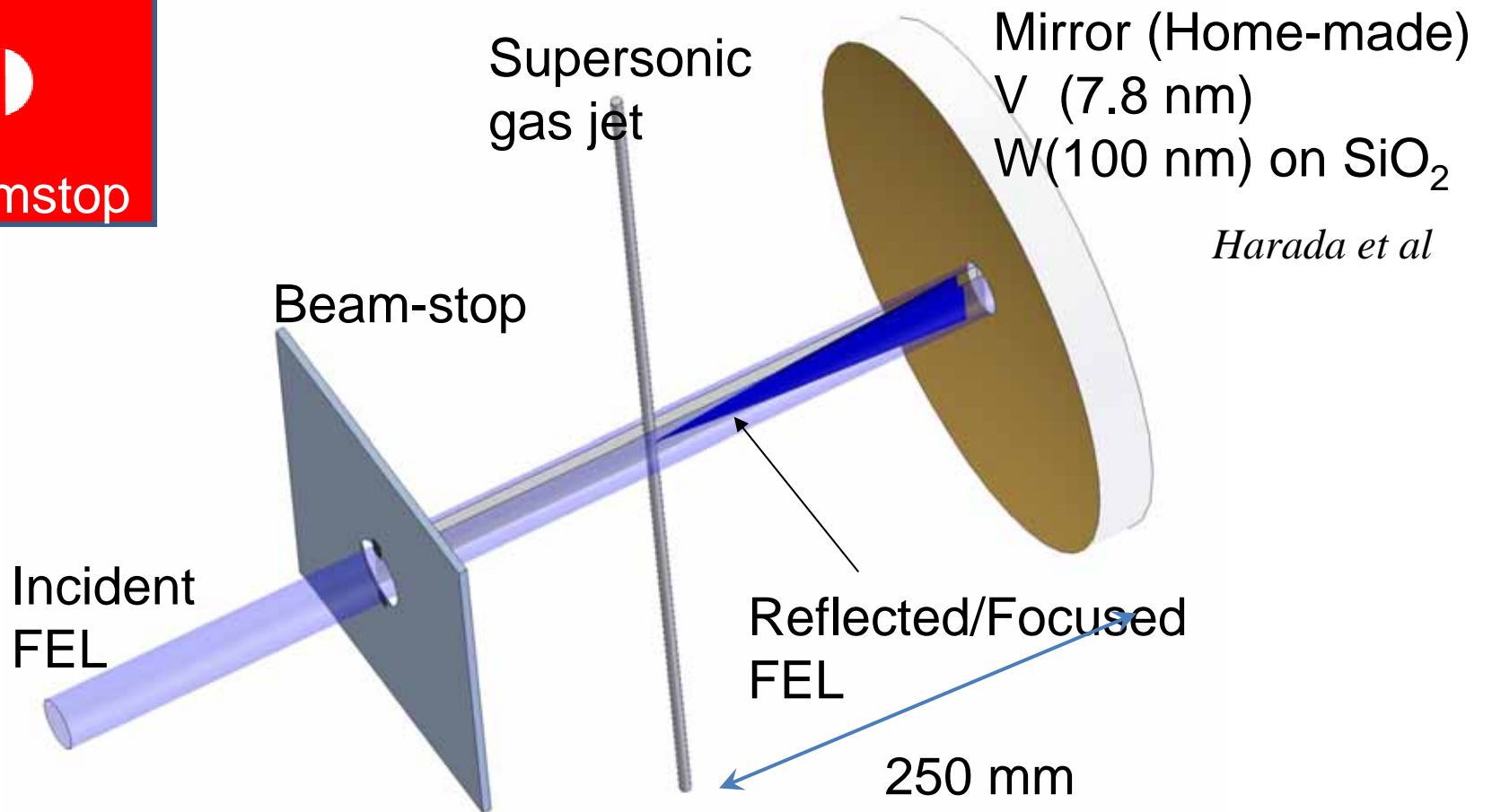


*FEL beam is broader than expected*

*Contributions from the second order light is one order more than expected*

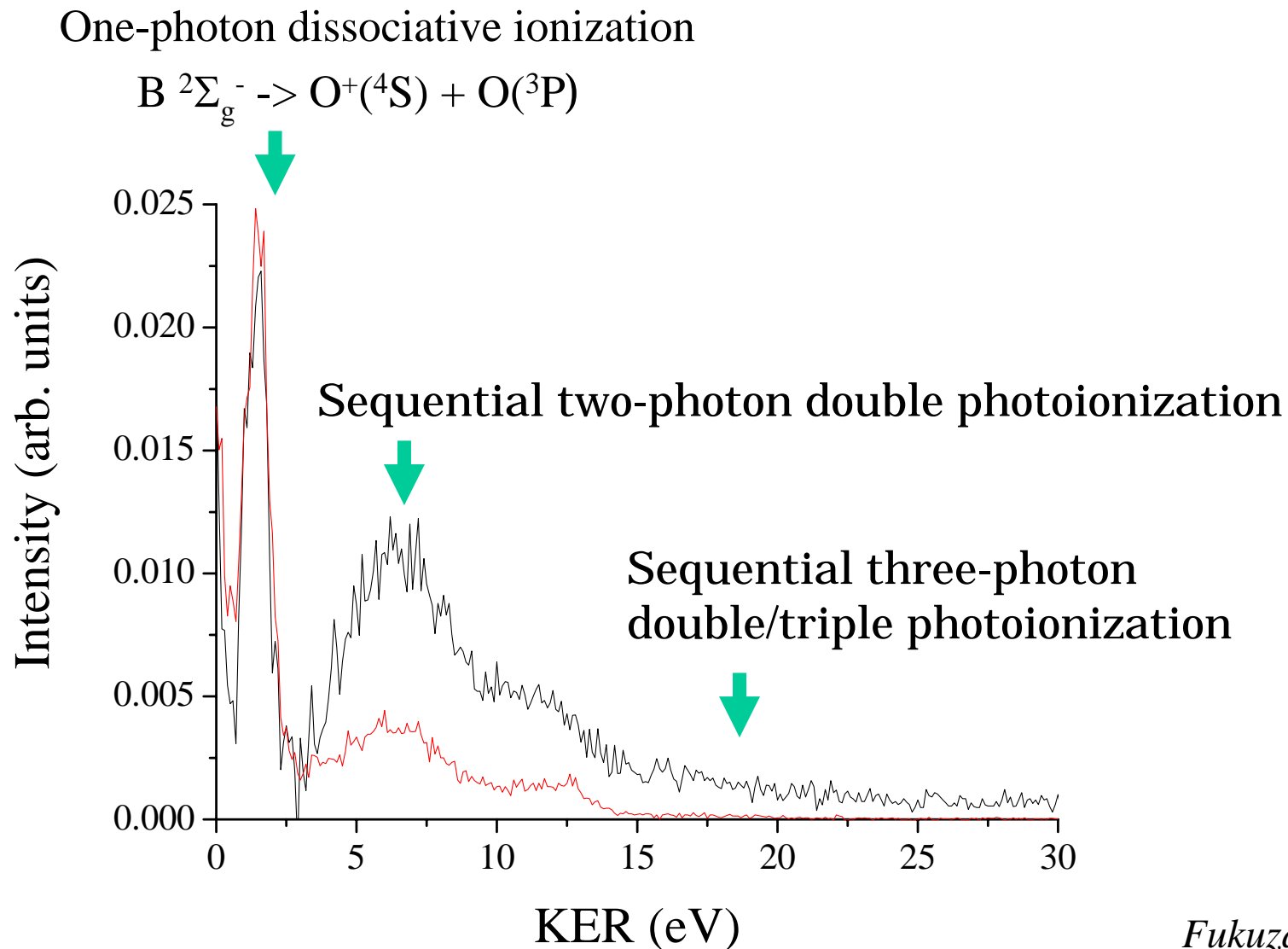
*The electron beam may not be completely focused: Instability of the electron beam?*

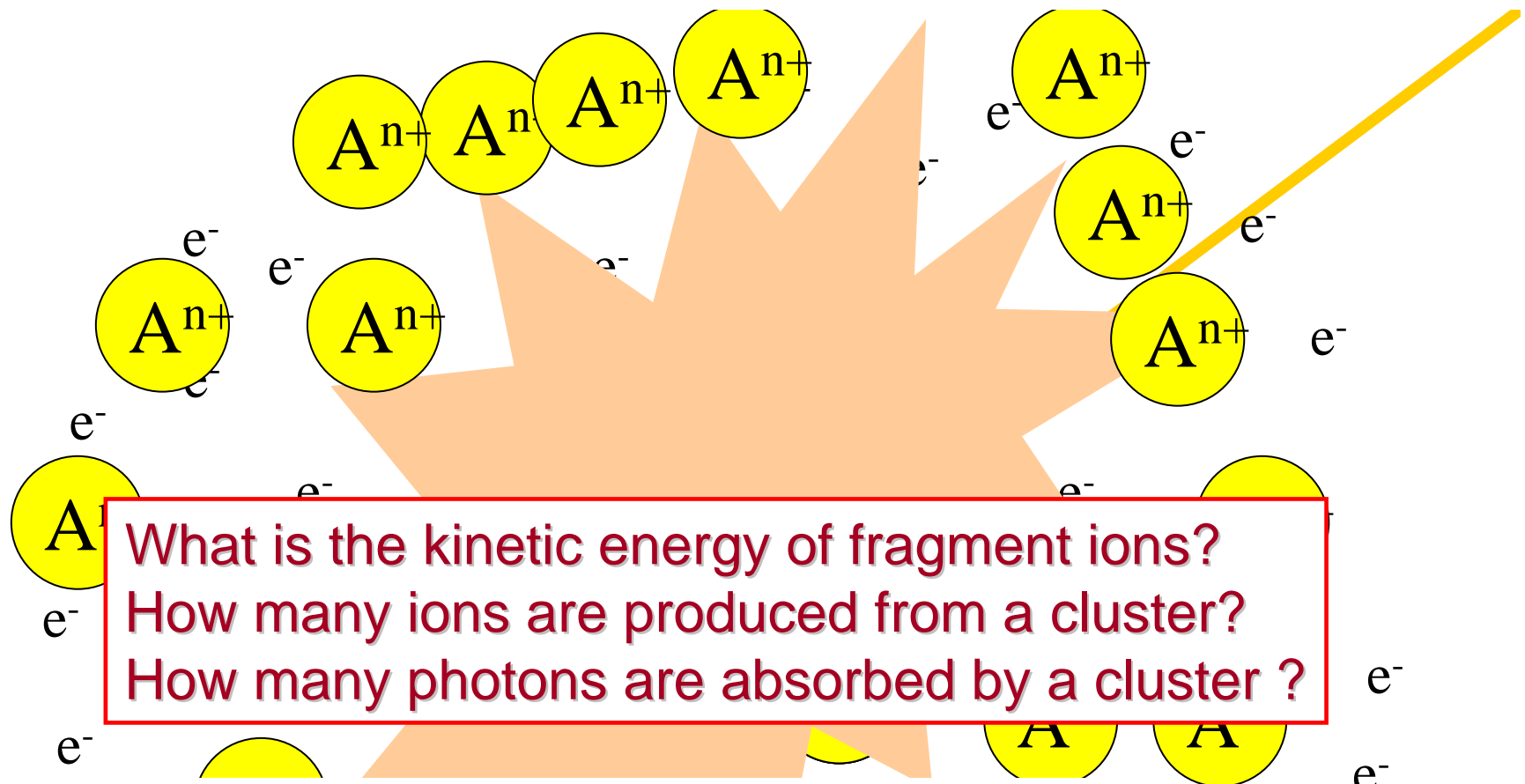
# Focusing the FEL



Beam-stop cuts the FEL along the gas jet so as not to cross the gas beam.

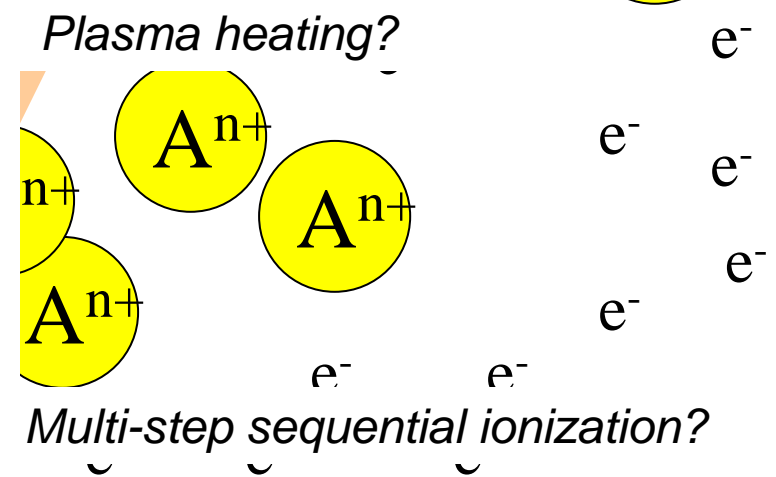
# Laser power dependence of KER distributions for $O_2$ irradiated by 51 nm fEL





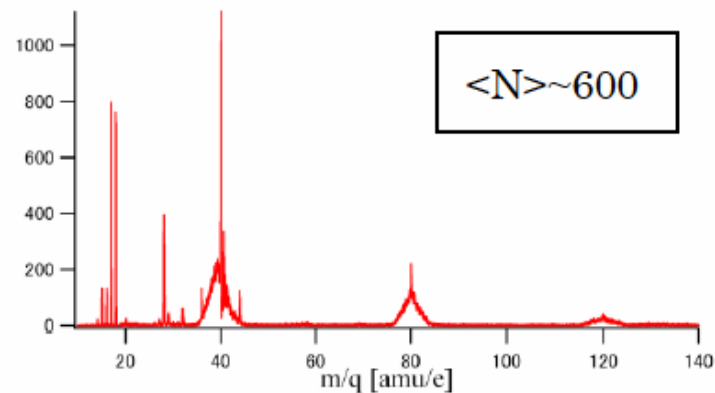
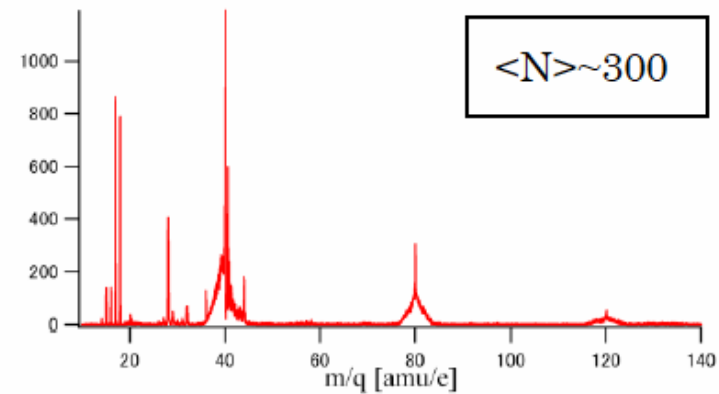
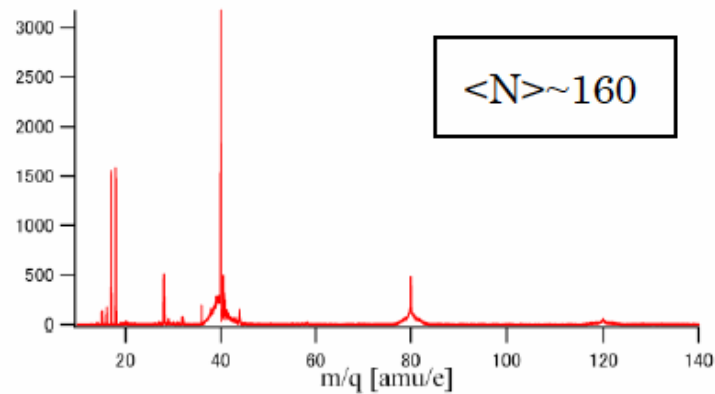
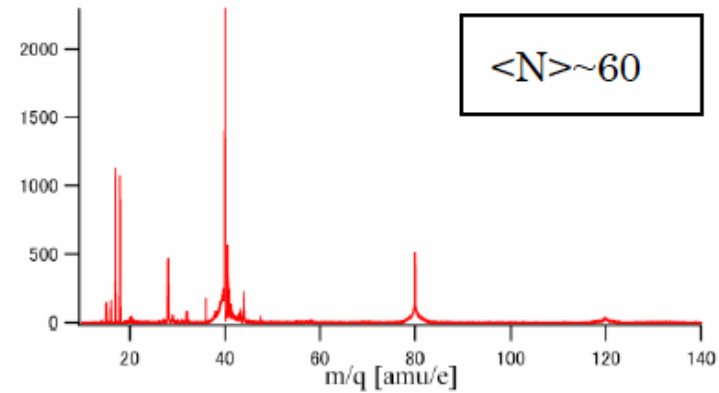
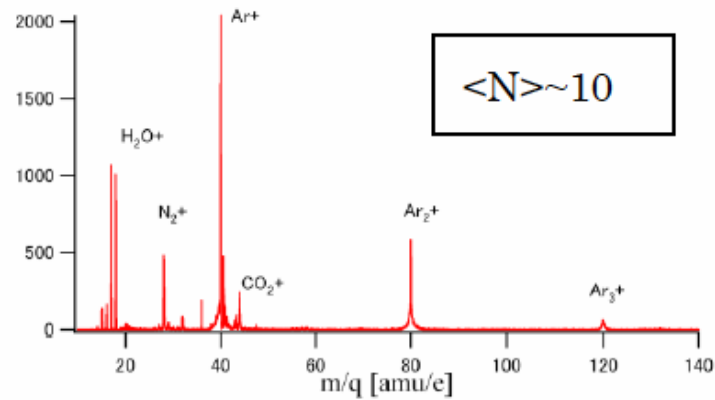
What is the kinetic energy of fragment ions?  
 How many ions are produced from a cluster?  
 How many photons are absorbed by a cluster ?

H. Wabnitz et al.,  
*Nature* 420, 482 (2002).  
 T. Laarmann et al.,  
*Phys. Rev. Lett.* 92, 143401 (2004).  
 T. Laarmann et al.,  
*Phys. Rev. Lett.* 95, 063402 (2005).  
 C. Bostedt et al.,  
*Phys. Rev. Lett.* 100, 133401 (2008).

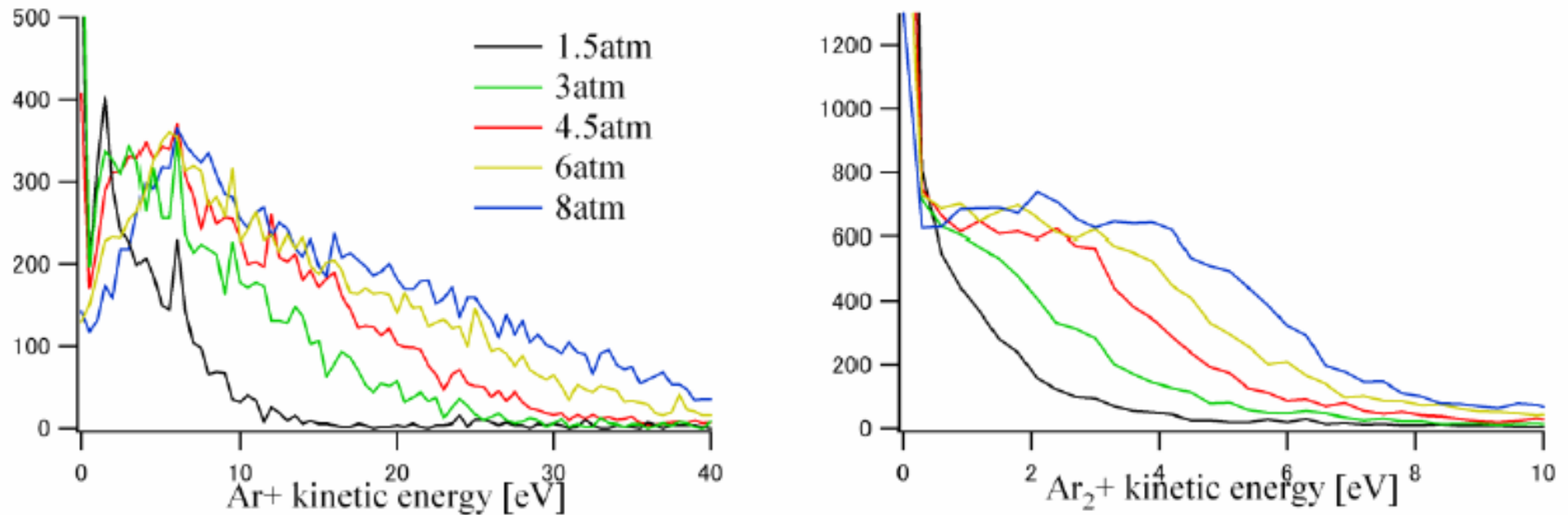


# Ar cluster mass spectra vs cluster size at 60 nm

Time-of-flight spectra



## *Kinetic energy distributions of $\text{Ar}^+$ and $\text{Ar}_2^+$ ions vs. cluster size measured at 60 nm*



*If the number of absorbed photons increases with an increase in the cluster size, then the charge state of the ion parent cluster and thus the kinetic energy release by Coulomb explosion increase.*

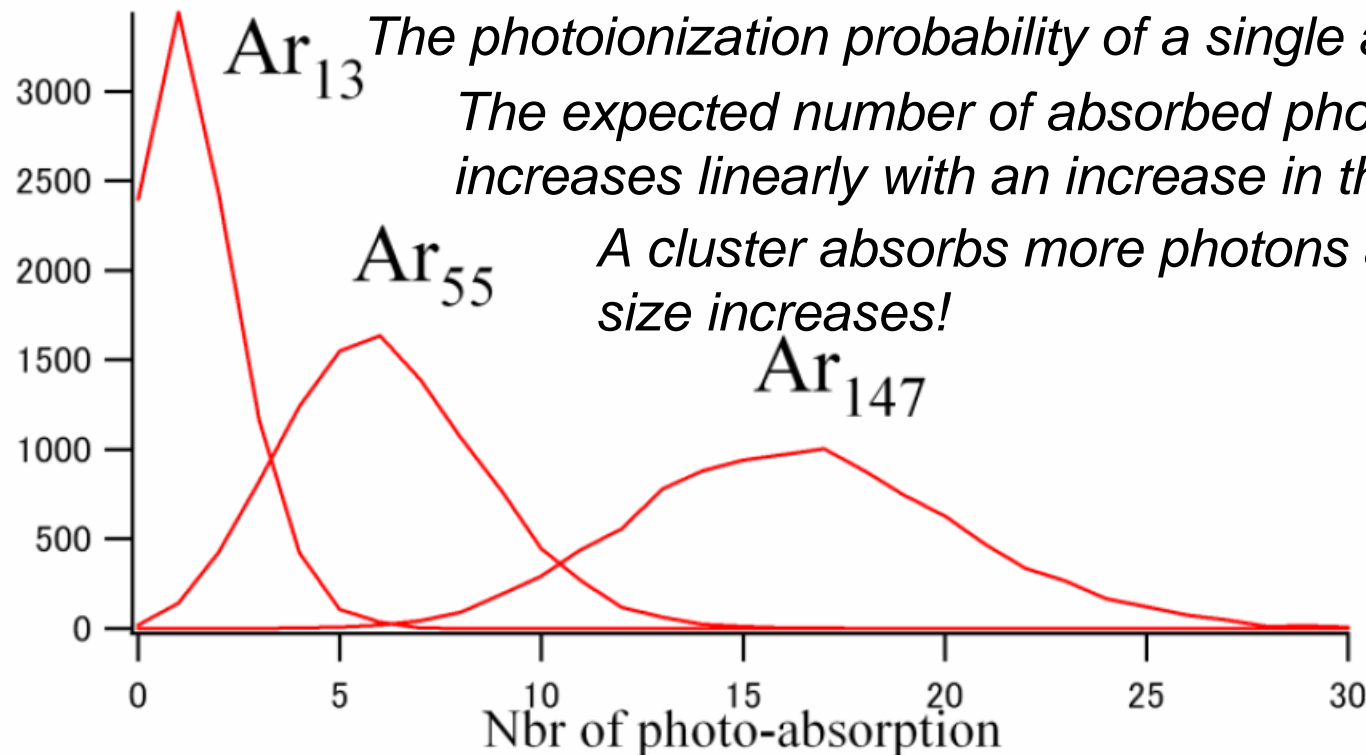
# Estimates for number of photons to be absorbed by a single cluster: cluster size dependence

FEL pulse duration : 100fs

Laser power :  $1 \cdot 10^{11}$  [W/cm<sup>2</sup>]

wavelength=60nm

cross section for 3p ionization : 35.7Mbarn



*Ar<sub>13</sub> The photoionization probability of a single atom is ~0.1.*

*The expected number of absorbed photons increases linearly with an increase in the cluster size.*

*A cluster absorbs more photons as the cluster size increases!*

*Ar<sub>147</sub>*

# Kinetic energy distributions of $Xe^+$ ions $Xe_{150}$ vs. laser power density, measured at 60 nm

Laser power density:

$1 \times 10^{11} \text{ W/cm}^2$

$4 \times 10^{11} \text{ W/cm}^2$

$10 \times 10^{11} \text{ W/cm}^2$

Photoionization probability  
of an Xe atom:

$P=0.095, 0.33, 0.63$

Expected number  $N$  of  
absorbed photons by  $Xe_{150}$ :

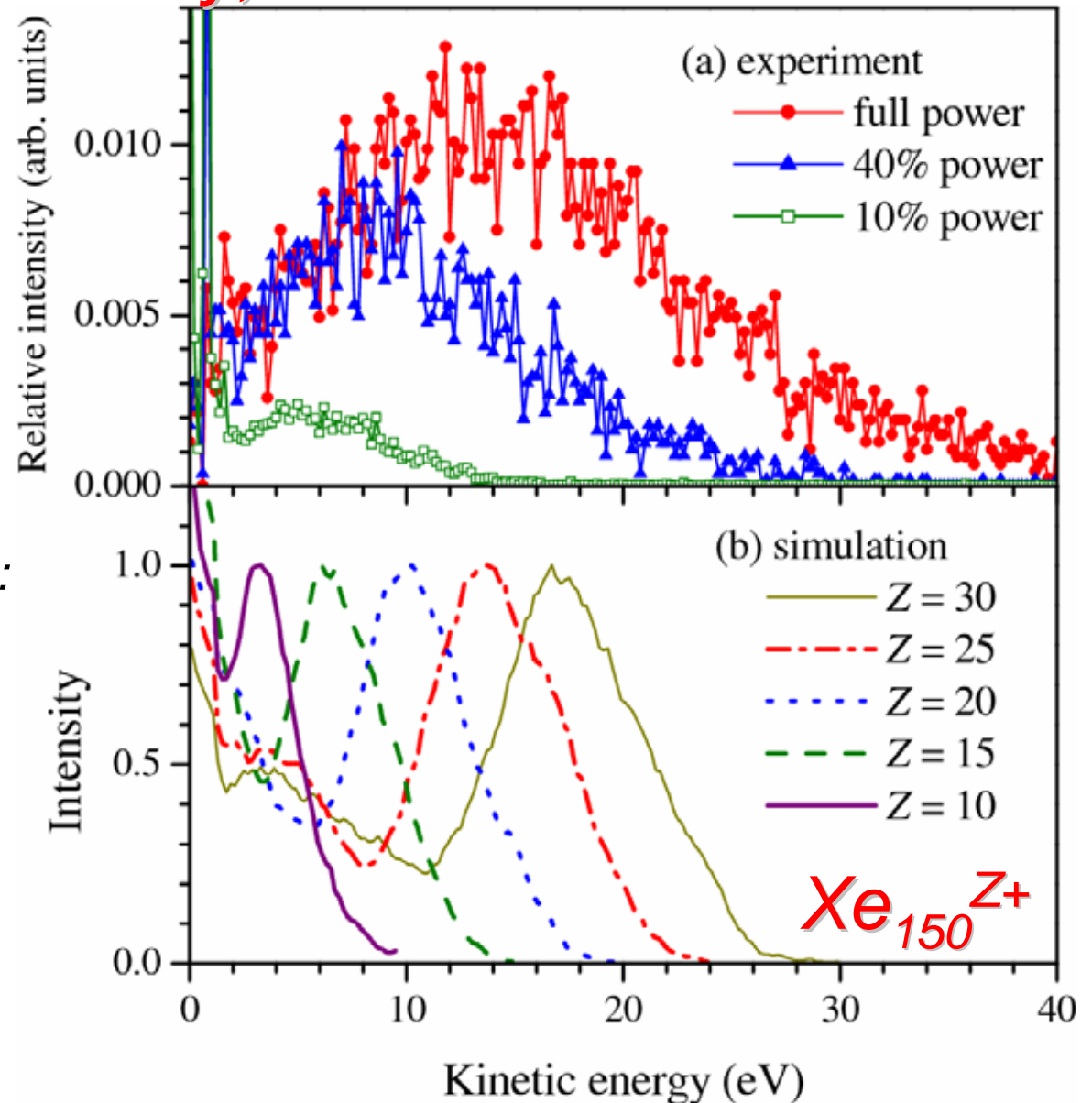
$N=14, 50, 95$

$Z/N = 1, 2/5, 1/4 < 1$

$Z/N < 1$

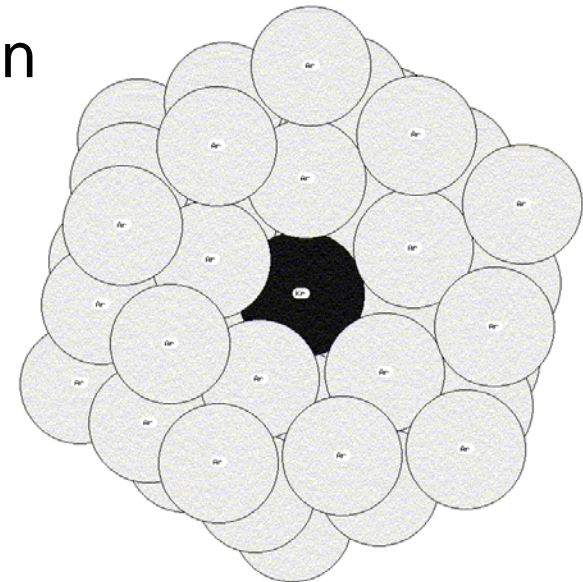
Suppression of cluster  
direct ionization

Photoabsorption of the  
individual atoms causes  
inner ionization at high  $Z$ .



# *MD simulation for estimates of initial kinetic energy distributions from given charged states*

- Cluster structure of  $\text{Xe}_{150}$  is constructed by adding 3 atoms on a perfect icosahedron structure of  $\text{Xe}_{147}$
- Randomly selected  $z$  atoms are singly charged ions ( $\text{Xe}_n^{z+}$ )
- Direct Coulomb potential and Lennard-Jones potential are taken into account
- Classical Molecular Dynamics simulation



# *X-FEL and EUV-FEL Facility in Japan*

X-FEL will be in operation in 2010!

EUV-FEL  
(in operation)

SPring-8



# Electron-ion momentum imaging apparatus for X-FEL

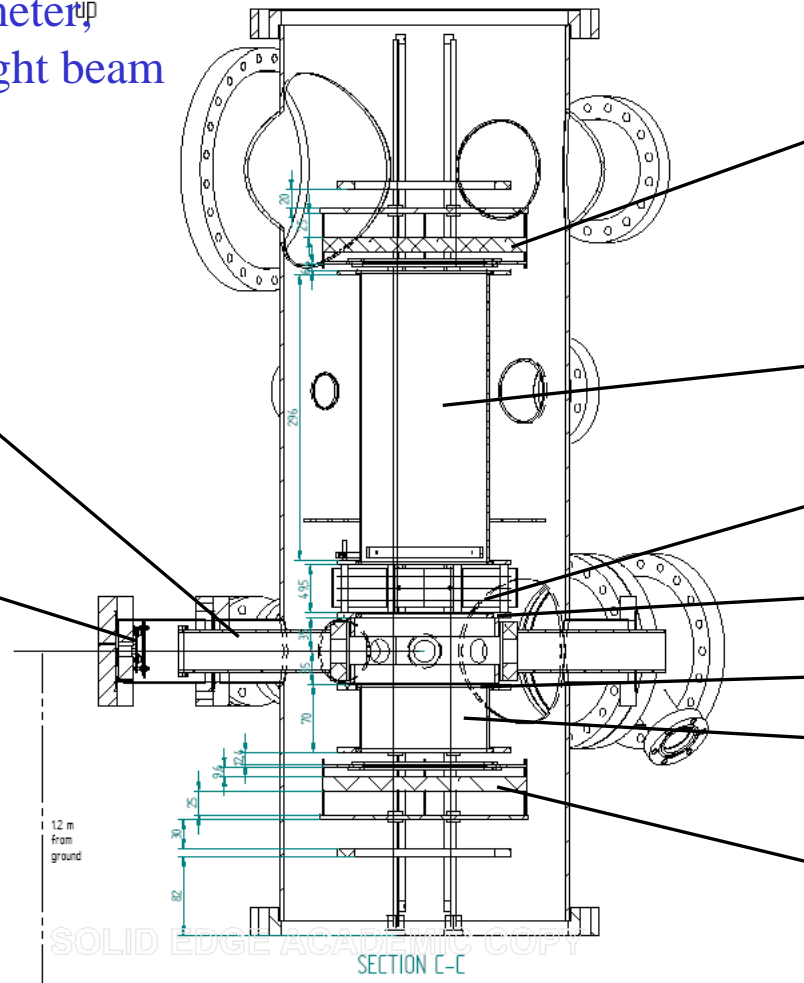
tested by laser 800nm, lens 50 mm focus outside chamber

4 x simple  
electron TOF spectrometer<sup>up</sup>  
2 x perpendicular to light beam  
2 x at magic angle  
(forward, backward)

ion detection  
pusher +320V  
extractor -320V  
ion drift tube -1000V  
120 mm diameter  
ion detector,  
hexagonal

4 x 50 mm diameter  
MCP stack

e-imaging:  
pusher +20V  
extractor -20V  
(electron DT -20V)  
B-field : 450  $\mu$ T



ion drift tube  
+ focussing lens

acceleration region  
0.75 Extractor +  
0.25 drift tube  
extractor

pusher

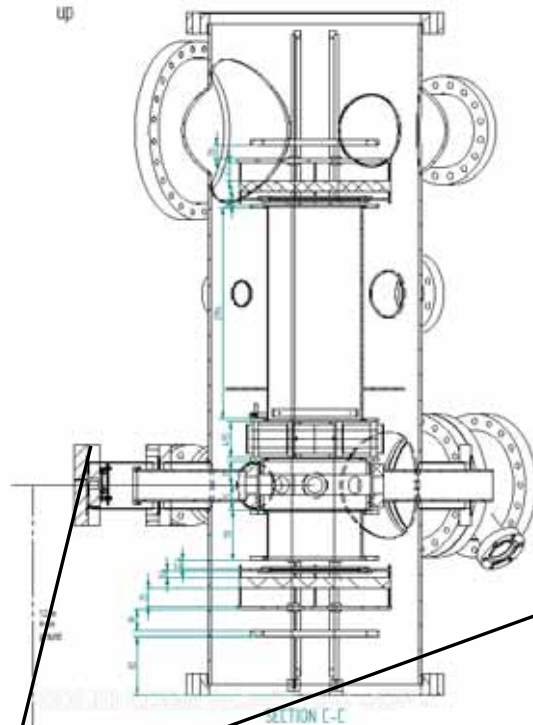
electron  
drift tube

80 mm diameter  
electron detector,  
hexagonal

dimensions: pusher - extractor 75 mm, electron DT 70 mm  
extractor - ion DT 40.5 mm, ion drift tube 296 mm

# Electron TOF spectrometer

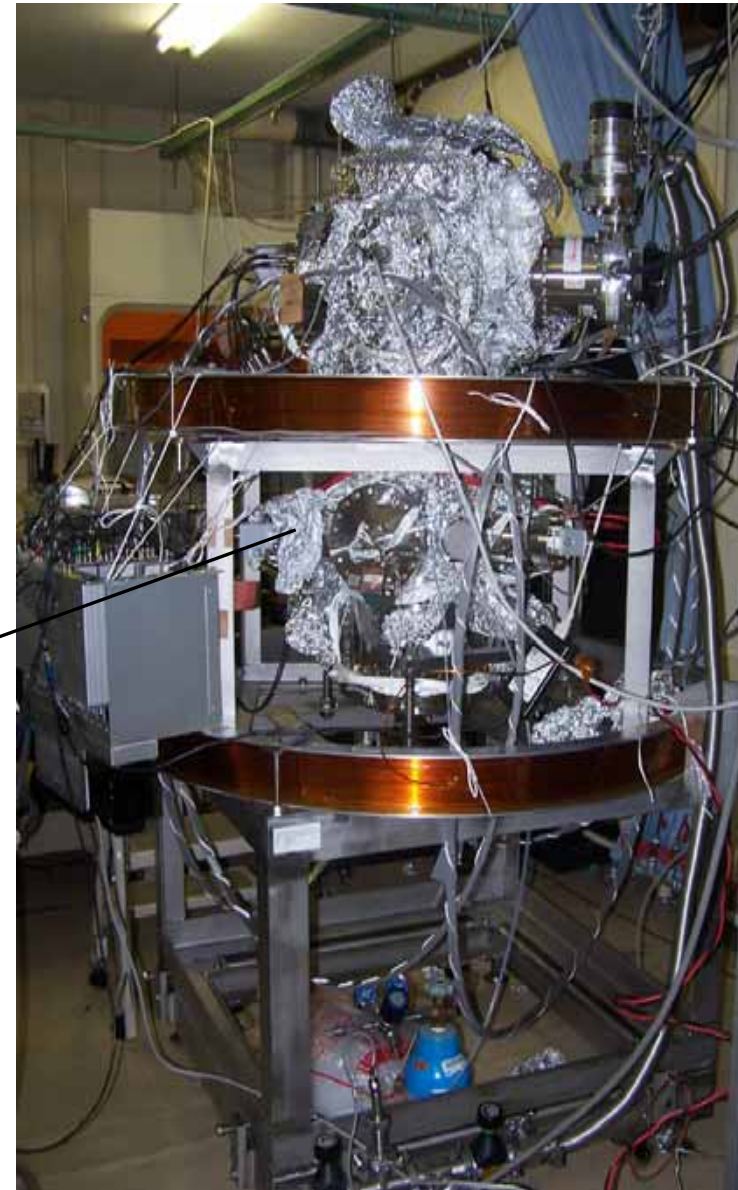
field free conditions  $0\text{V/cm}$   $B < 5\mu\text{T}$



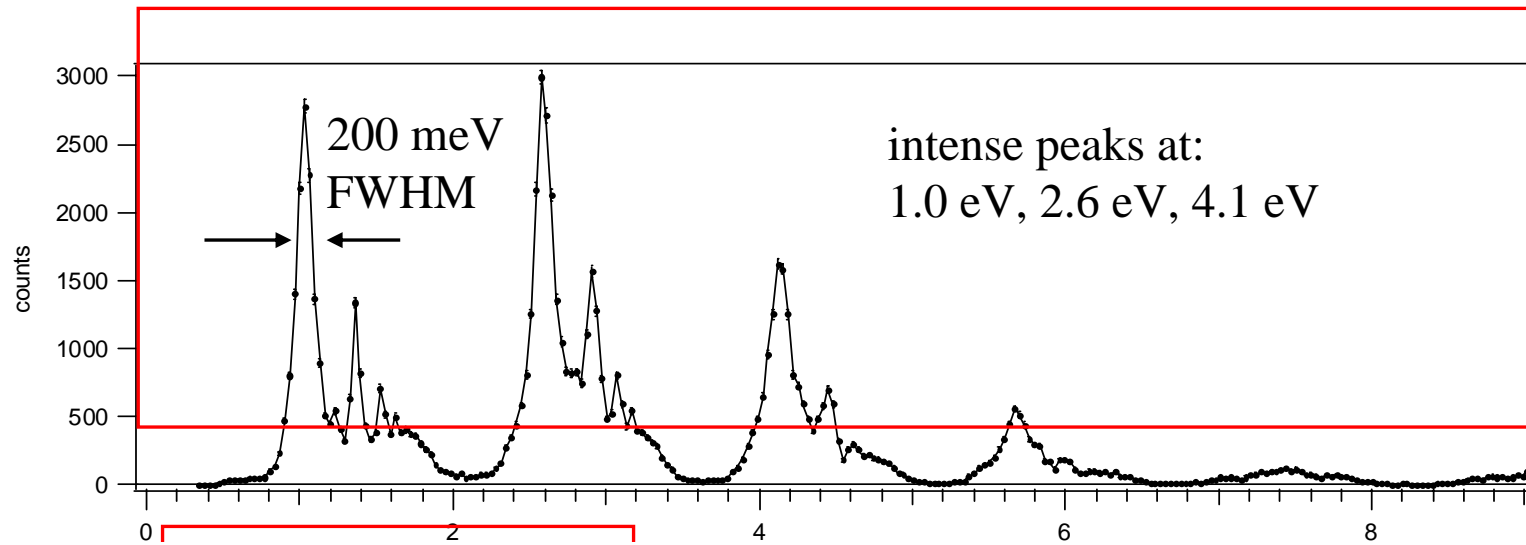
High-resolution  
High-intensity  
measurements

4 directions at the  
same time.

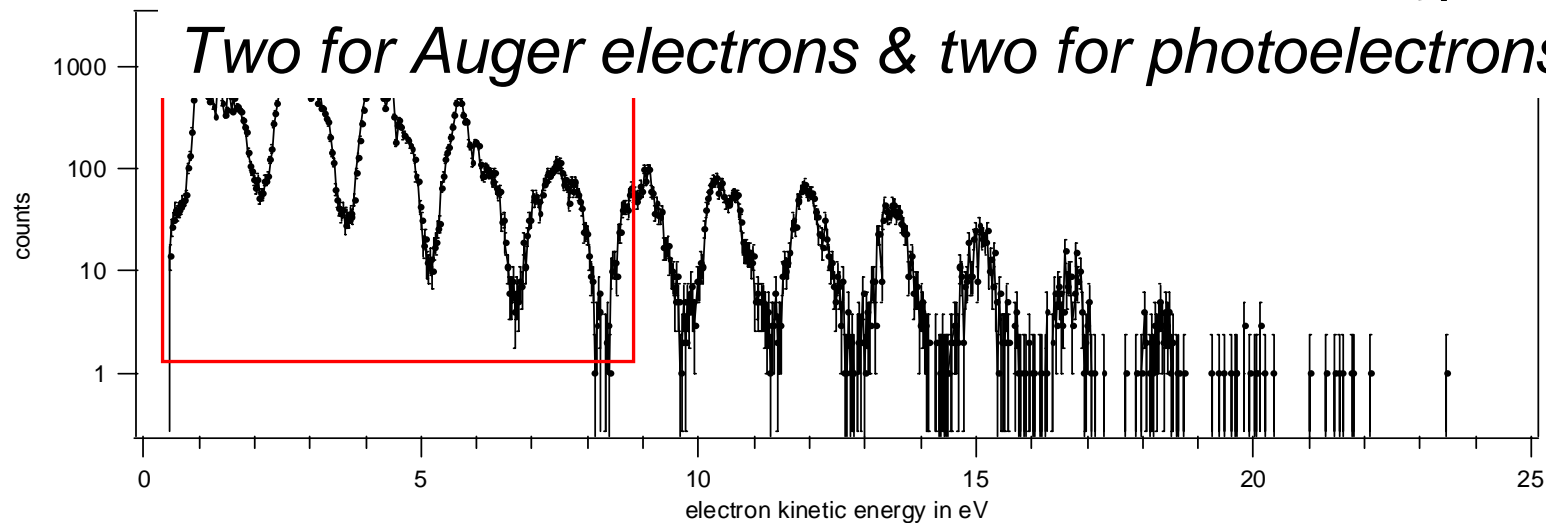
counting mode or 1 spectrum per shot mode



*ATI electron spectrum of Xe by 800nm, 40 mW laser pulses recorded by one of TOF spectrometers (limited acceptance angles) under the field-free conditions. Freeman resonances appear. The spectrum shows good resolution, good energy range.*

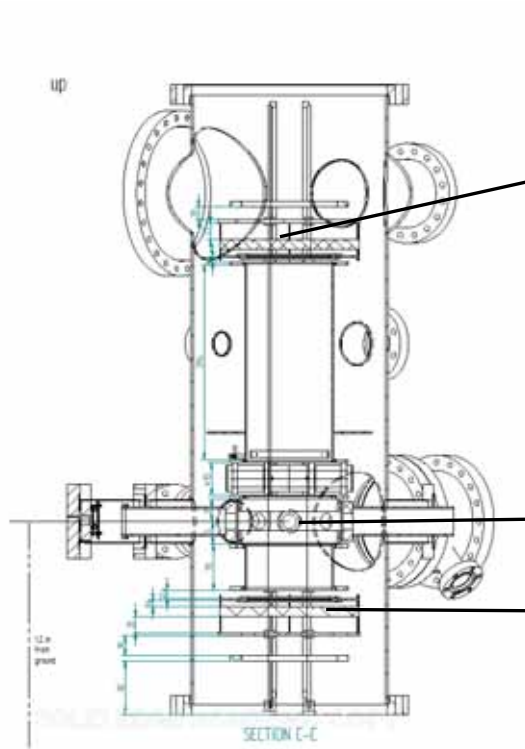


*Two of four will be modified to have retarding fields.  
Two for Auger electrons & two for photoelectrons.*



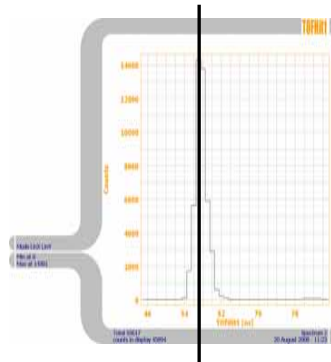
# *Electron & ion momentum imaging spectrometers*

*ion imaging*

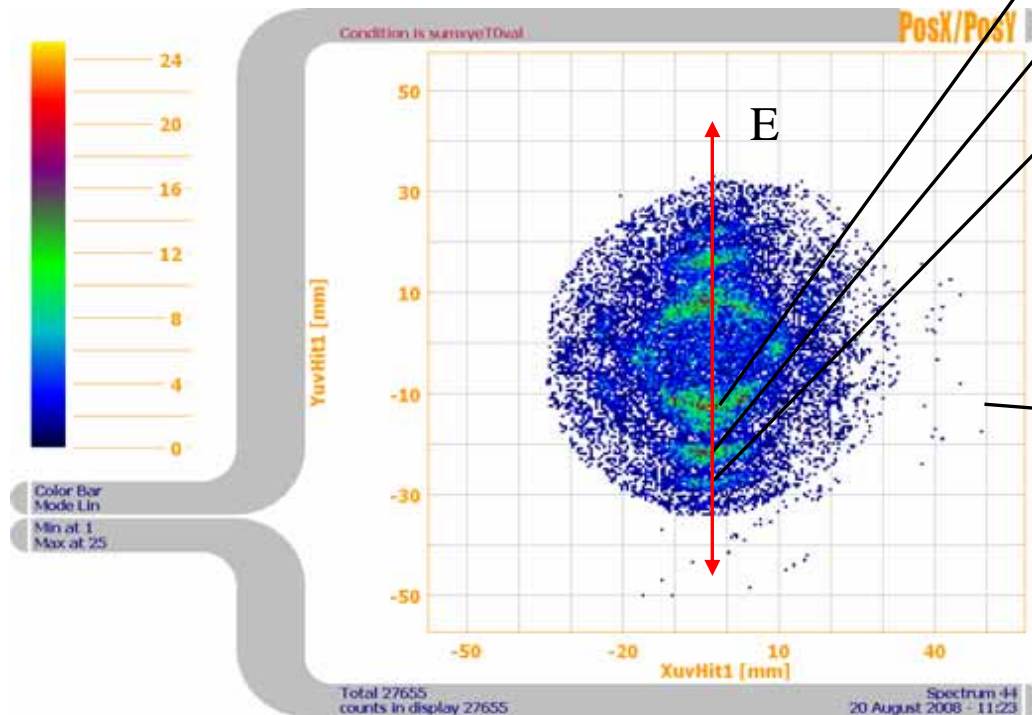
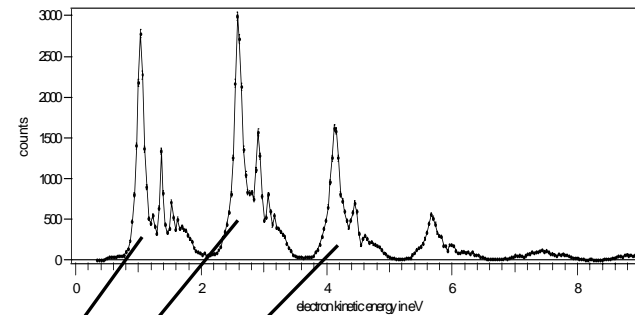


*electron imaging*

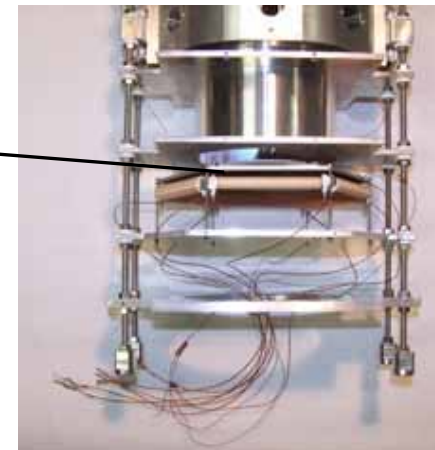
# Electron 2D imaging with DC-extraction field



cut in electron TOF,  
Momentum ( $p_x$ ,  $p_y$ )  
only perpendicular to detector  
laser: horizontal polarization



+100V at pusher / extractor  
get all electrons up to 8 eV  
without magnetic field

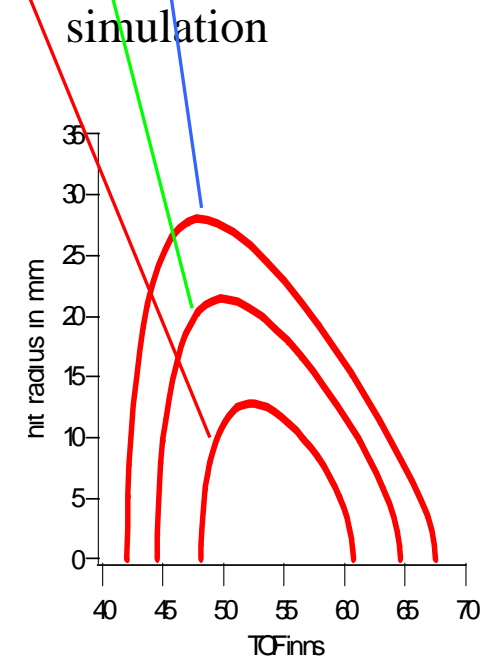
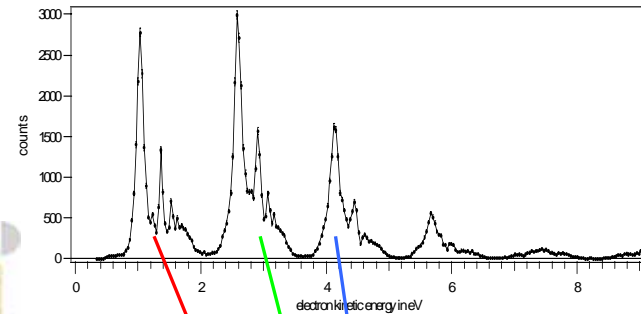
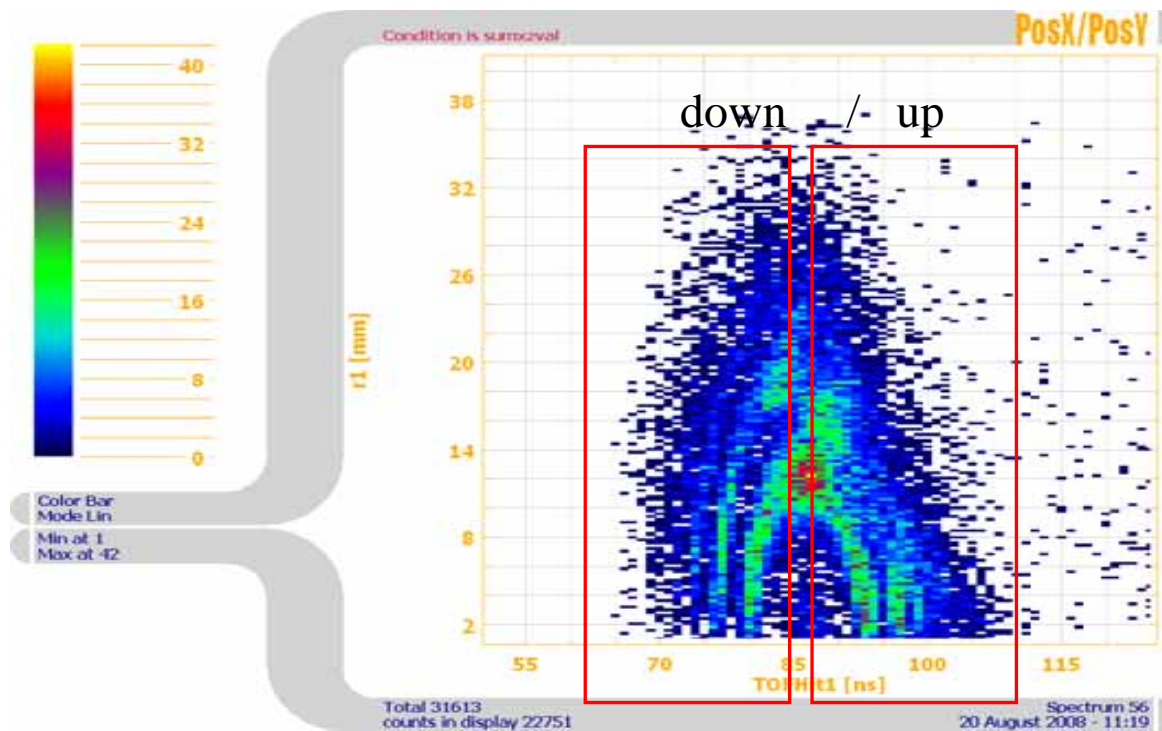


*Can be used for molecules and clusters (for e-ion coincidence)*

# Electron 3D imaging (2D imaging+ TOF)

4  $\pi$  detection efficiency  
moderate resolution, 1 or more electrons per shot  
+/-20V at pusher / extractor, 450  $\mu$ T (4.5 G)  
get all electrons up to 5 eV,  
all direction with reasonable resolution

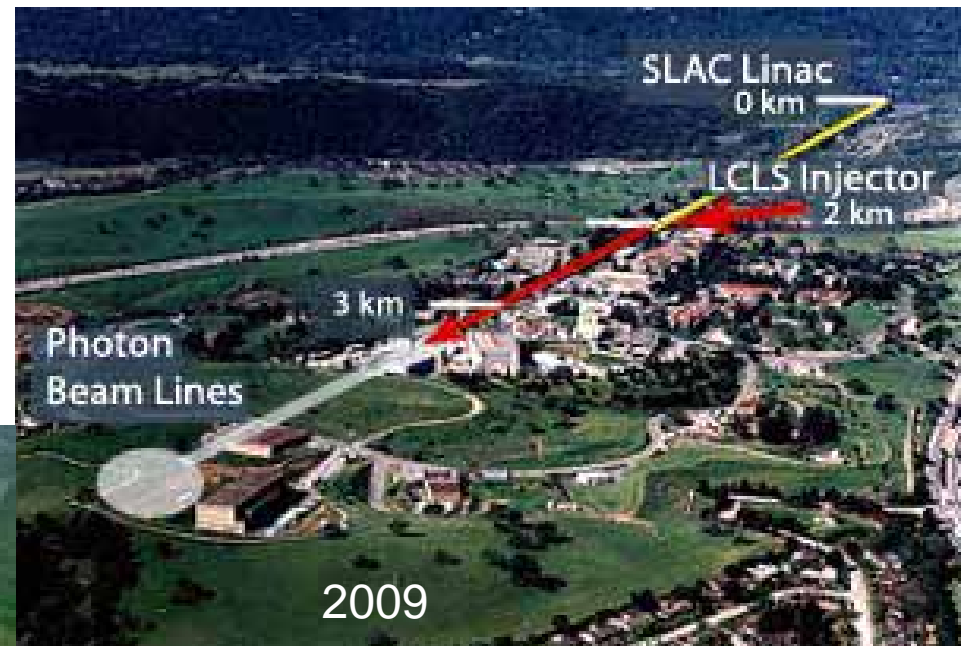
intense peaks at:  
1.0 eV, 2.6 eV, 4.1 eV



(laser: horizontal + vertical polarization)

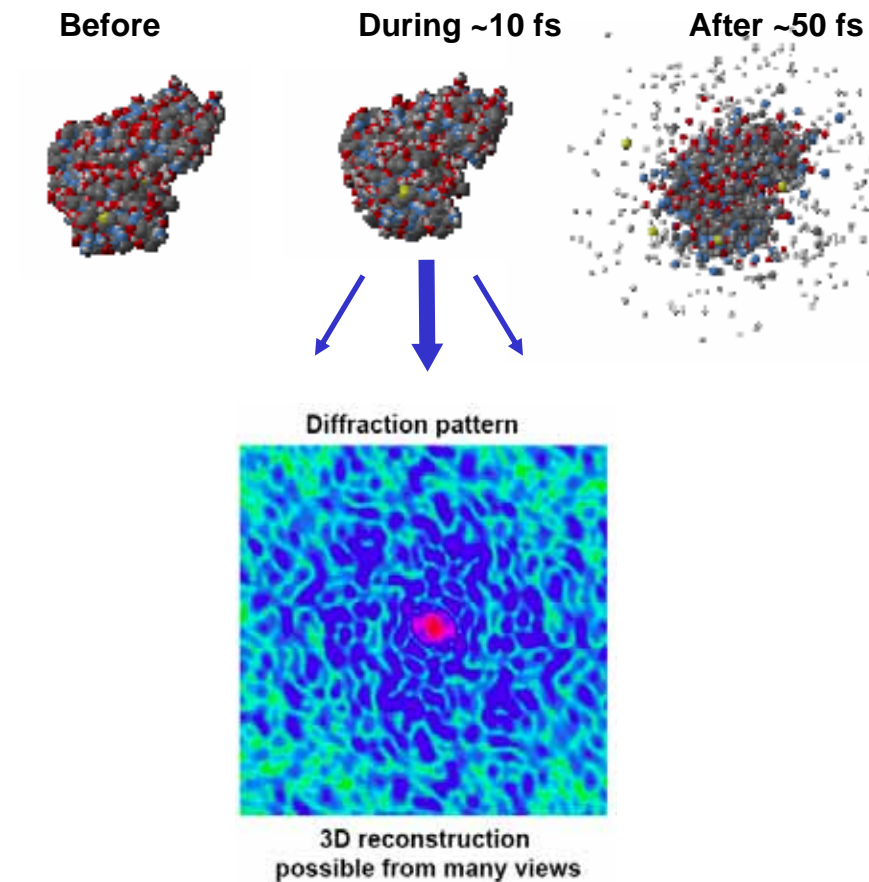
*Can be used for (He and) Ne recoil momentum coincident with e*

# Single-shot imaging by X-FEL



# Structure analysis for a single biomolecule

Neutze, Wouts, van der Spoel, Weckert, Hajdu Nature 406, 752 (2000)



Should record the X-ray diffraction pattern before Coulomb explosion....  
Change of the electron density may be even faster....  
Developing the X-ray detectors is the most important issue....



*The end*

Thank you very much for your attention!



*Sendai  
in summer*



*Tanabata festival*