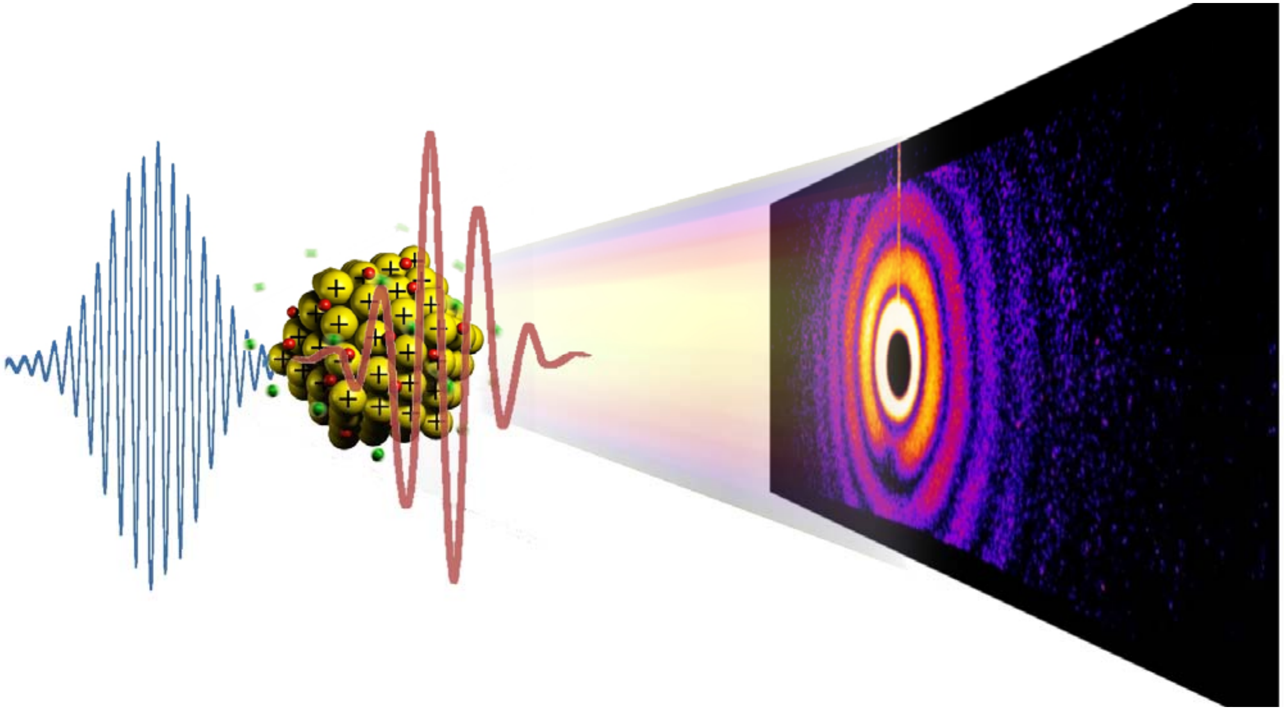
X-ray FEL experiments on clusters:
Ultrafast x-ray induced dynamics

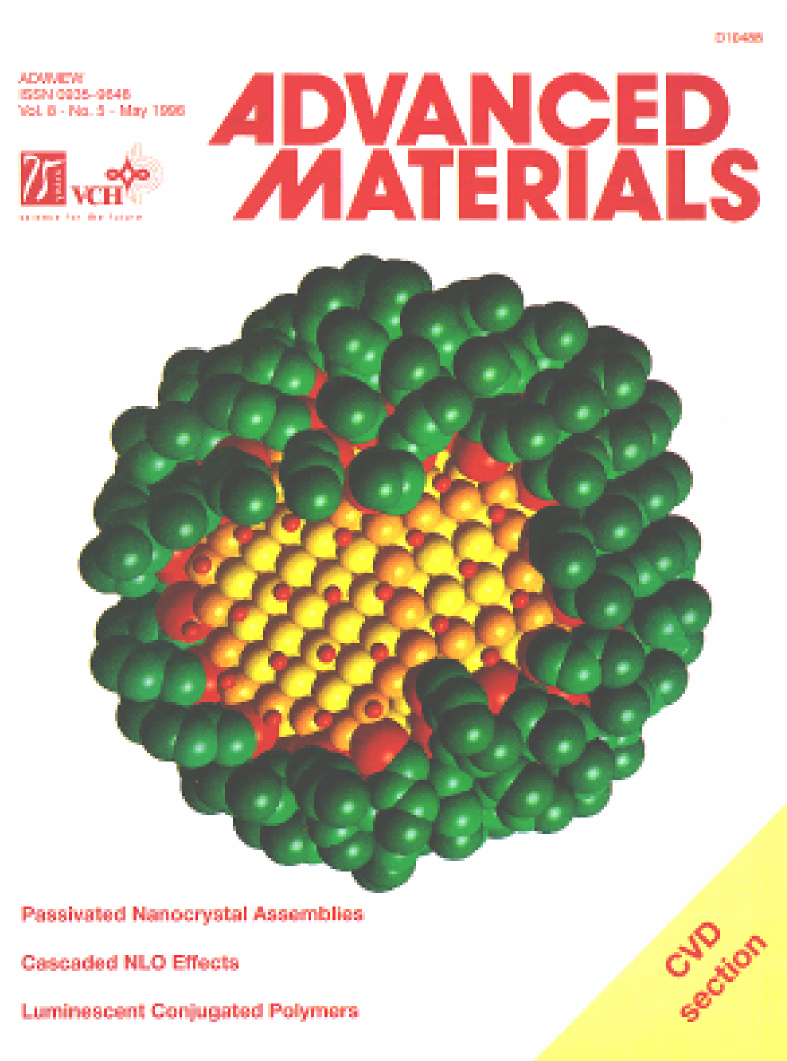
Thomas Möller, Technische Universität Berlin

Bad Honnef, April 12, 2012

Structure and light induced dynamics



Issues and Questions



Clusters and Nanocrystals are new materials

Size dependent properties

- Optical properties
- Catalytic activity
- Magnetic properties
- Photochemical processes
- **Phase transitions**
- **Light induced dynamic and dissociation**

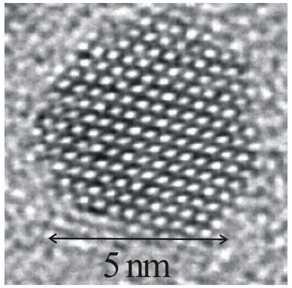
geometric structure

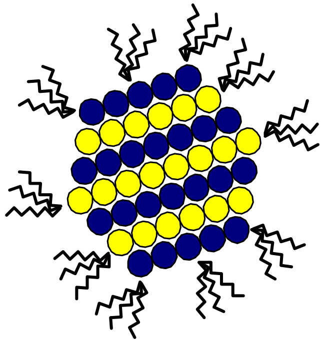
positions of the individual atoms, shape

So far: mass spectroscopy,
spectroscopy, (TEM)

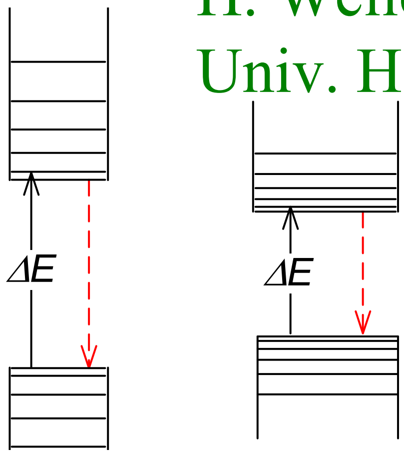
X-ray scattering

Clusters and nanocrystals are new materials with size dependent properties

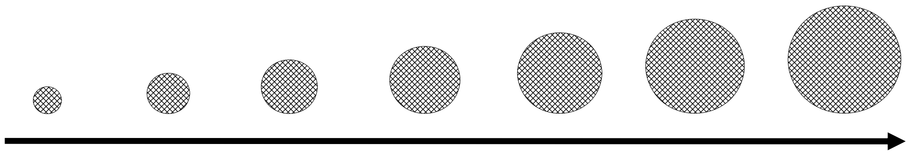




H. Weller
Univ. Hamburg



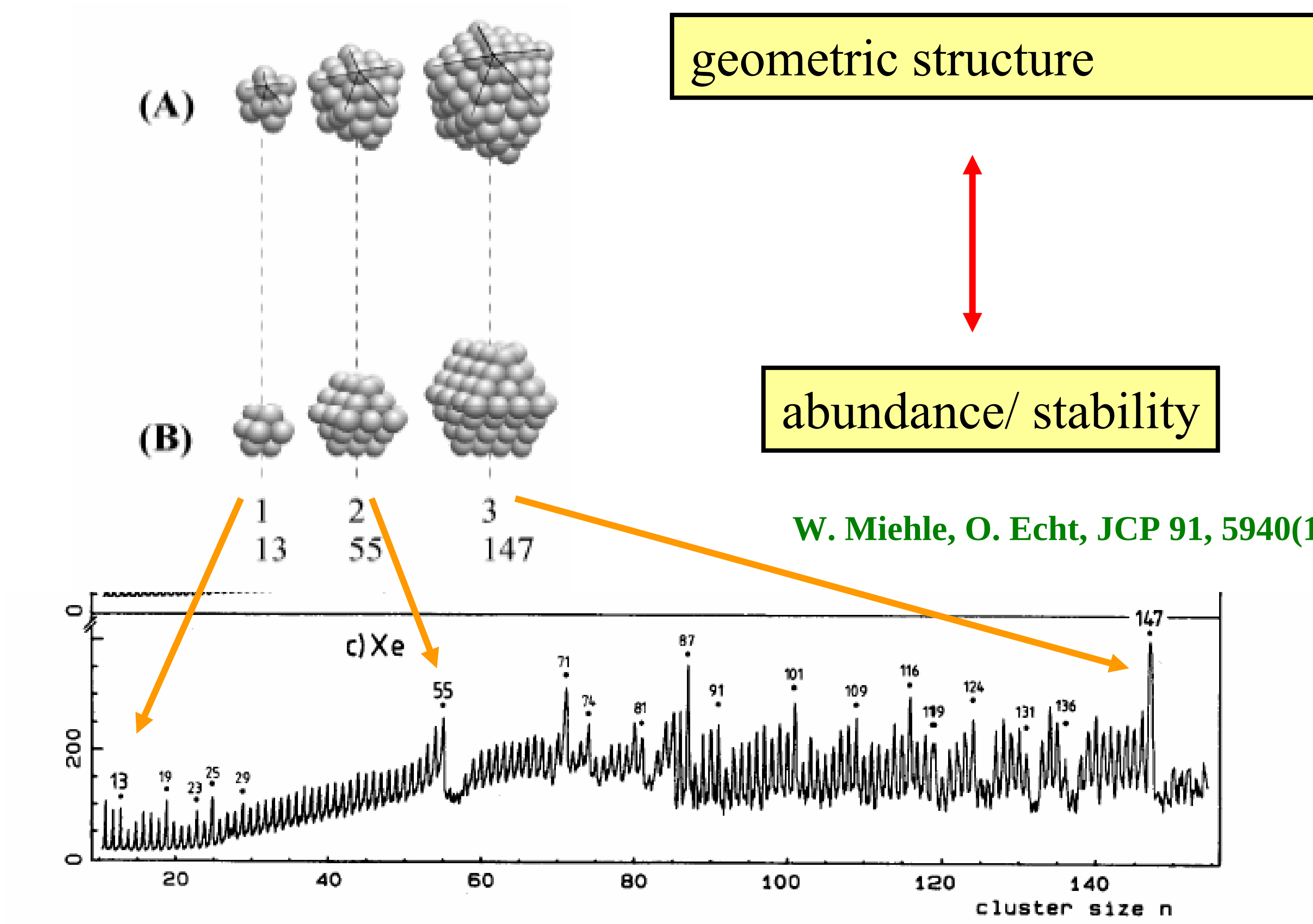
$$\Delta E = E_g + \hbar^2/8R^2(1/m_e + 1/m_h)$$



Applications:

Luminescent biological labels

I. Geometry and Stability of Clusters

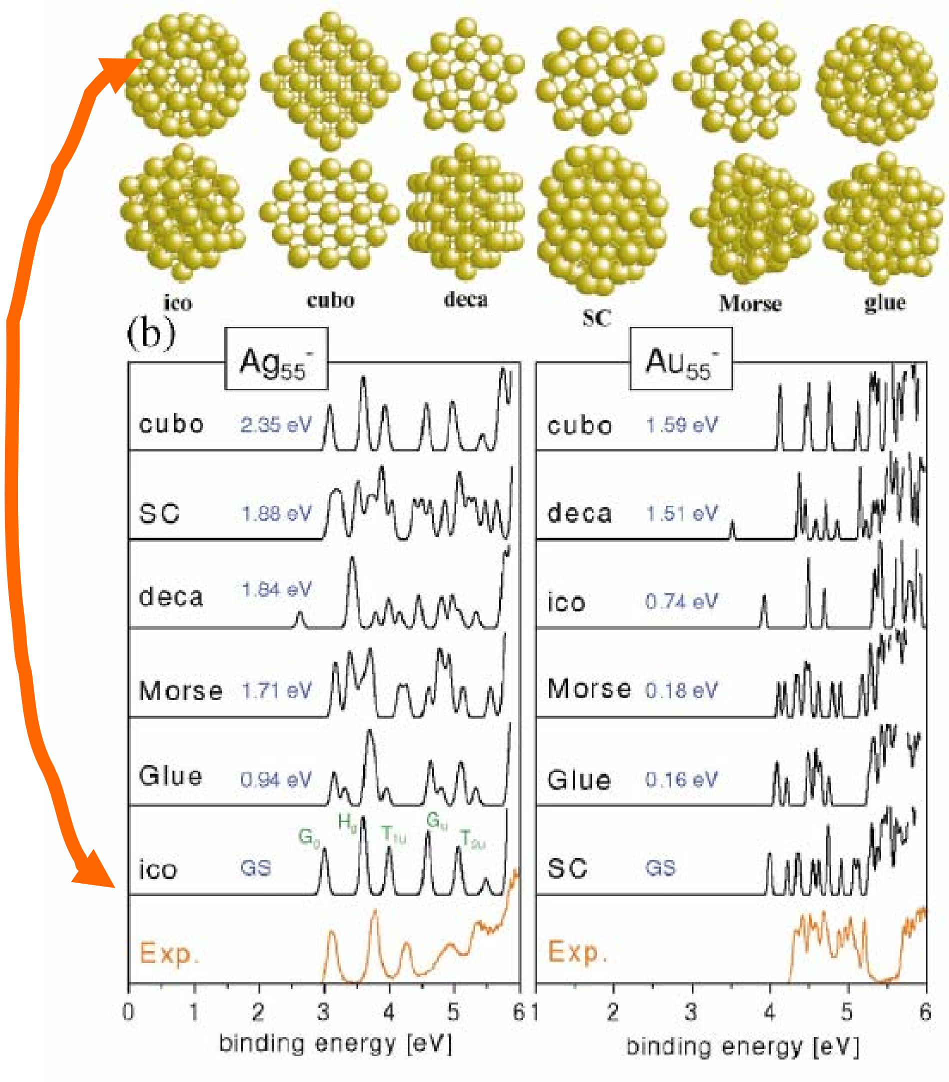


geometric structure

abundance/ stability

W. Miehle, O. Echt, JCP 91, 5940(1989)

Geometrical and electronic structure of medium size clusters



Structure determination from valence band photoelectron spectra

- structural model needed
- highly symmetric structures
- a few element clusters
- small clusters (less than a few hundred atoms)

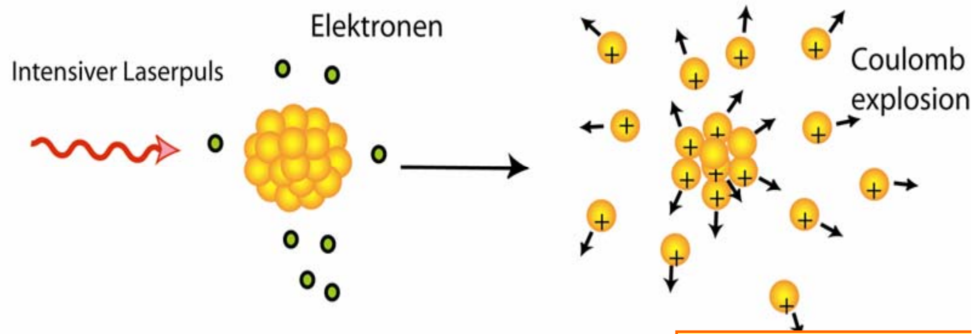
Works for few systems only!

H. Häkkinen, B. Issendorff et al. PRL 93, 93401 (2004)

II. Cluster - light interaction at short wavelength

Cluster: „nano-lab"

- isolated objects, bulk density
- intra/interatomic effects



At IR regime:
T. Ditmire et al.
M. Lezius et al.
Meiwes-Broer et a
M. Mudrich
....

Driving questions:

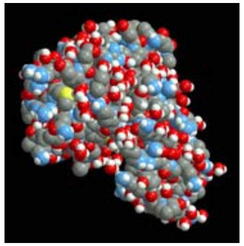
- mechanism of absorption and ionization
- nanoplasma formation, electron-ion recombination
- time scale of electron removal / explosion dynamics, hydrodynamic expansion
- radiation damage in different systems, covalently bound, metallic, ionic

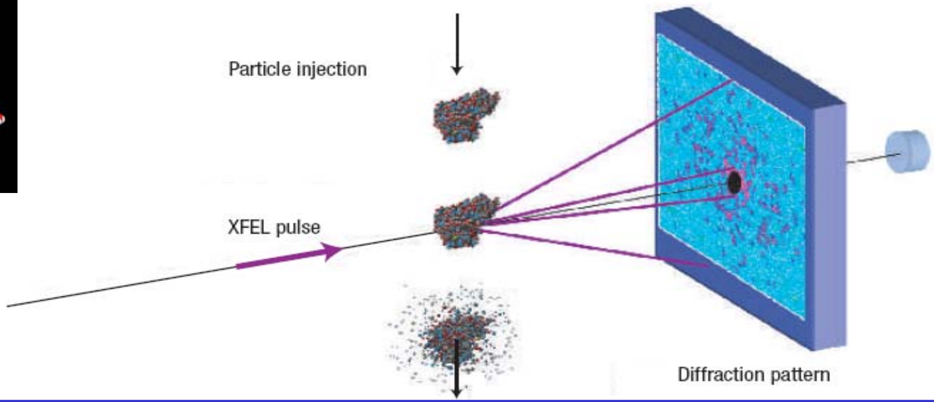
→ imaging of particles and their dynamics

Structure determination with a FEL

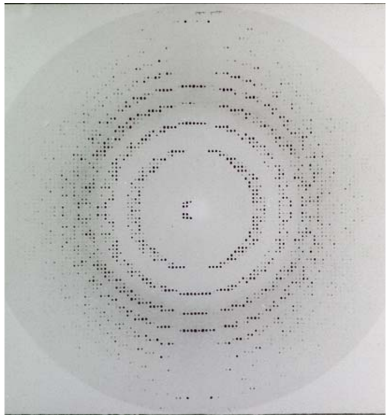
direct method

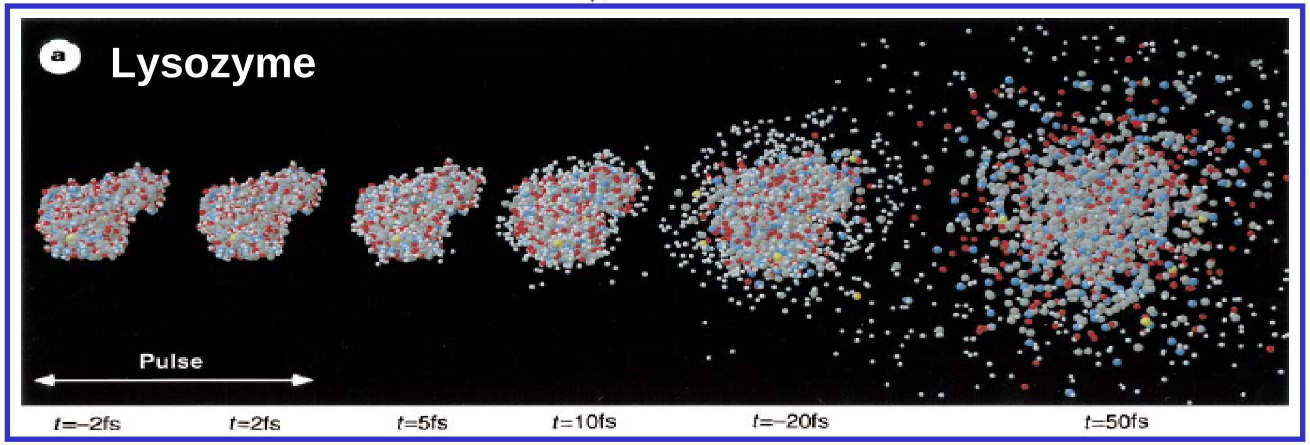
Molecules atomic resolution





Crystal



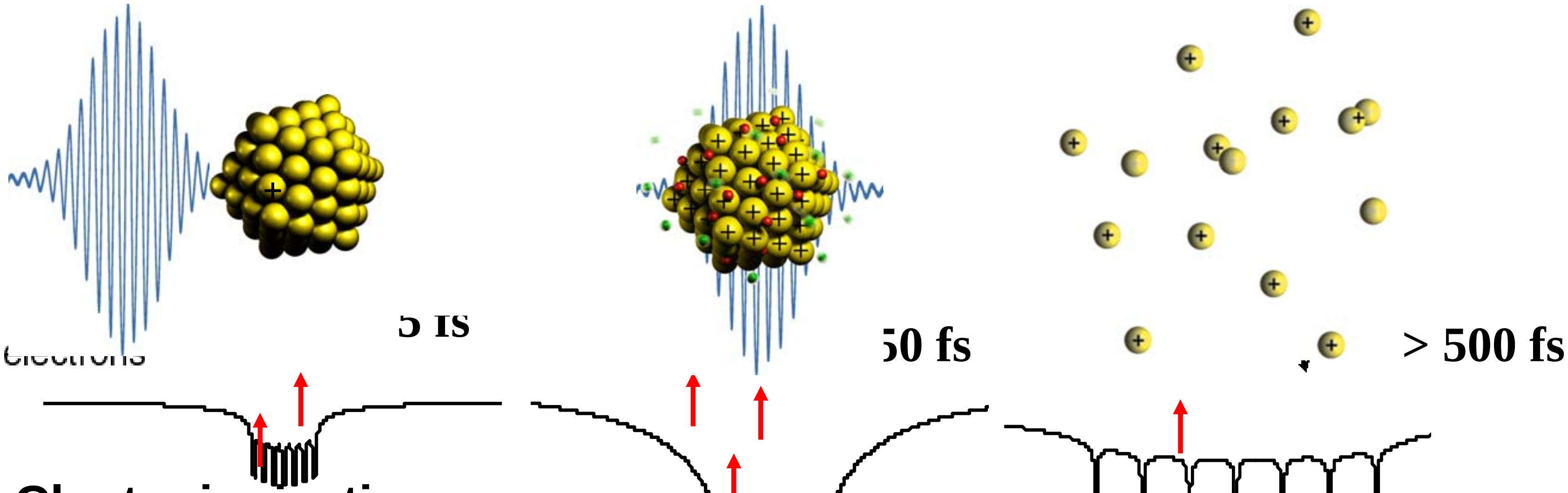


R. Neutze, J. Haidu et al., Nature 406, 752 (2000)

Radiation damage and Coulomb explosion

Intense light pulse - cluster interaction

Beginning of the pulse **During the pulse** **After the pulse**



Cluster ionisation

Single photon
Ionization
Few electrons are
removed from cluster

Disintegration,

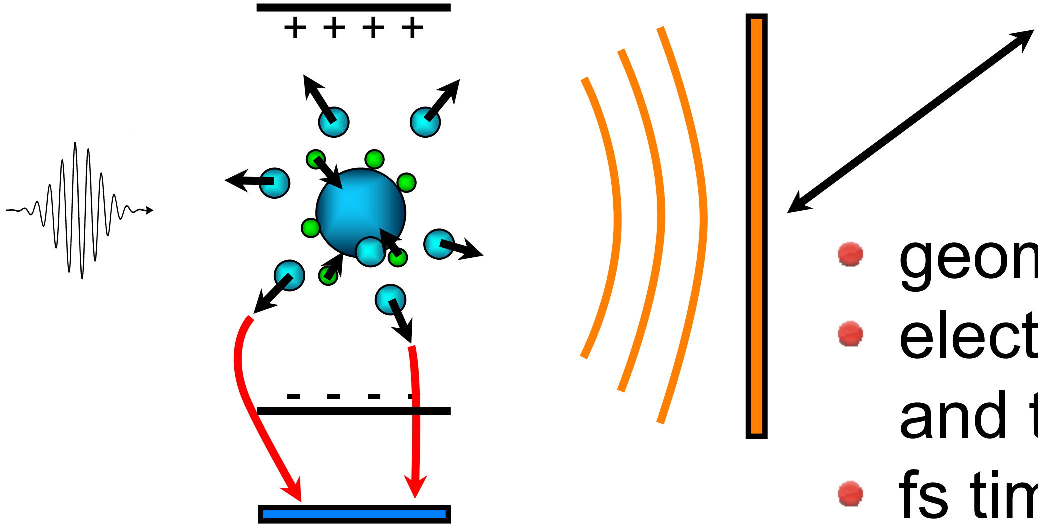
Strongly dependent on wavelength and power density!

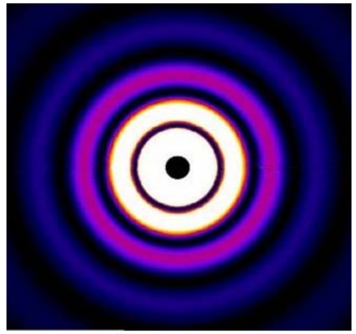
Inne
Electron trapping
Nano plasma, neutral core

Experiments Wabnitz et al, Nature 420, 482 , Laarmann et al, PRL 92, 143401, PRL 95, 063402(2005)

Theory R. Santra, PRL 91, 233401 (2003), Siedschlag, Rost, PRL 93, 43402 (2004), Ziaja, Phys. Rev. Lett. 102, 205002 (2009)

Spectroscopy and light scattering of clusters



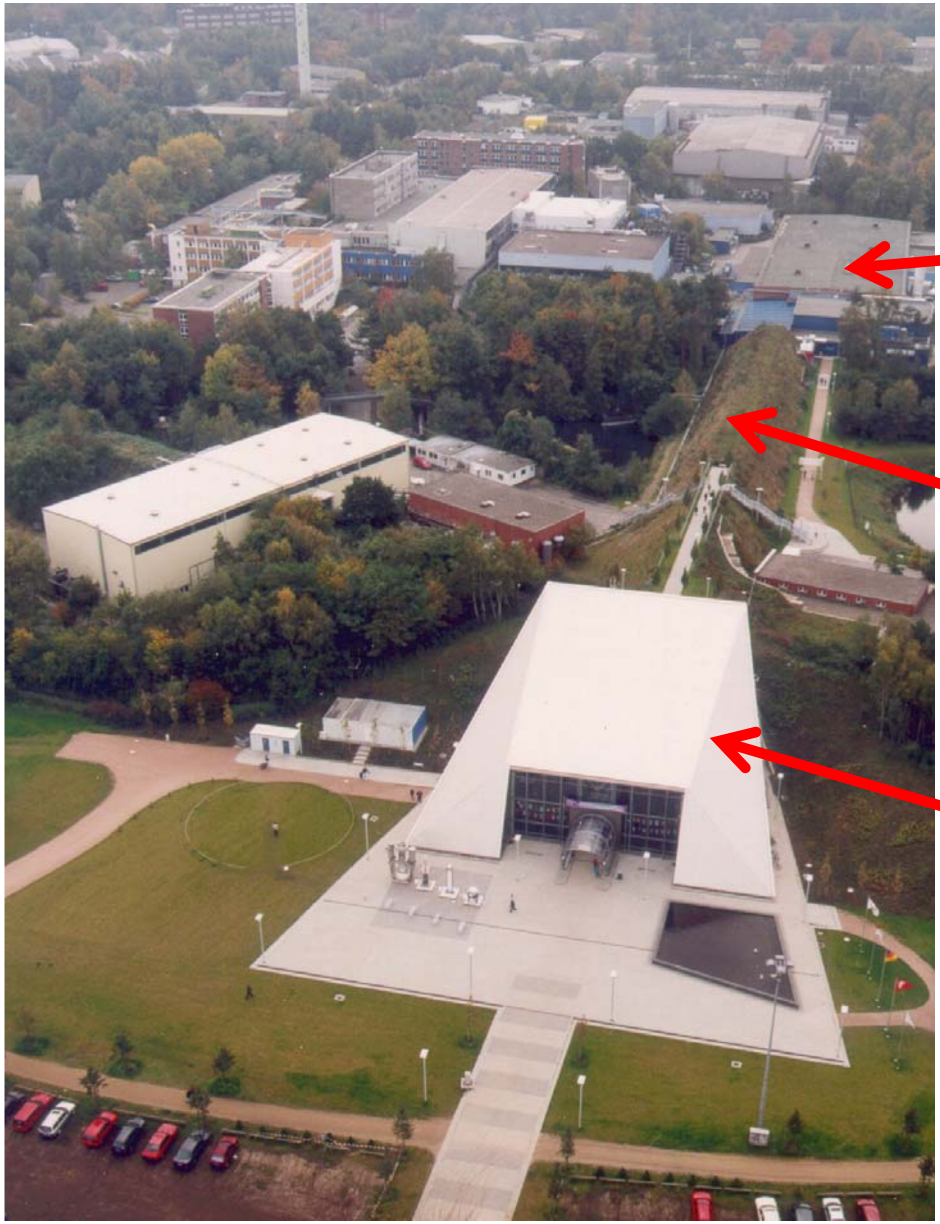


- geometry of the cluster
- electronic processes and transient states
- fs time scale
- during the pulse

- Ion/electron spectroscopy
- fluorescence
- ionisation, relaxation, recombination
- ps-µs time scale after the pulse

Different time scales!

FLASH FEL at DESY (Hamburg)



Electron gun

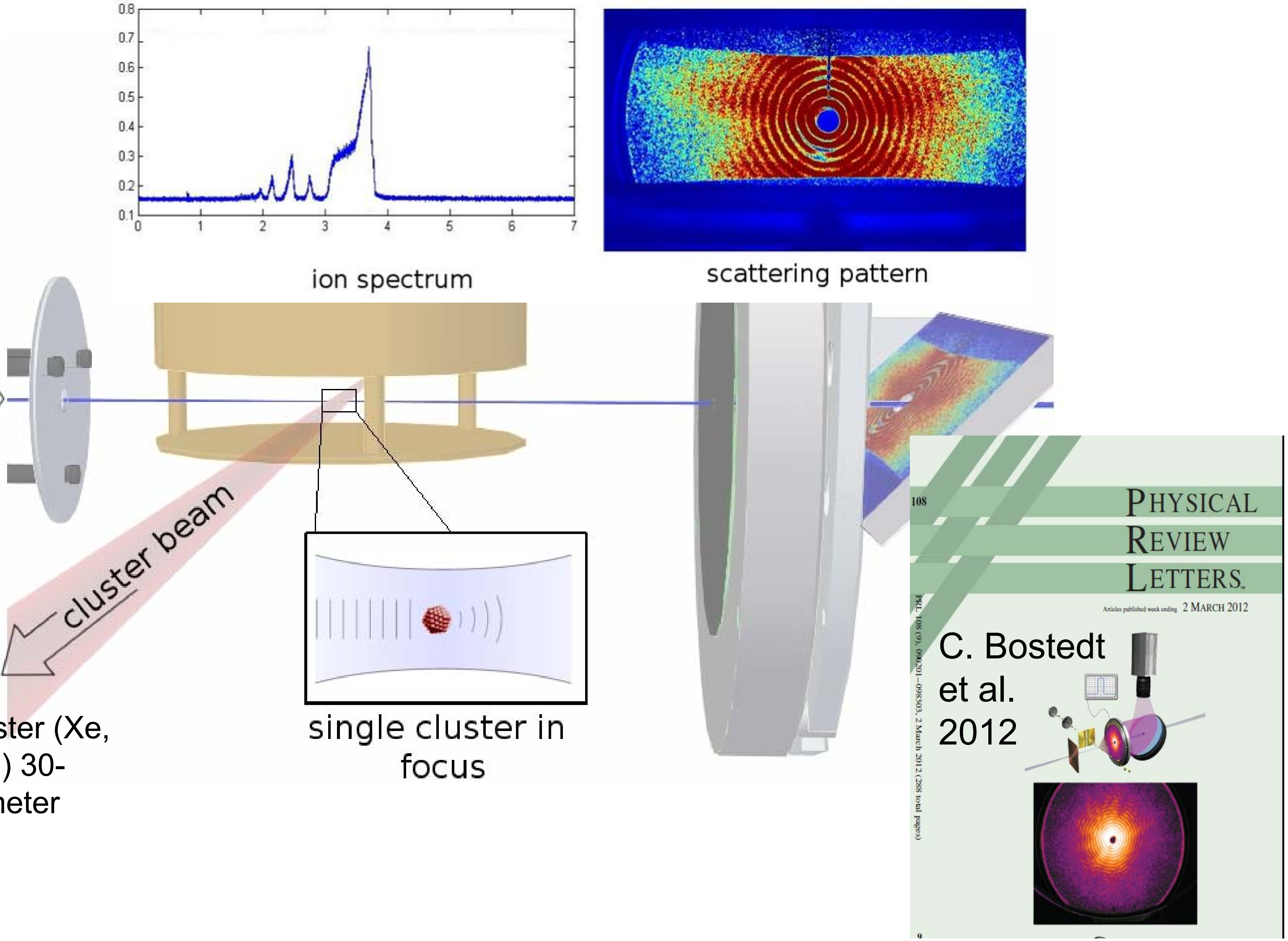
Linac and FEL undulator

Experimental hall (User Facility started July 2005)

- 4,5- 50 nm
- 10-300 µJ
- 1 GW_{peak}
- 10-200 fs

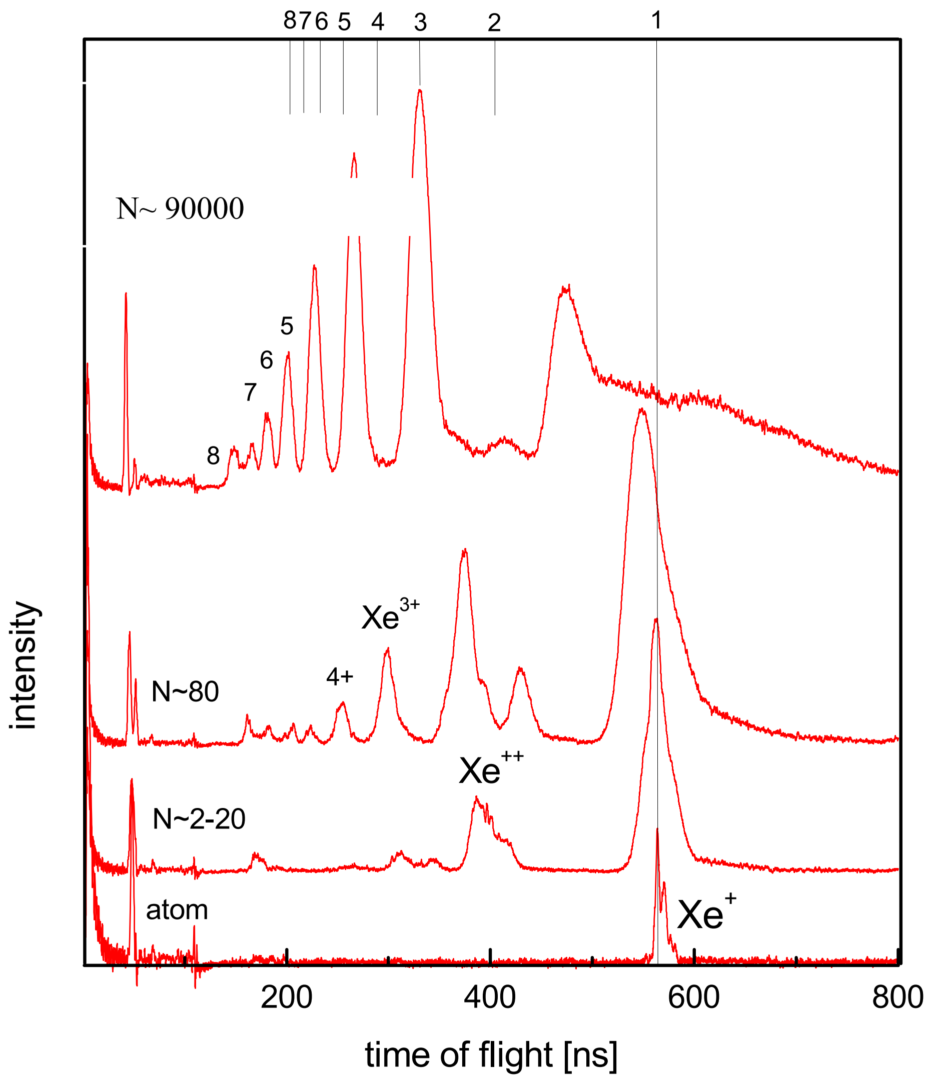
Simultaneous light scattering and ion spectroscopy on individual clusters

FLASH Free-Electron Laser 90eV, ~100fs, ~10e15W/cm²



Rare gas cluster (Xe, Ar, core-shell) 30-1500nm diameter

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 atoms**
- **detailed theoretical work to explain the enhanced absorption**

Plasmabsorption (IB)

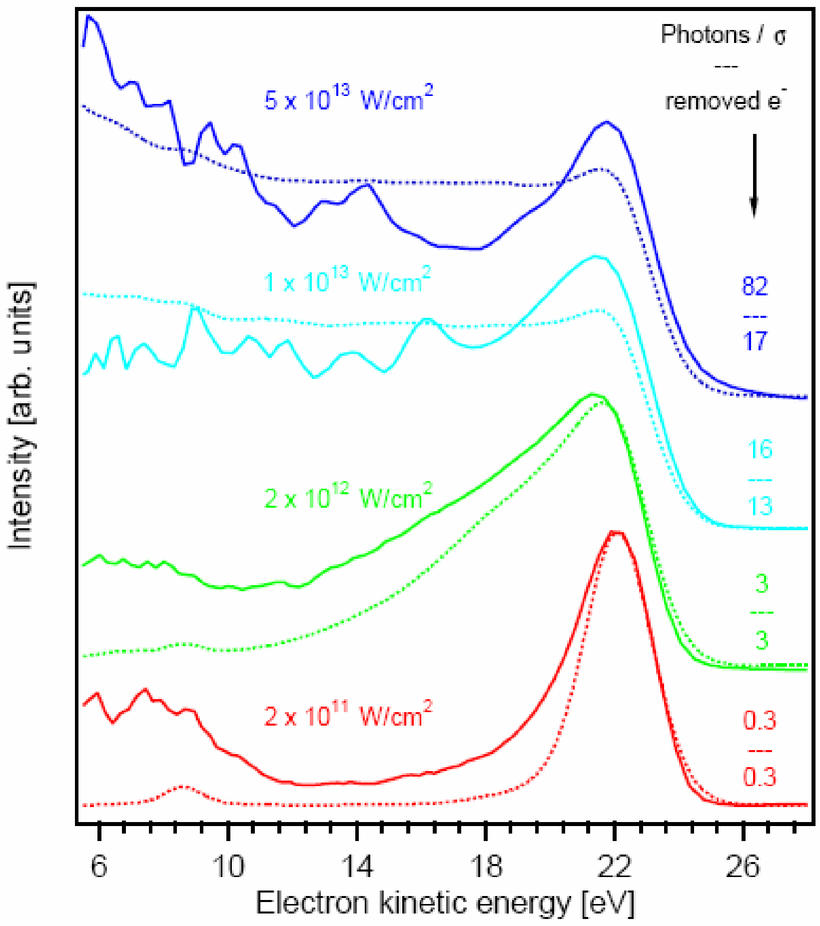
C. Siedschlag, J. M. Rost , PRL 93, 43402 (2004)

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

Ionisation studied with electron spectroscopy

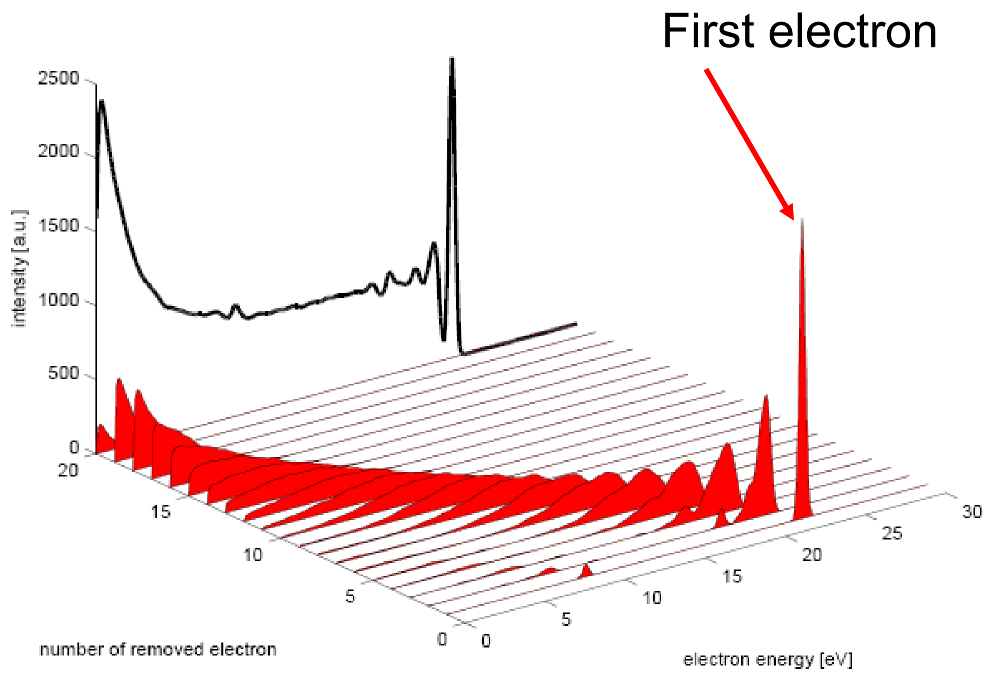
- experiment

····theory



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

Ar_{150} clusters, 32 nm,
a few $10^{11} \sim 10^{14}$ W/cm^2



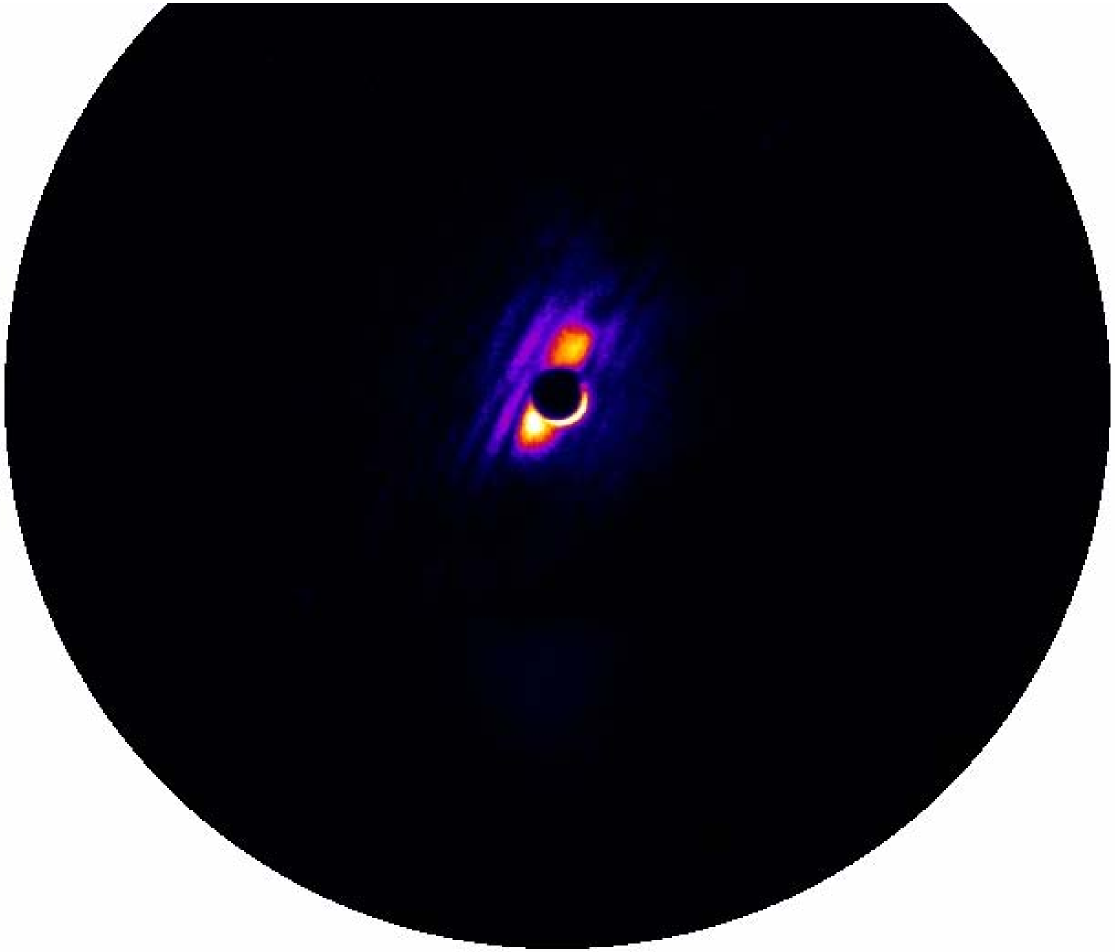
Theory (T. Fennel, Rostock)

sequential emission of electrons

only a small percentage of generated photoelectrons can leave the cluster

Single shot scattering of individual clusters

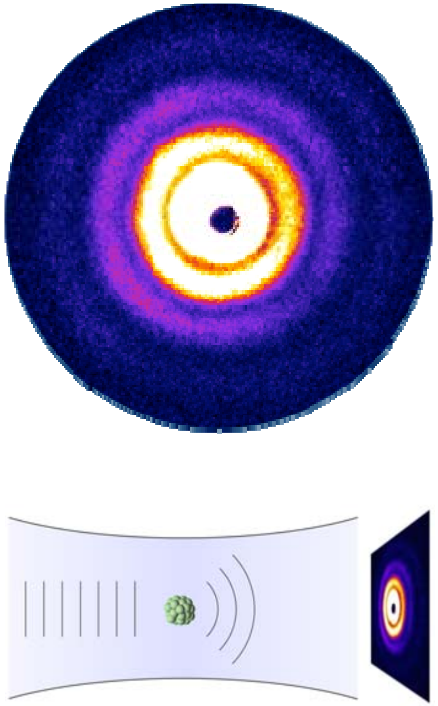
Large Xenon clusters



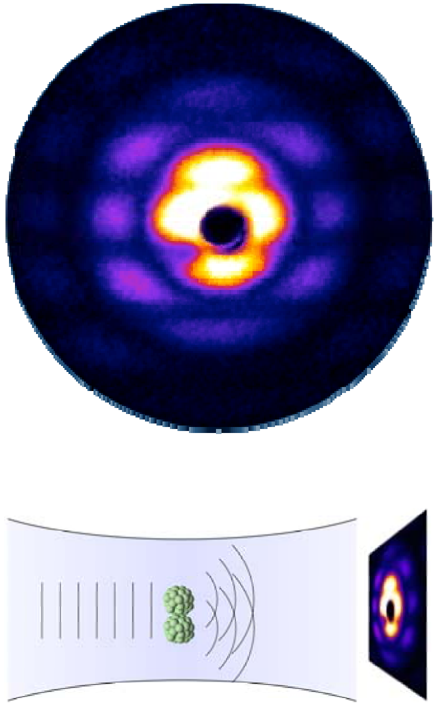
5 Hz

Clusters stay intact
during exposure
$\Delta R < 3Å$

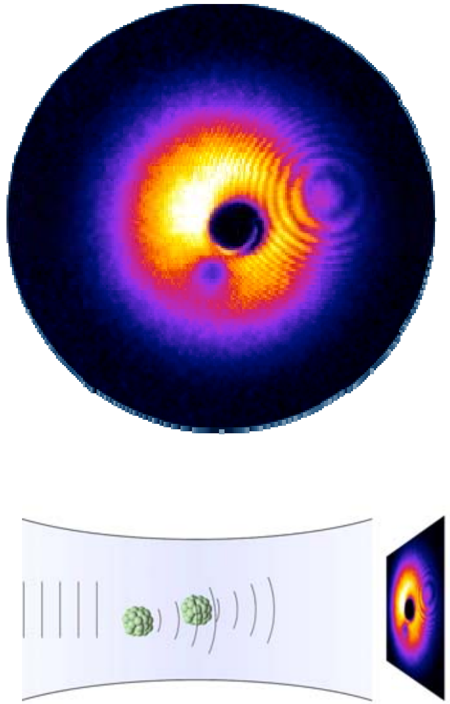
Single shot cluster imaging



one cluster in focus



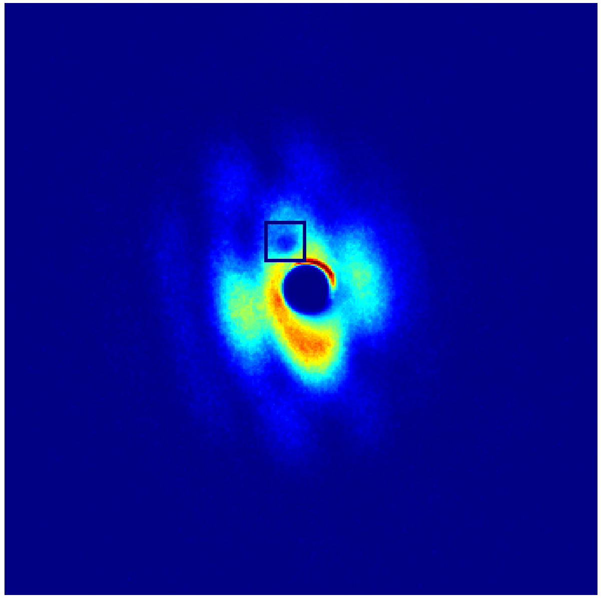
twin clusters in focus



two clusters in focus

Reconstruction of scattering patterns

Fengline Wang,
Henry Chapman, Beata Ziaja

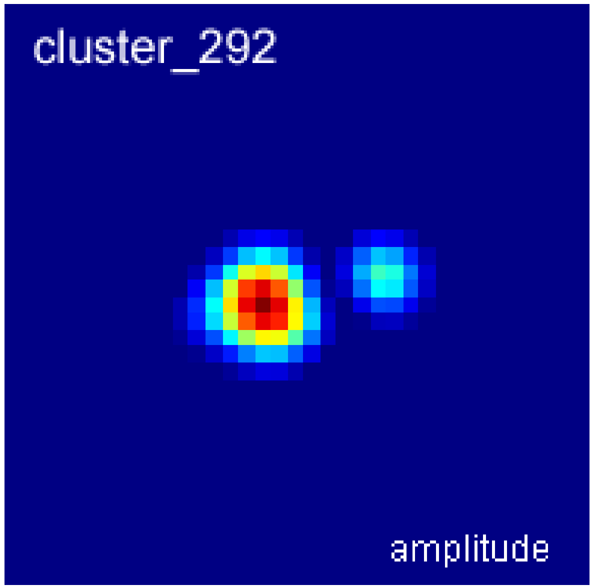


scattering pattern

two clusters in
direct contact

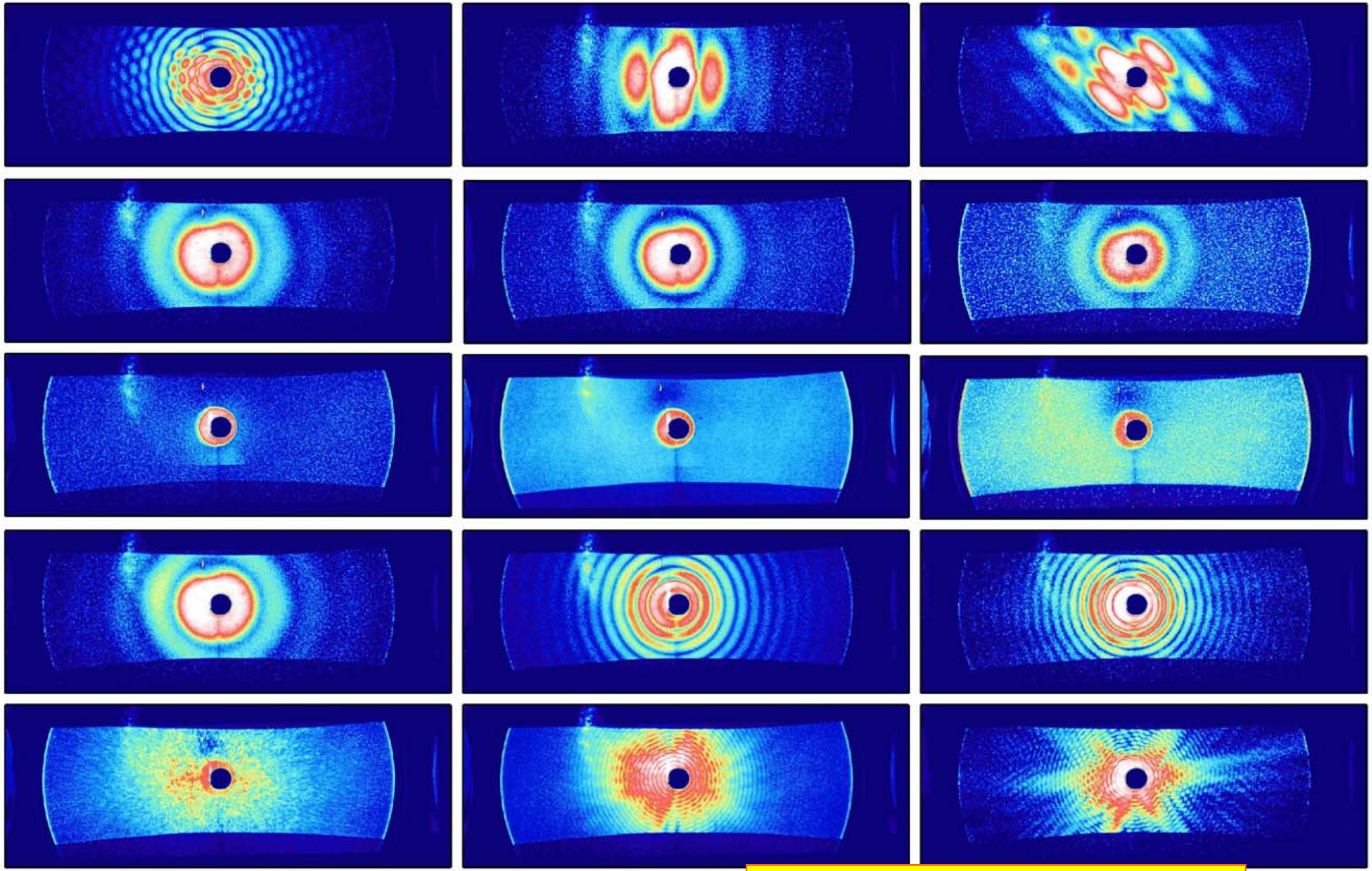
5-15 % twins !

cluster growth by coagulation

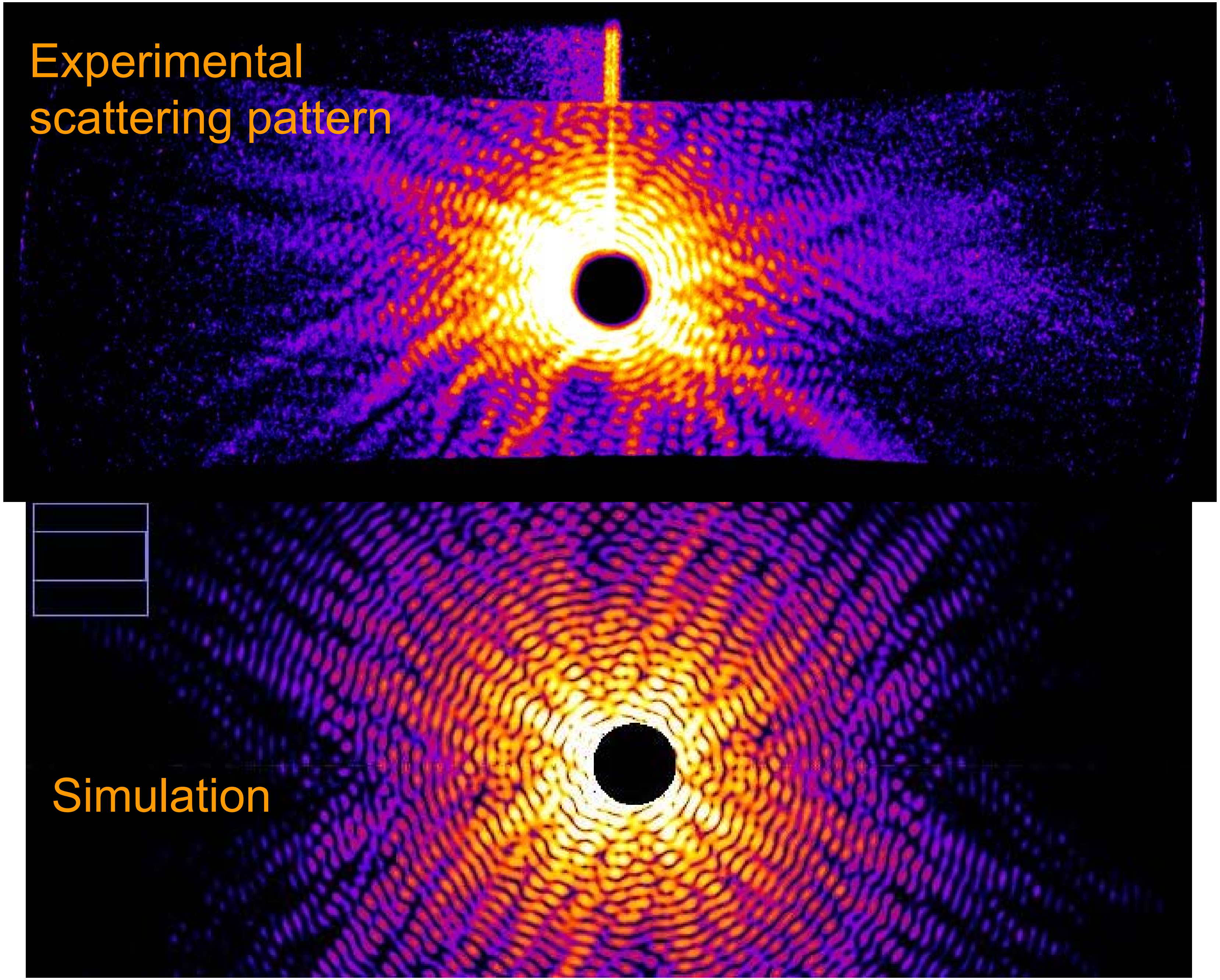


reconstructed image

Scattering pattern and cluster structure



Poster L. Fückiger

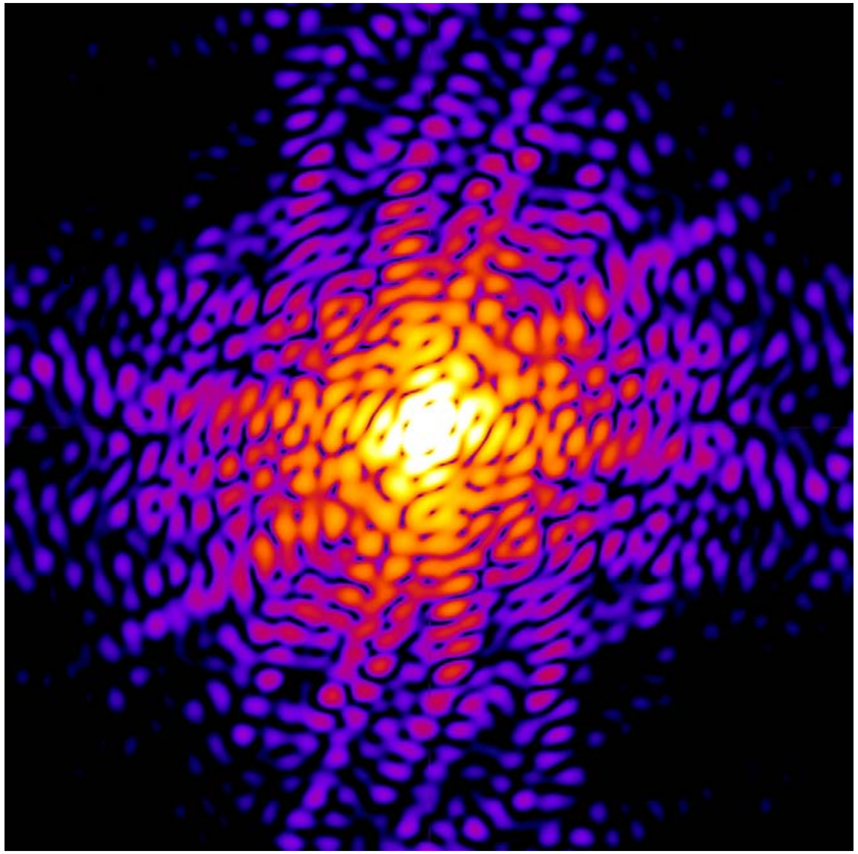

Experimental
scattering pattern
Simulation

Shape of large gas phase clusters

700 nm

Grainy structure
hailstones = snapshots of
cluster growth by coagulation

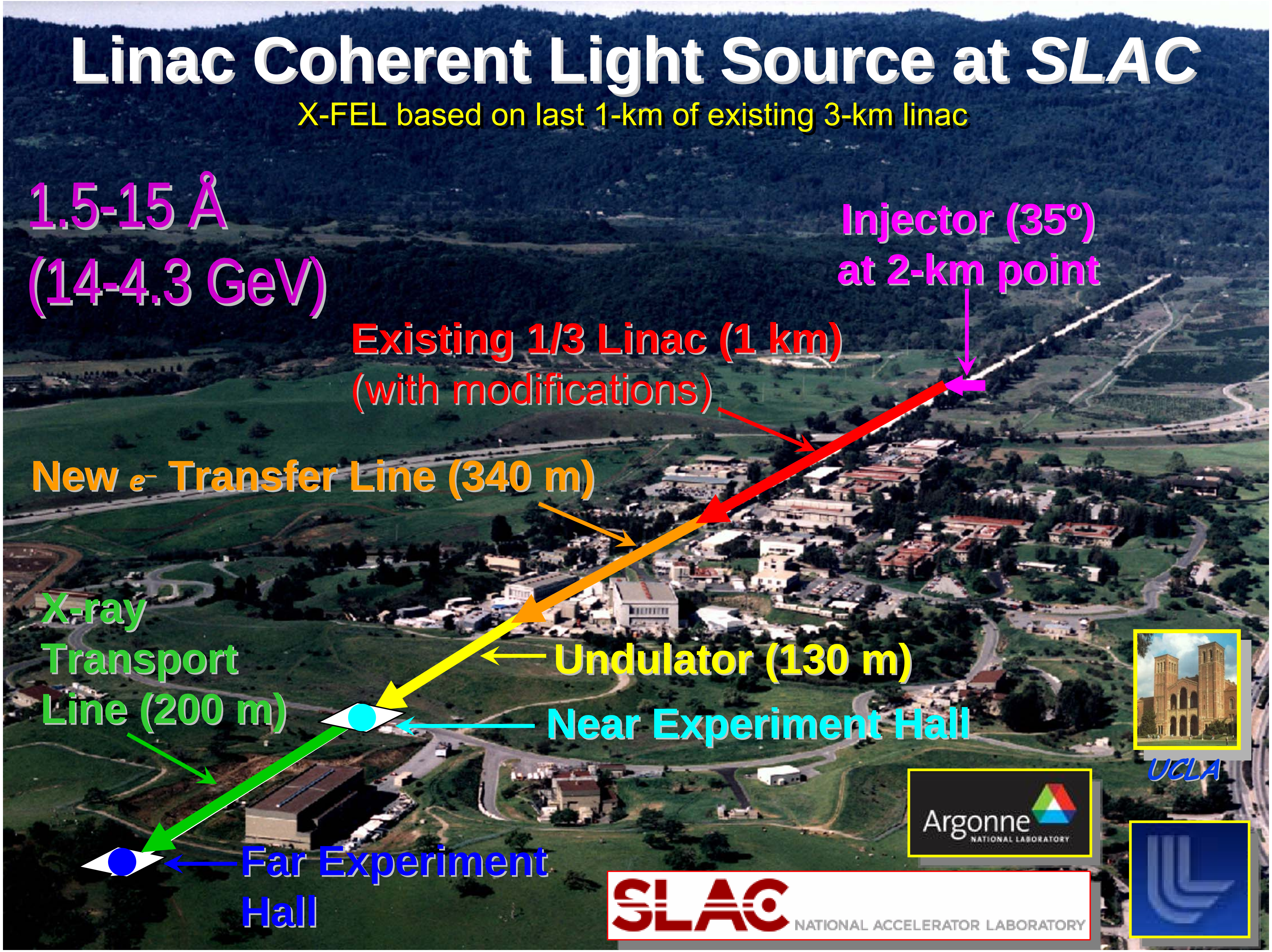
Simulation of scattering pattern
with 2D Fouriertransform



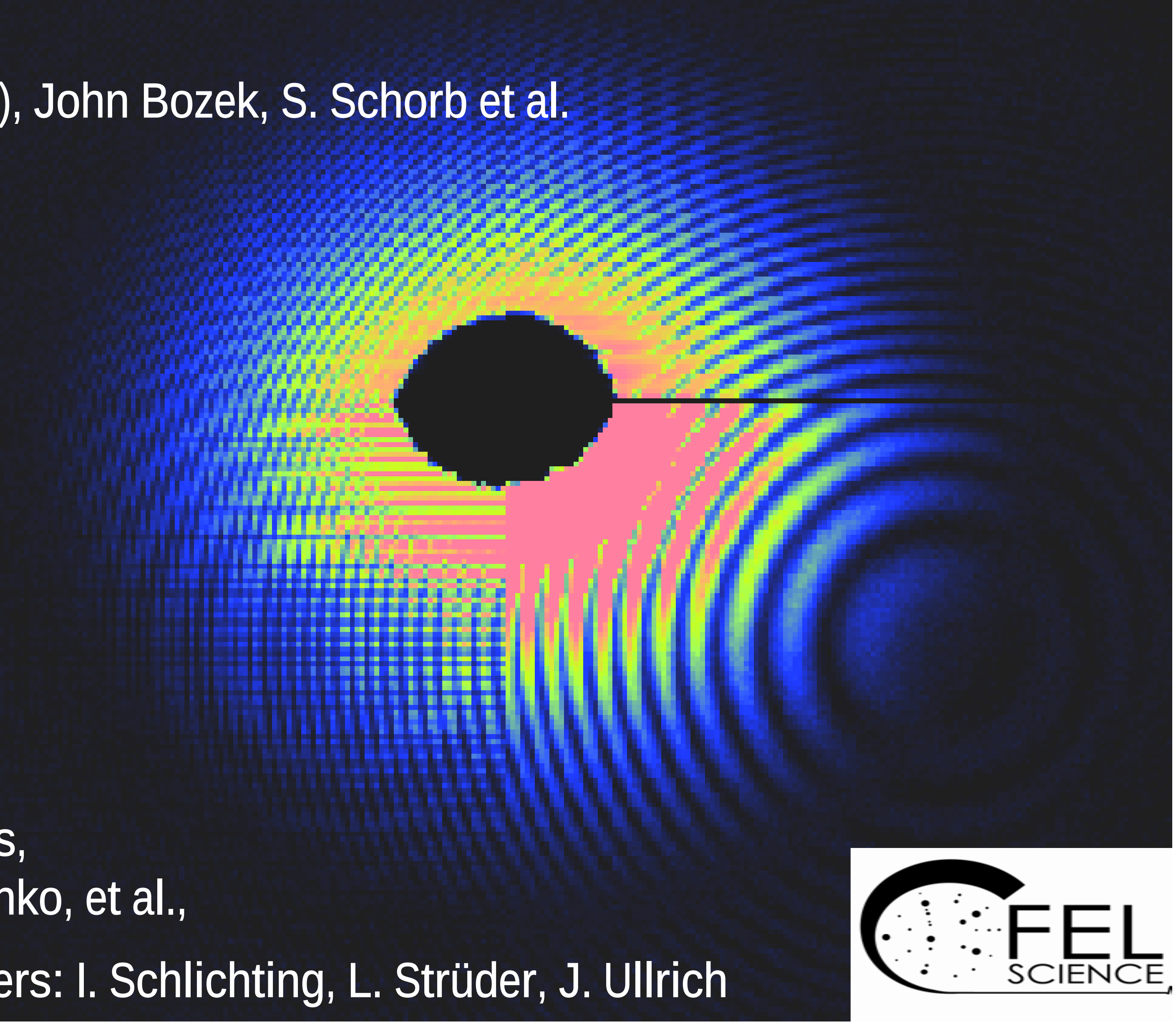
Poster D. Rupp

Linac Coherent Light Source at *SLAC*

X-FEL based on last 1-km of existing 3-km linac



Dynamics in Clusters



LCLS / SLAC

Christoph Bostedt (PI), John Bozek, S. Schorb et al.

TU-Berlin

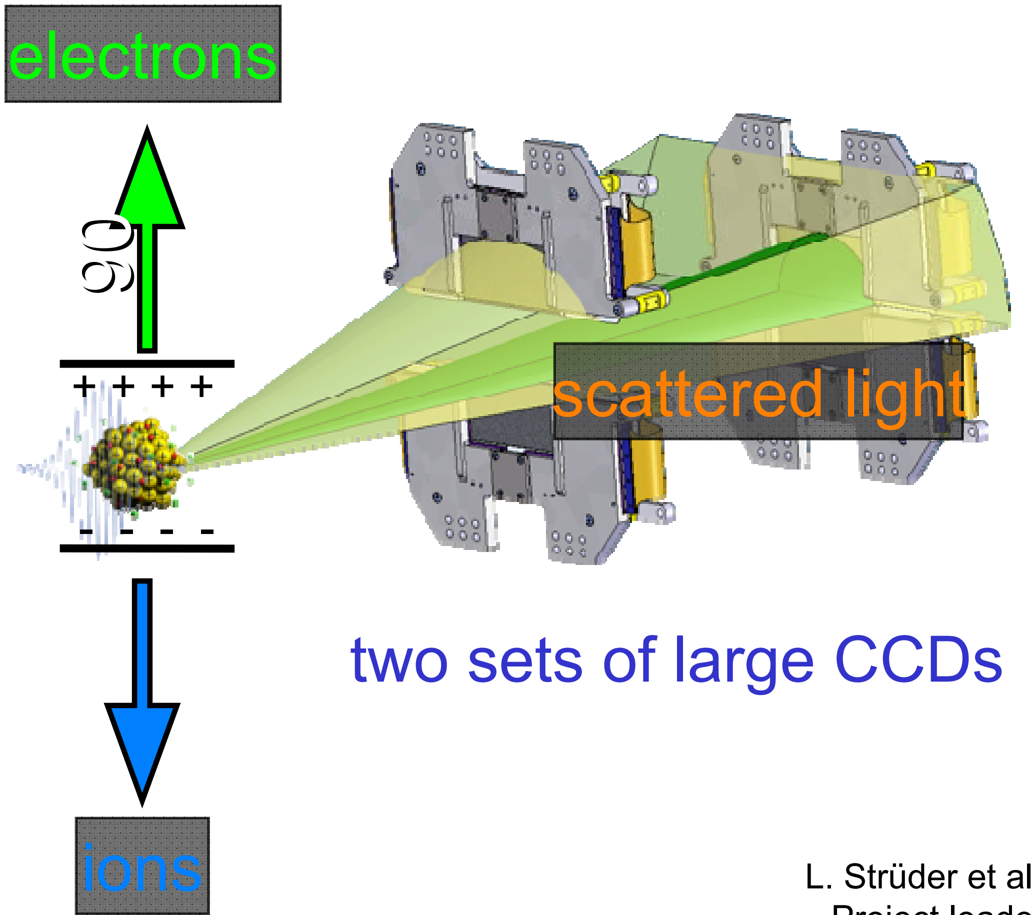
Marcus Adolph,
Daniela Rupp,
Sebastian Schorb,
Tais Gorkover,
Thomas Möller

CAMP Team

Sascha Epp,
Lutz Foucar,
Robert Hartmann,
Daniel Rolles,
Artem Rudenko, et al.,

Project leaders: I. Schlichting, L. Strüder, J. Ullrich

Single cluster coincident measurements



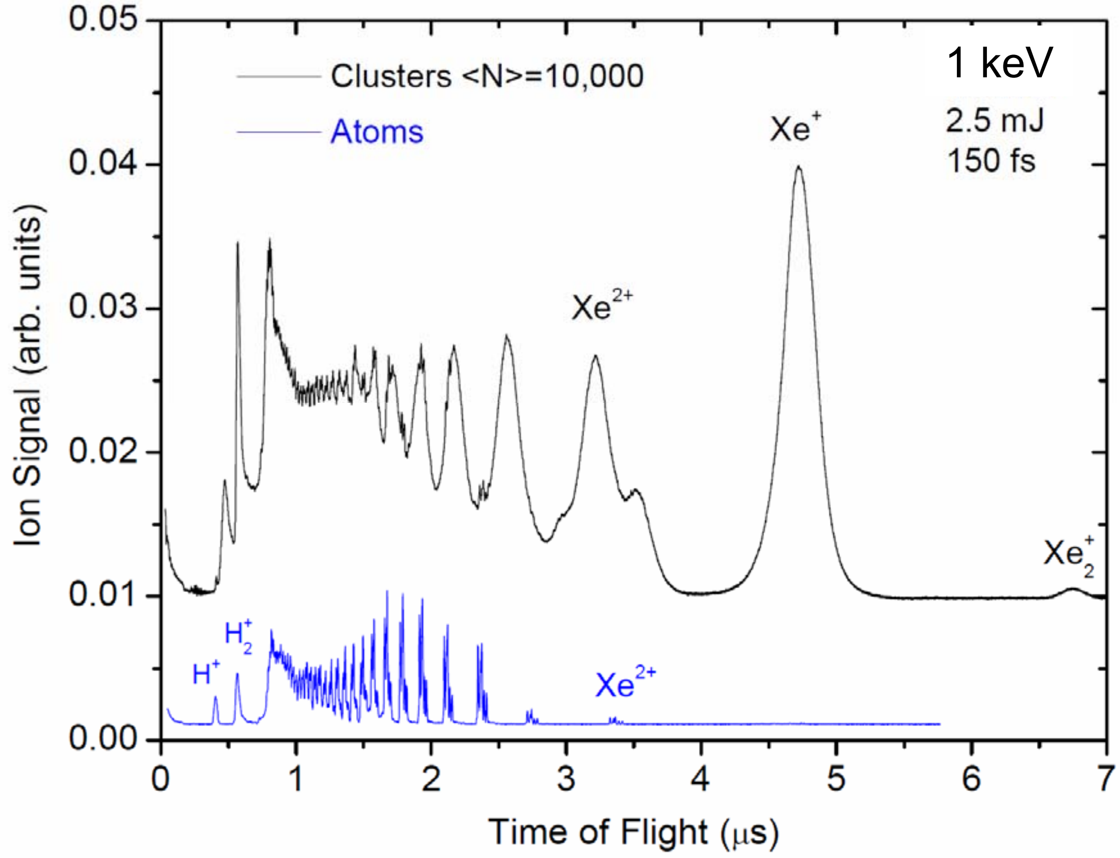
two sets of large CCDs

simultaneous detection
- ions
- electrons
- scattered light

CFEL-ASG-Multipurpose-Camber (CAMP)

L. Strüder et al. Nucl. Instr. Meth. A 610, 483 (2010)
Project leaders: J. Ullrich, L. Strüder, I. Schlichting

Rare gas clusters and atoms in the LCLS focus



AMO station

K. Hoffmann[1*], N. Kandadai[1], H. Thomas[1], A. Helal[1], J. Keto[1], T. Ditmire[1], B. Iwan[2], N. Timneanu[2], J. Andreasson[2], M. Seibert[2], D. van der Spoel[2], J. Hajdu[2], , S. Schorb[3], T. Gorkhover[3], D. Rupp[3], M. Adolph[3], T. Möller[3], G. Doumy[4], L.F. DiMauro[4], C. Bostedt[5], J. Bozek[5], M. Hoener[6], B. Murphy[6], N. Berrah[6]

sequential multi-photon ionization up to Xe^{25+}

Collaboration with T. Ditmire, J.Hajdu

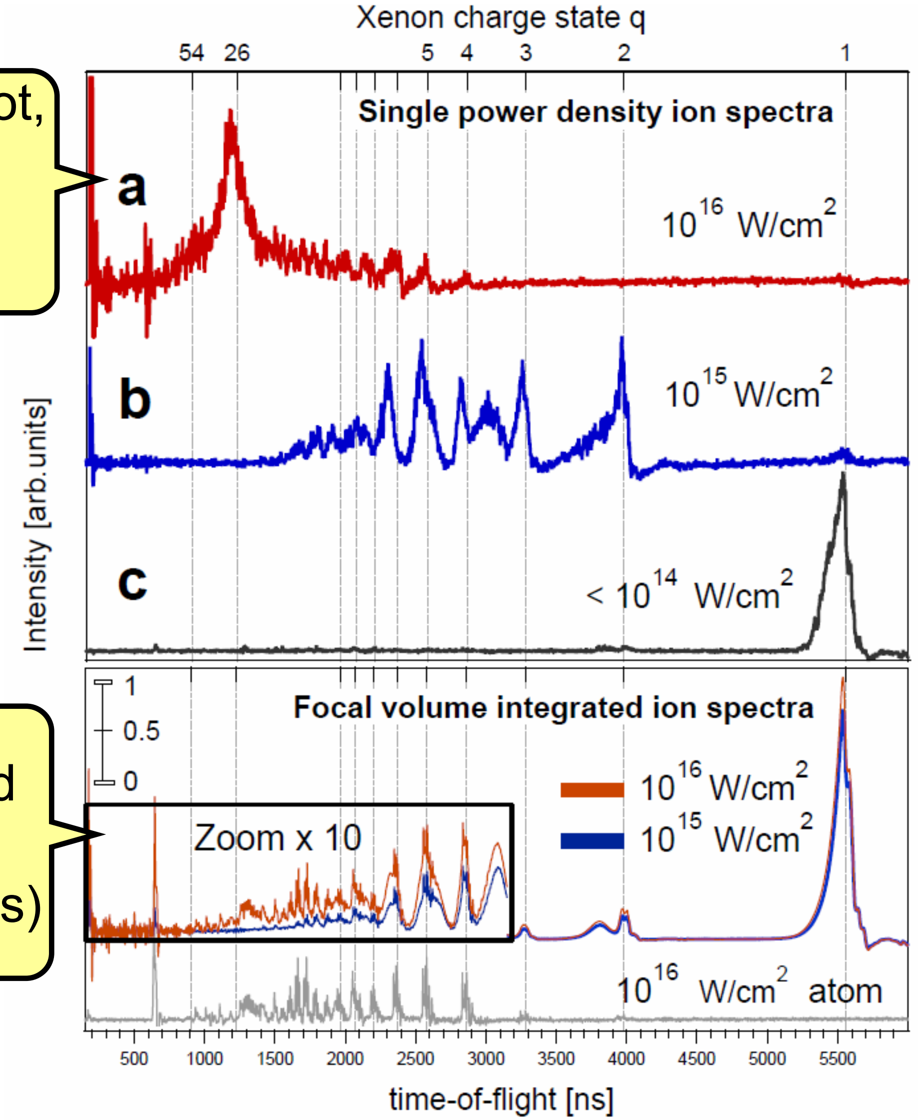
PRL 108, 133401 (2012)

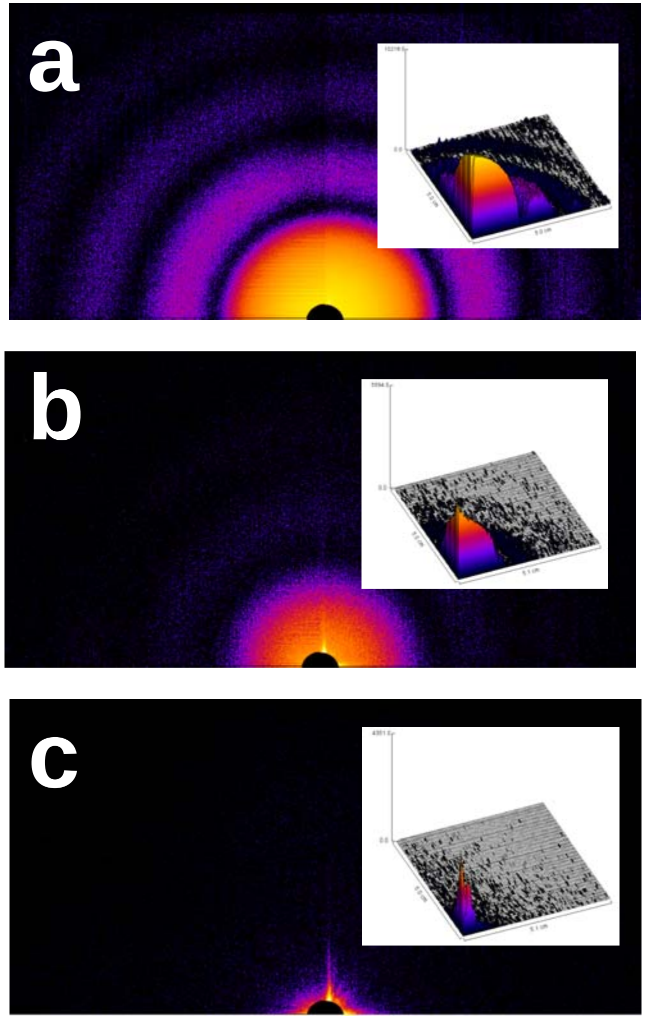
Single clusters coincident measurements

Xenon Clusters, 800.000 atoms, 800 eV, 3 mJ

Single shot, single cluster spectra

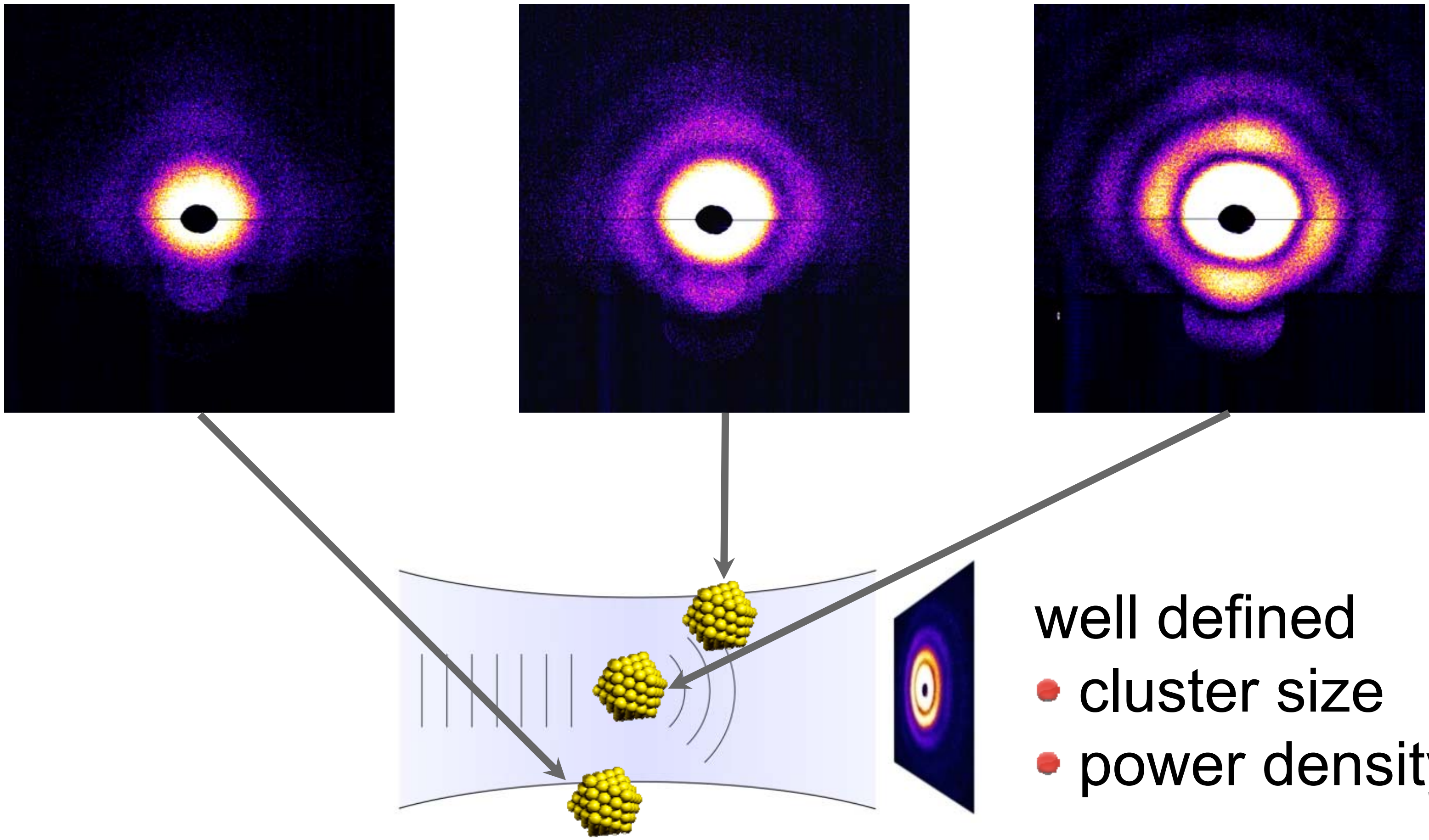
averaged spectra (300 Shots)





PI Christoph Bostedt

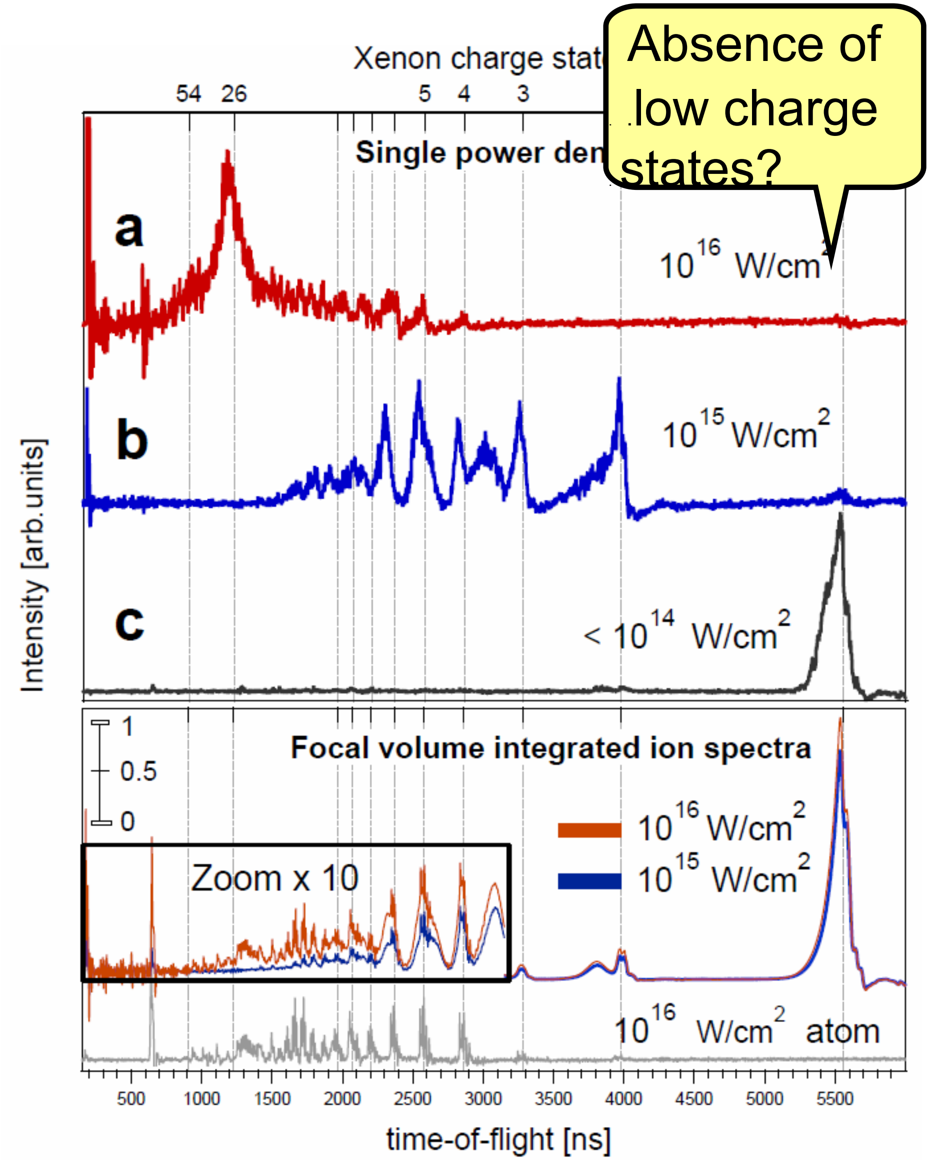
Position of single cluster in FEL beam

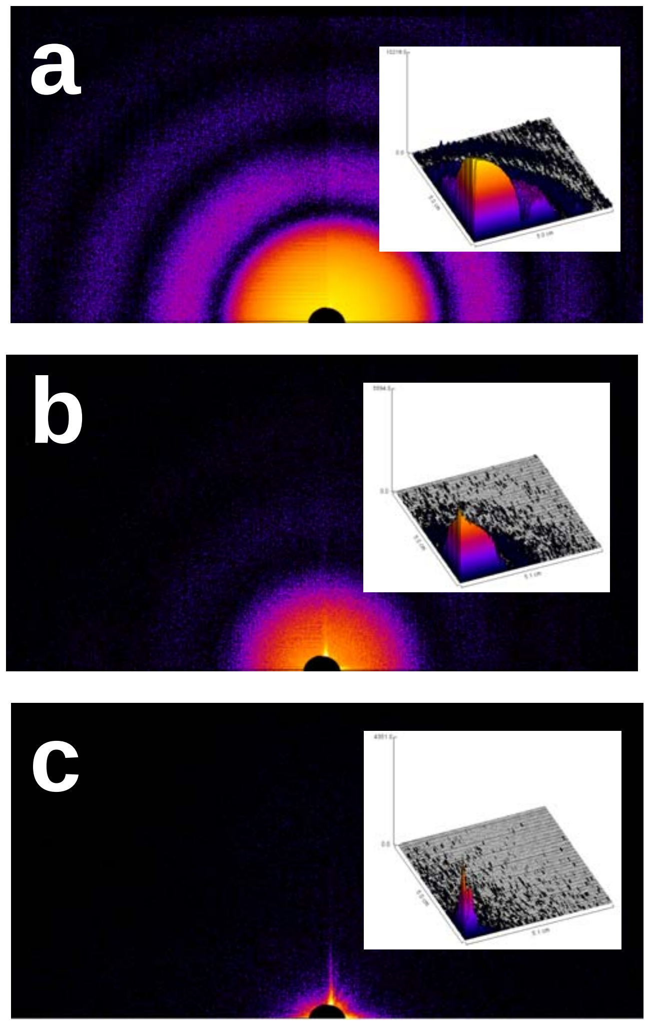


well defined
- cluster size
- power density

Single clusters coincident measurements

Xenon Clusters, 800.000 atoms, 800 eV, 3 mJ





Electron removal from a highly charged cluster

Total binding energy of (N*z) cluster electrons (removal of z Electrons per atoms), R cluster radius, N number of atoms,

$E_{tot} = e^2 / 4\pi\varepsilon_0 * (Nz)^2 / 2R$, $\sim N^2 z^2$

Example $N=10^6$, $z=25$, $E_{tot} = 10^{13}$ eV

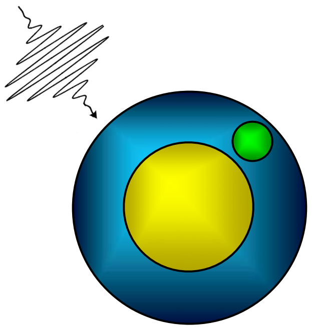
X-rays, 1000 eV photon, Absorption of 10^{10} photons,

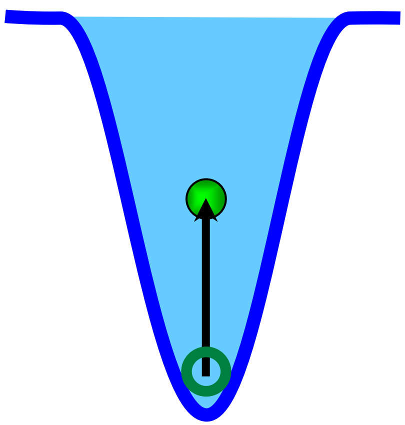
10^4 Photons/atom are needed ?

Only a small fraction (few %) of electrons can be removed of an intact cluster

Absence of low charge states

Electron trapping





Xe cluster 20 nm, 10^6 atoms
0.8 keV

only 3 % of electrons can be directly emitted (electrostatic)

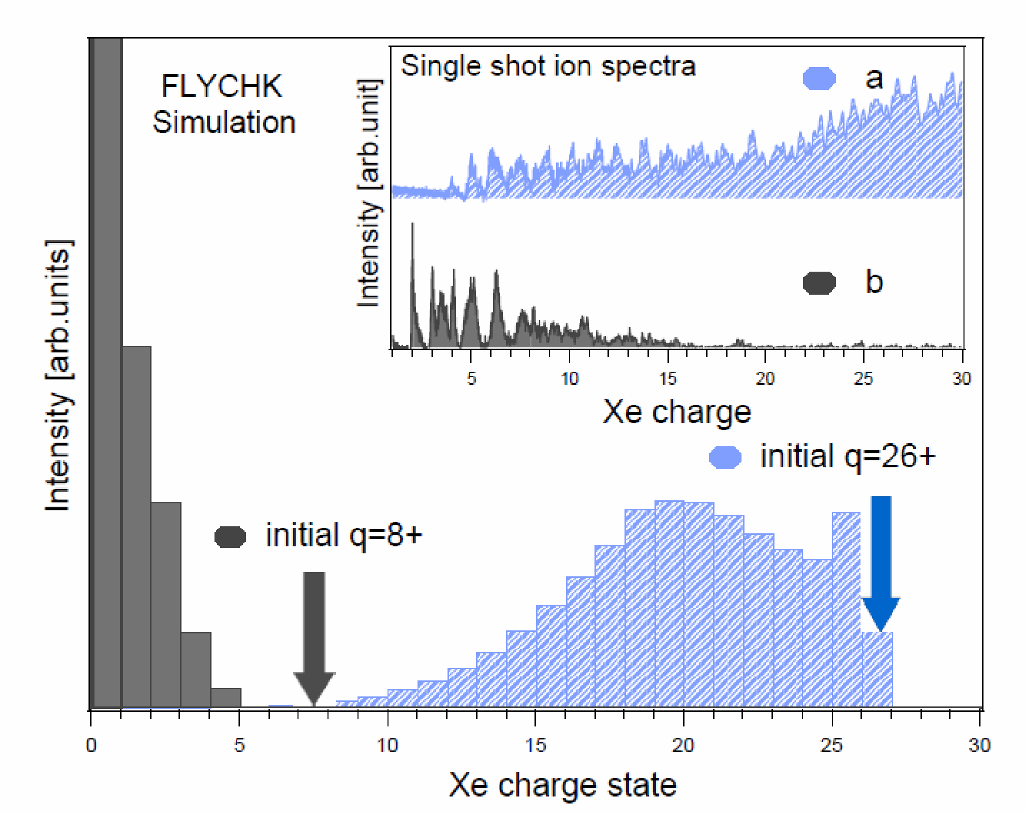
- only highly charged ions from the surface layer explode off, core full recombination?

- **no recombination? no neutral atoms?**

recombination rate ~ $T^{-4,5}$ very hot plasma small recombination

VUV: almost complete recombination in large clusters

Simulation with plasma code FLYCHK:



weak recombination at the highest power density (only very highly excited states)

up to 6 ps

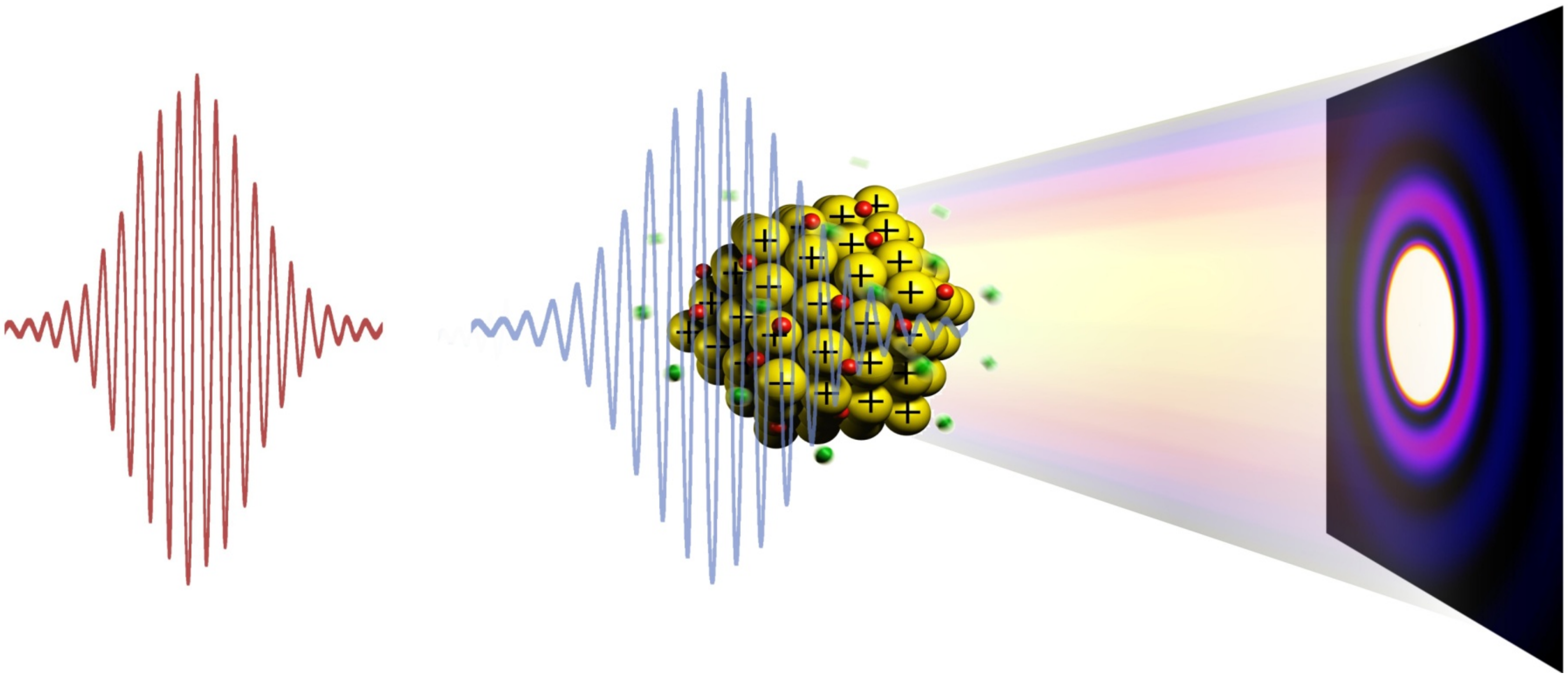
PRL, submitted

- **cluster explosion from FEL is treated as expansion from a highly excited, ultra dense plasma state** T. Ditmire, J. Zweiback, V.P. Yanovsky et al., Nature **398**, 489 (1999).

- **simplified hydrodynamic expansion fed into FLYCK, a classic plasma code** H.-K. Chung, M.H. Chen, W.L. Morgan , Y. Ralchenko, R.W. Lee, High Energy Density Physics 1 3-12 (2005)

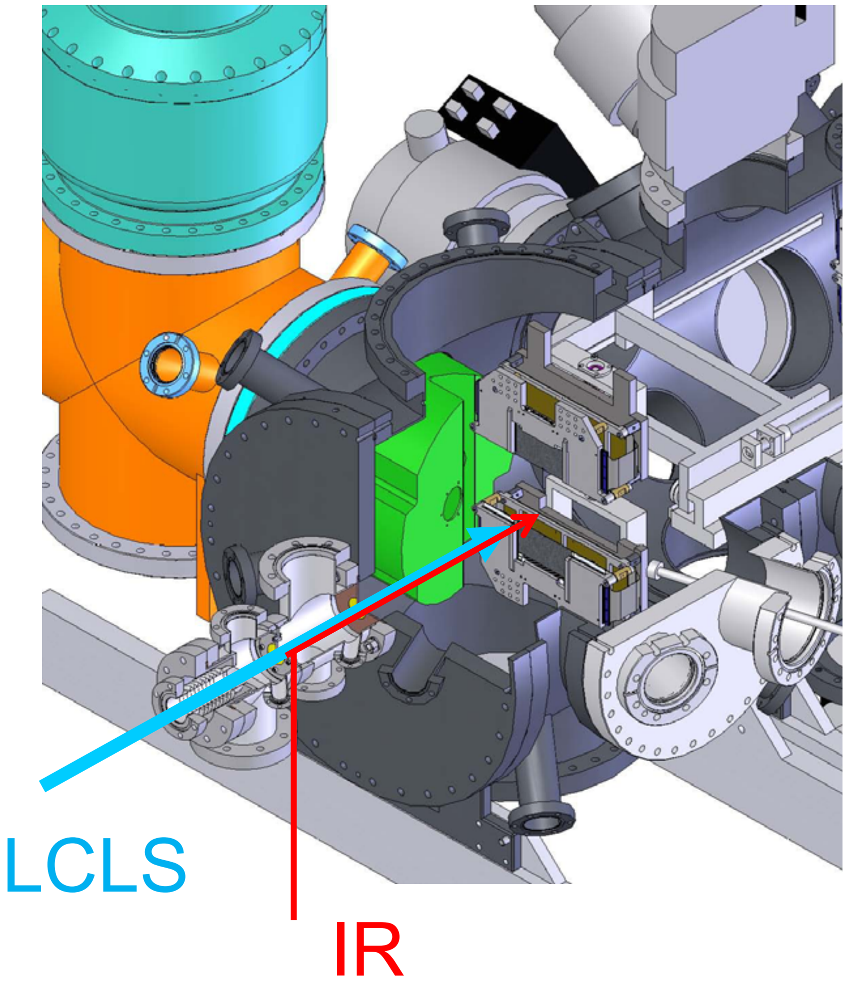
Time resolved imaging of exploding clusters

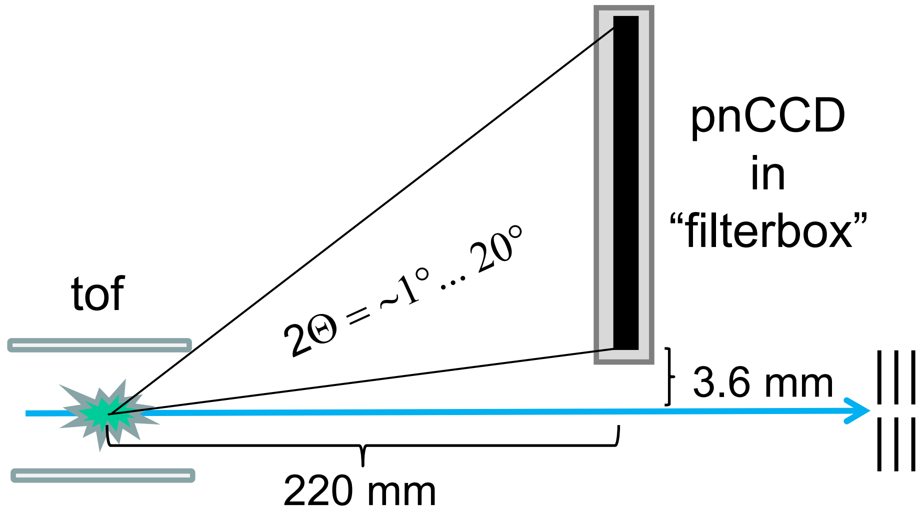
- Study how ultrafast ionization dynamics influence scattering process
- Scattering sensitive to both, changes in electronic and geometric structure



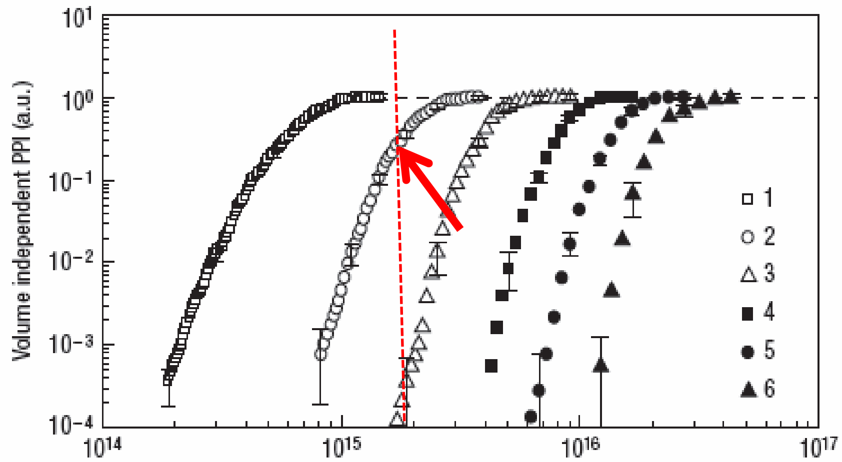
IR pump + FEL probe pulse (LCLS), CAMP

Experimental layout





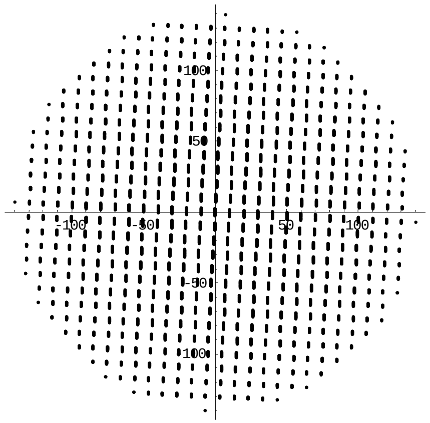
IR laser: 50 fsec, 2 mJ, $2 \cdot 10^{15}$ W/cm^2

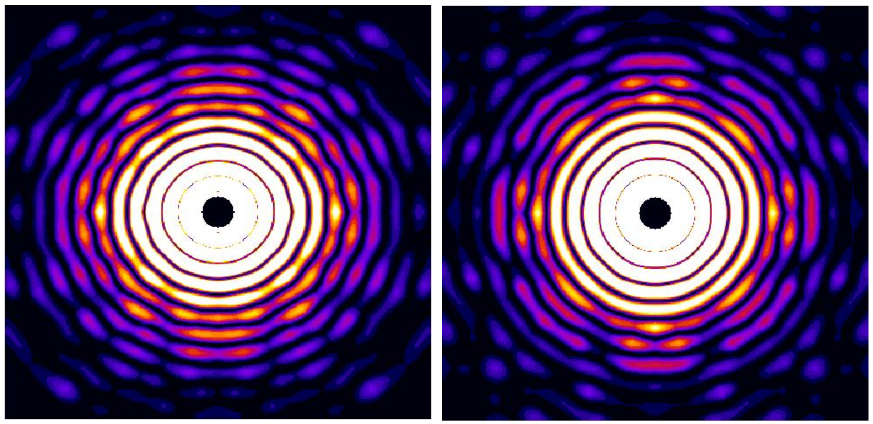


Bryan et al., Nature Physics 2, 379 (2006)

Imaging: What can be expected with nm x-ray radiation?

Xenon cluster with ellipsoidal shape
long half axis 15 nm, short 14 nm
lattice constant 0.5 nm
Wavelength 0.7 nm



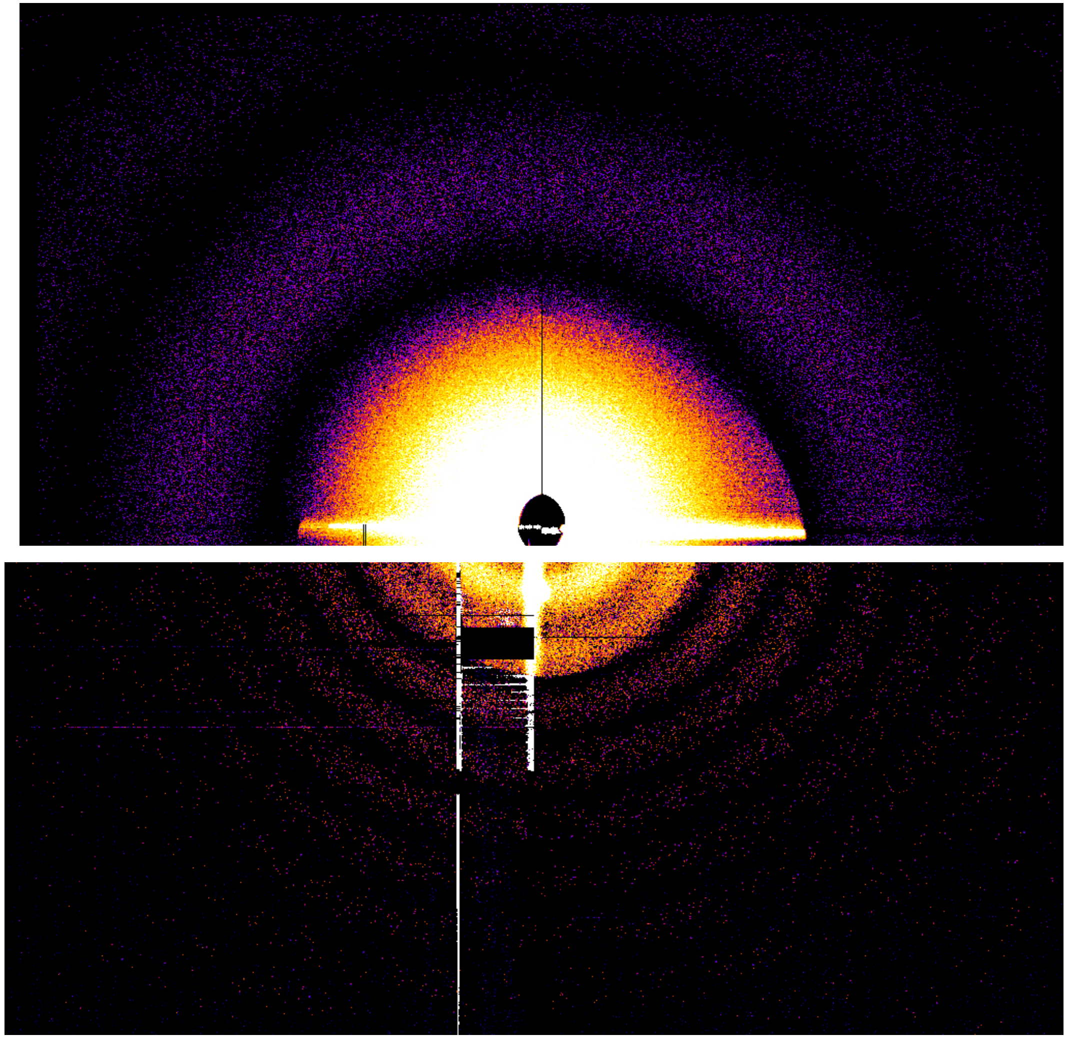


long axis perpendicular to light beam long axis parrallel to light beam

Facetts become visible

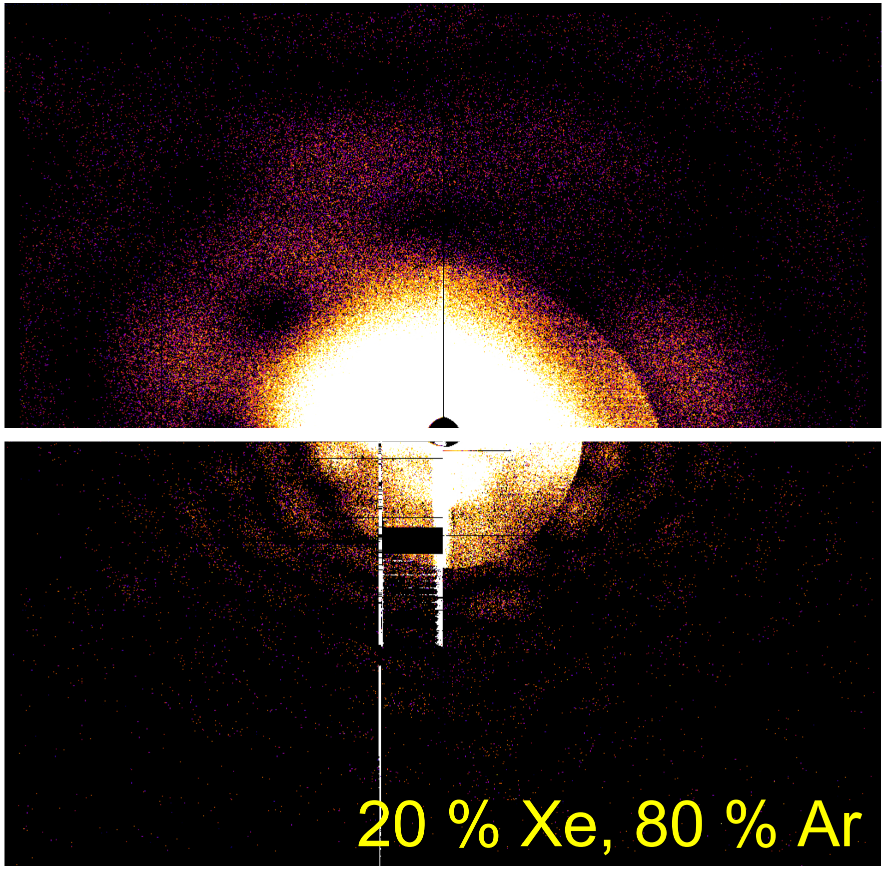
Cluster symmetry

Ar cluster, radius 23 nm, 1 nm wavelength, 3 mJ



Imaging of clusters with X-rays: Ar-Xe core shell cluster

1,5 keV
2 mJ



Complicated scattering patterns

~20 nm

- twin clusters
- internal structure

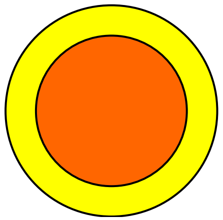
Spatial resolution:

$\Delta x = \lambda \; N_a$

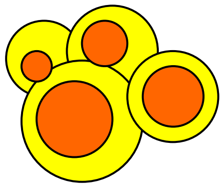
N_a numerical aperture

trade off between wavelength and cross sections

Core shell cluster

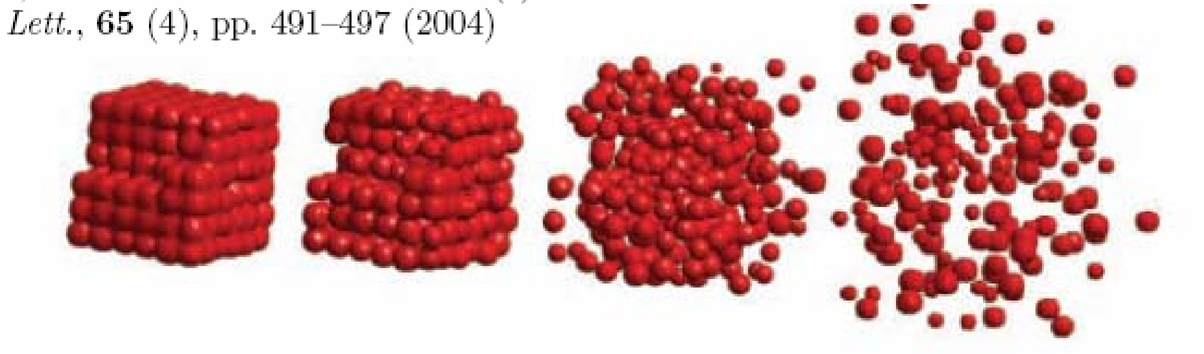


mixed core shell clusters



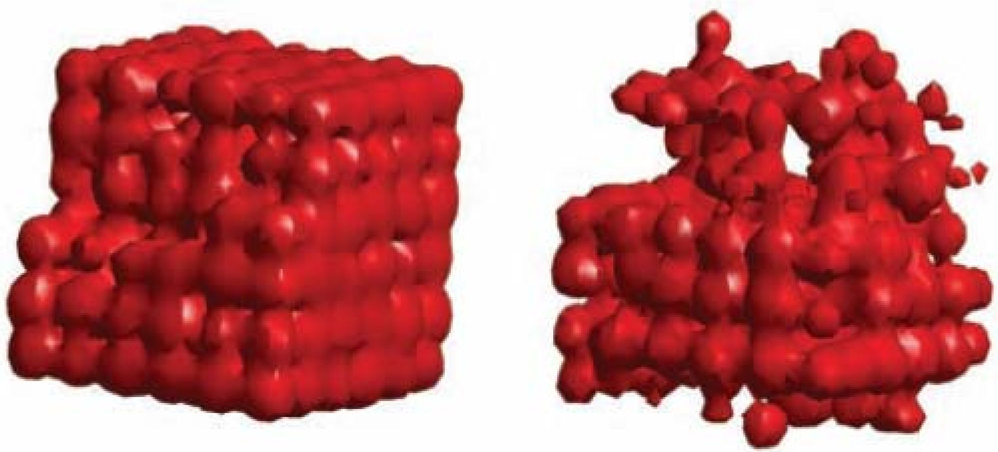
Vision: Cluster structure determination with atomic resolution?

Z. JUREK, G. OSZLÁNYI and G. FAIGEL(*)
Europhys. Lett., **65** (4), pp. 491–497 (2004)



10 fs   12 fs   15 fs   18 fs

200 atom cluster in an 20 fs X-ray pulse (1 Å)



12 fs   18 fs

Reconstructed images

- minimum cluster size ?
- heavy elements ?

Summary

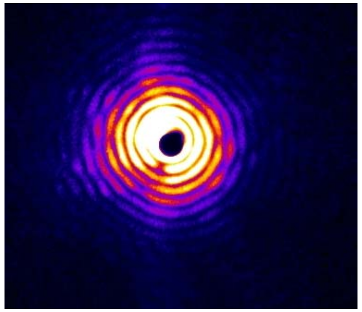
Ionisation of clusters:

- absorption of many (single) photons

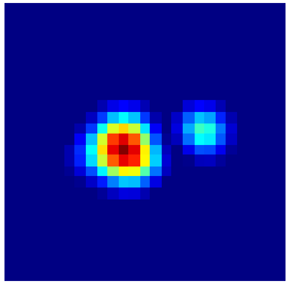
  sequential electron emission, exchange of energy

- single shot imaging of single particles

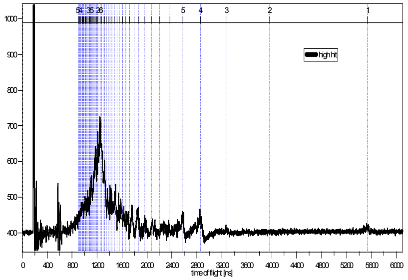
  cluster structure and shape



better resolution



- scattering can probe transient states



- new perspectives at X-ray lasers, coincidence

  inefficient ion recombination at highest power density

time resolved studies: electron dynamics

lot´s of exciting physics ahead of us!

Acknowledgement

Funding by BMBF and DFG

TU Berlin + MPHII @ FLASH

K.H. Meiwes-Broer,
J. Tiggesbäumker, T. Laarmann,
T. Ditmire, J. Hajdu

DESY/ FLASH Team
LCLS Team

Cooperation with theory
T. Fennel (Rostock)
U. Saalman, J. Rost (Dresden)
B. Ziaja, R. Santra (CFEL/DESY)

LCLS
C. Bostedt
S. Schorb,

TU Berlin:
M. Adolph,
D. Rupp,
L. Flückiger
T. Gorkhover
M. Müller
T. Oelze
E. Ovcharenko
M. Sauppe
M.Krikunova

CAMP Team

Sascha Epp,

Lutz Foucar,

Robert Hartmann,

Daniel Rolles,

Artem Rudenko, et al.,

Project leaders:

I. Schlichting, L. Strüder, J. Ullrich

CAMP @ LCLS

And thank you for your attention!