Structure and dynamics of nanoparticles in intense short wavelength light pulses

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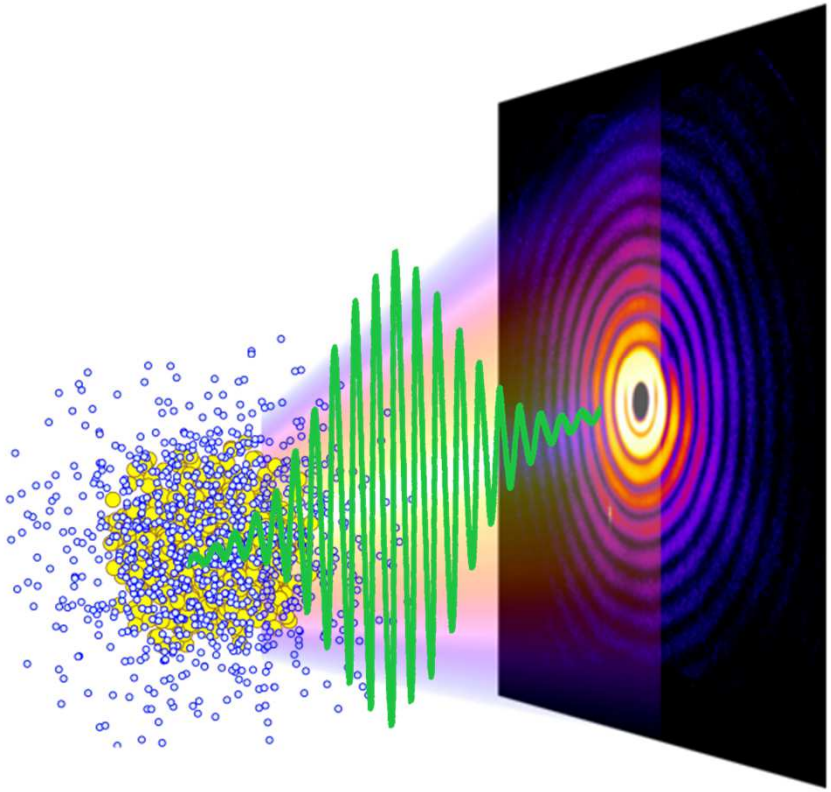
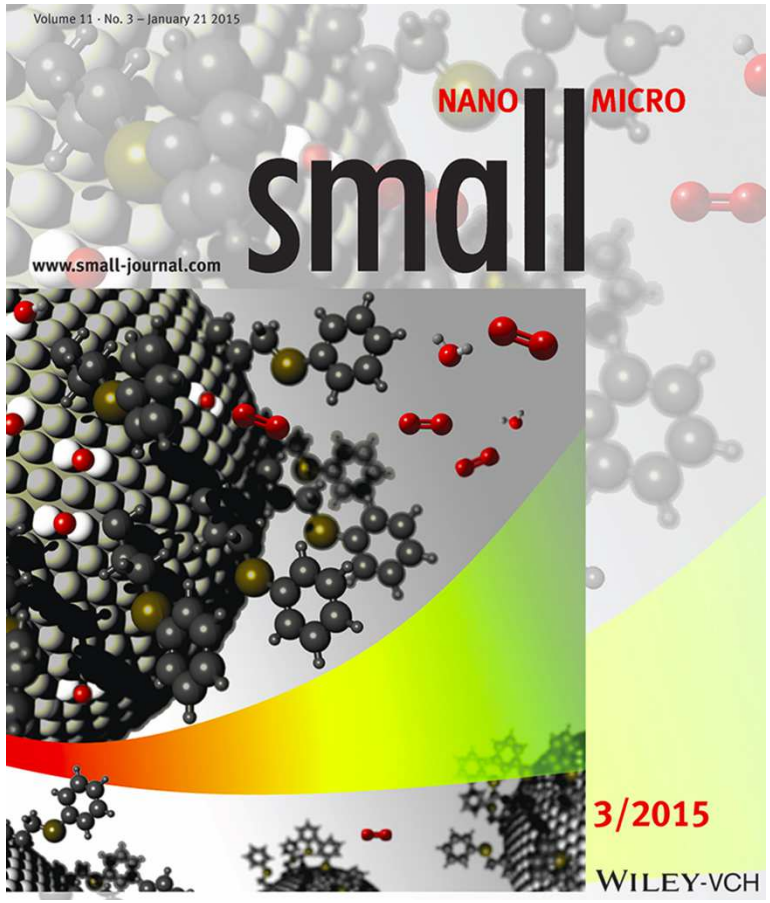
Nobel Symposium on Free Electron Laser Research
Sigtuna, June 14-18, 2015

Interaction of intense short wavelenght pulses with clusters





Nanoparticles and clusters: Issues and Questions

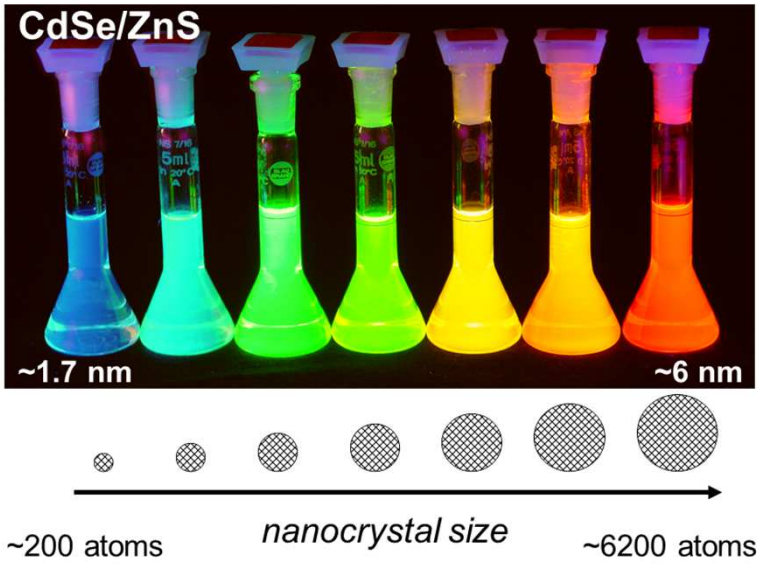
Clusters and nanocrystals are new materials

Size dependent properties

- catalytic activity
- magnetic properties
- photochemical processes
- light induced dynamics
- geometry and shape

Size dependent colour

Novel pigments in tv-screens



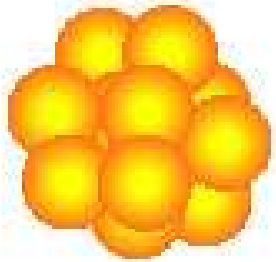
Courtsey: H. Weller, Universität Hamburg

Atoms / clusters in intense x-ray pulses

What are the differences?

Atom

Cluster



absorption into continuum states

→ ionization

→ inner ionization: electron removal from a cluster atom

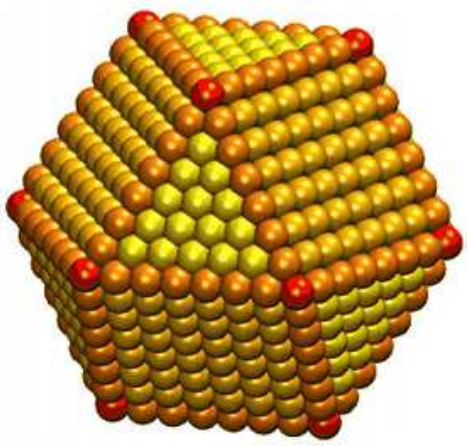
→ outer ionization: electron removal from the cluster

Last, Jortner, Phys. Rev. A, 62, 013201(2000)

→ Plasma formation

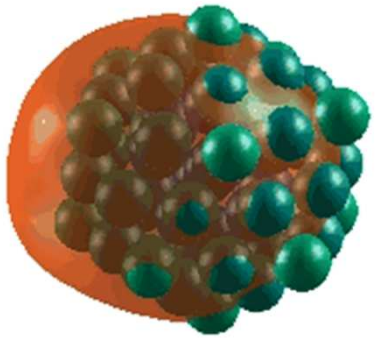
Properties of clusters / Driving questions

- Shape and structure of individual particles
  - Regular shape, non-equilibrium structures ?



W.Zhu et al,
JACS **2013** *135* (45), 16833

- Light induced dynamics
  - ion motion, electron motion
  - collective motion, plasma dynamics?
  - Phase transitions, melting, surface melting

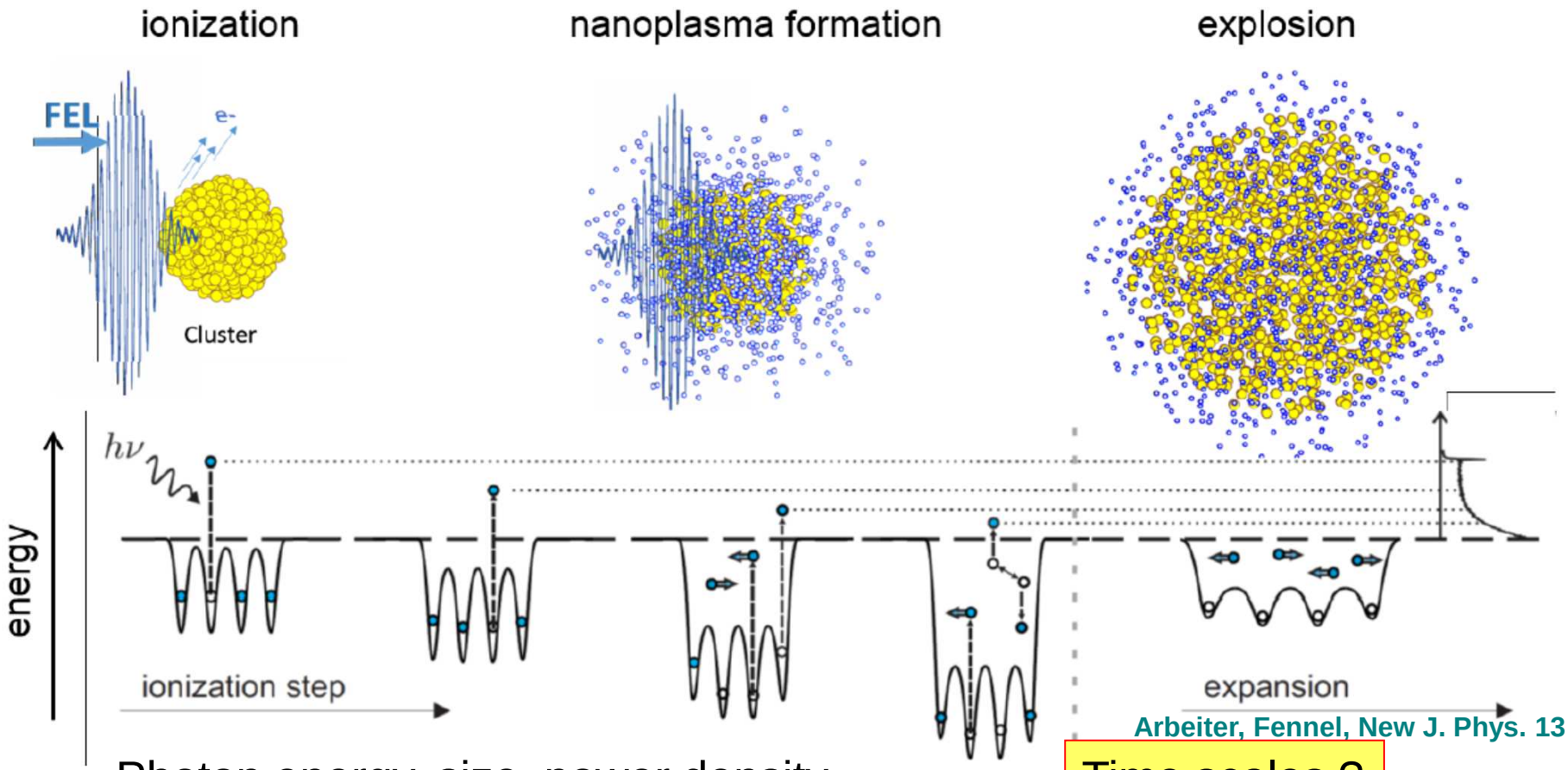


Curtesy of T. Fennel

Cluster: Nanolab for laser-matter-interaction

Rare gas cluster : simple structure, detailed studies with IR light

„Three step model“



Arbeiter, Fennel, New J. Phys. 13

Photon energy, size, power density

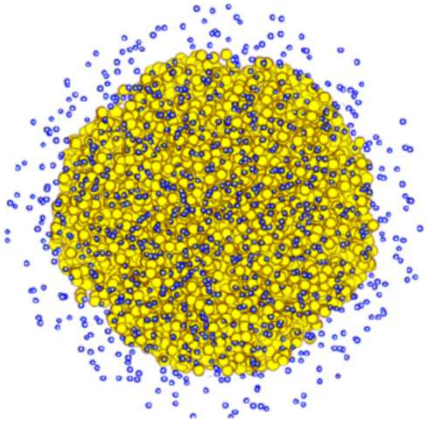
Time scales ?

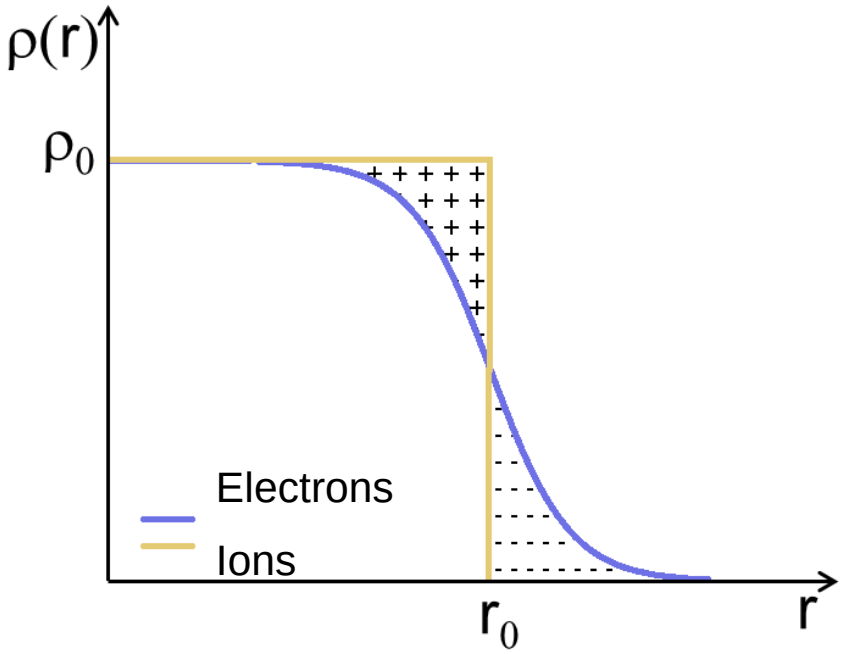
Experiments Wabnitz Nature 420, 482 (2002) , Laarmann , PRL 92, 143401, PRL 95, 063402 (2005), Bostedt PRL 100, 133401

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

Nanoplasma formation

What is a nanoplasma?





Questions:

- internal structure
- particle surface / expansion
- electron-ion recombination
- electron and ion dynamics, consequences for imaging?

Outline

- How we got started: initial experiments at TTF-FEL
- Nanoplasma formation
  - Ar clusters, autoionization of He clusters
- Imaging with soft X-rays
  - Single clusters, spatial evolution of plasma, shape of metal clusters, in flight holography
- Time resolved studies
  - IR –X-ray Pump-probe Xe clusters
- New opportunities

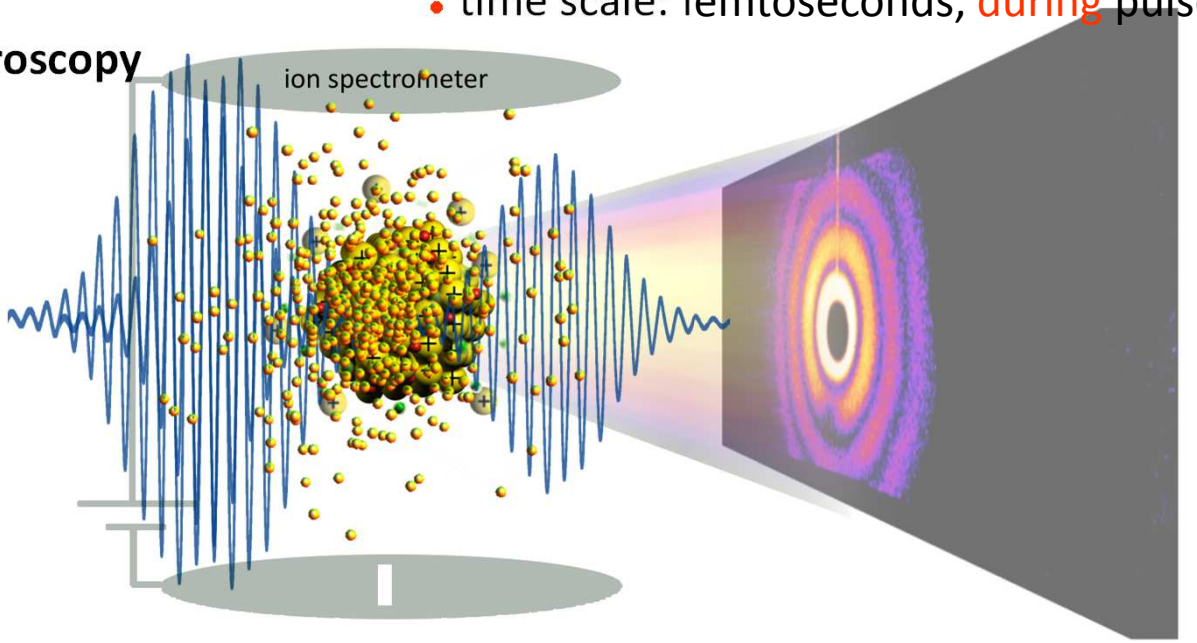
Method:
Simultaneous imaging and spectroscopy

Soft x-ray scattering

- size and shape of the clusters
- electronic configuration during interaction
  → change of refractive index
- time scale: femtoseconds, during pulse

Ion and electron spectroscopy

- ionisation and recombination
- kinetic energies, expansion process
- time scale: fs up to hundreds of ps after the pulse

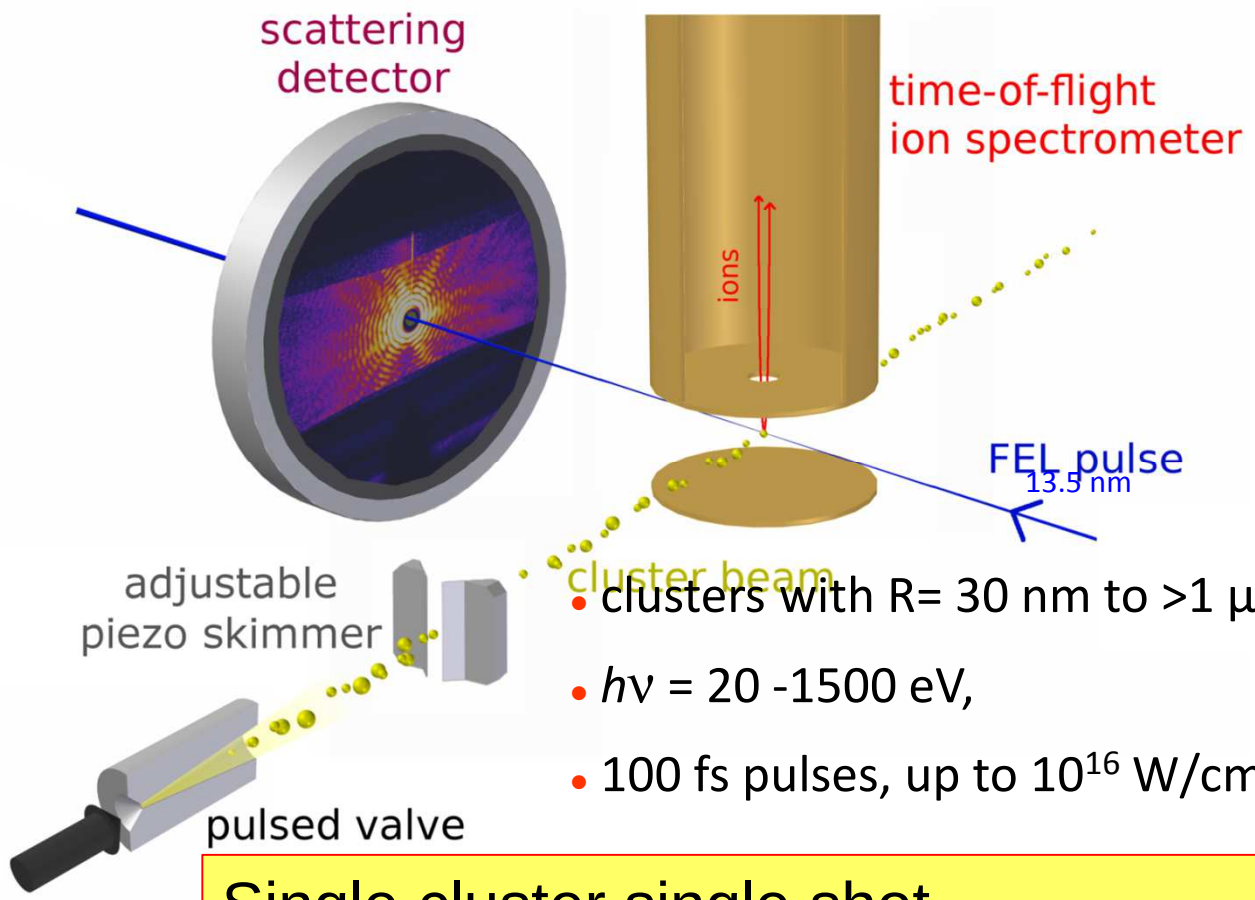


Pump-probe techniques

- excite cluster with a pump pulse (NIR/XUV)
- probe explosion with a delayed XUV pulse
- time scale: resolve full range from sub-ps to ns

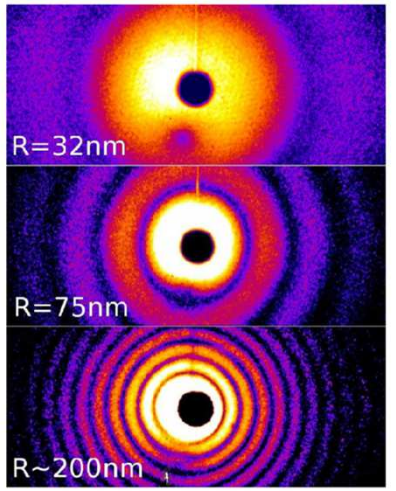
Complementary methods looking into different timescales!

Setup: Simultaneous imaging and ion spectroscopy: Single cluster intensity distribution: no averaging

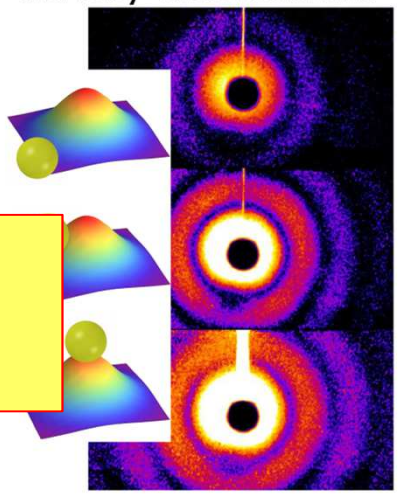


- clusters with R= 30 nm to >1 µm
- $h\nu$ = 20 -1500 eV,
- 100 fs pulses, up to 10^{16} W/cm²

resolve size distributions



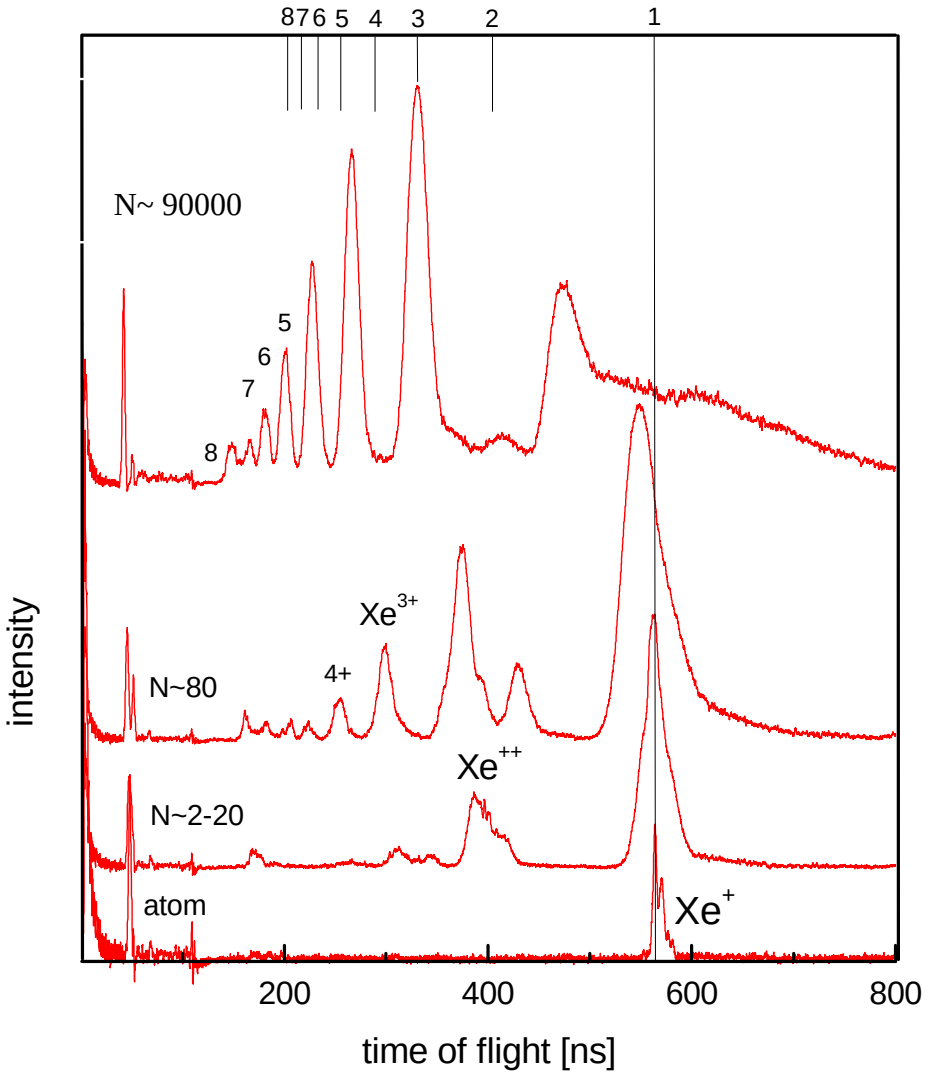
resolve power density distributions



Single cluster single shot
well defined size and power density !

T. Gorkhover et al., Phys. Rev. Lett. 108, 245005 (2012)
C. Bostedt et al., J. Phys. B 43, 194011 (2010)

First results from the TTF-FEL at DESY (98 nm): Ion spectra of Xenon atoms and clusters



$1*10^{13}$ W/cm²

E_{phot}= 12.8 eV
Ip_{Xe} = 12.1 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)
ionisation contiuum lowering

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

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

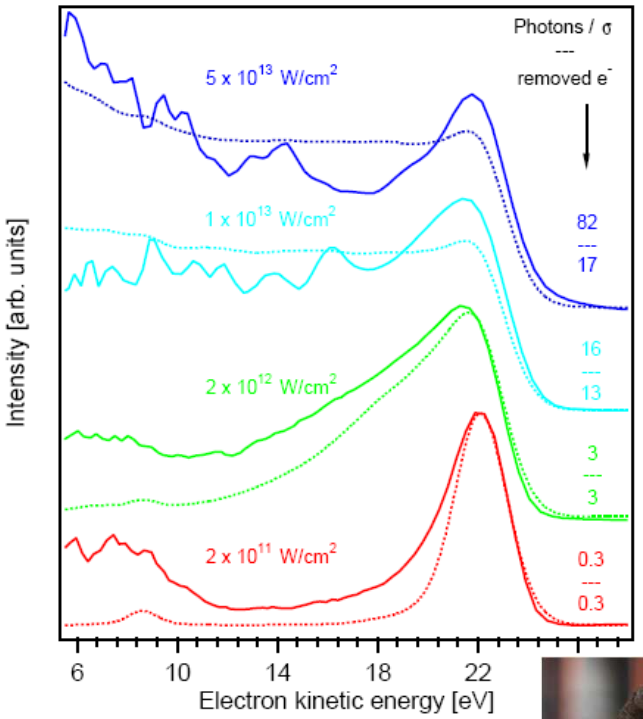
B. Ziaja et. al PRL 102, 205002 (2009).

Cluster ionisation and nanoplasma formation: Electron spectra of Ar clusters:

- experiment

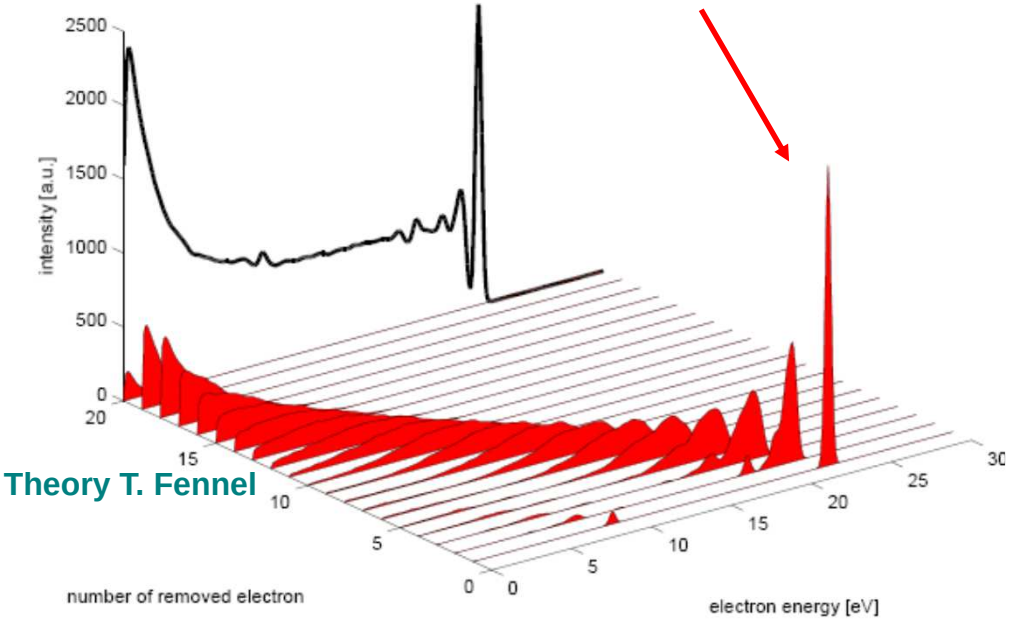
····theory

Ar_{150} clusters, 38 eV,
a few 10^{11} ~ 10^{14} W/cm²



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

First electron



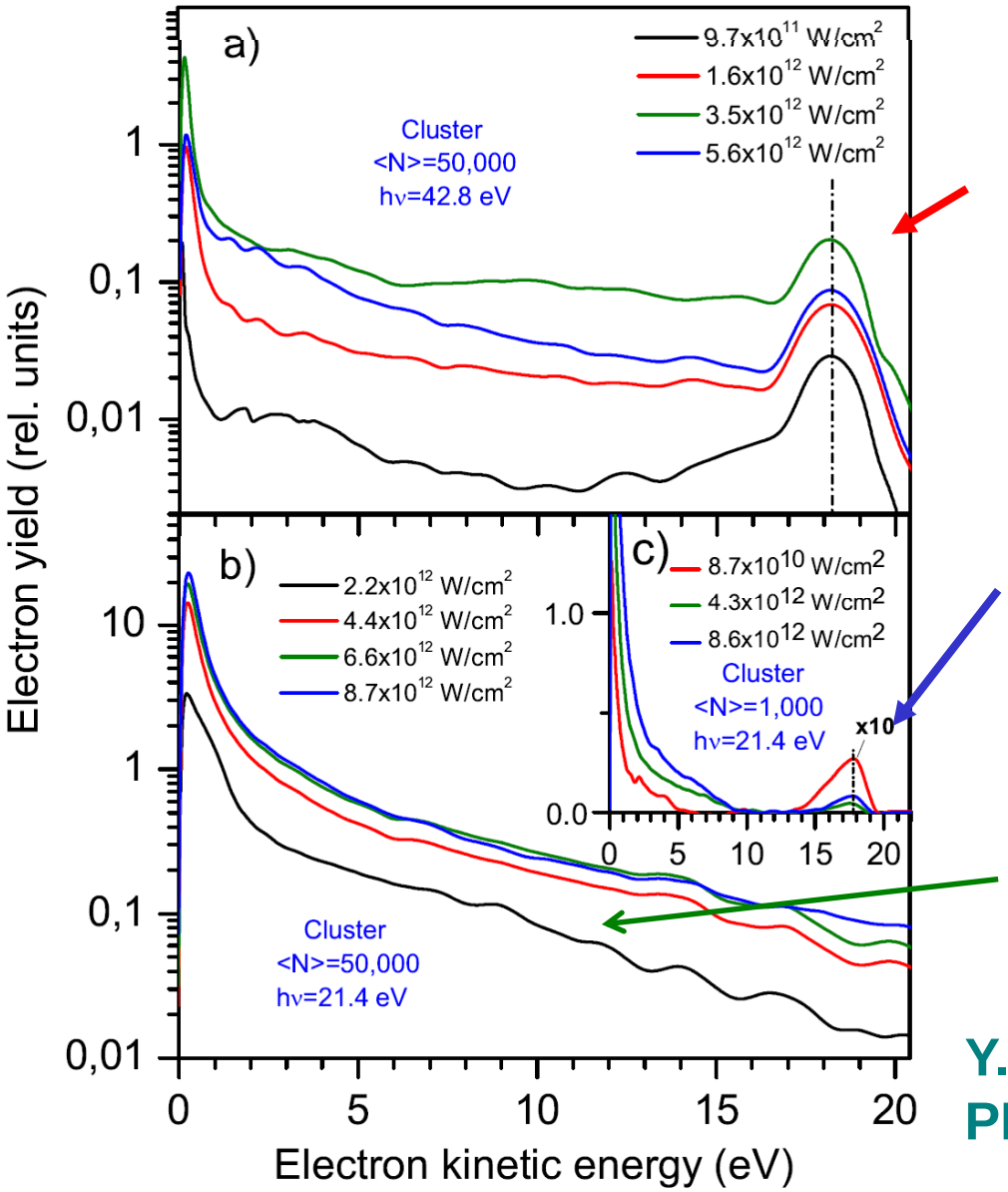
Theory T. Fennel

sequential emission of electrons

only a small percentage of generated photoelectrons can leave the cluster

→ nanoplasma

Ionisation below and above Ip: Electron spectra of He clusters



FERMI

Direct photo emission
42.8 eV (2x 21.4 eV)

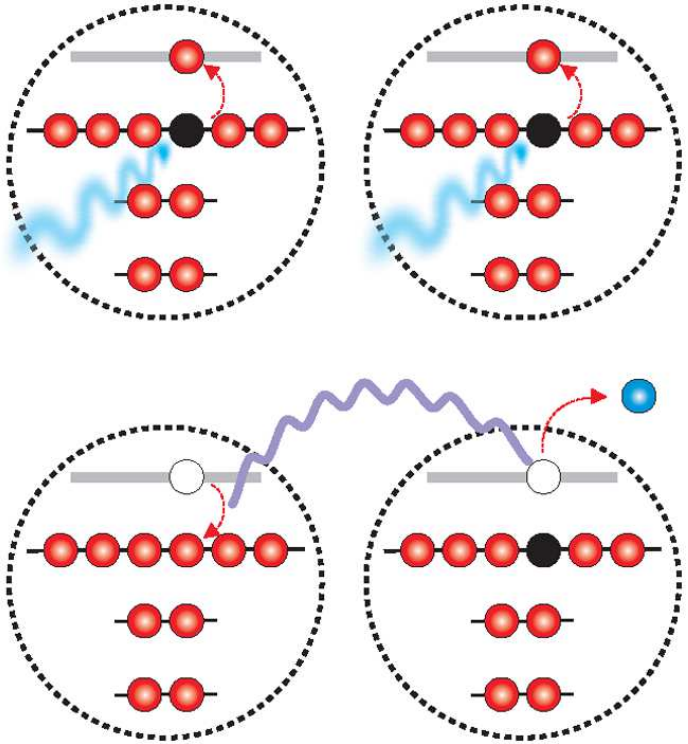
ICD, 1s → 2p, 21.4 eV

Collective autoionisation

Y. Ovcharenko et al.
PRL 112, 073401 (2014)

ICD type Autoionisation

Proposed for Ne clusters

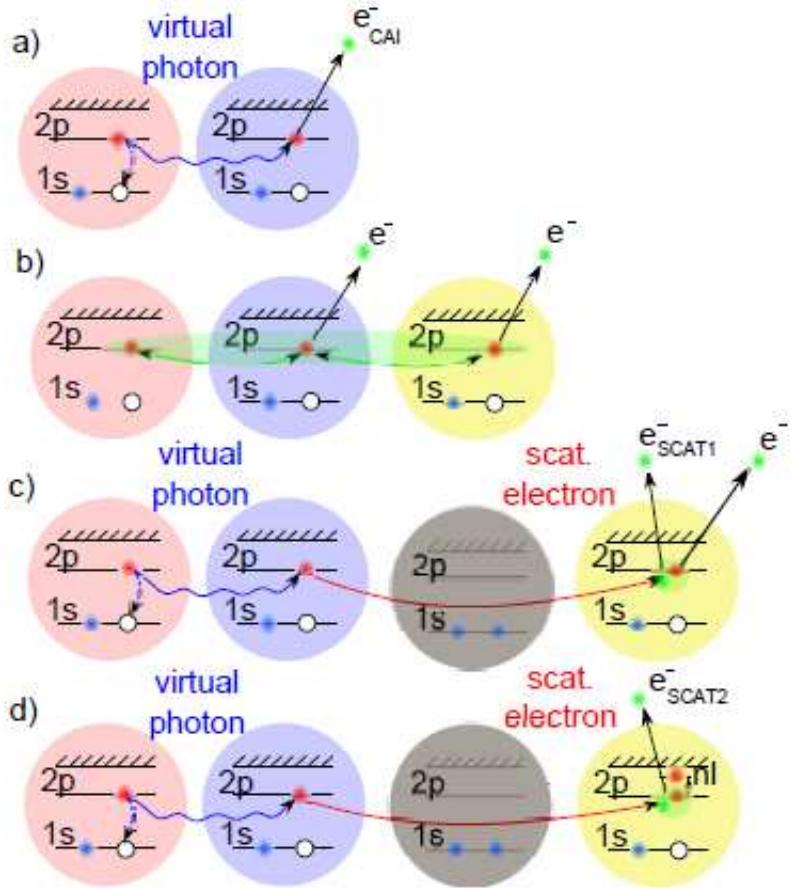


A. Kuleff et. al.

Phys. Rev. Lett. 105, 043004 (2010)

- With intense light sources, multiple atoms in the cluster can be excited, 2p> 3s
- ICD between neighboring atoms leads to ionization of one of the atoms
- Ionization rate through ICD sequential one photon absorption (linear process) >> 2 photon ionization (nonlinear)

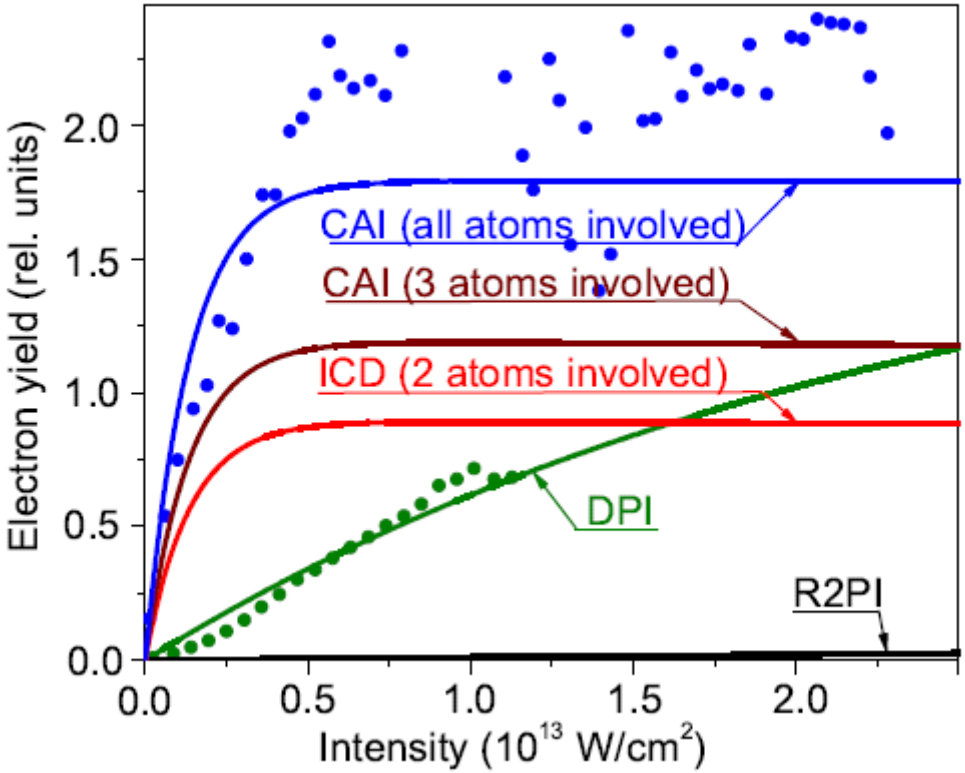
Different autoionisation processes



Inelastic collision between electrons and excited atoms

- A new type of nanoplasma is formed
- Many excited atoms are involved at the same time

Collective autoionization extremely efficient



- much more efficient than **direct** photoemission
  at least two photon process

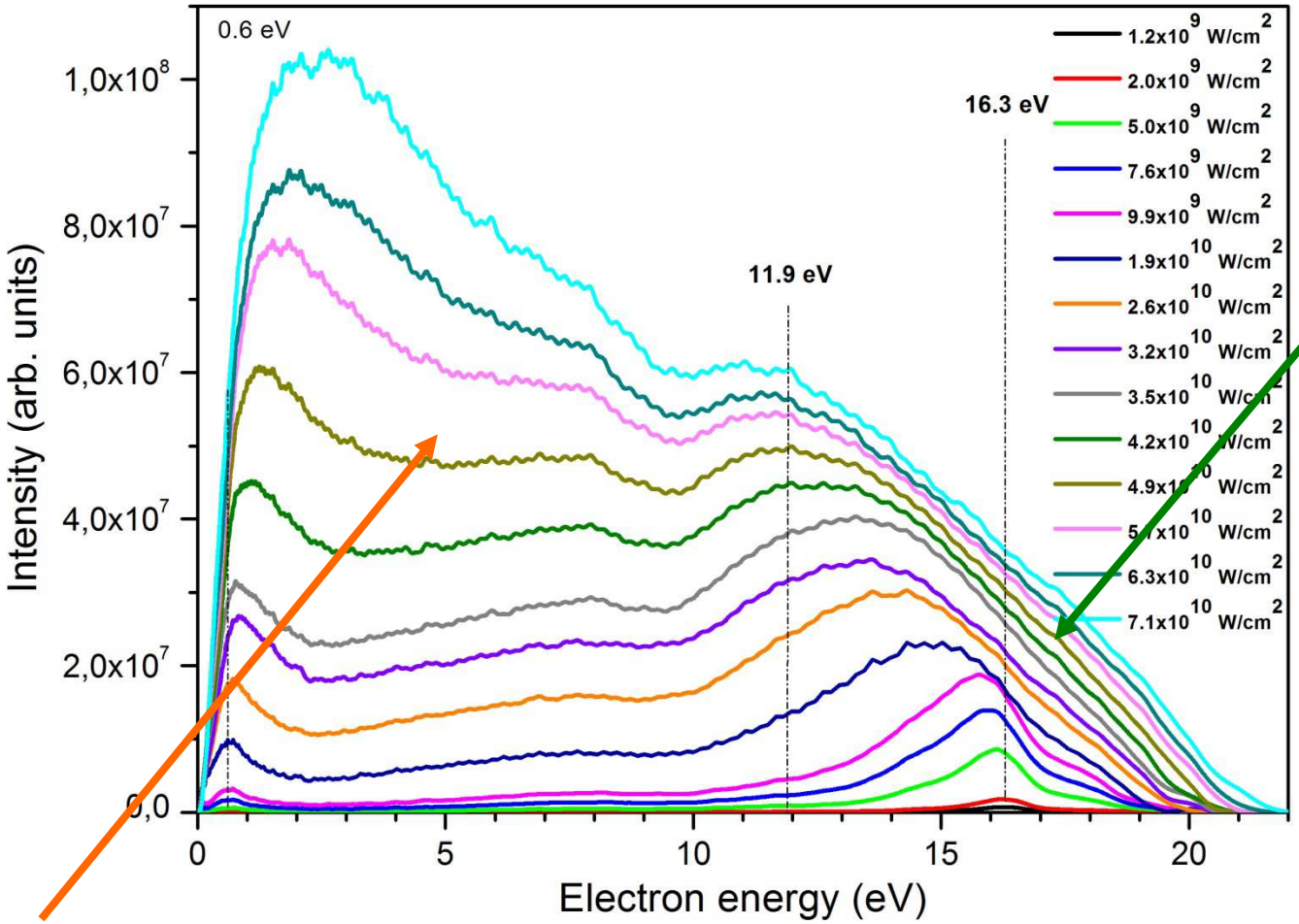
- electron yield linear at ‘low‘ power density
- saturation at high power density

A. LaForge et al.
Scientific Reports 4, 3621 (2014)

Y. Ovcharenko et al.
PRL 112, 073401 (2014)

Transition from ICD type to collective autoionization

He cluster N= 50000



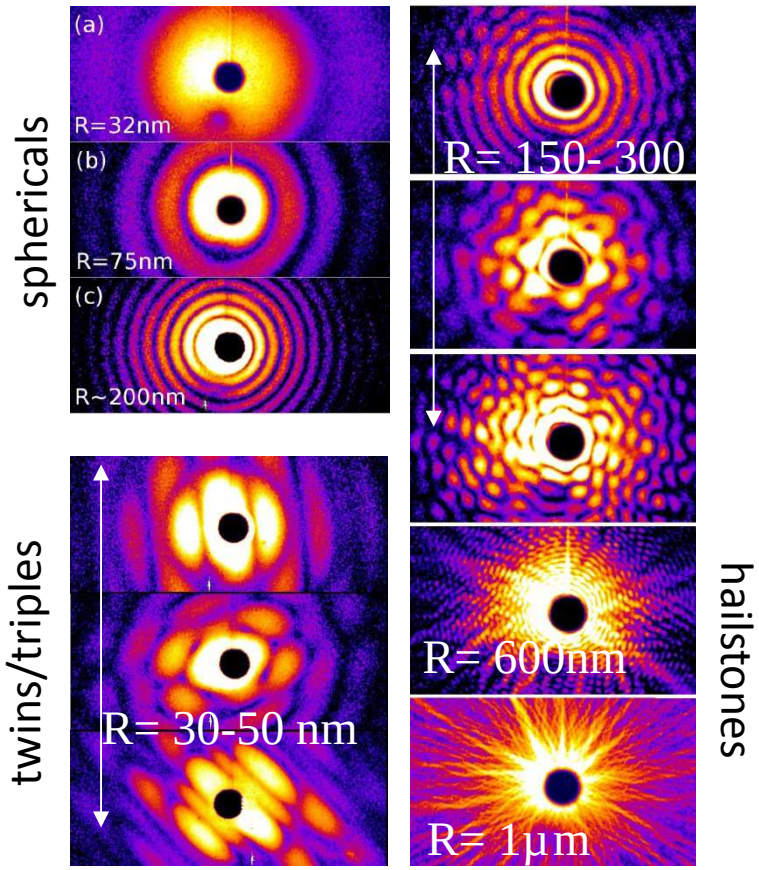
ICD,
two atoms
Next to each
other, isolated

Time scale
sub fs?

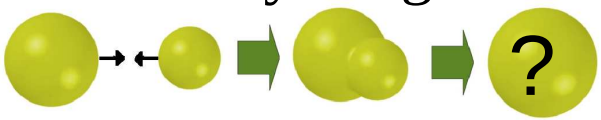
He 1s > 2p, collective autoionisation,
network, plasma

Y. Ovcharenko, M. Mudrich, A. LaForge, et al. in preparation

Morphology of large xenon clusters



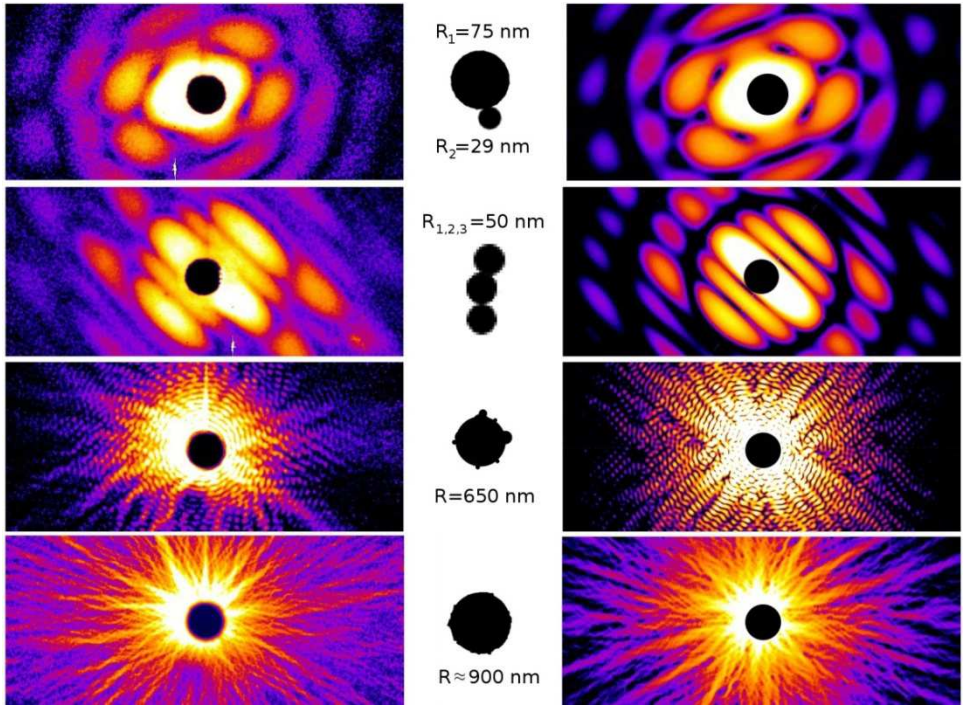
Growth by coagulation



D.Rupp et. al, J. Pys.B 43,194011(2010).
JCP 141, 044306 (2014)

Experimental pattern
2D-Fourier transformation

2D-projection



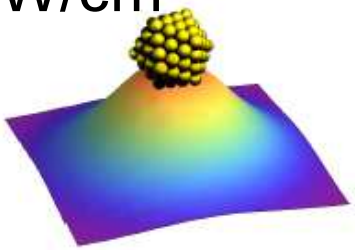
- Direct imaging of growth by coagulation
- Non-spherical shapes freeze out (“hailstones”)

Single cluster intensity distribution: no averaging

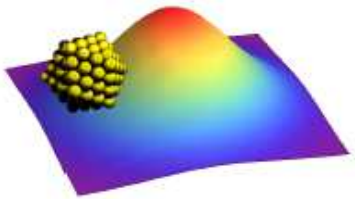
Focal density distribution

Hit in focus

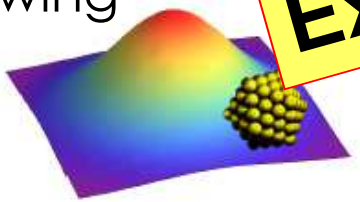
10^{14} W/cm^2



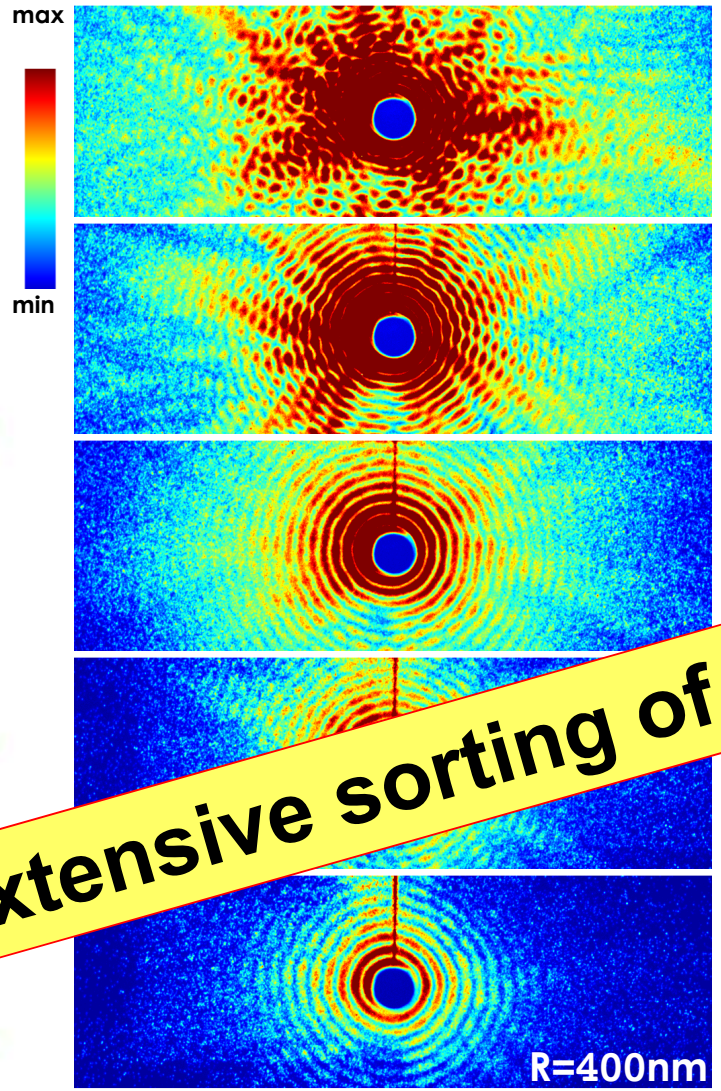
Hit in side



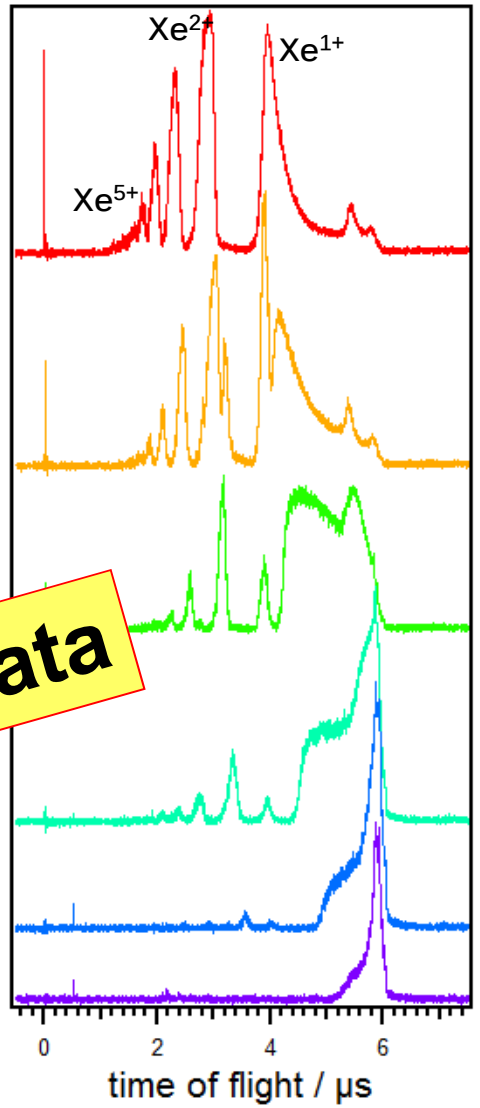
Hit in wing



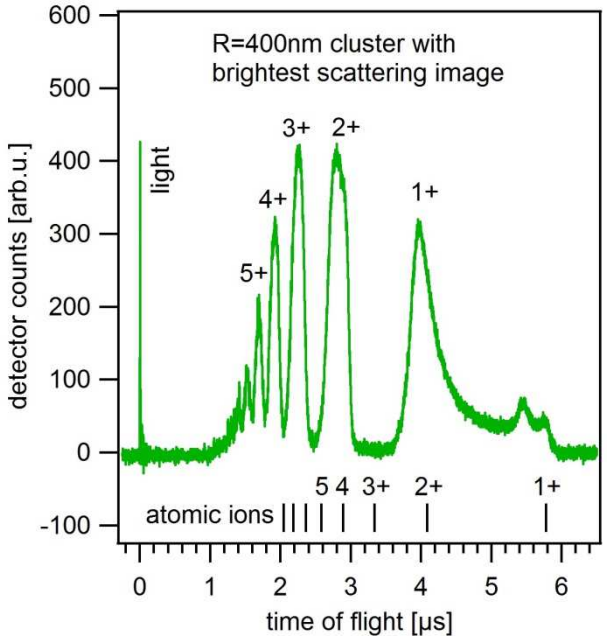
5x 10^{12} W/cm^2

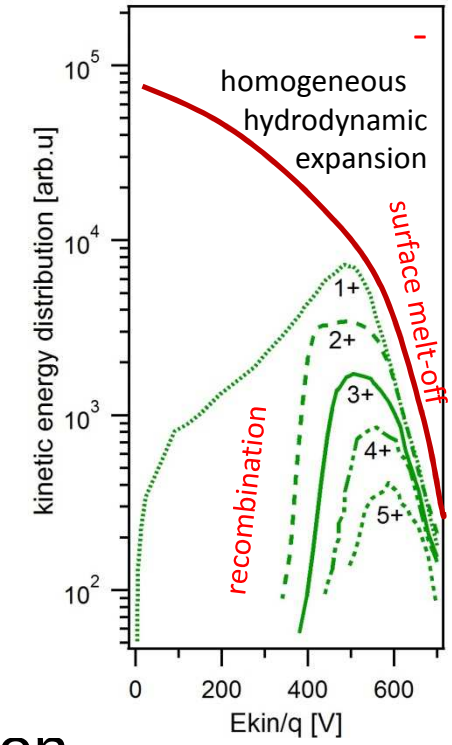


Extensive sorting of data

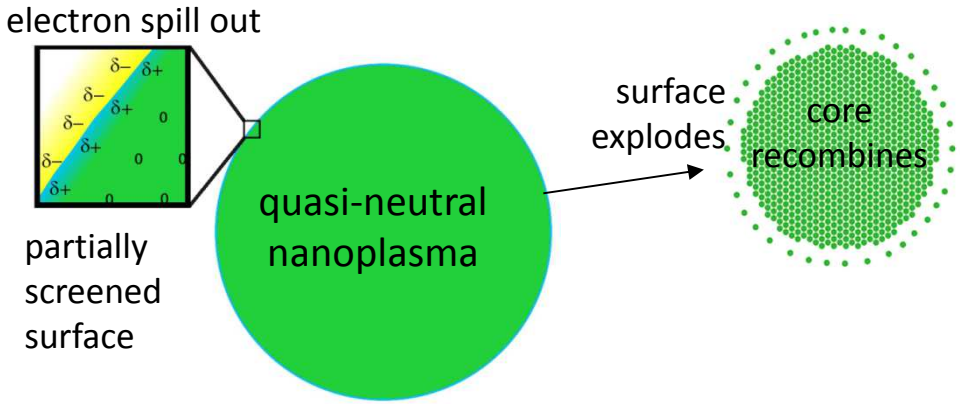


Single cluster ion spectra: Signatures of strong recombination





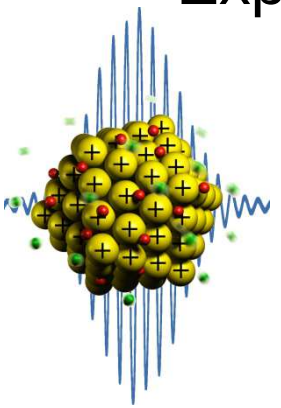
Very sharp lines;
narrow kinetic energy distribution

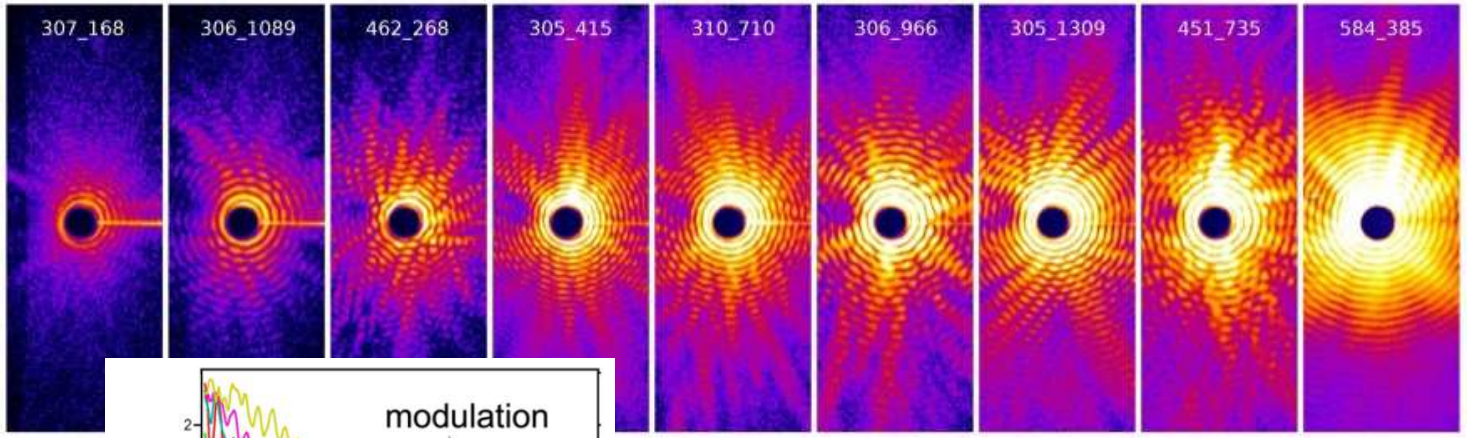


D. Rupp

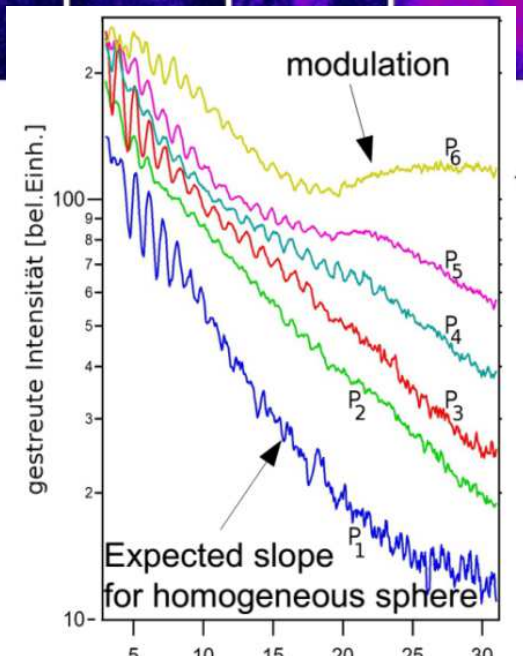
Single cluster scattering patterns

- Exposure power density (Intensity at position of the cluster)



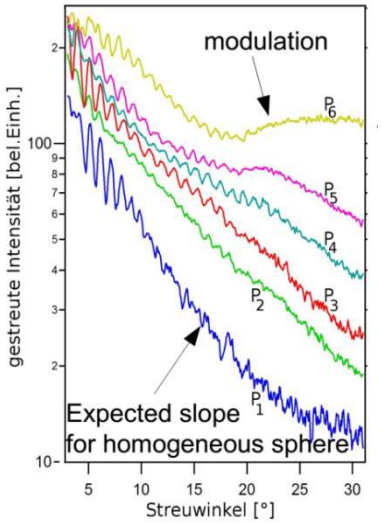


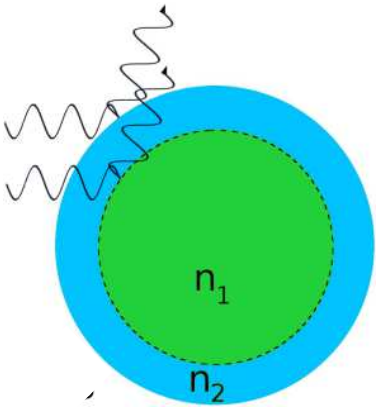
400 nm radius

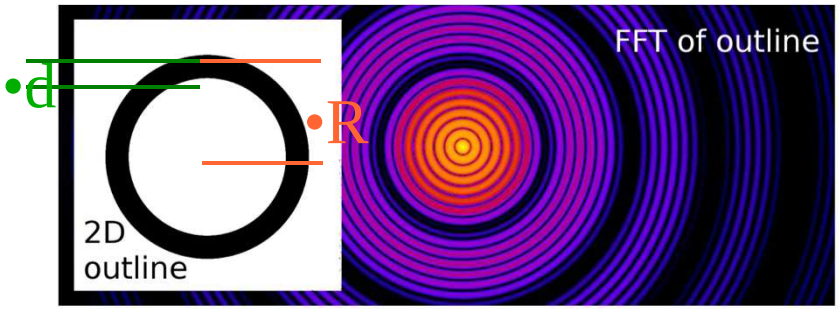


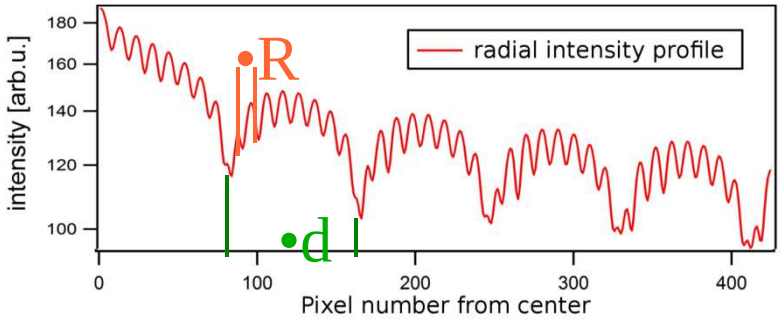
- Ultrafast electronic changes during the 100 fs light pulse

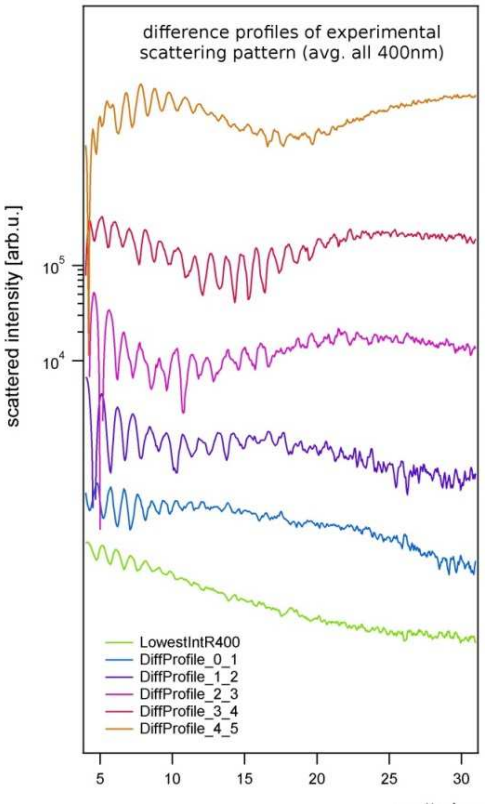
Nanoplama-shell with different refractive index

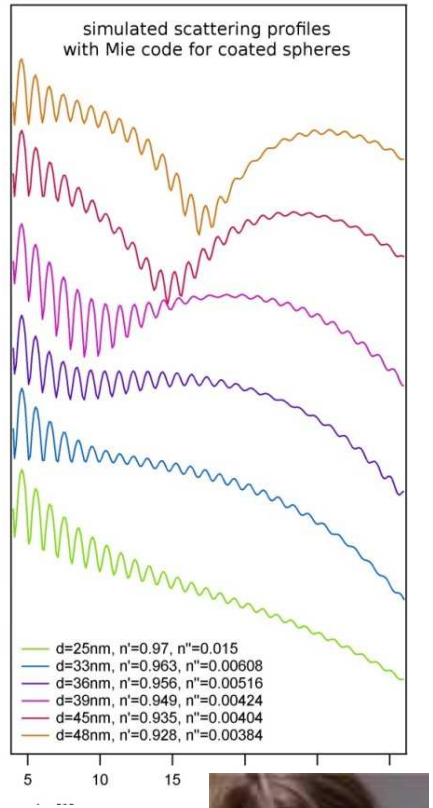










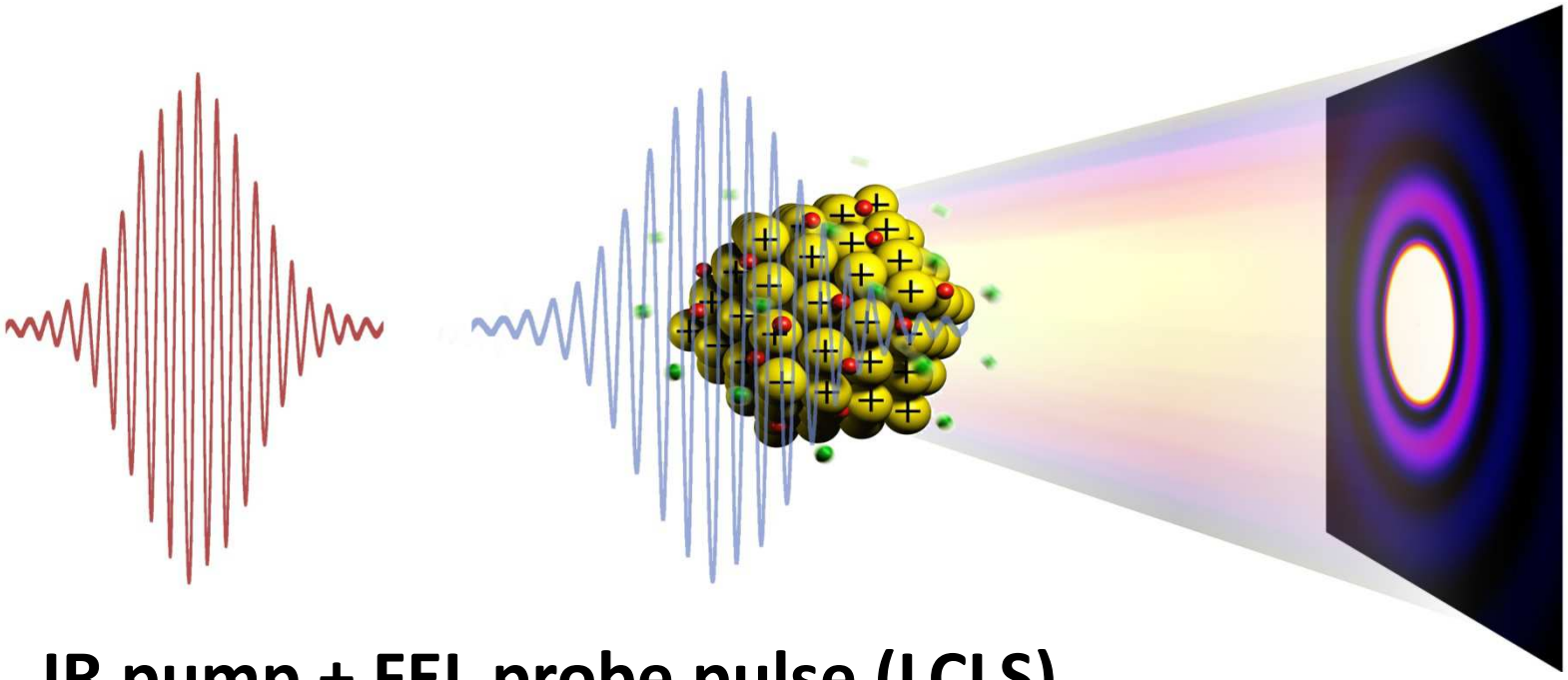


From bottom to top:
Weakly absorbing outer shell
Increasing thickness 25-50nm
Decreasing real part of the refra
index

D. Rupp PhD thesis

Time resolved imaging of exploding clusters

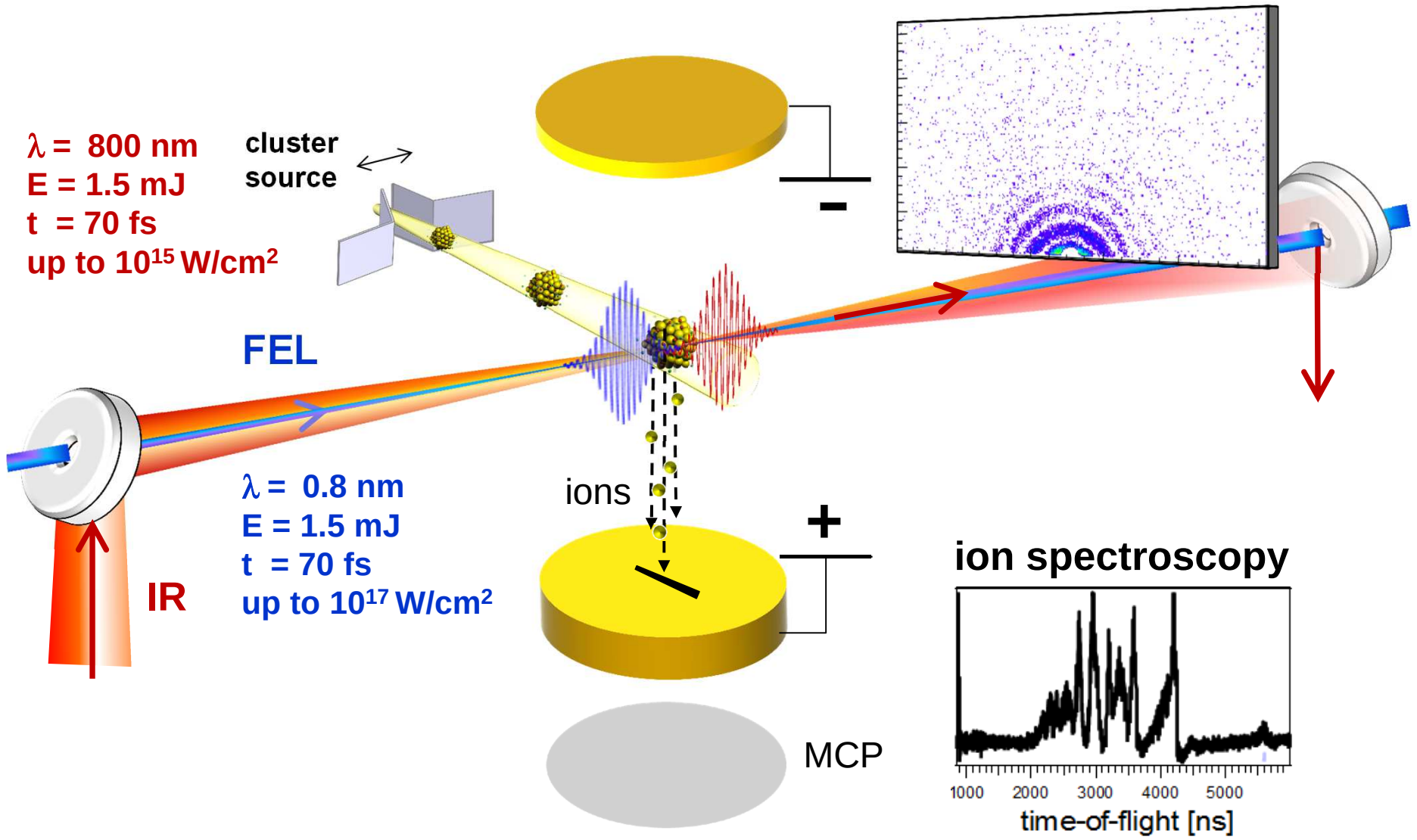
Scattering sensitive to both, changes in electronic and geometric structure



IR pump + FEL probe pulse (LCLS),

CAMP L. Strüder et al. Nucl. Instr. Meth. A 610, 483 (2010)

Experimental setup in CAMP

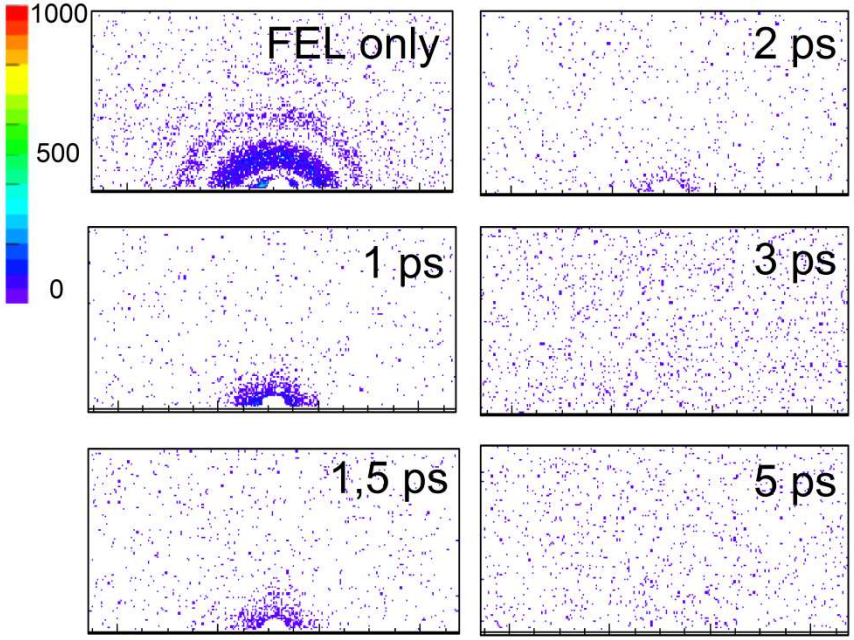


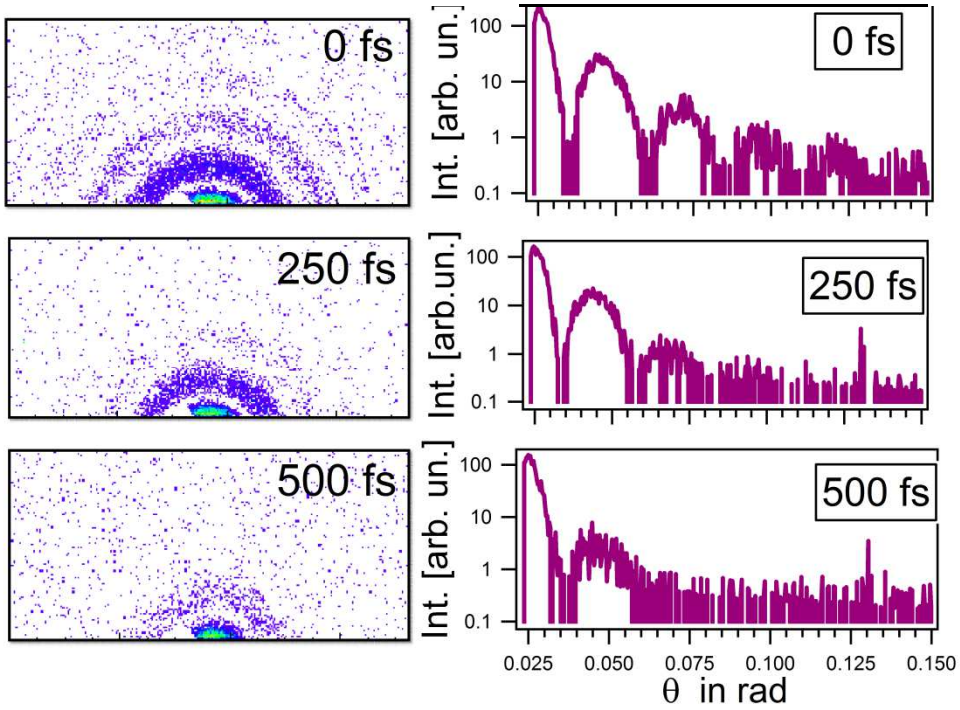
X-ray only: T. Gorkhover et al., Phys. Rev. Lett. 108, 245005 (2012)
S. Schorb, T. Gorkhover, et al., Appl. Phys. Lett. 100, 121107 (2012)

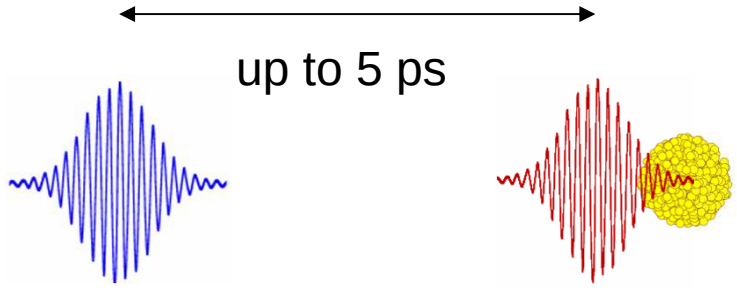
Delay dependent X-ray diffraction

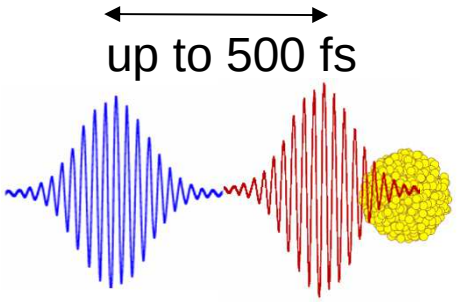
Xe clusters 20 nm radius
X-ray pulse 1.5 mJ, 1.5 nm

T.Gorkhover, PhD. thesis,

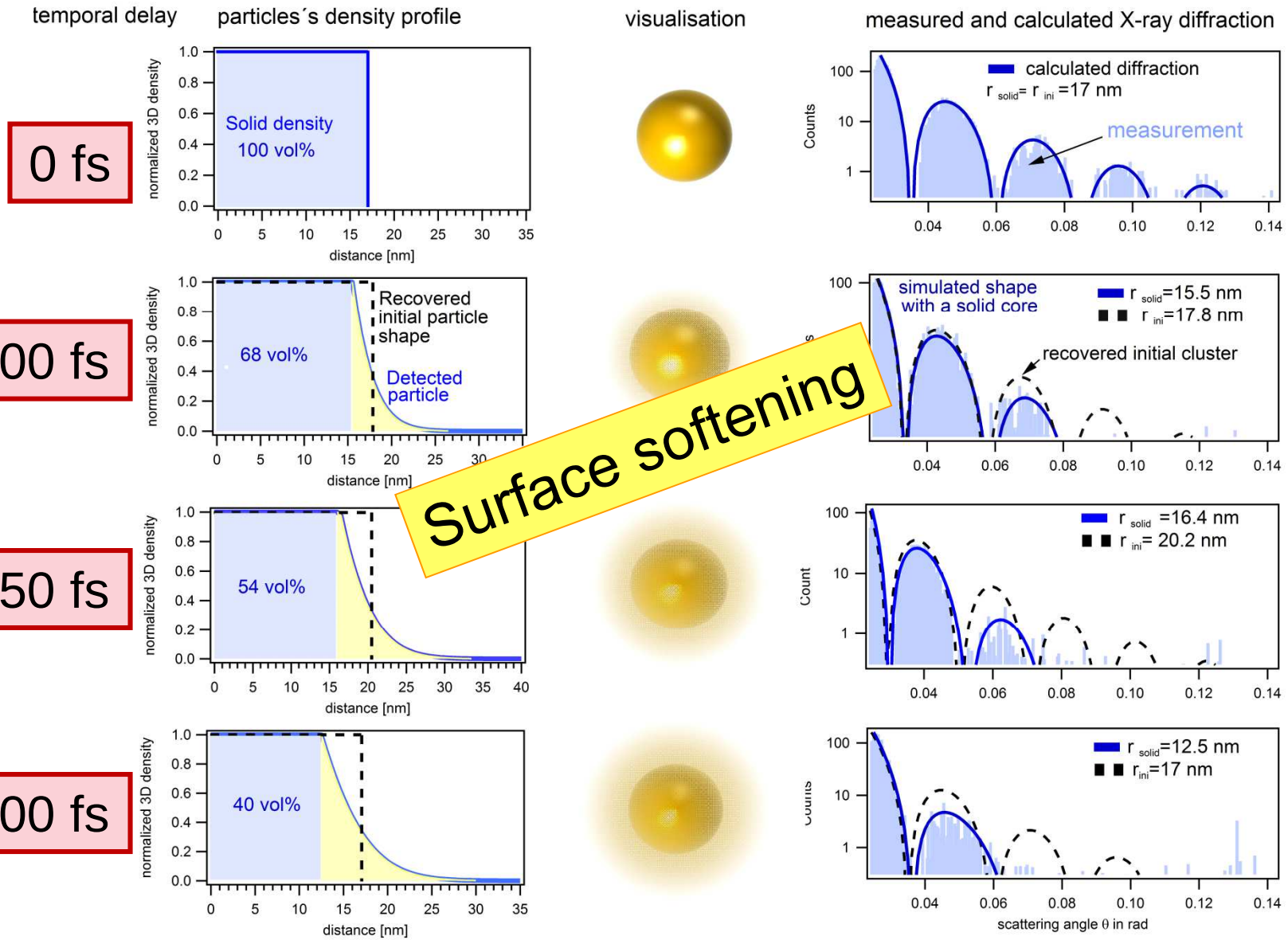








Comparison with simulation



T. Gorkhover et al., Nature photonics, under review

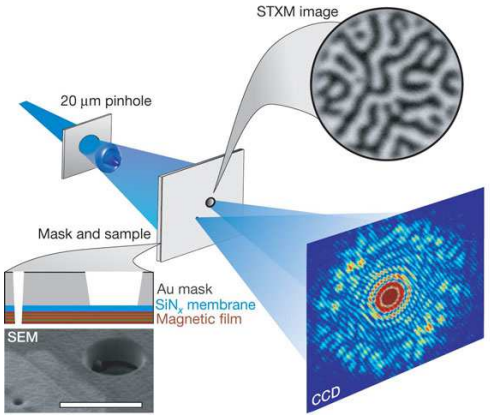
New imaging approaches

Holography: overcoming the phase problem

„In-flight“ holography

Tais Gorkhover, C. Bostedt et al

High resolution imaging of single gas phase nanoparticles



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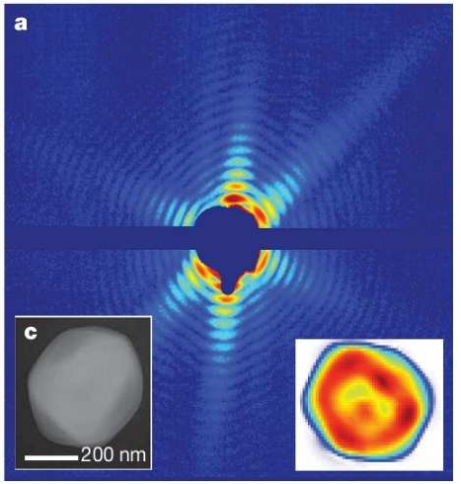


Eisebitt,S., et al., Nature **432**, 885 (2004)

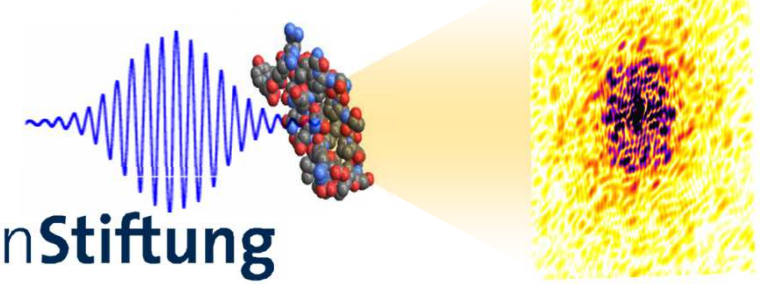
Geilhufe,J. et al., Nature Communications 5, 3008 (2014)

M. M. Seibert et al., Nature 470, 78 (2011)

X-ray Fourier holography

?????

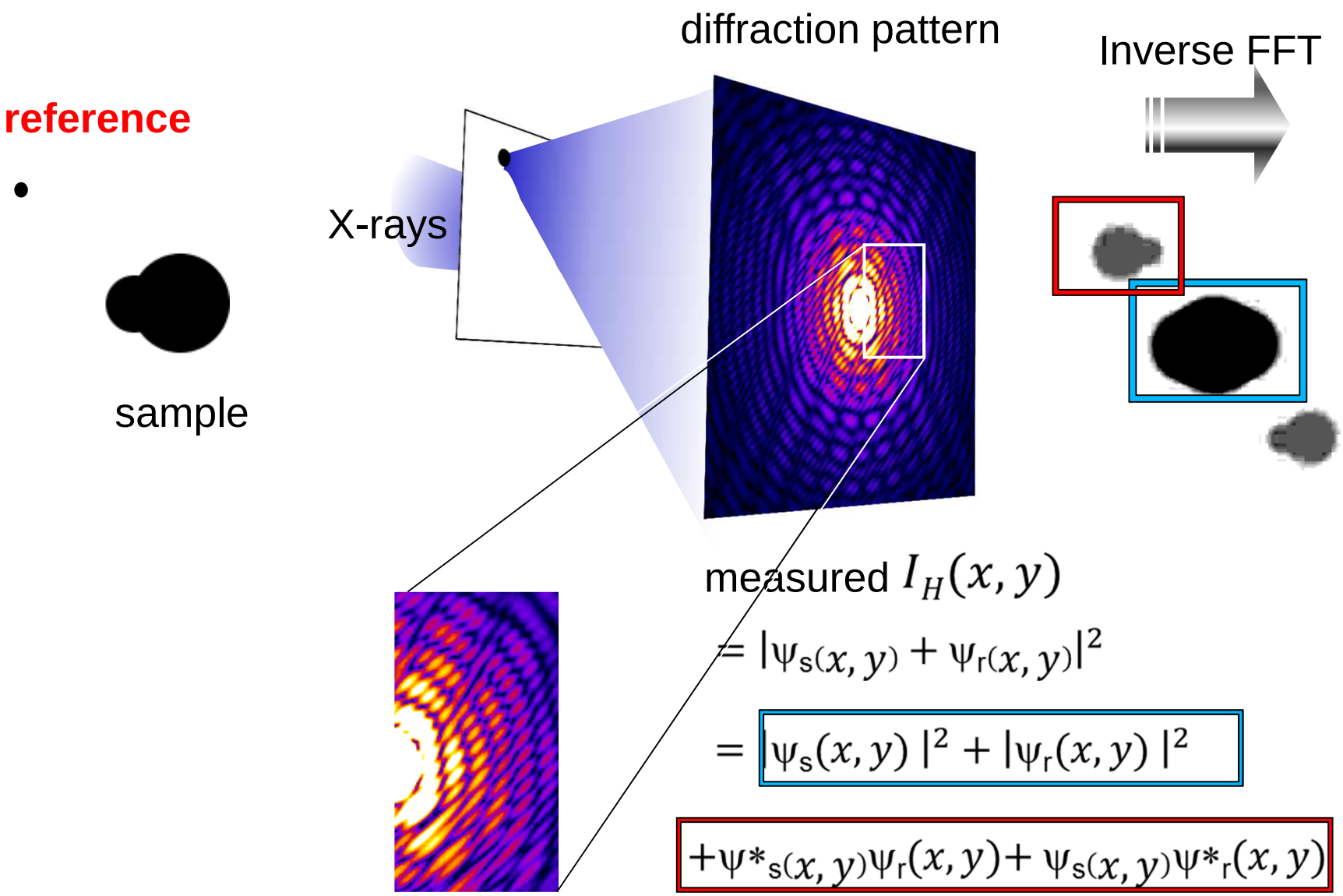
Single nanoparticle imaging



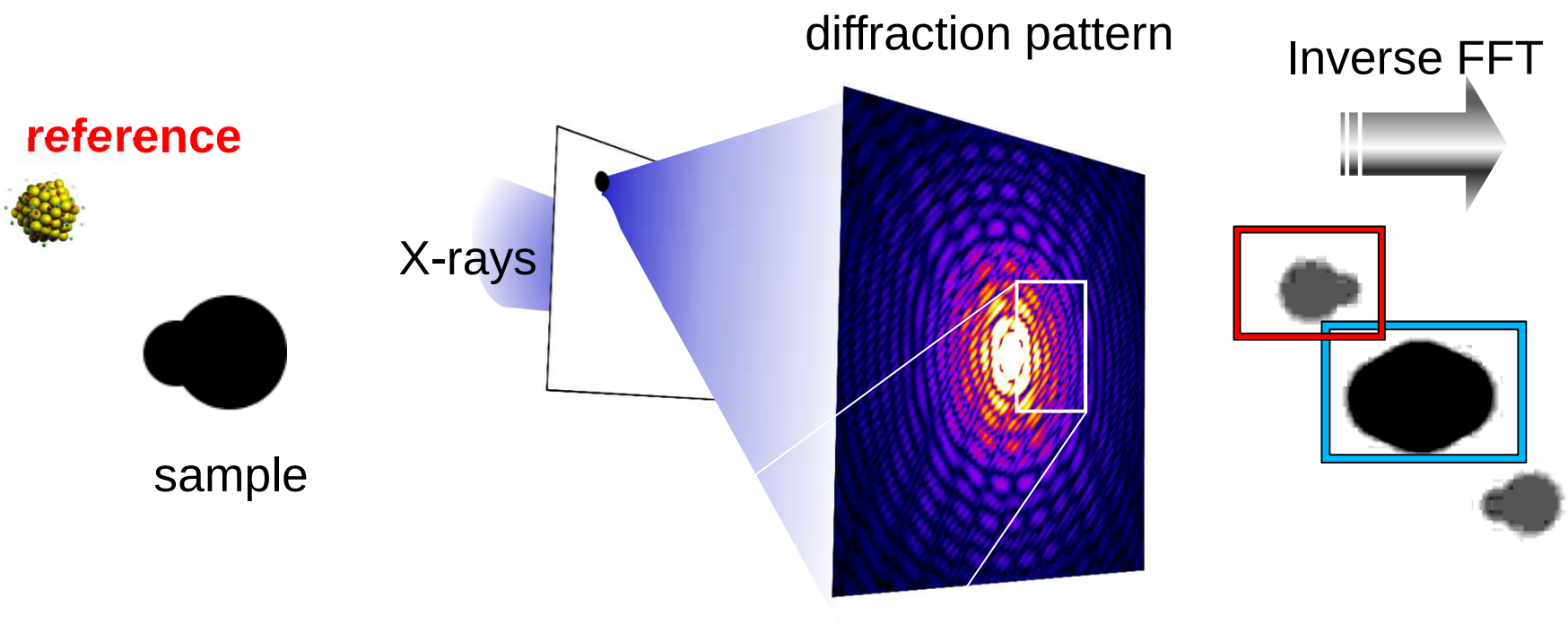
T. Gorkhover

VolkswagenStiftung

X-ray Fourier holography

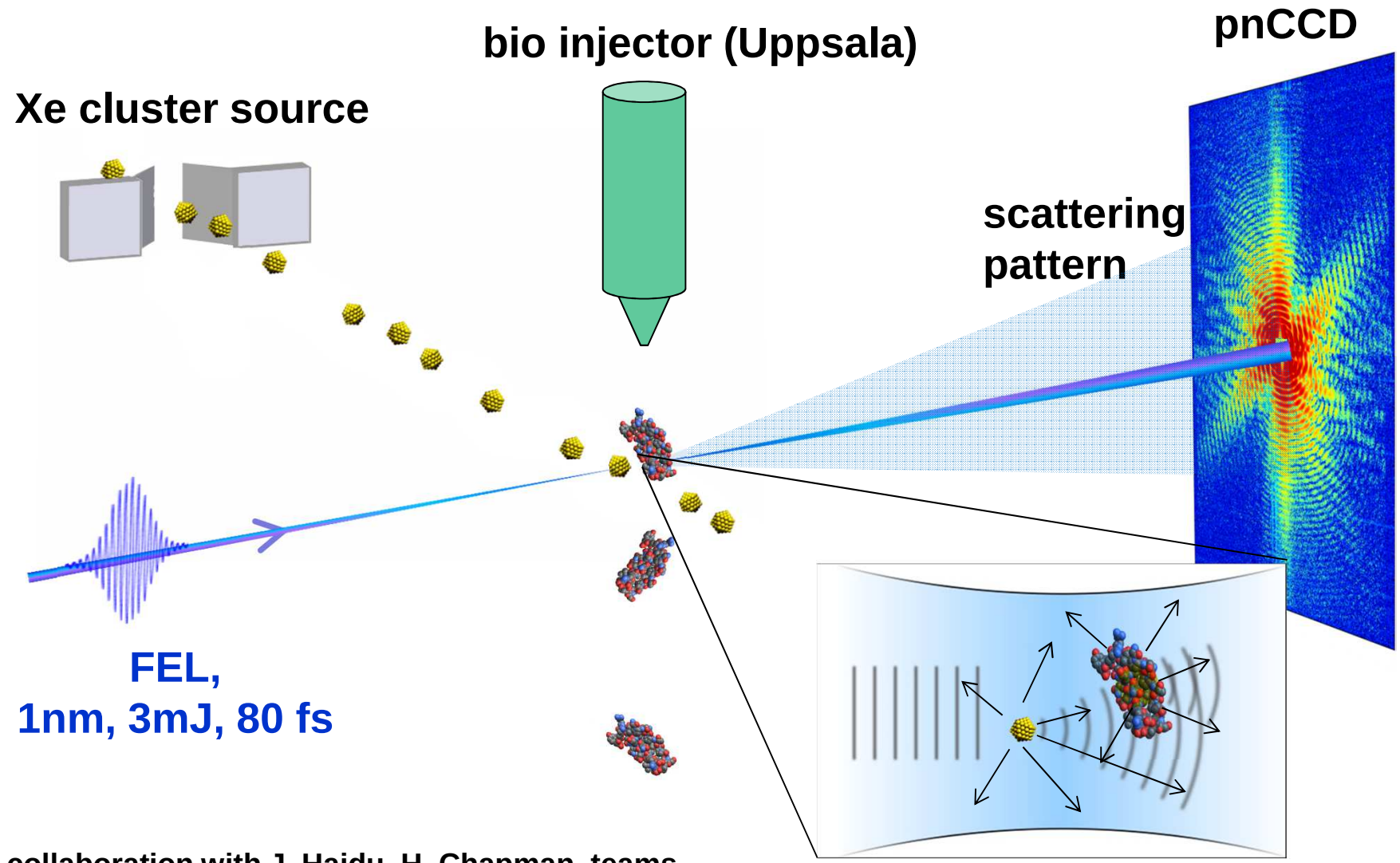


„In-flight“ X-ray Fourier holography



Gas phase single particle holography:
instead of a fixed mask, use randomly injected Xe clusters

Experimental setup in LAMP



collaboration with J. Hajdu, H. Chapman teams

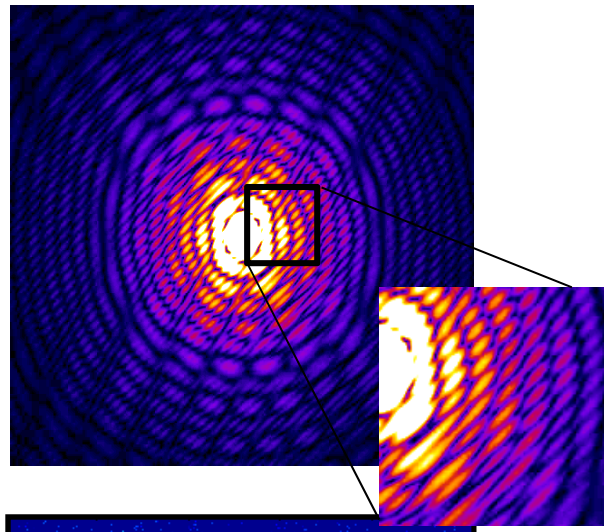
Holograms of twin particles

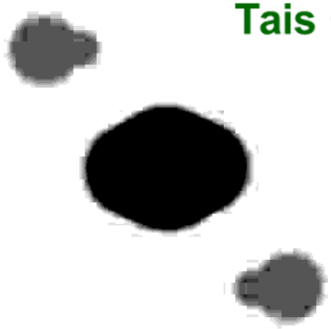
diffraction pattern

inverse 2D FFT

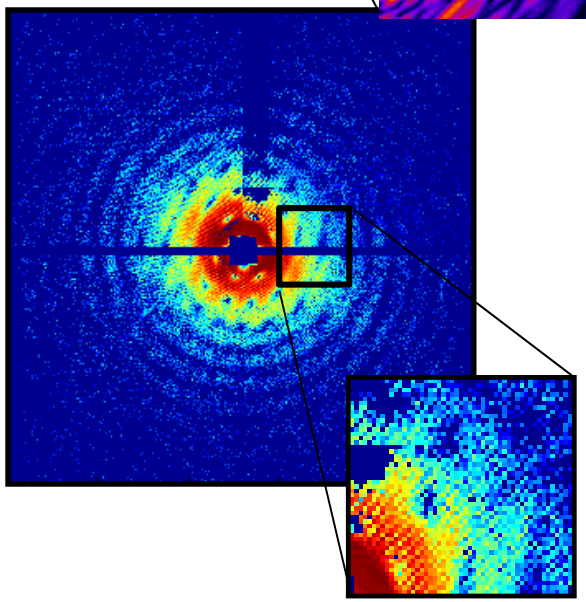
simulation

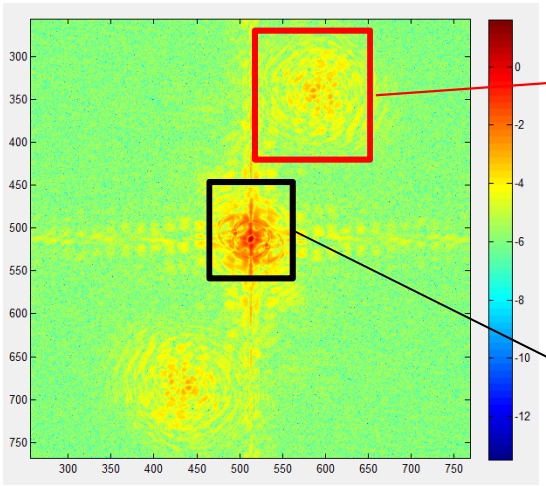
experiment

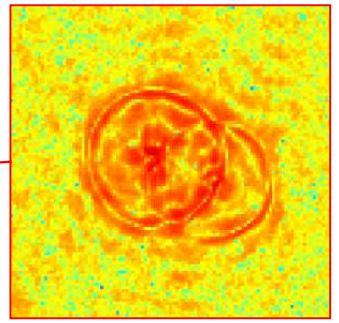


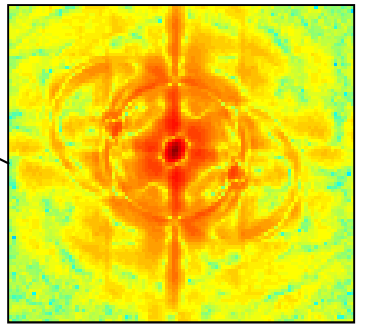


Tais Gorkhover, C. Bostedt et al





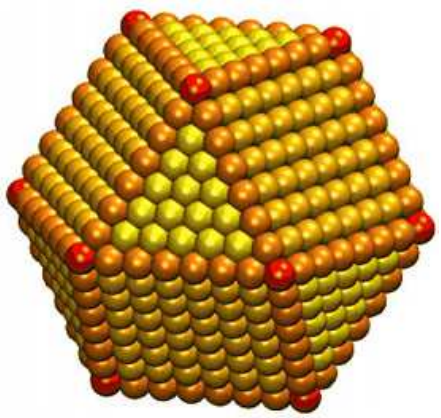




Imaging of metal clusters

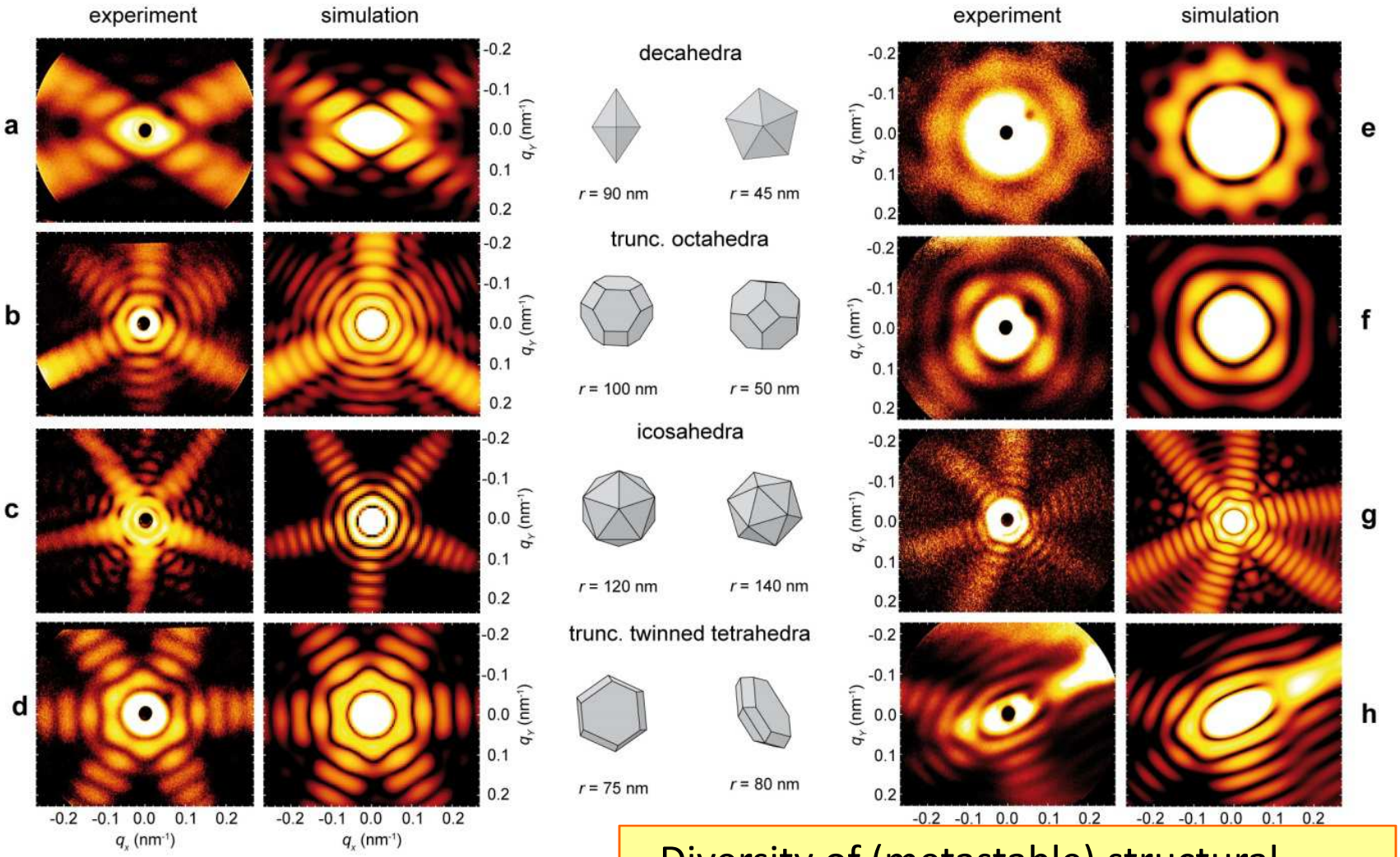
- Nanoplasma effects ?

- Regular shape, non-equilibrium structures ?



W.Zhu et al,
JACS **2013** *135* (45), 16833

Morphology of large gas-phase silver clusters



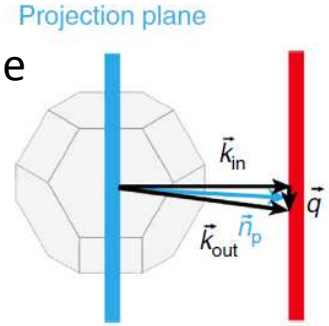
I. Barke, et. al, Nature Communications 7187 (2015)

collaboration with Rostock

- Diversity of (metastable) structural motives
- 3D information in a single-shot image

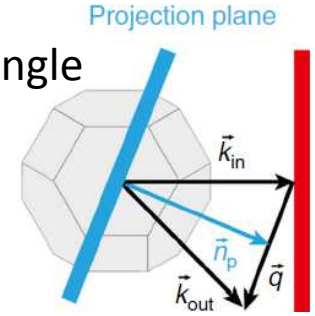
Key to 3D sensitivity: large angle scattering

small angle



$\mathbf{n}_p \| \mathbf{k}_{in}$

large angle



variation of $\mathbf{n}_p$ with $\mathbf{q}$
(single shot tomography)

$$I(\mathbf{q}) \propto \left| \int \rho(\mathbf{r}) e^{i\mathbf{q}\mathbf{r}} d^3r \right|^2$$

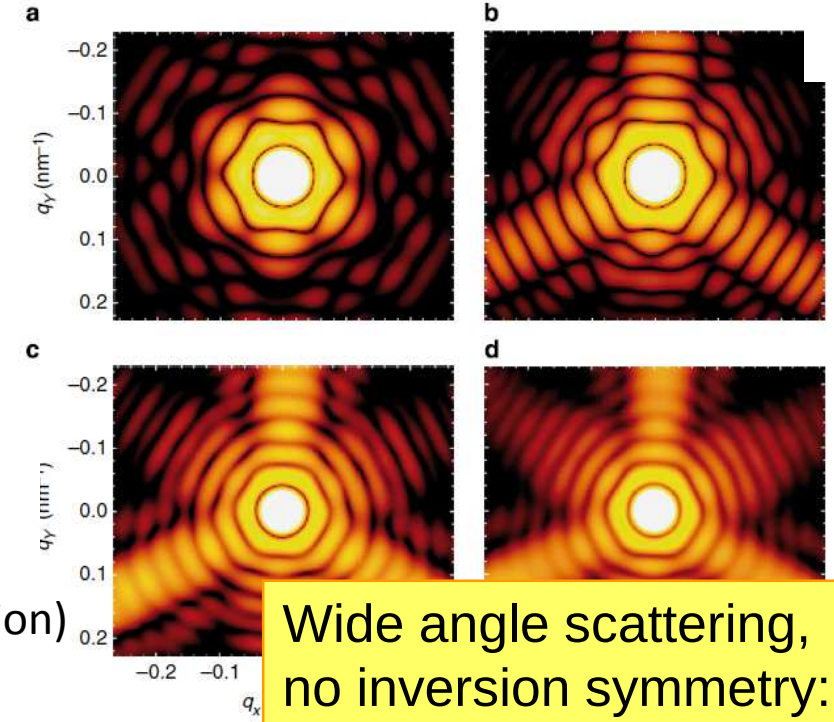
$$\mathbf{n}_p = \mathbf{k}_{in} + \mathbf{q}/2$$

$$\rho(\mathbf{r}_\perp) = \int \rho(\mathbf{r}) dr_{||}$$

$$I(\mathbf{q}) \propto \left| \rho(\mathbf{r}_\perp) e^{i\mathbf{q}\mathbf{r}_\perp} d^2 r_\perp \right|^2$$

trunc. oct., r=120, λ=13.5nm

Born 2D projected



Born 3D

Born 3D
+ effective absorption

(used for quick identification)

full solution of continuum Maxwell Eq. via FDTD

(used for refinement)

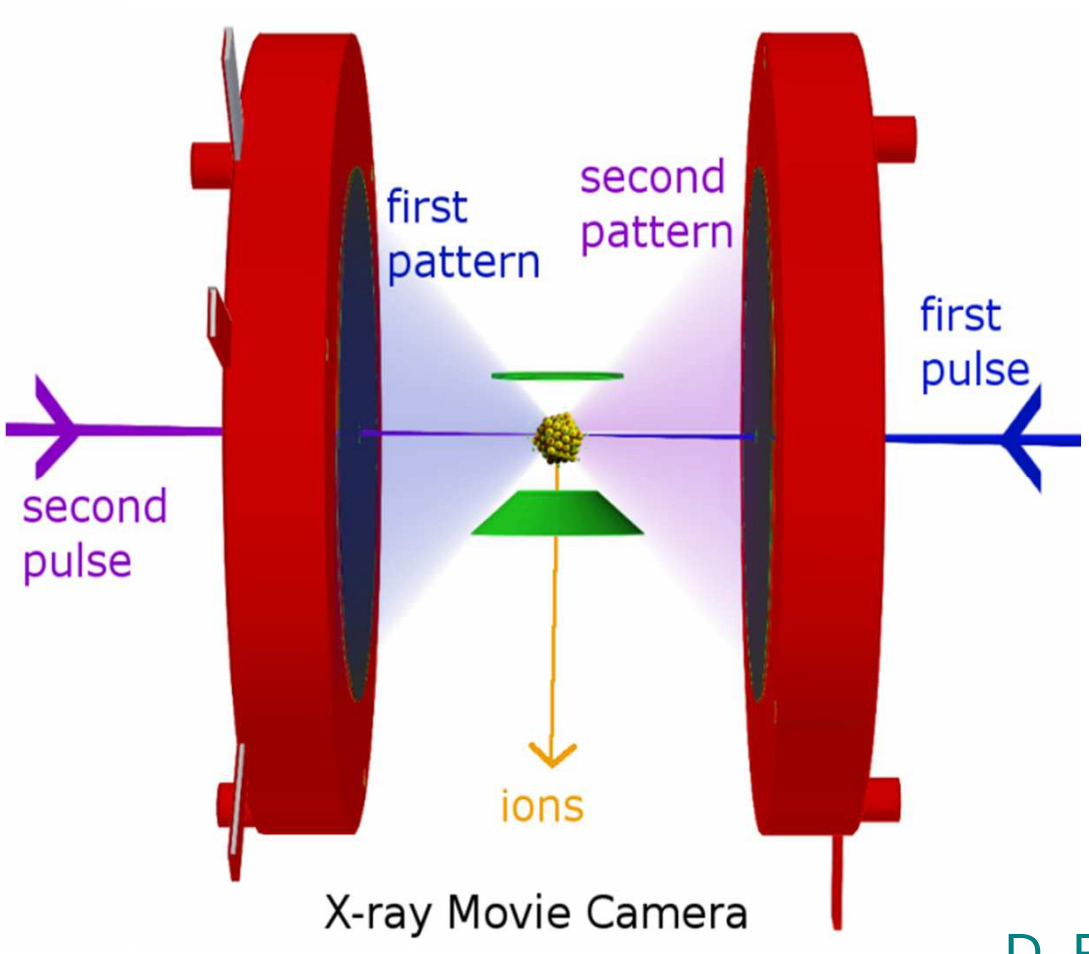
Wide angle scattering,
no inversion symmetry:
→ 3D Structure

I. Barke et. al,
Nature communications 7187 (2015)

Outlook

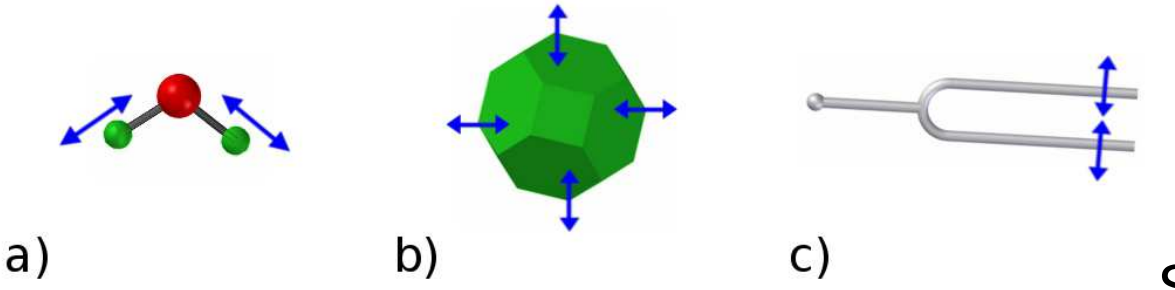
Novel approach for time resolved imaging

Two images from a single clusters at different times

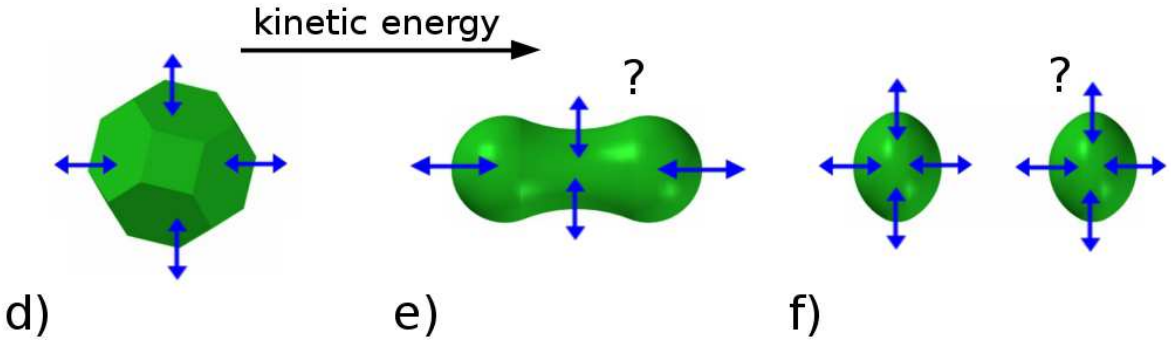


D. Rupp, TU-Berlin

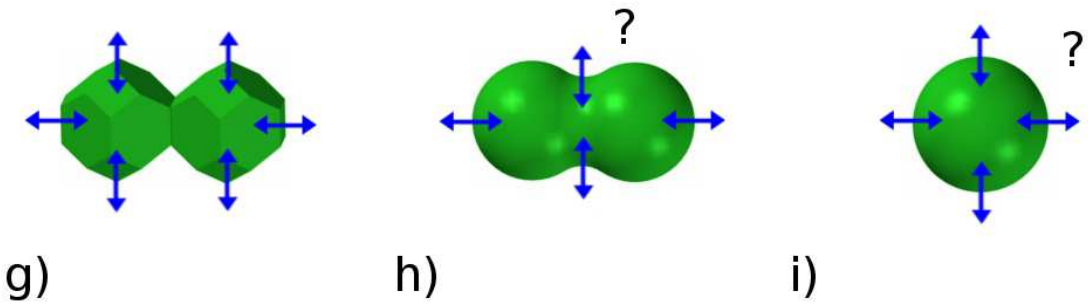
Collective oscillations/dynamics in nanoparticles, surface melting



Size selected



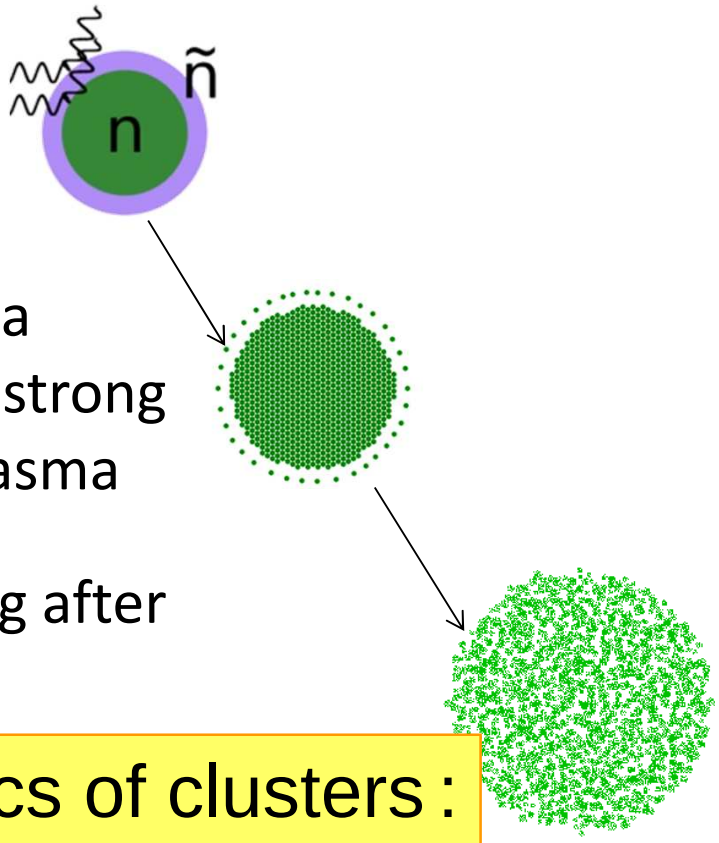
Damping?



Vis /XUV pump probe

Summary and outlook: Clusters in intense X-ray pulses

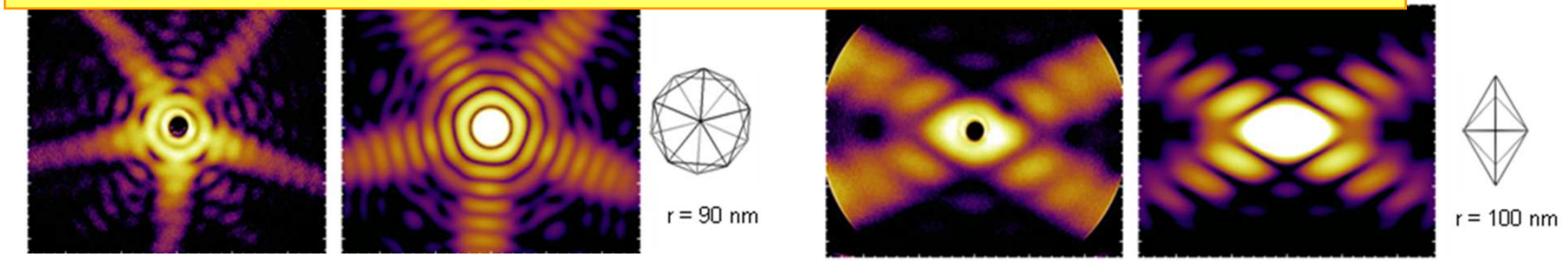
- Scattering pattern: ultrafast electronic changes due to plasma generation



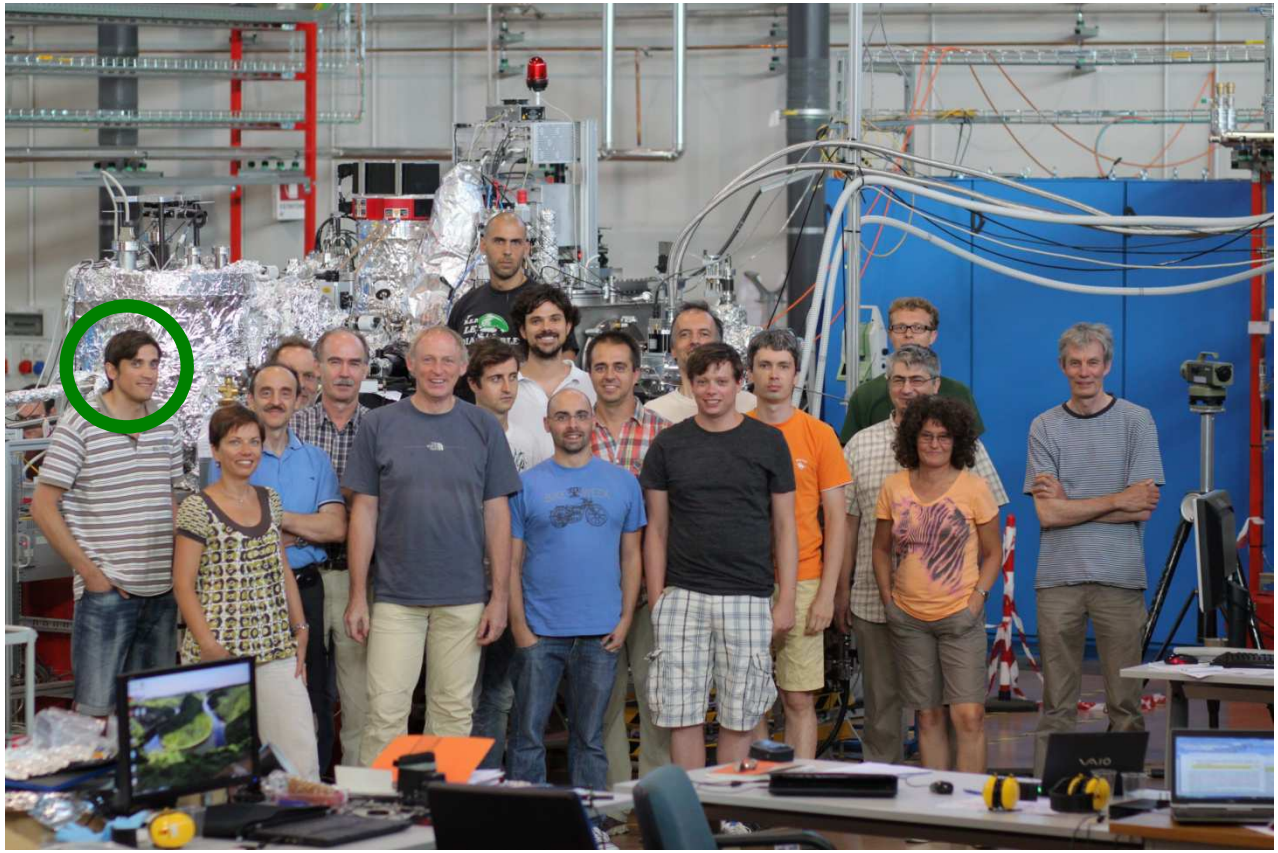
- electron and ion spectra: nanoplasma formation explosion of a thin surface, strong recombination in the quasi-neutral plasma

- Time-resolved: image surface melting after tens of ps, debris after ps-ns

Imaging structure and dynamics of clusters : A lot of exciting physics ahead of us!



Experiments at FERMI / LDM collaboration: He cluster



Carlo Callegari, Aron Laforge, Y. Ovcharenko, Paolo Piseri, Victor Layamayev, Ravael Katzky, Paola Finetti, Oksana Plekan, Marcello Coreno, Robert Richter, Marcel Drabbels, Kevin Prince, Thomas Möller, Frank Stienkemeier , from ERMI Flavio Capotondi, Gerardo D'Auria, Giuseppe Penco,Emiliano Principi, Marco Zangrando.

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Thank you for your attention!