Clusters structure and dynamics studied with X-ray FEL pulses

Thomas Möller, Technische Universität Berlin
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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
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 only for few systems!

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

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

At IR regime:
T. Ditmire et al.
M. Lezius et al.
Meiwes-Broer et al.
M. Mudrich
Intense light pulse - cluster interaction

Cluster ionisation
- Single photon ionization
- Few electrons are removed from cluster

Inner ionisation
- Electron trapping
- Nano plasma, neutral core

Strongly dependent on wavelength and power density!

Experiments

Theory
Spectroscopy and light scattering of clusters

- Ion/electron spectroscopy
- Fluorescence
- Ionisation, relaxation, recombination
- ps-μs time scale after the pulse

Different time scales!

- Geometry of the cluster
- Electronic processes and transient states
- Fs time scale
- During the pulse
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

ion spectrum

scattering pattern

single cluster in focus
Simultaneous light scattering and ion spectroscopy on individual clusters

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

Xe clusters
Experimental scattering pattern

Simulation
Shape of large gas phase clusters

Simulation of scattering pattern with 2D Fourier transform

Grainy structure
hailstones = snapshots of cluster growth by coagulation

700 nm
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

electrons

g1
+++++

scattered light

two sets of large CCDs

ions

simultaneous detection
• ions
• electrons
• scattered light

CFEL-ASG-Multipurpose-Camber (CAMP)

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

Absence of low charge states?
Absence of low charge states

Xe cluster 20 nm, 10^6 atoms

\( h\nu = 1.5 \text{ keV} \)

Pulse energy: \( 2.9 \pm 0.2 \text{ mJ} \)

Pulse intensity: \( 5 \times 10^{16} \text{ W/cm}^2 \)

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

- only highly charged ions from the surface layer explode off?
- no recombination? no neutral atoms?

recombination rate \( \sim T^{-4.5} \) very hot plasma small recombination

VUV: almost complete recombination in large clusters
Simulation with plasma code FLYCHK:

• 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).


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

up to 6 ps
Imaging of clusters with X-rays: Ar-Xe core shell cluster

Complicated scattering patterns

- twin clusters
- internal structure

Spatial resolution:

\[ \Delta x = \lambda N_a \]

\( \lambda \) numerical aperture

Trade off between wavelength and cross sections

20 nm

20 % Xe, 80 % Ar

1.5 keV
2 mJ
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/cm²

Next steps I: Time resolved dynamics

- Ionisation and plasma dynamics of size selected particles

- Phase transitions in clusters
  - liquid-solid transitions time resolved
  - surface melting
  - ...

Next steps II: Complex samples

- nanocrystals, nanodroplets, geometric structure, shape
- heterogeneous clusters, core shell structures, fractal clusters
- mass selected cluster ions
  - almost exact number of atoms
  - precise control of temperature
- surface structure

Water clusters, Ice nanoparticles
What can be expected with nm radiation?

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

Facetts become visible

long axis perpendicular to light beam long axis parallel to light beam

Cluster symmetry
Vision: Cluster structure determination with atomic resolution?

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

Reconstructed images

- minimum cluster size ?
- heavy elements ?
Challenge

- cluster structure with nearly atomic resolution
- two colour pump-probe (1th and 3rd harmonic) 'before' and 'after' light induced changes
- time resolved electron dynamics
  THz streaking
Wish List / Requirements

1. Direct beam with maximum power, 250eV – 2 (6 keV), > 5 mJ
   • small focus, 100 nm, variable
   • pulse length 5- 200 fs
   • option for extensions

2. Synchronized lasers and THz pulses (electron streaking)
   • IR-laser 50 fs puls length, a few mJ pulse energy
   • THz pules, 100 fs puls length, interesting option

3. X-ray split and delay unit (1th and 3rd harmonic)
   • 3rd harmonic more than 1%?
   • Energy dispersive pixel detector

4. Flexible experimental chamber
   Sources for nanoparticles and size selected clusters (users)
Summary

Fundamental x-ray induced processes in clusters:

- ionization, plasma formation, recombination, desintegration

Single shot imaging of single particles

cluster structure and shape

better resolution

- maximum information thanks to *coincidence* detection

Time resolved studies with pump-probe:

electron dynamics, phase transitions
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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

DESY/FLASH Team
K.H. Meiweis-Broer,
J. Tiggesbäumker, T. Laarmann,
T. Ditmire, J. Hajdu

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And thank you for your attention!