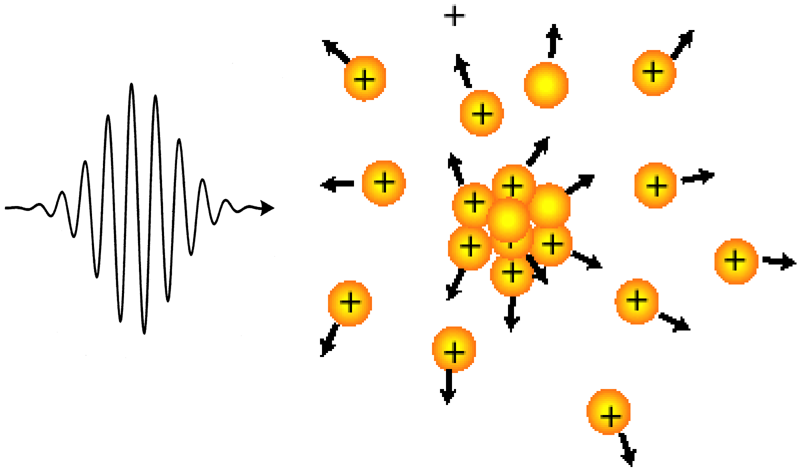
Ultrafast processes of clusters in intense X-ray beams

Thomas Möller, Technische Universität Berlin
Aarhus, October 29, 2008

- **Motivation: Issues in cluster physics**
- **Interaction of intense VUV- and X-ray pulses with clusters**



Mechanisms of photoabsorption

Ionisation dynamics

Coulomb explosion

- **Electronic processes, innershell ionistion, electron migration**
- **Cluster imaging: Geometric structure (~1Å)**

Clusters exhibit interesting properties

Materials properties

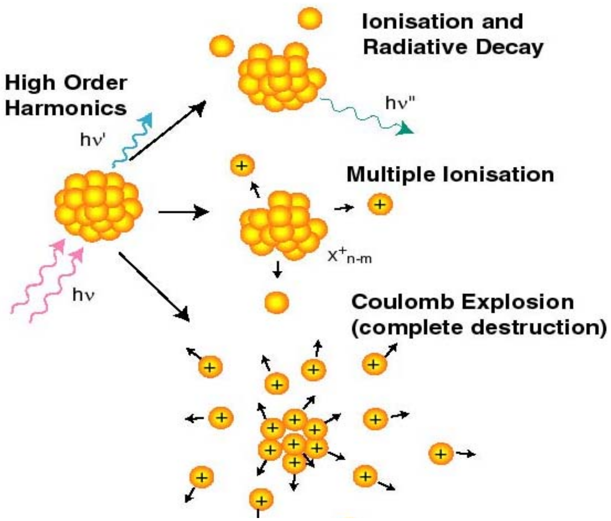
- electronic structure
- optical properties
- reactivity, catalysis
- structure – property relationship

CdSe clusters of different size

Ideal sample systems

- sizeable
- solid state density
- collective phenomena
- „nanolab“ for plasma physics

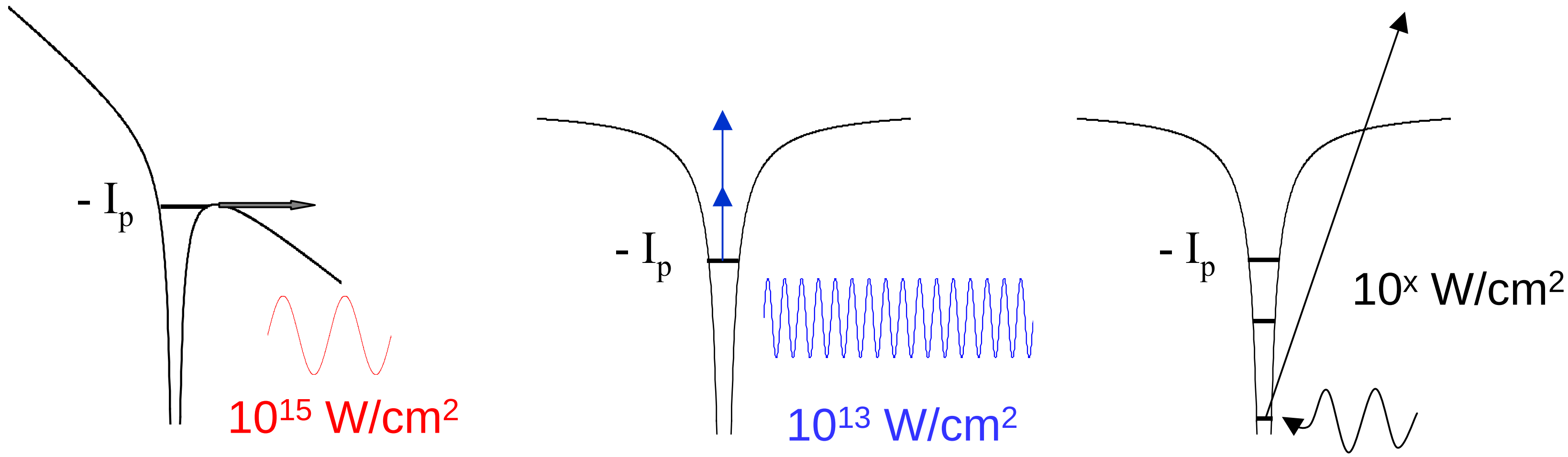
IR : T. Ditmire



Clusters bridge the gap from atomic to solid state physics

Radiation–matter interaction

IR: quasistatic description | VUV FEL: intense photon source | X-ray FEL: highly ionizing



IR: quasistatic description

- **Keldysh parameter $\gamma \ll 1$**
- **Tunnel / over the barrier ionisation**
- **Ponderomotive energy 10 – 100 eV**

VUV FEL: intense photon source

- **Keldysh parameter $\gamma \gg 1$**
- **Multi-photon ionisation**
- **Ponderomotive energy 10 meV**

X-ray FEL: highly ionizing

- **Angstrom wavelength**
- **Direct ionisation**
- **Secondary processes**

Keldýsh parameter

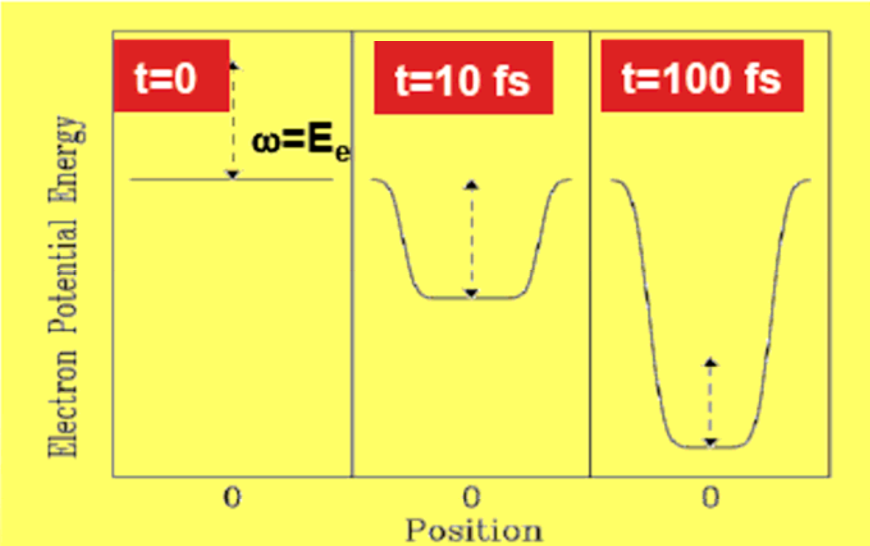
$$\gamma = \sqrt{\frac{E_I}{2U_p}} \sim 1/\lambda$$

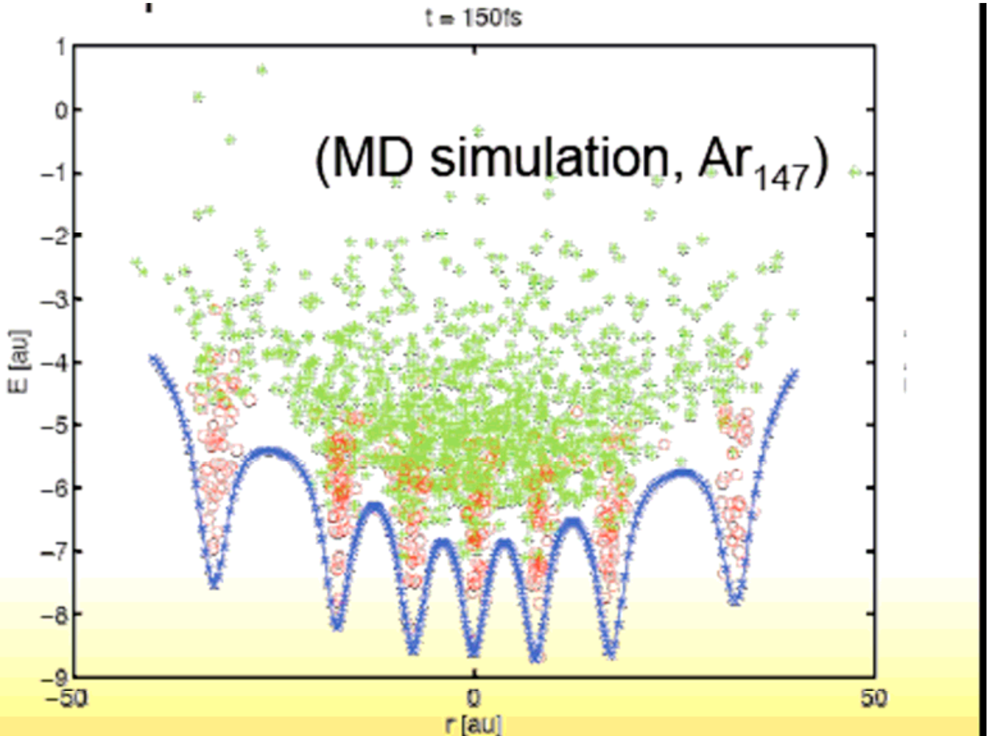
E_I Ionisation potential, U_p ponderomotive engergy

Cluster ionisation, electron delocalisation and electron migration

photoabsorption leads to loss of electrons
- ionic charge builds up
- further ionisation by: photoabsorption, electron impact ionisation field ionisation at surface
- non-screened surface ions explode

strongly wavelength depending!





courtsey, J.M. Rost, Dresden

Future perspectives with X-rays

- innershell processes, ionisation form inside

- ultrafast dynamics, pump probe

- imaging of single/few particles

Going to shorter wavelength: Will sequential single photon processes dominate?

M. A. Kornberg et al.
J. Synchrotron Rad. 9, 298 (2002)

ionisation yield

1Å

- Single photon regime often valid (per atom!)
- In heavy elements ionisation yield close to one
- plasma absorption ~ λ^2 cross sections ~10^{-5}Mbarn at 0.1 nm

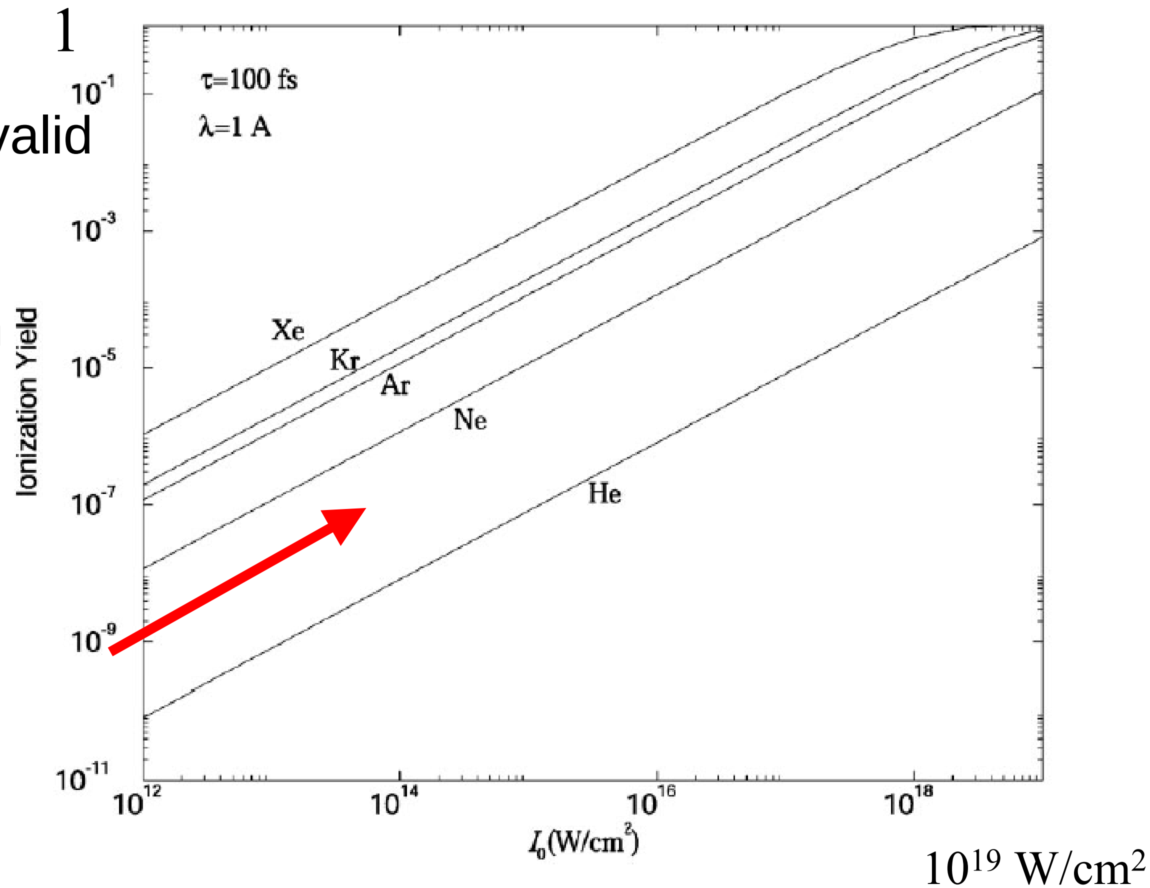
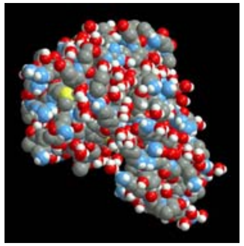
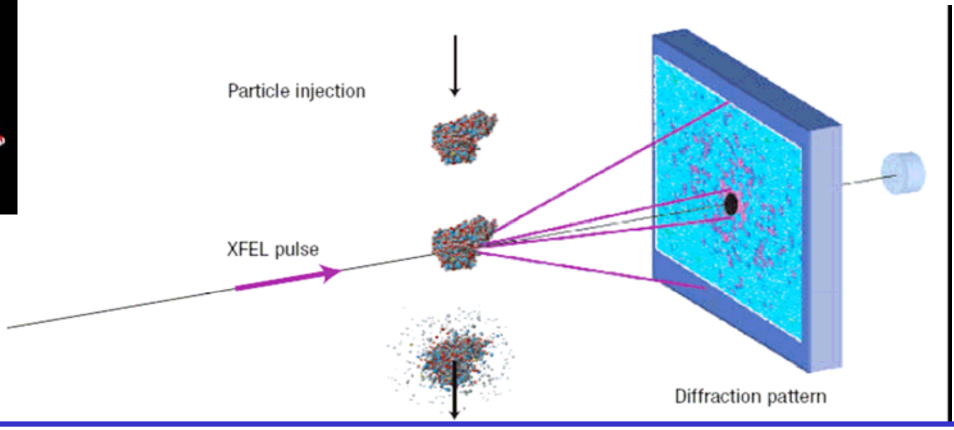


Figure 1
Ionization yield as a function of peak intensity for the rare gases. The photon wavelength is 1 Å, and the pulse duration is 100 fs.

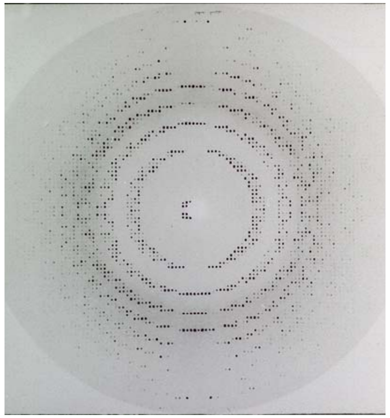
Structure determination with a FEL: The issue of radiation damage

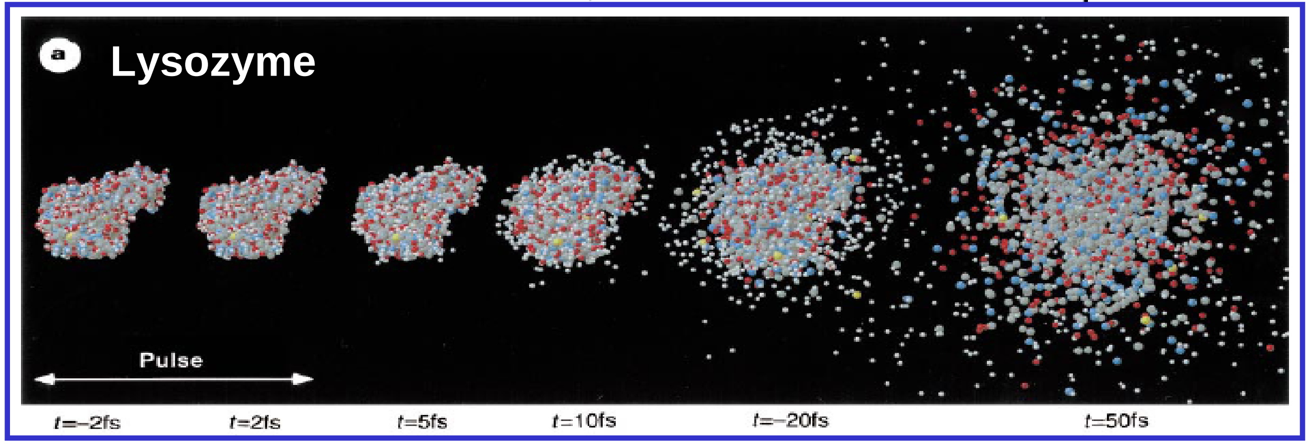
Molecules atomic resolution





Crystal





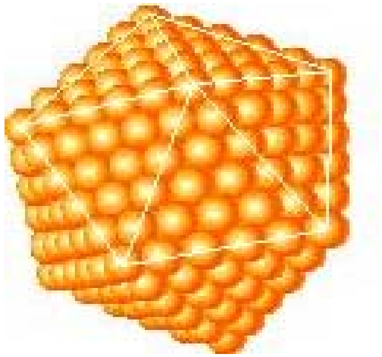
R. Neutze, J. Haidu et al., Nature 406, 752 (2000)

Radiation damage and Coulomb explosion

Structure determination of clusters with a FEL

Clusters

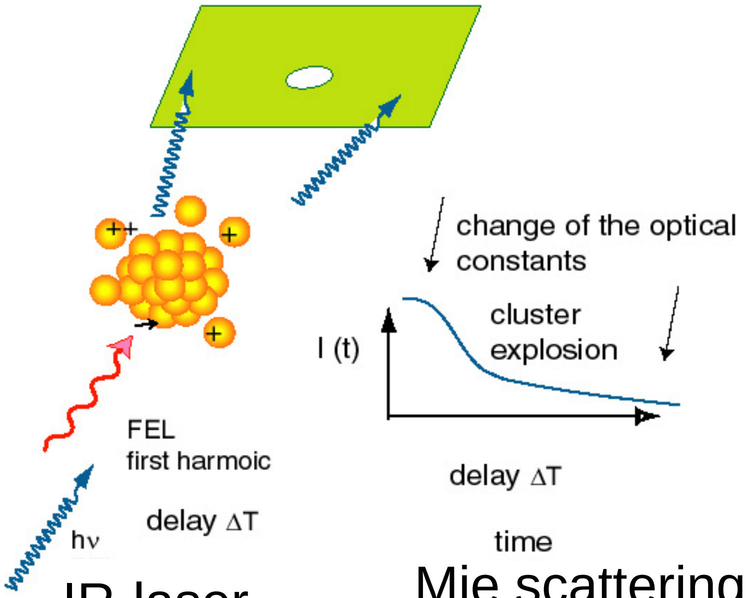
Different/ Advantages:



Low and **high** Z-Elements
C, Si, Au, Pt (catalytic properties)

Some translational symmetry

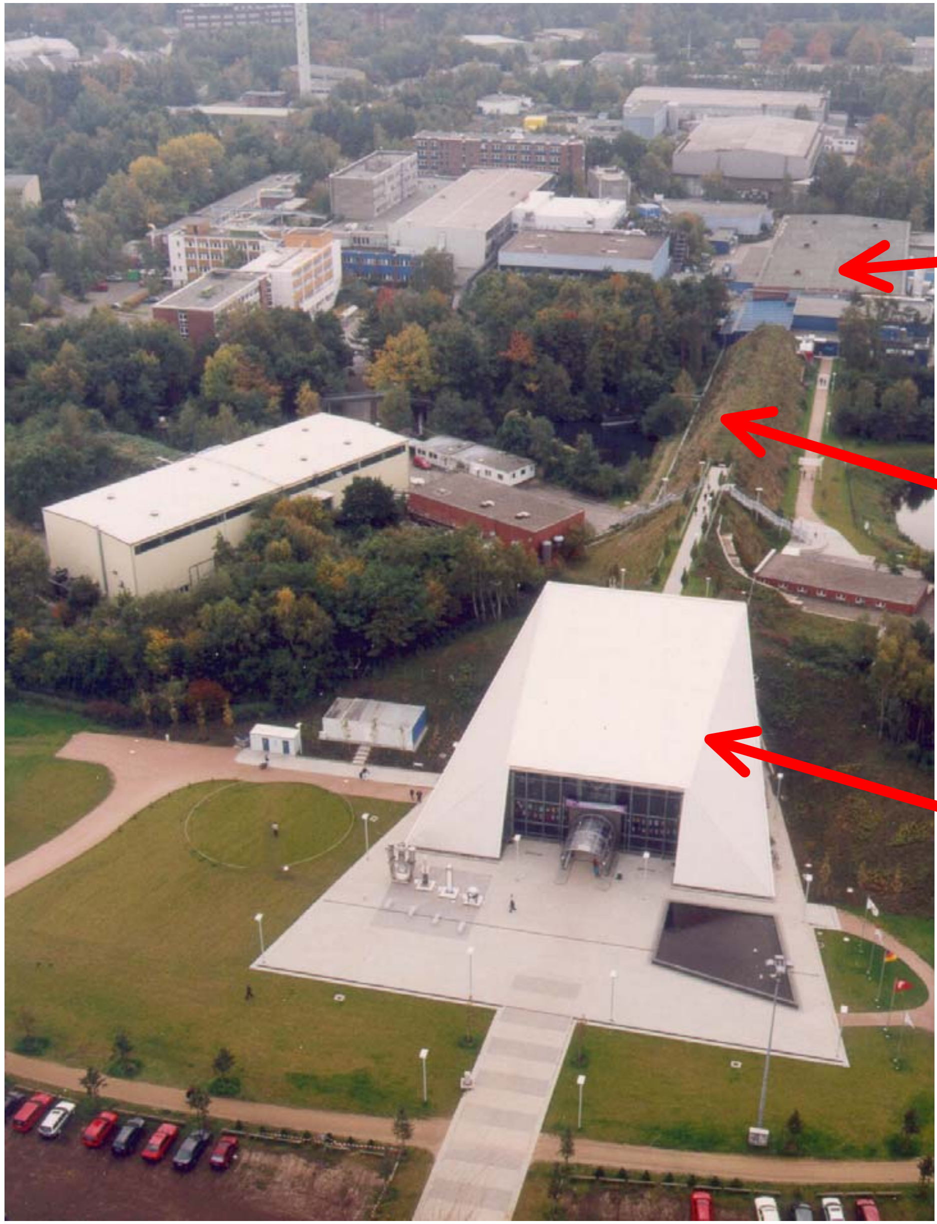
Opportunities:



IR-laser

Mie scattering
Diffraction imaging
Real time dynamics

FLASH FEL at DESY



Electron gun

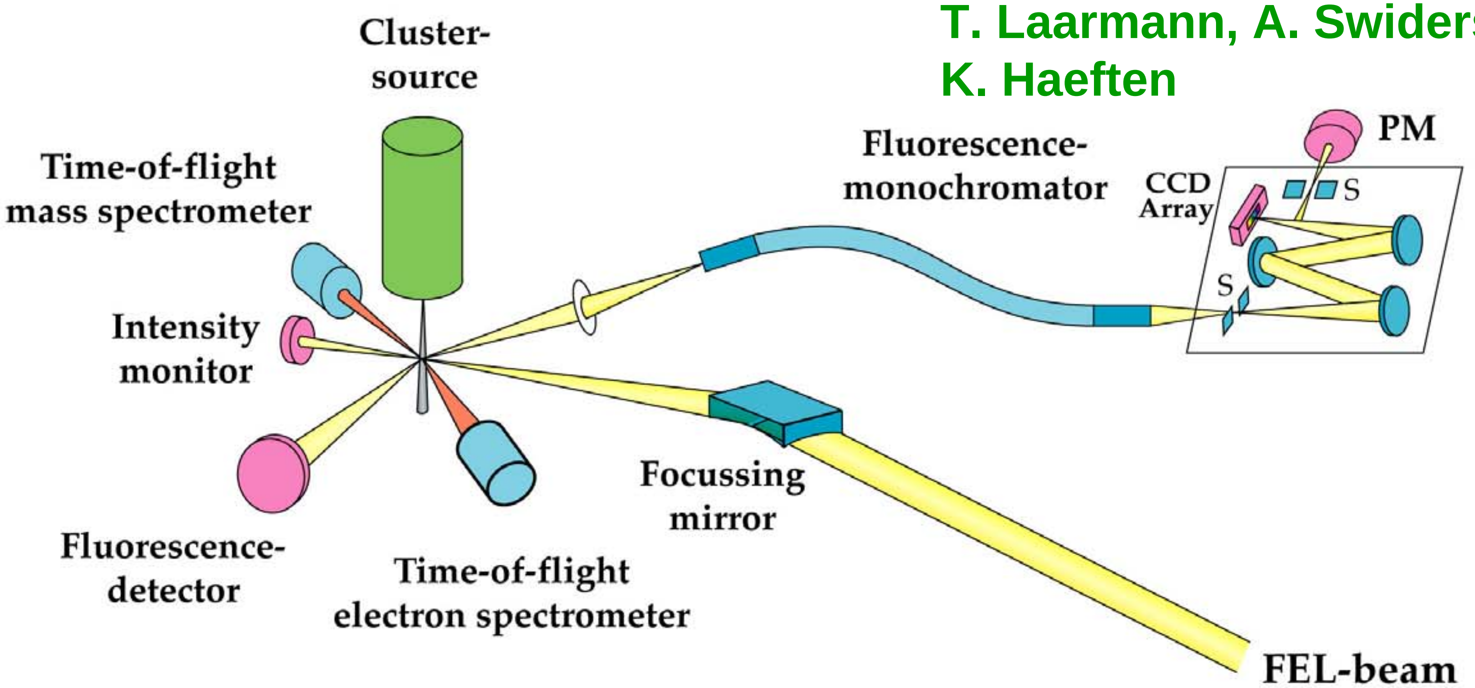
Linac and FEL undulator

Experimental hall (User Facility started July 2005)

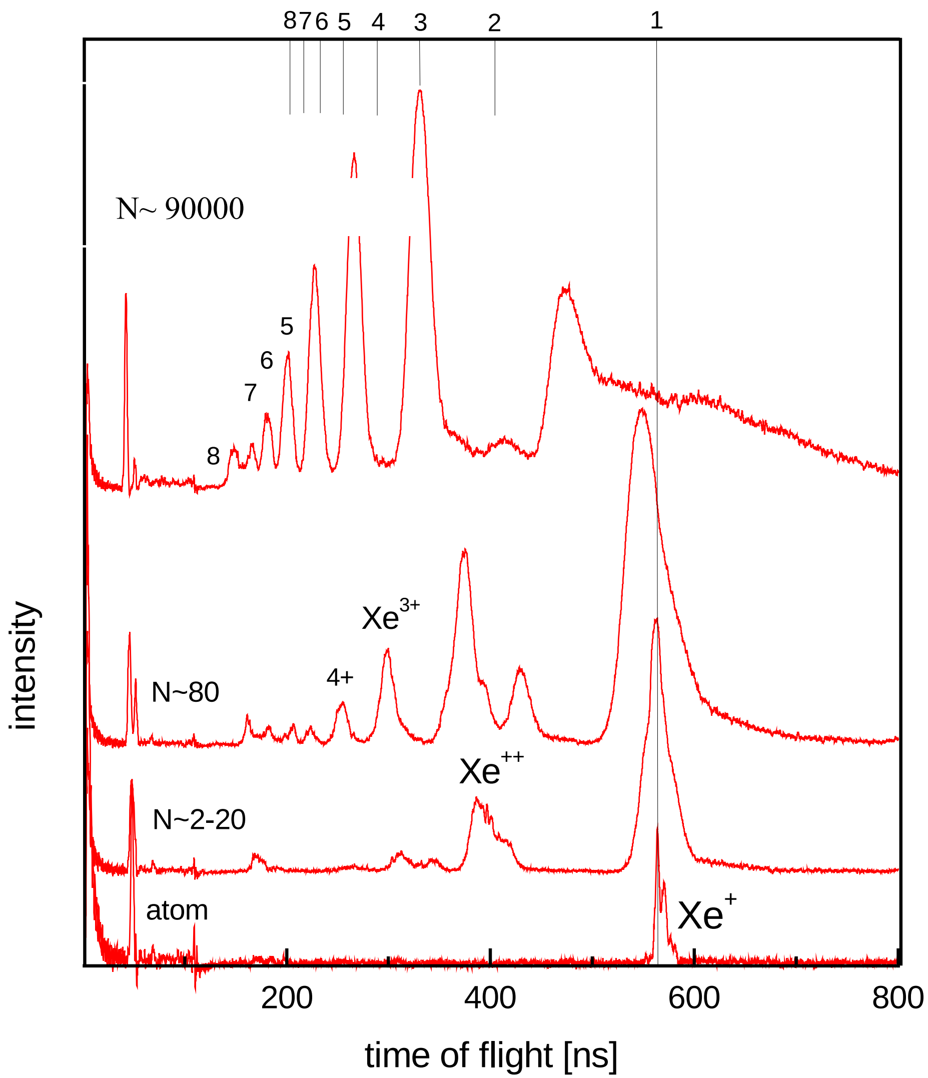
- 6,5- 50 nm
- 10-100 µJ
- 1 GW_{peak}
- 10-100 fs

A cluster experiment for studies with a FEL radiation

H. Wabnitz J. Schulz
P. Gürtler, W. Laasch
T. Laarmann, A. Swiderski
K. Haeften



Time of flight mass spectra of Xe atoms and clusters



$1*10^{13}$ W/cm^2

Ip_{Xe} = 12.1 eV
E_{phot}= 12.8 eV

H. Wabnitz et al, Nature 420, 482(2002)

- **multiply charged ions from clusters, keV energy**
- **singly charged ions from atoms**
- **detailed theoretical work to explain the enhanced absorption**

Plasmabsorption (IB)

R. Santra, Ch. H. Green, PRL 91, 233401 (2003)

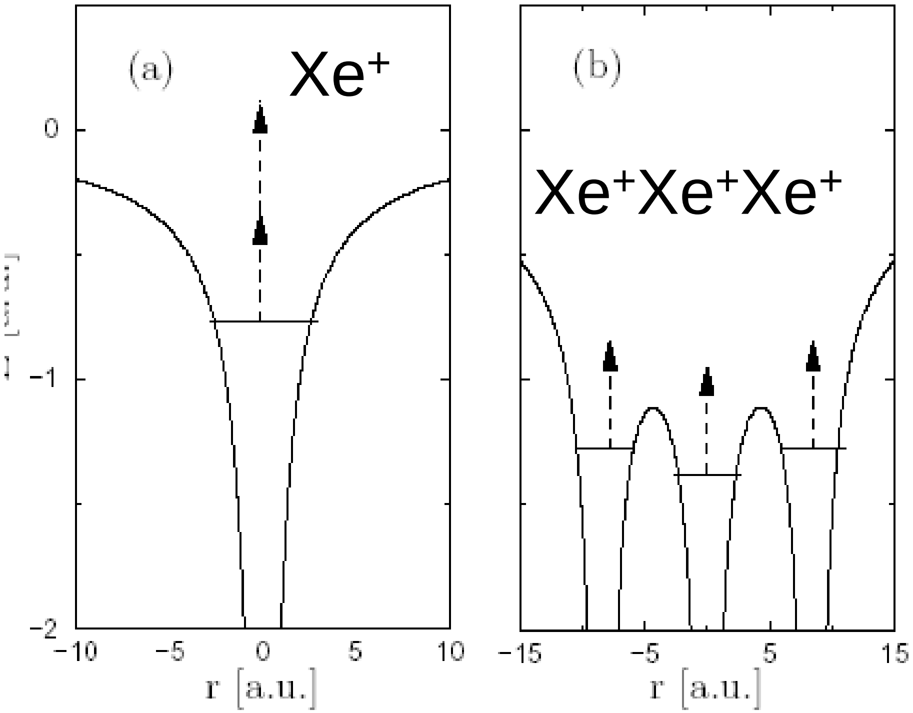
C. Siedschlag, J. M. Rost , PRL 93, 43402 (2004)

Z. Walters, R. Santra, C. Green PRA, 74 43204(2006)

Enhanced ionisation: Recent theoretical work

Mixed quantum-classcial model (λ=98 nm)

Siedschlag, Rost, PRL 93, 43402 (2004)



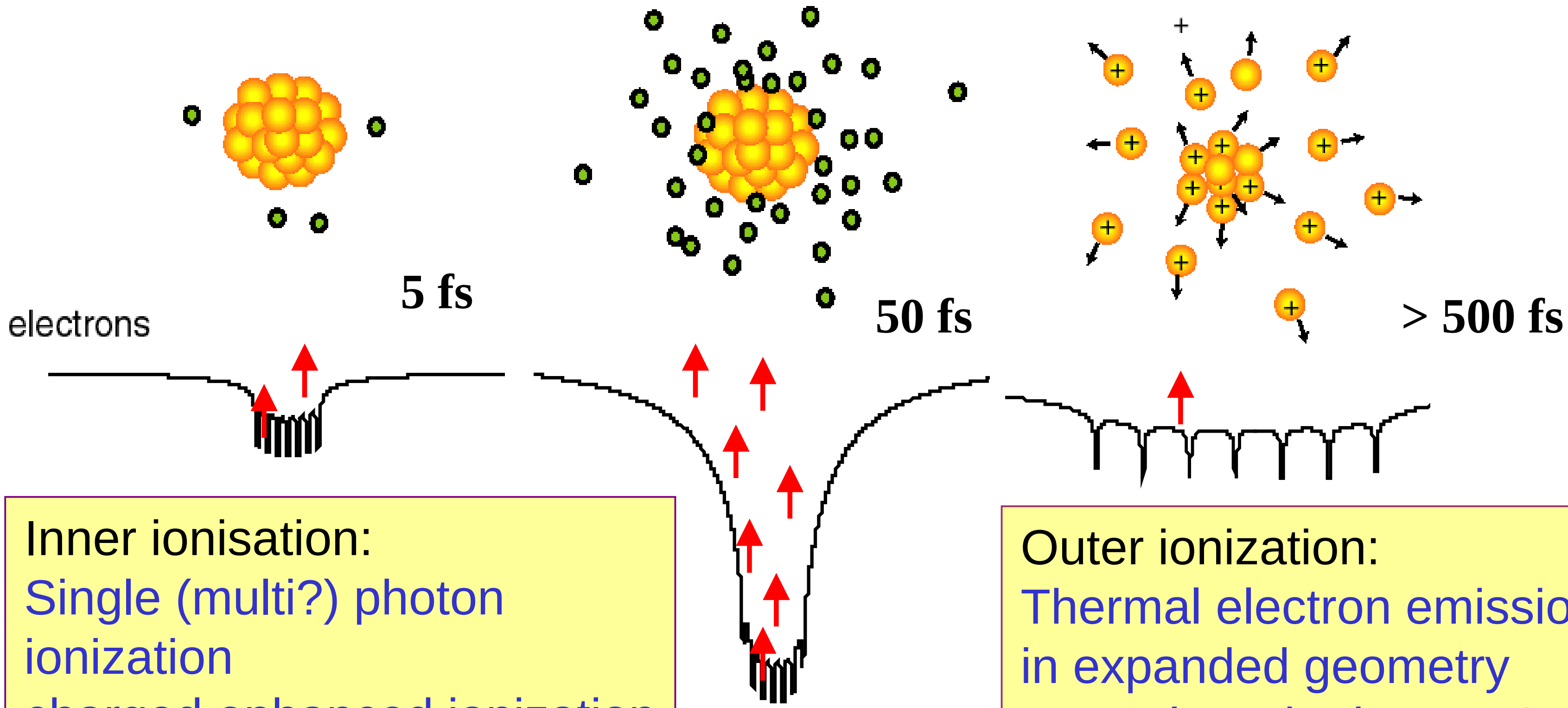
one effect:

Barriers pulled down by surrounding charges

'charged enhanced ionistion'

- **higher inner ionisation, Xe^{2+} with 13 eV**
- **enhanced absorption**

Model for Coulomb explosion

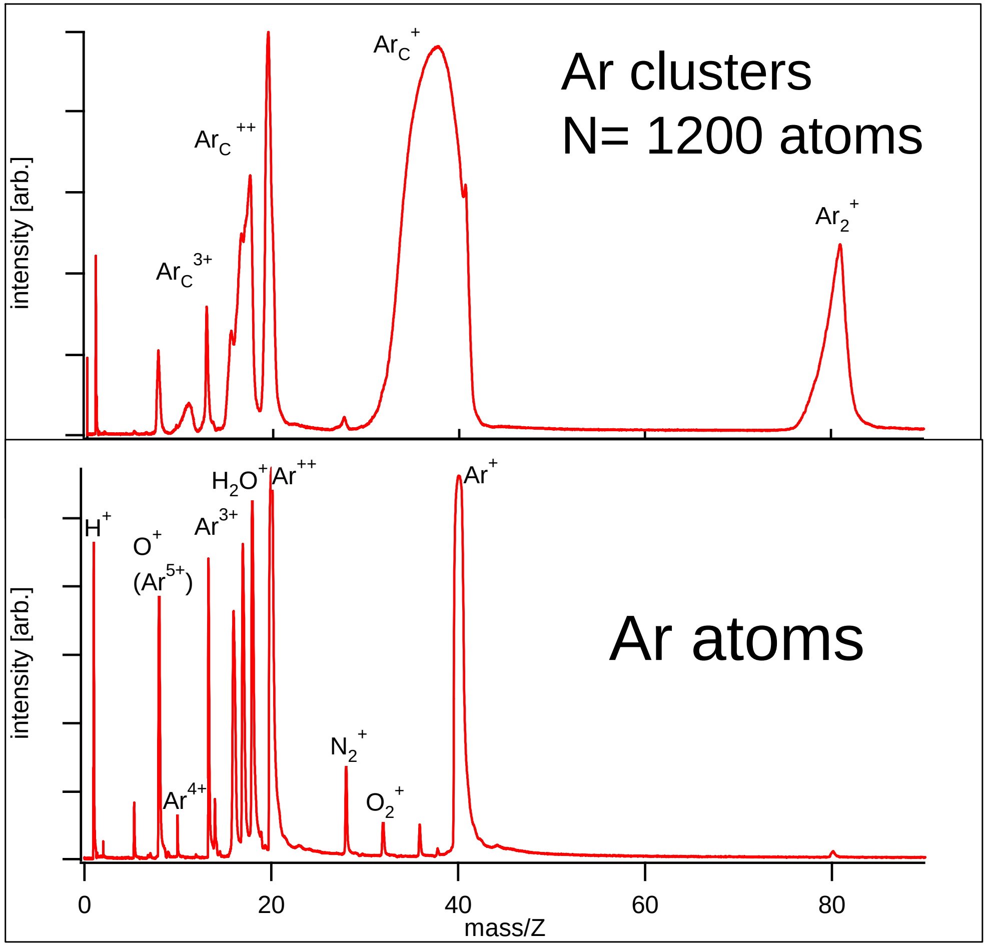


Inner ionisation:
Single (multi?) photon ionization
charged enhanced ionization

Outer ionization:
Thermal electron emission in expanded geometry
Neutral, excited atoms?

Cluster heating: plasma absorption
Enhanced rate due to electrons close to nuclei

Towards soft X-rays: Coulomb explosion at 32 nm



38.5 eV

30 fs, $2*10^{13}$ W/cm^2

cluster beam:
atomic ions, frament ions
kinetic energy ~ 25eV

highly charged atoms
up to Ar^{4+}

no strong enhancement of absorption in clusters

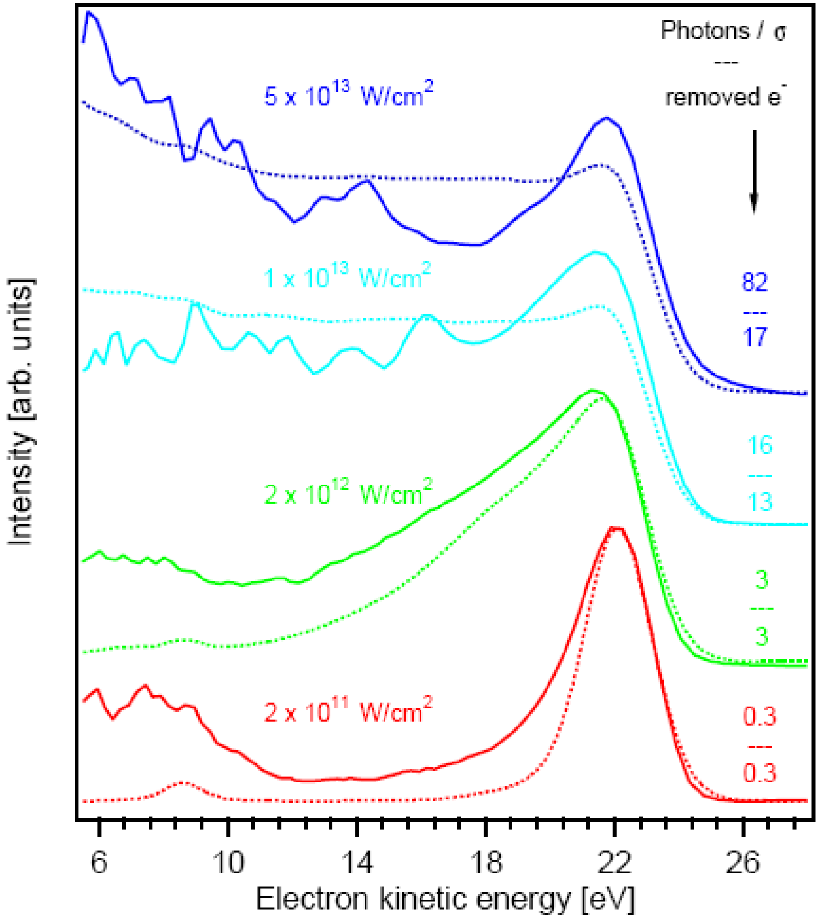
recent theory paper: @20 eV IB still strong

I. Georgescu et al. (Phys.Rev. A 76, 043203 (2007))

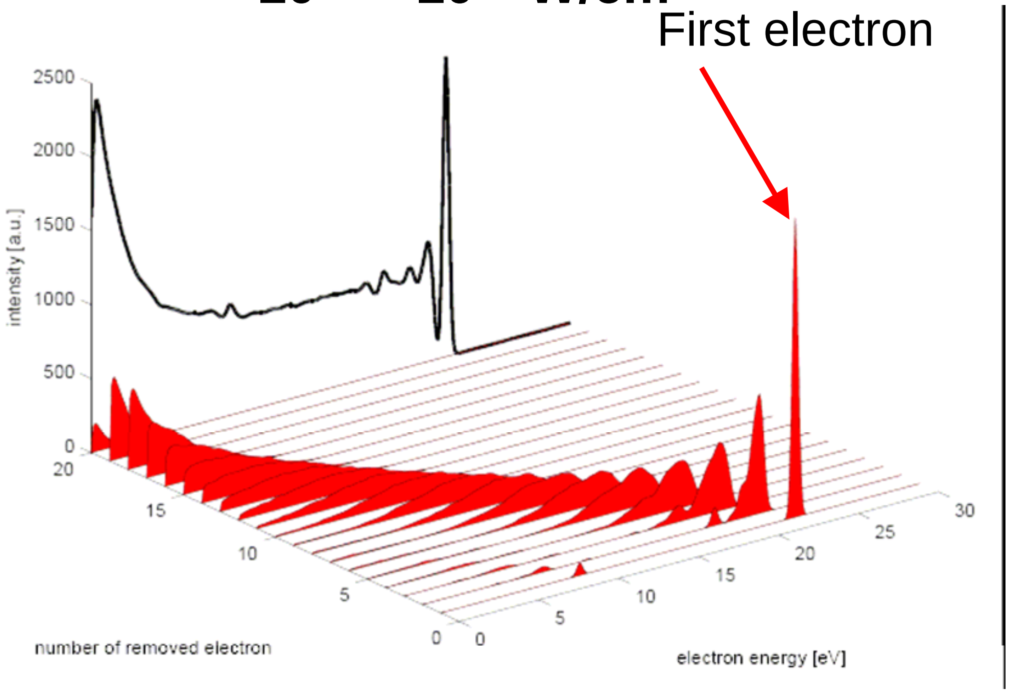
Ionisation studied with electron spectroscopy

- experiment

····theory



Ar_{150} clusters, 32 nm, $10^{11} \sim 10^{14}$ W/cm²



Theory (T. Fennel, Rostock)

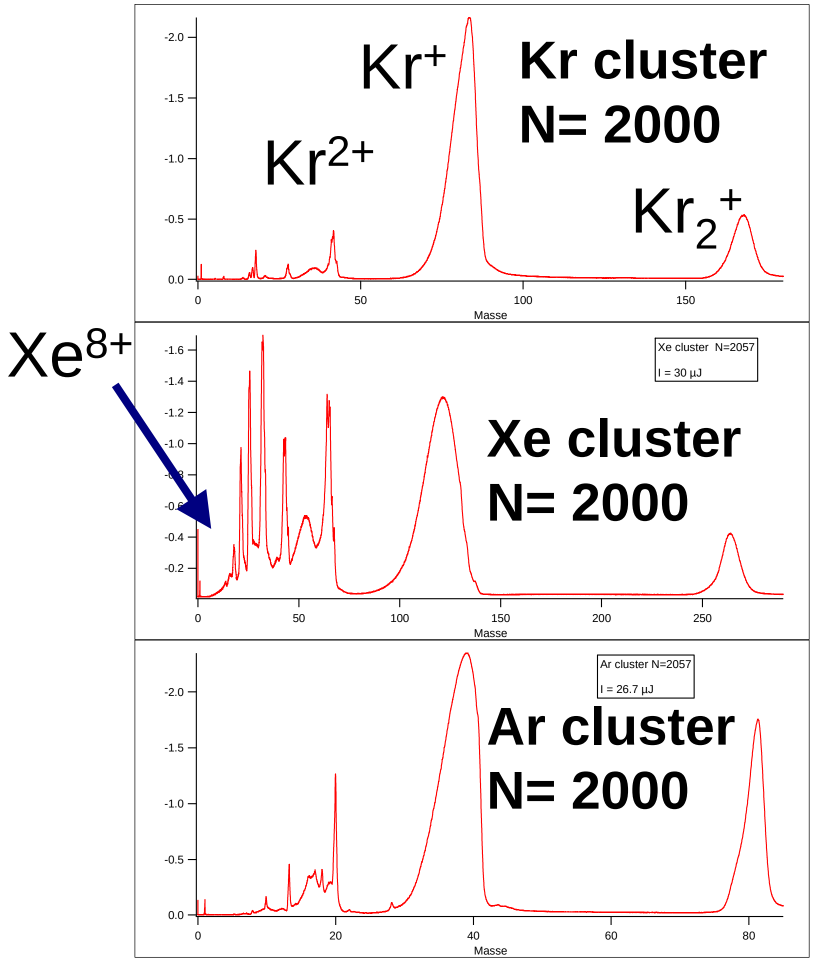
sequential emission of electrons

only a small percentage of generated photoelectrons can leave the cluster

C. Bostedt et al.
Phys. Rev. Letters 100, 133401 (2008)

Photon energy 90 eV (λ=13,5 nm): Comparison Ar, Kr and Xe cluster

time of flight mass spectra



30 fs, ~5*10^{13} W/cm^2

- much higher charge states for Xe, Xe^{7+} (clusters); Xe^{8+}(atoms)
- absorption in Kr smaller than Ar

σ_{abs} at 90 eV

0,5 Mbarn Kr	4p valence band
20 Mbarn Xe	4d Innershell
1 Mbarn Ar	3p valence band

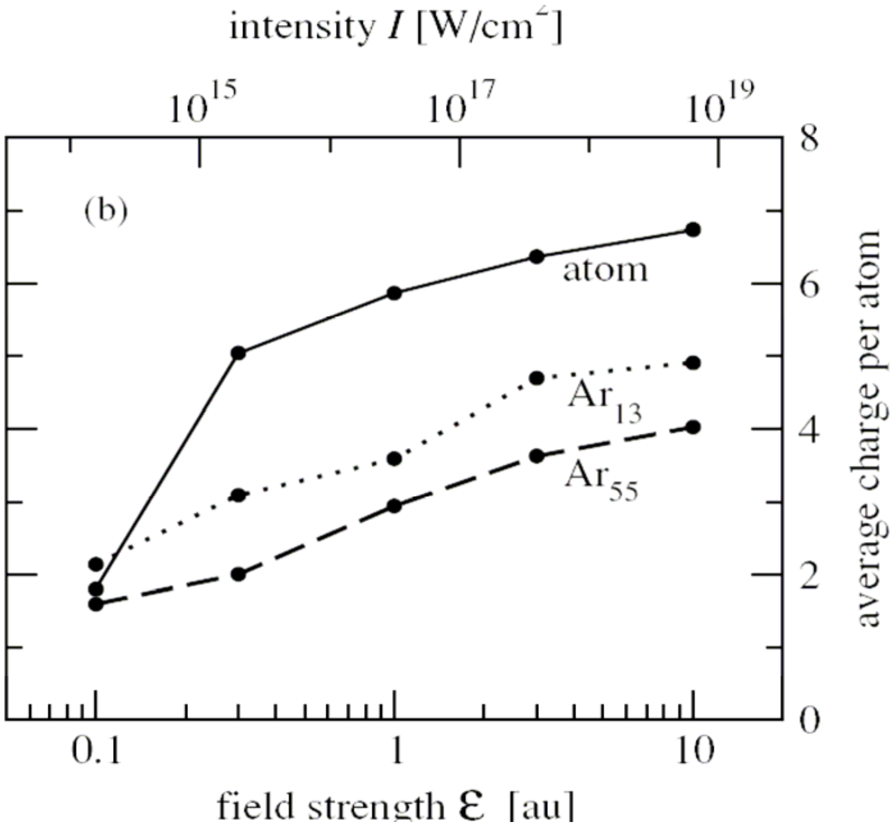
- ionisation controlled by absorption of the first photon?

Innershell ionisation: Theory

U. Saalmann, J.M. Rost, PRL, 143401 (2002)

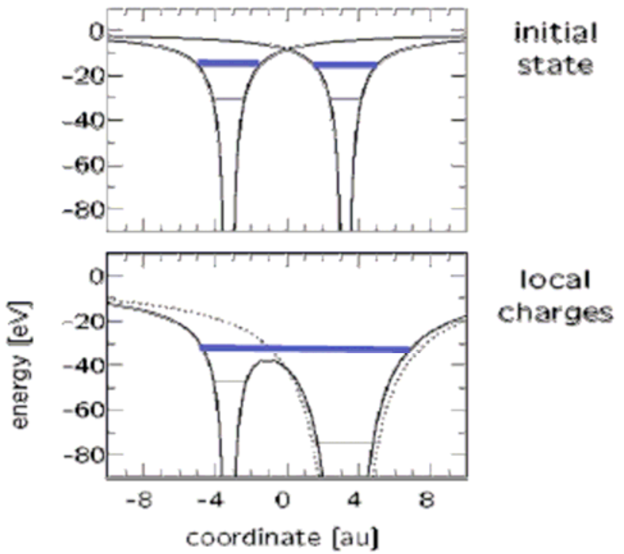
Prediction:

- **absorption in clusters strongly reduced compared to free atoms**
- **low Auger rates due to delocalized valence electrons**



theory for Argon 2p

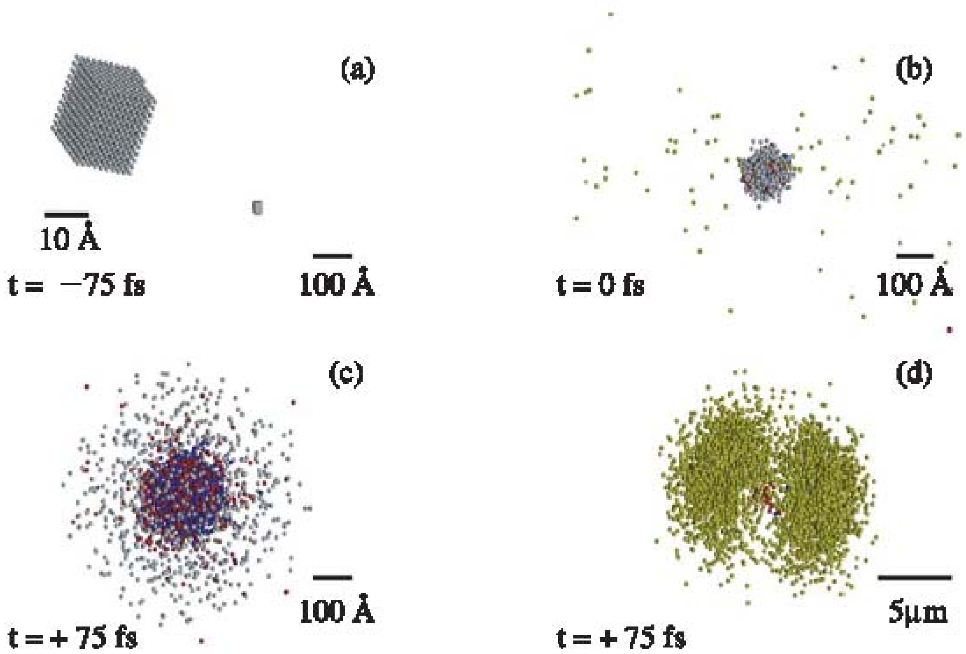
350 eV ~ 10^{16} W/cm^2

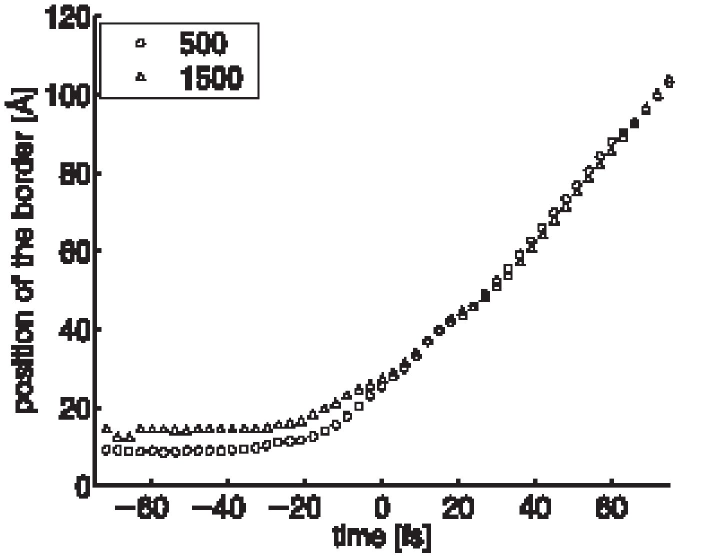


X-rays: Time evolution of exploding clusters

Z. Jurek, G. Faigel, M.Tegze
Eur. Phys. J. D 29, 217(2004)

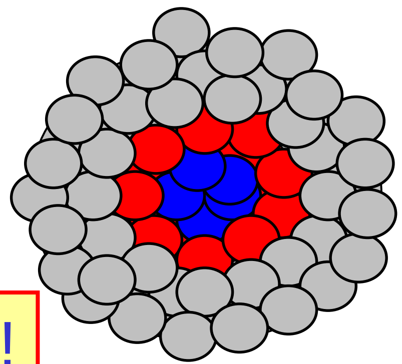
1500 atom carbon cluster, **λ= 0,1nm**
100 nm focus, 5*10^{12} Photons,
50 fs pulse





. 9. Radius of the inner part of the cluster vs. time.

Cluster inside a large droplet:
Delayed explosion?



no trapping of photoelectrons, 12 keV energy!

Delaying the expansion of exploding clusters

PRL **98,** 198302 (2007) PHYSICAL REVIEW LETTERS week ending 11 MAY 2007

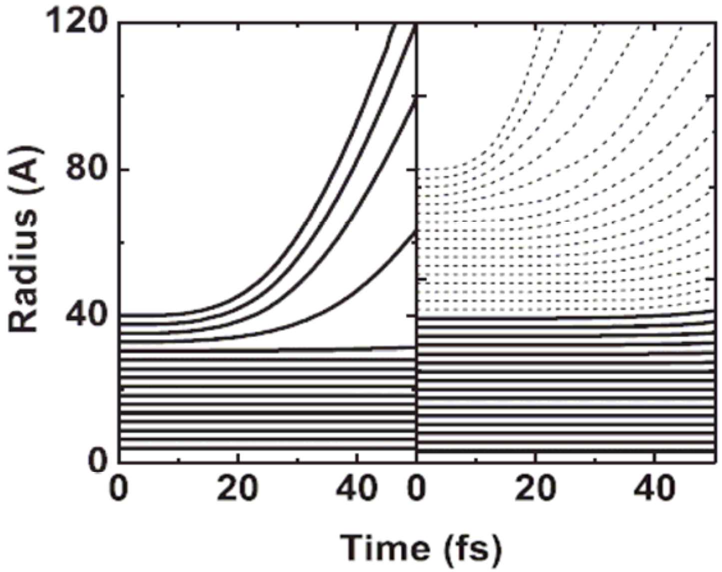
Encapsulation and Diffraction-Pattern-Correction Methods to Reduce the Effect of Damage in X-Ray Diffraction Imaging of Single Biological Molecules

Stefan P. Hau-Riege,* Richard A. London, Henry N. Chapman, and Abraham Szoke
Lawrence Livermore National Laboratory, P.O. Box 808, Livermore, California 94551, USA

Nicusor Timneanu

hydrodynamic model

explosion of an 8 nm carbon cluster



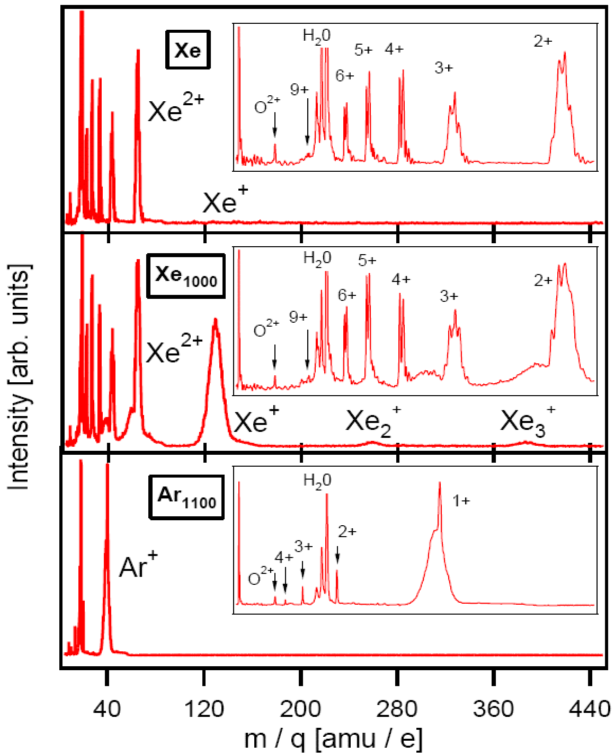
with tamper

FIG. 1. Motion of the atomic shells for the case of an 80 Å diameter carbon cluster (left) without and (right) with a tamper. The tamper shells are dashed lines.

Controlling the Coulomb explosion of core – shell clusters

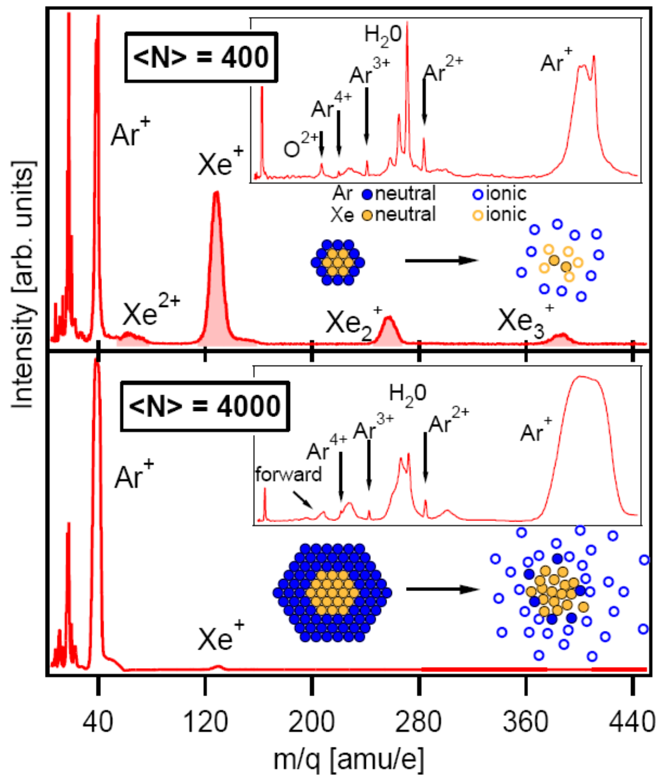
13 nm, 10^{14} W/cm^2

Pristine clusters



- **High charge states, fast ions**

Core – shell systems



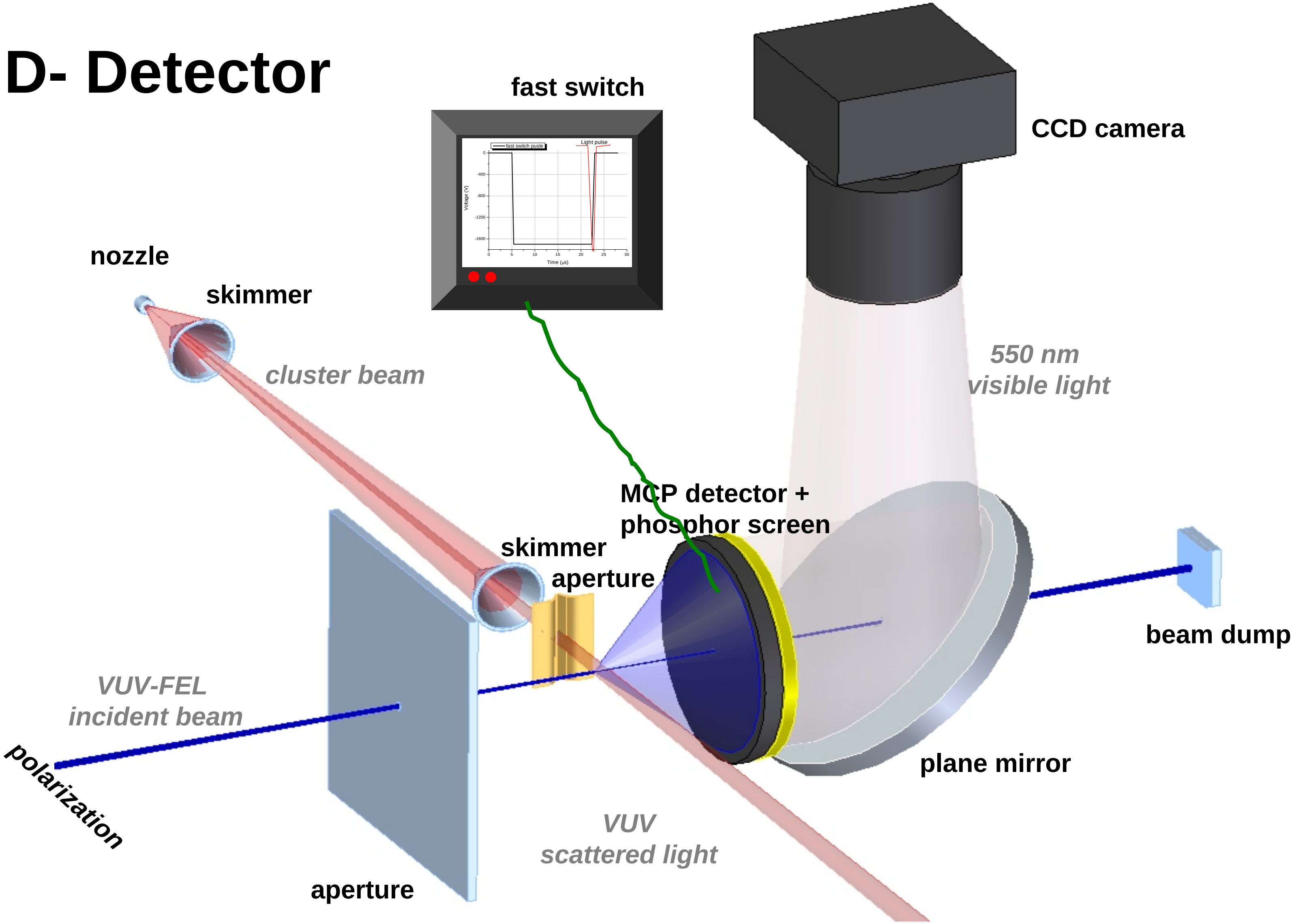
- **Xe core signal suppressed due to overlayer**

Recombination of cluster nanoplasma core – tamper for bio-molecule imaging

M. Hoener,et al J. Phys. B 41, 181001 (2008)

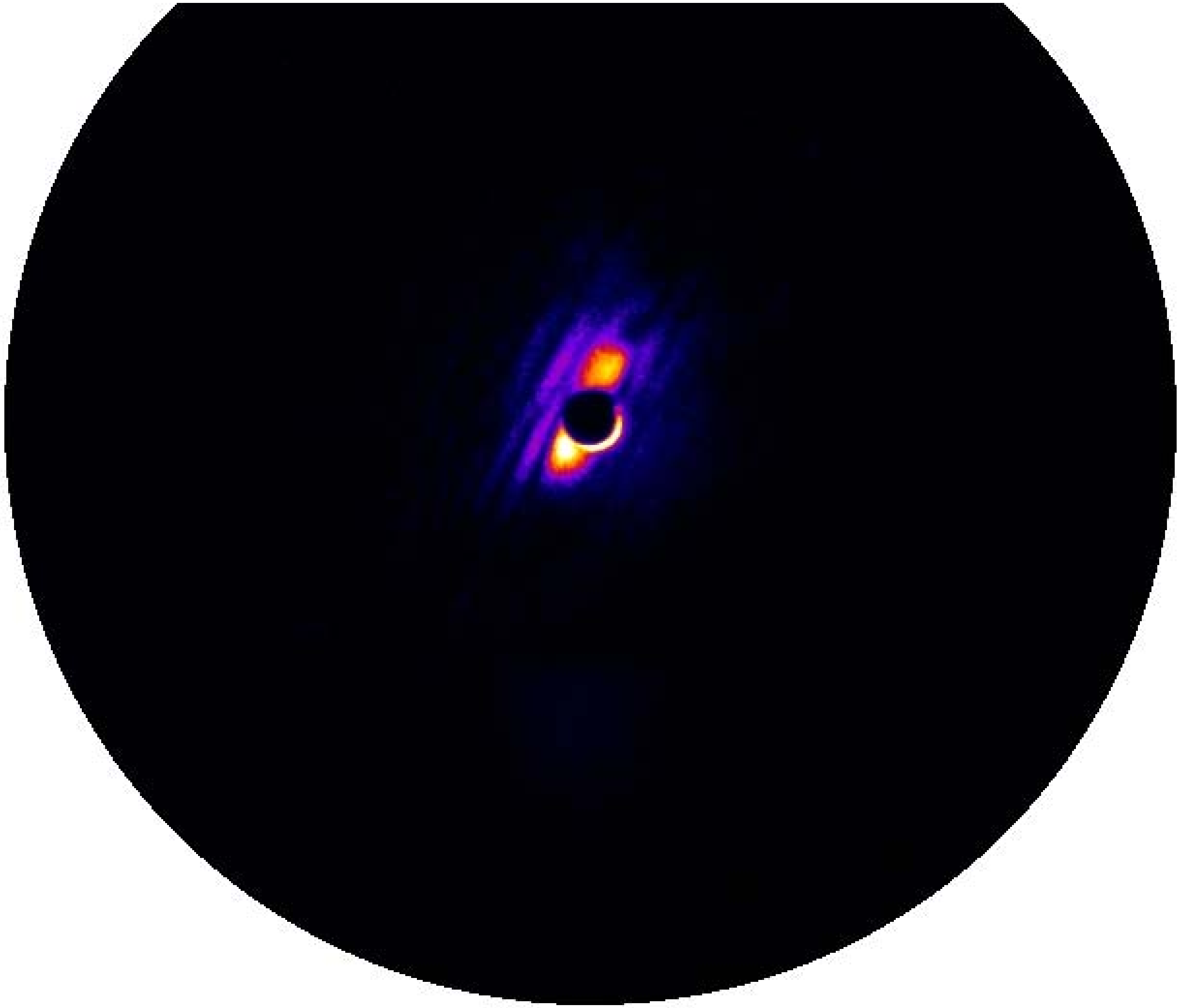
Scattering and Imaging

2 D- Detector



Single shot scattering of individual clusters

Large Xenon clusters

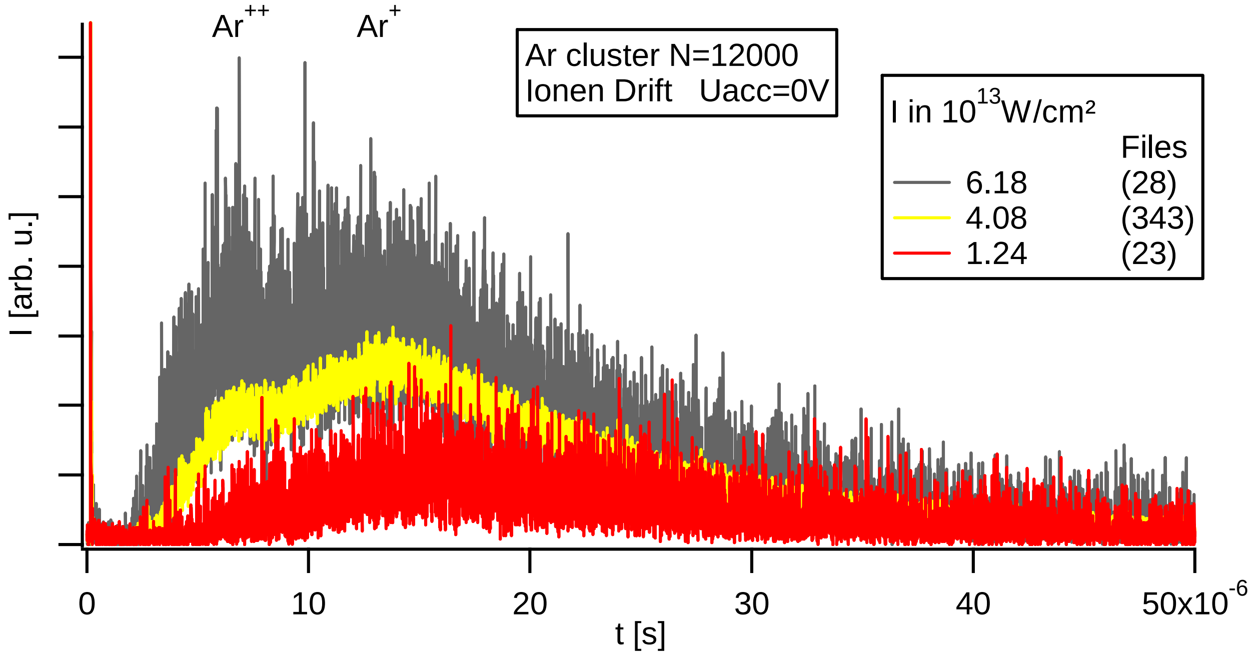


5 Hz

Do the clusters
stay intact
during exposure?

Do the clusters stay intatact during exposure during exposure ?

Ion Drift Times



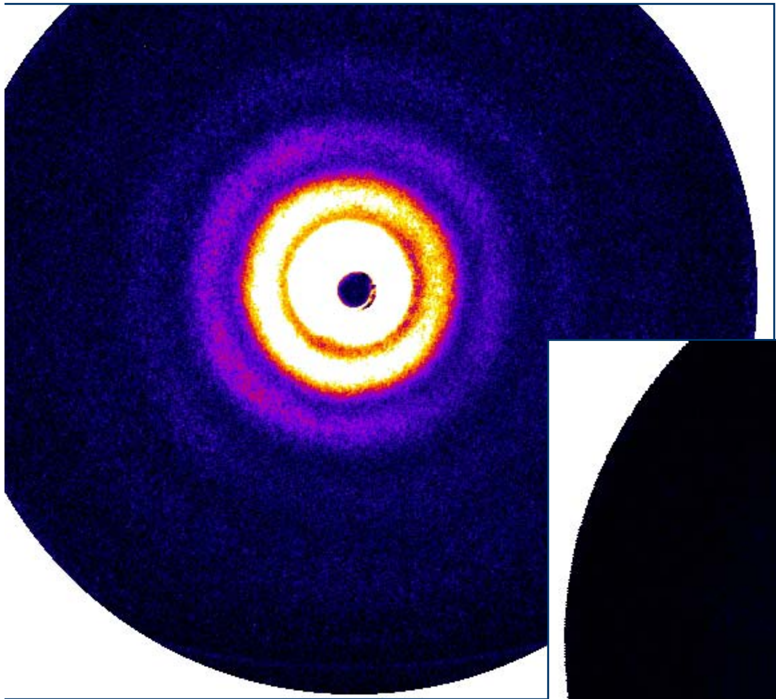
$V_{ar+} = 1.1 \cdot 10^4$ m/s (≅ 25 eV) fastes ions move ~ 3.3 Å in 30 fs

$V_{ar++} = 2.0 \cdot 10^4$ m/s (≅ 85 eV)

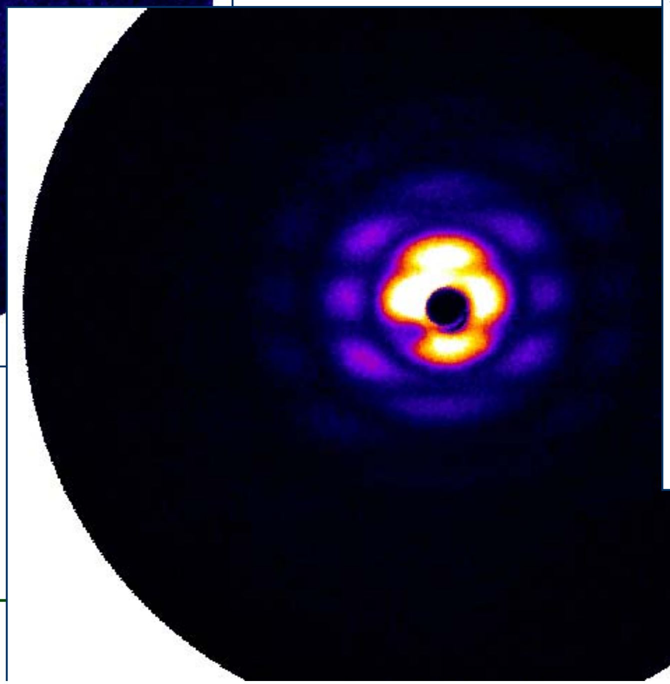
clusters stay intact during pulse

Simple cases

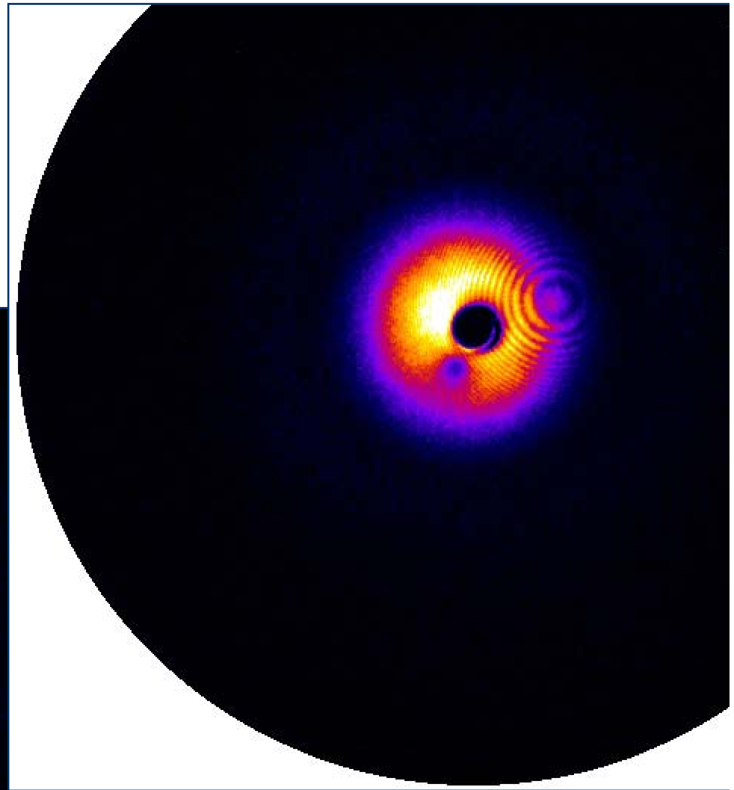
one cluster in focus



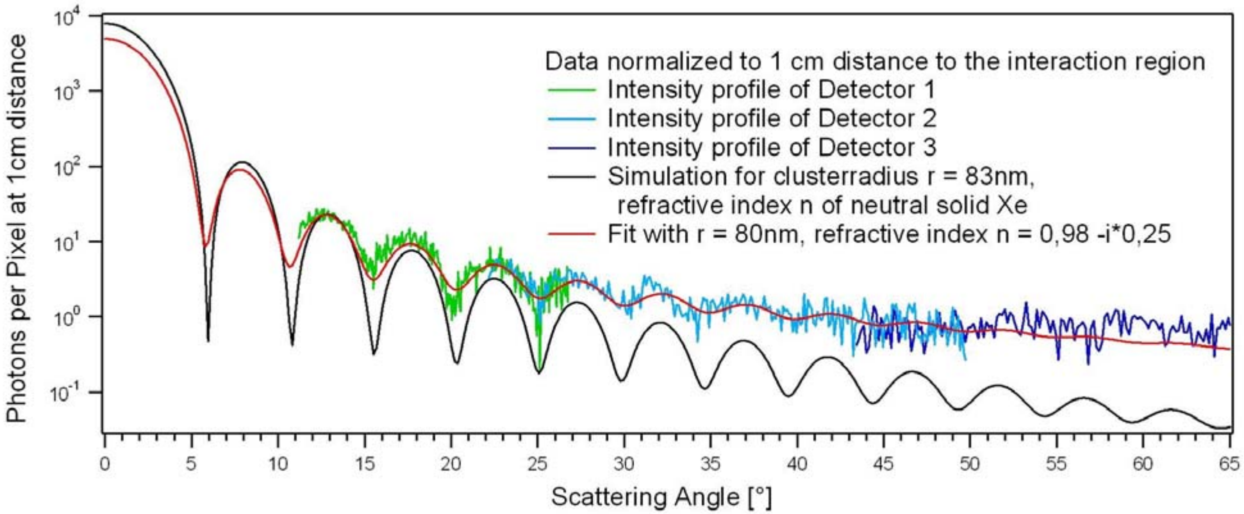
twin clusters
in focus



two clusters in focus
(„Newton‘s Rings“)



Electronic structure information scattering data



Fit optical constants in Mie theory to describe scatter data

Fits indicate strongly increased absorption, similar to plasma calculations

optical constants correspond to a plasma with average charge state 1.5 (Xe^{+} - Xe^{++})

Novel route to probe ultra-fast electron dynamics

Rupp, Adolph, Bostedt, et al, in preparation

Technical issues

Cluster sources:

- van der Waals systems/ hydrogen bonded
- Metal clusters
- Semiconductors
- pure/doped clusters, core shell structure

Detectors:

Ionisation processes :

- Tof-mass spectrometer, X-ray fluorescence,

  COLTRIMS, Thomson parabola, velocity map imaging

Electron emission:

- electron spectrometer, COLTRIMS, velocity map imaging
- 2D detectors for scattered light

Technical issues II

Mirrors:

- spot size below 1 μm
- differential pumping

Time resolved studies

- pump-probe techniques; IR, optical laser/ X-rays,
- high power IR lasers

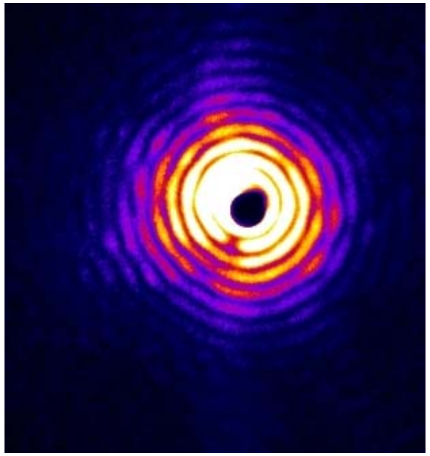
Summary

Clusters in intense x-ray beams

- interesting, complex, many body processes
- strongly size, time- and wavelength dependent dynamics

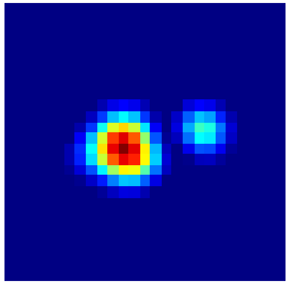
- neutral plasma in the core

  neutral atoms, delayed expansion?
- new processes?



direct link to single shot imaging of single particles

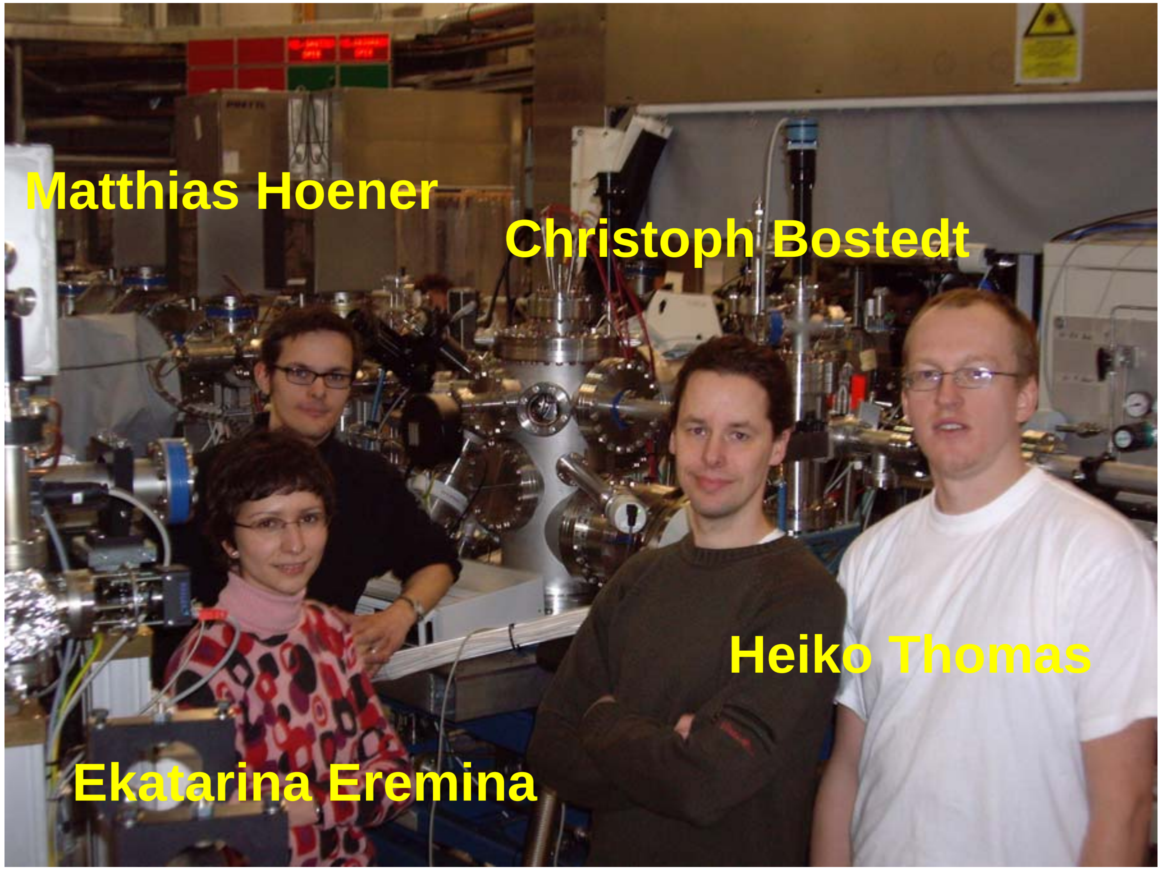
reconstruction of structures



shorter wavelength: higher lateral resolution

The group

Technische Universität Berlin



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