Kinetic equation approach to describe dynamics of irradiated samples

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in collaboration with
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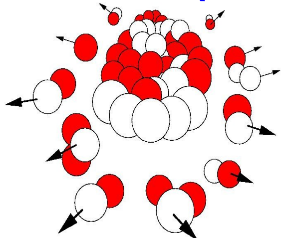
T. Laarmann, R. Santra, F. Wang, E. Weckert (DESY)
and T. Mőller (TU Berlin)



X-Ray Science in the 21st Century, Kavli Institute of Theoretical Physics, 2-6 August 2010

HELMHOLTZ ASSOCIATION

CFEL SCIENCE

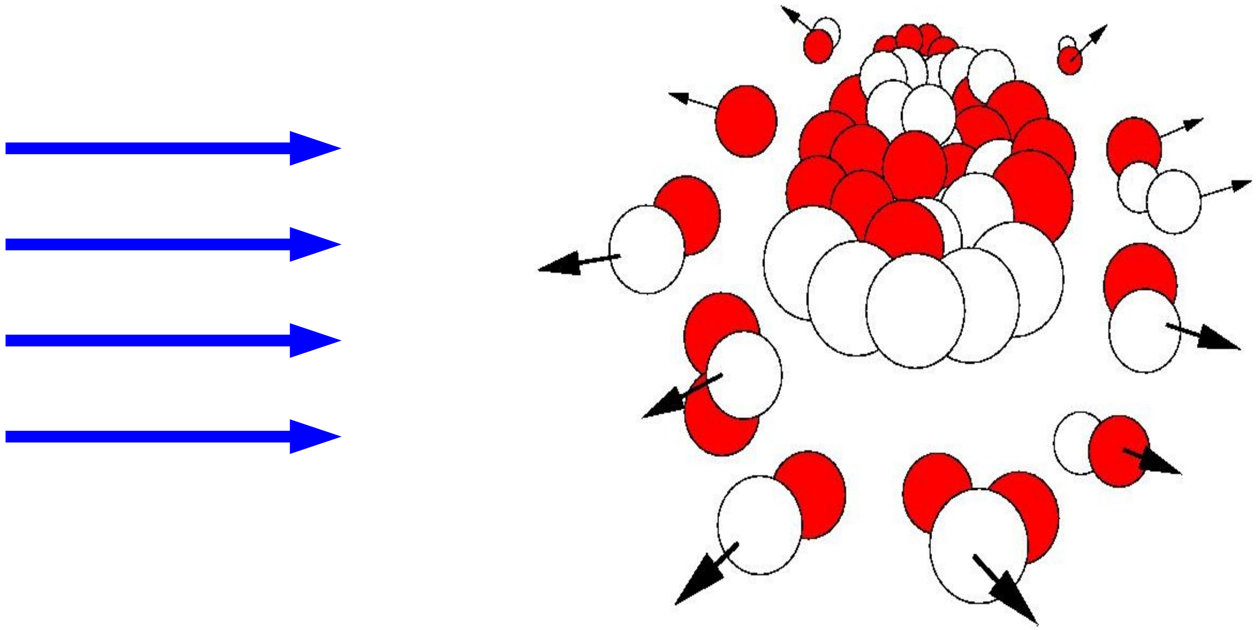
DESY

Motivation for theoretical study

Important for: (i) experiments with FEL → cluster experiments, single particle imaging, warm dense matter etc.

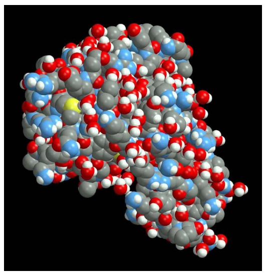
(ii) construction of the laser → FEL optics,

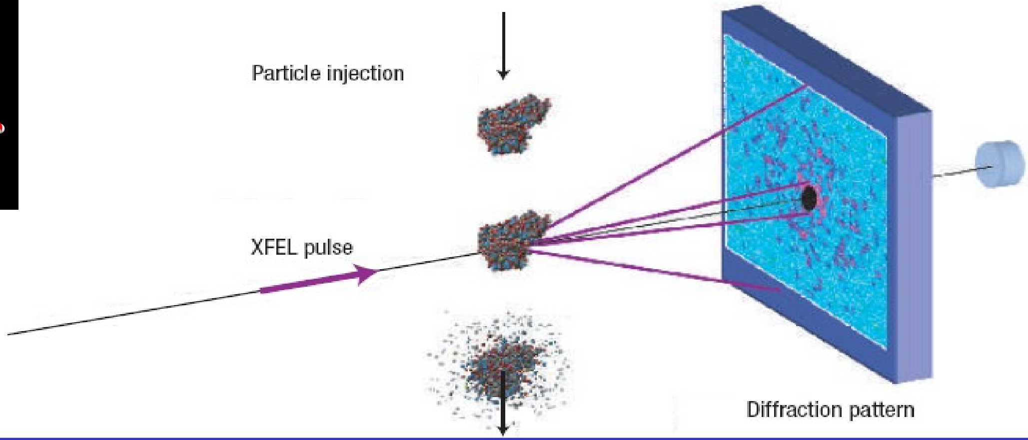
(iii) test of various theoretical models



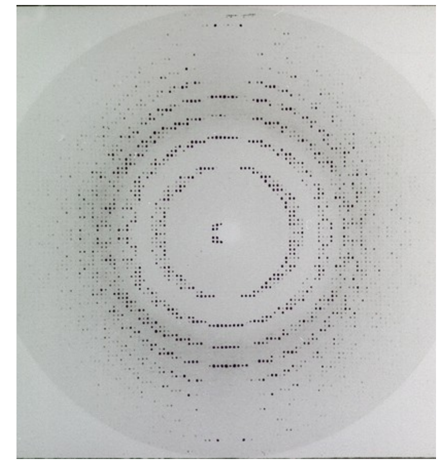
Structure determination through single particle diffraction imaging?

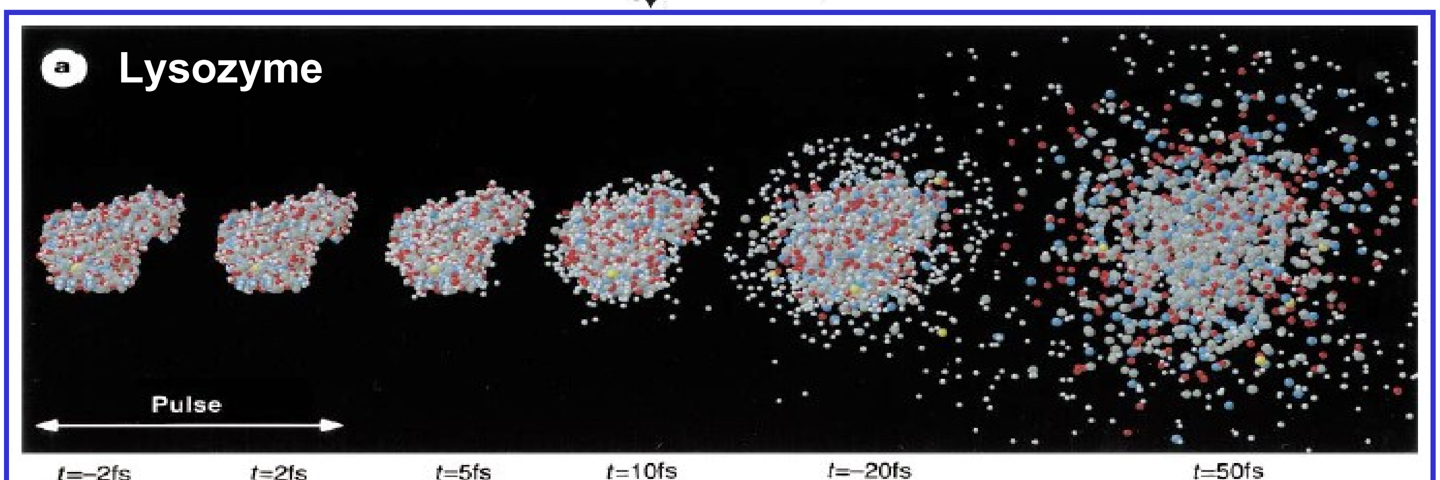
Molecules at atomic resolution





Crystal



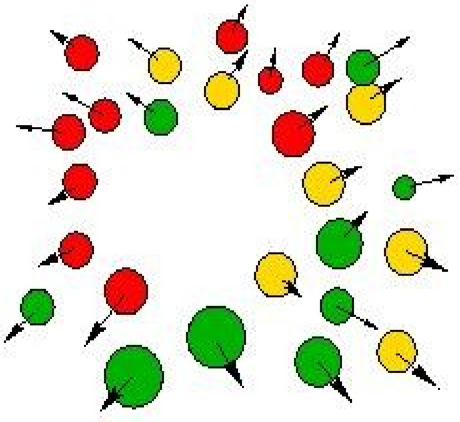


R. Neutze, R. Wouts, D. van der Spoel, E. Weckert, J. Hajdu Nature 406, 752 (2000)

Radiation damage and Coulomb explosion

Tool: statistical Boltzmann approach

Evolution of larger systems described in terms of statistical density function, ρ(r,v,t), in phase space:



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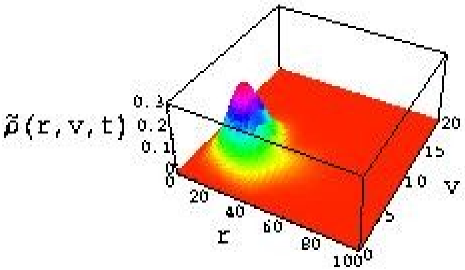


Tool: statistical Boltzmann approach

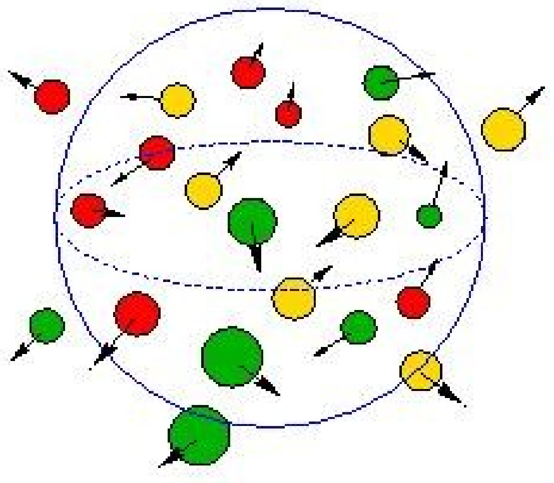
- first-principle approach

Boltzmann equations are able to follow

- single-run method

non-equilibrium processes

- computational costs do not scale with number of atoms



Difficulty:

- requires advanced numerical methods

Solving Boltzmann equations

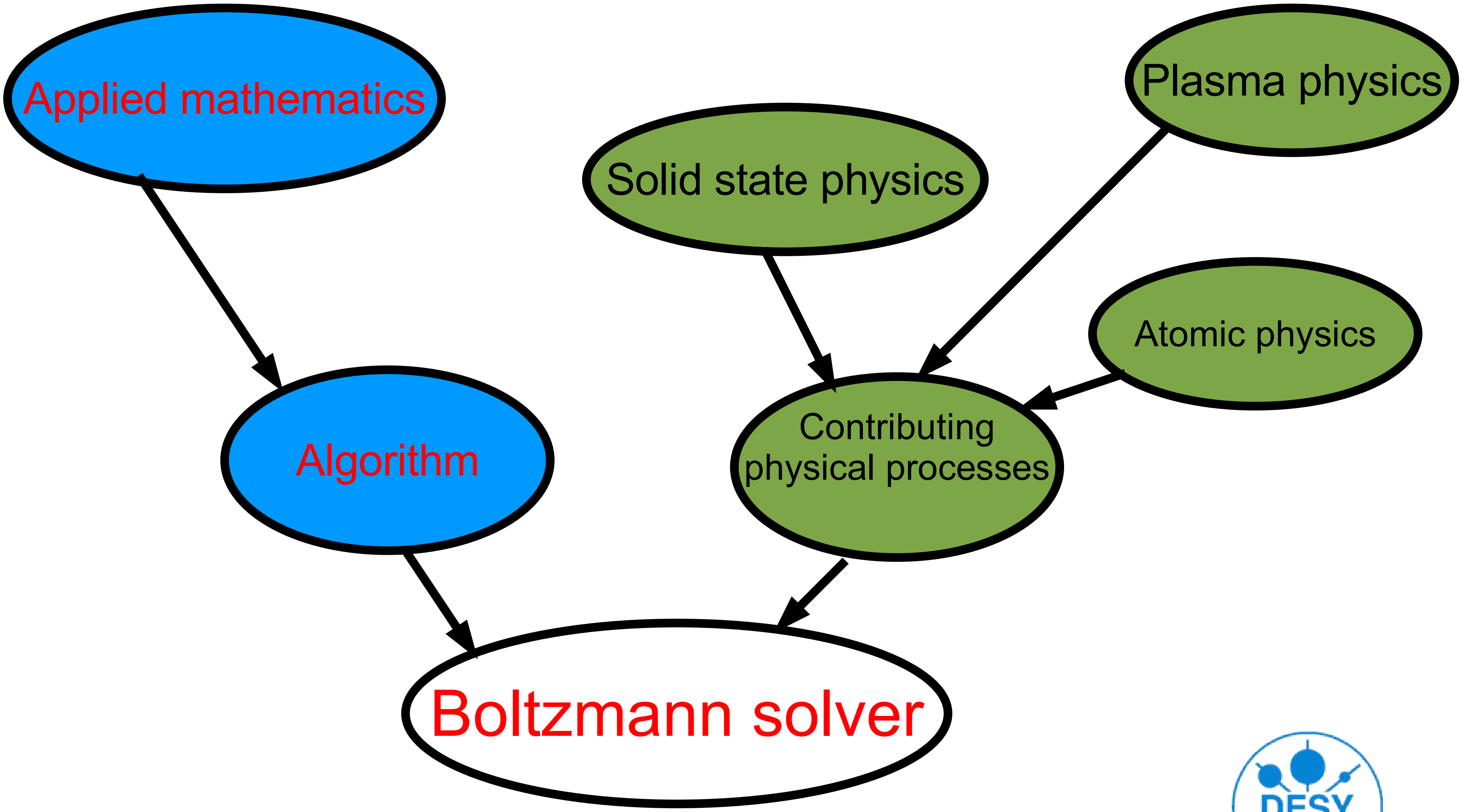
The general coupled Boltzmann equations for electron, $\rho^{(e)}(\mathbf{r},\mathbf{v},t)$, and ion densities, $\rho^{(i)}(\mathbf{r},\mathbf{v},t)$, where $i=0,1,\ldots,N_J$ denotes the ion charge, and N_J is the maximal ion charge in the system are:

$$\partial_t\rho^{(e)}(\mathbf{r},\mathbf{v},t)+\mathbf{v}\cdot\partial_{\mathbf{r}}\rho^{(e)}(\mathbf{r},\mathbf{v},t)+\frac{e}{m}\left(\mathbf{E}(\mathbf{r},t)+\mathbf{v}\times\mathbf{B}(\mathbf{r},t)\right)\cdot\partial_{\mathbf{v}}\rho^{(e)}(\mathbf{r},\mathbf{v},t)=\Omega^{(e)}(\rho^{(e)},\rho^{(i)},\mathbf{r},\mathbf{v},t),$$

$$\partial_t\rho^{(i)}(\mathbf{r},\mathbf{v},t)+\mathbf{v}\cdot\partial_{\mathbf{r}}\rho^{(i)}(\mathbf{r},\mathbf{v},t)-\frac{ie}{M}\left(\mathbf{E}(\mathbf{r},t)+\mathbf{v}\times\mathbf{B}(\mathbf{r},t)\right)\cdot\partial_{\mathbf{v}}\rho^{(i)}(\mathbf{r},\mathbf{v},t)=\Omega^{(i)}(\rho^{(e)},\rho^{(i)},\mathbf{r},\mathbf{v},t).$$

These equations include the total electromagnetic force acting on ions and electrons. Collision terms, $\Omega^{(e,i)}$, describe the changes of the electron/ion densities of velocities $(\mathbf{v},\mathbf{v}+d\mathbf{v})$ measured at the positions $(\mathbf{r},\mathbf{r}+d\mathbf{r})$ with time. These changes are due to short-range processes, e. g. collisions, photoabsorptions. The number of processes involved in the sample dynamics depends on the radiation wavelength.

Solving Boltzmann equations



Boltzmann solver

- Investigates the non-equilibrium phase of evolution of an irradiated sample until thermalization of electrons and saturation of ionization is reached

It uses the angular moment expansion for density function, ρ:

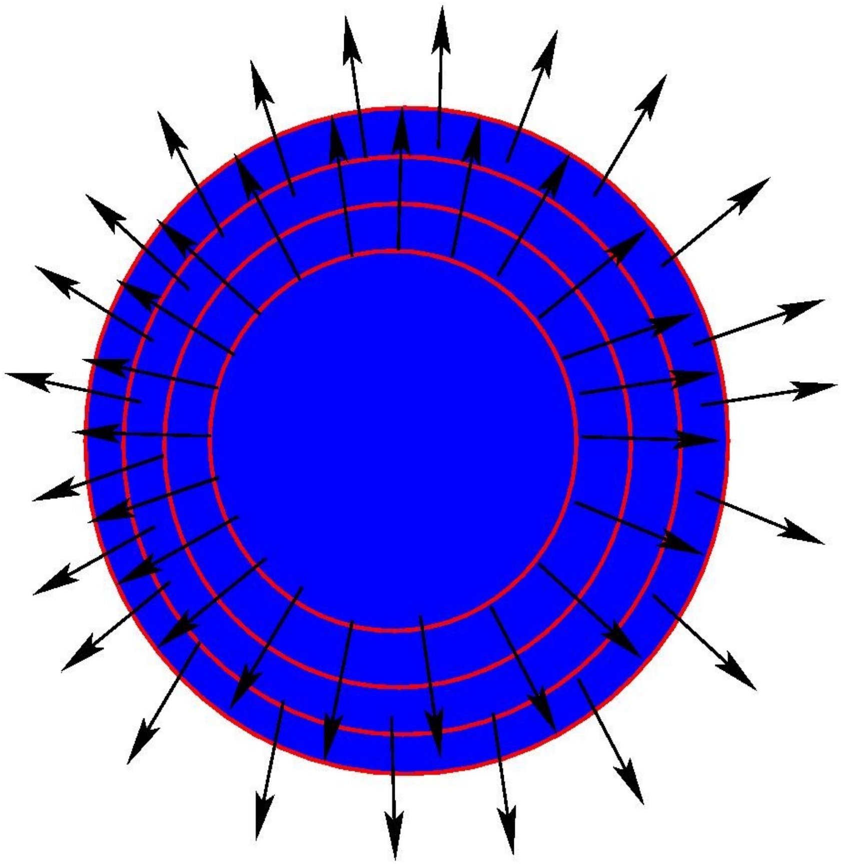
$$\rho_e \sim \rho_0 + \rho_1 \cdot \cos\theta_{vr} + \rho_2 \cdot \cos\theta_{v\varepsilon}$$

with dominating isotropic component of ρ. This is appropriate for the non-equilibrium phase of sample evolution

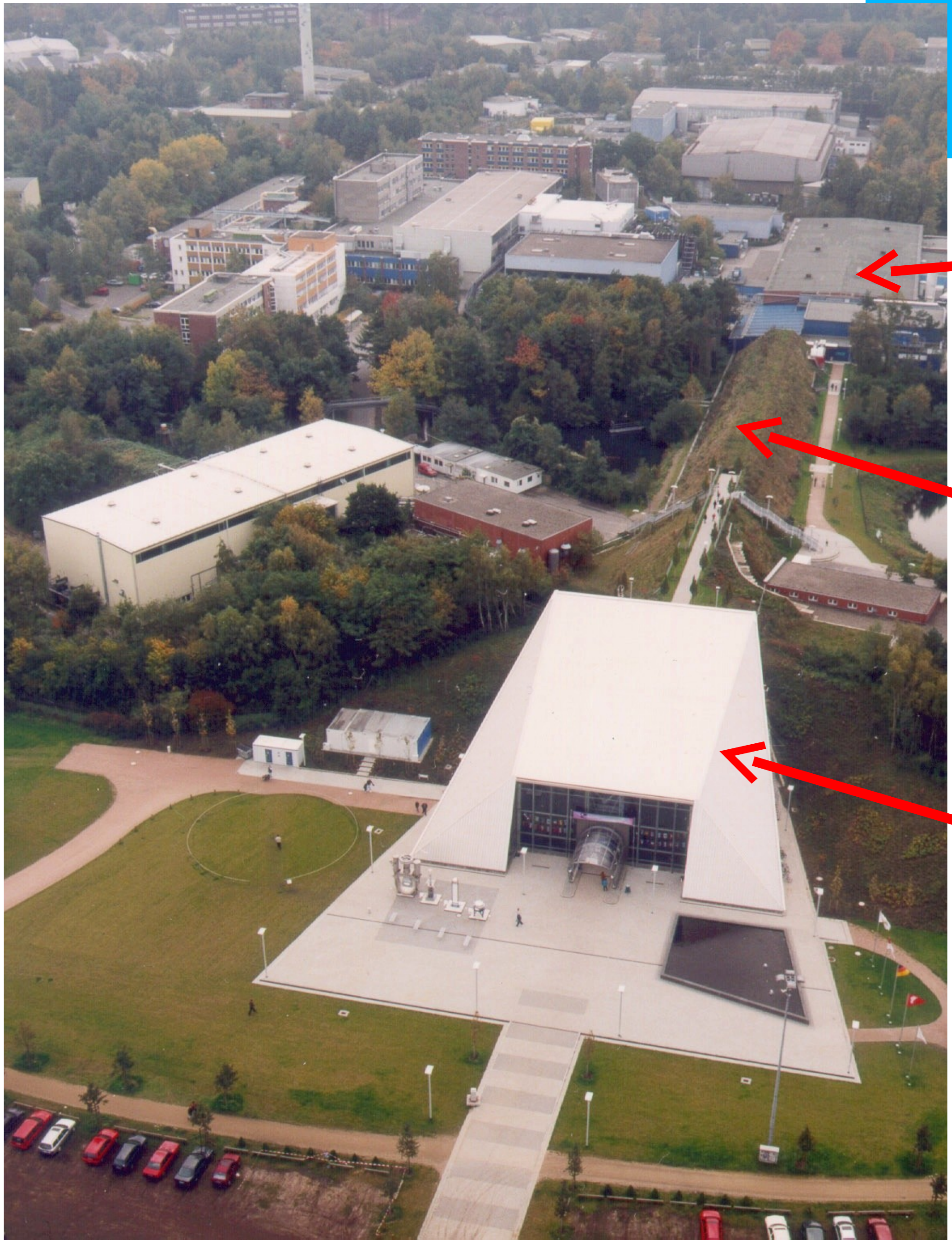
Boltzmann solver: accuracy tests

- Flow control in real space → check how many particles and how much energy escaped from the simulation box → accuracy < 5%
- Energy and number of particles conserved with a good accuracy (< 1%) in collisionless case
- Collisionless motion simulated with Boltzmann solver checked with an analytical model
- Accuracy of pseudospectral approximation checked with an independent method

Four spheres of flow control



FLASH FEL at DESY



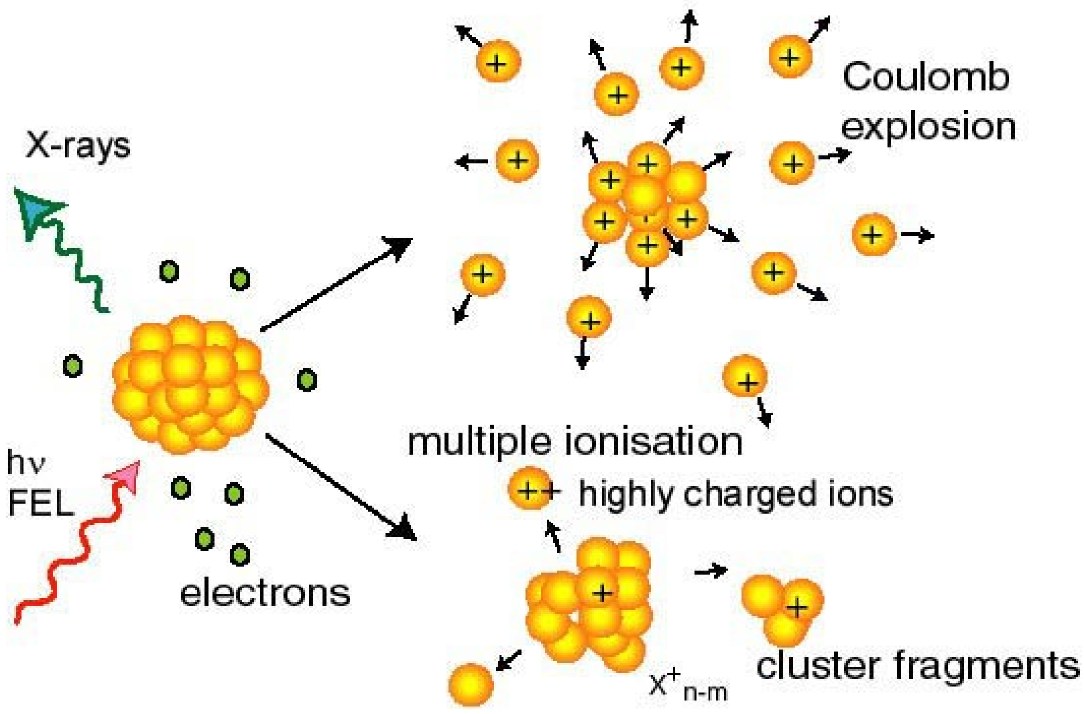
Electron gun

Linac and FEL undulator

Experimental hall (User Facility started July 2005)

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

Experimental studies on clusters irradiated with intense FEL pulses

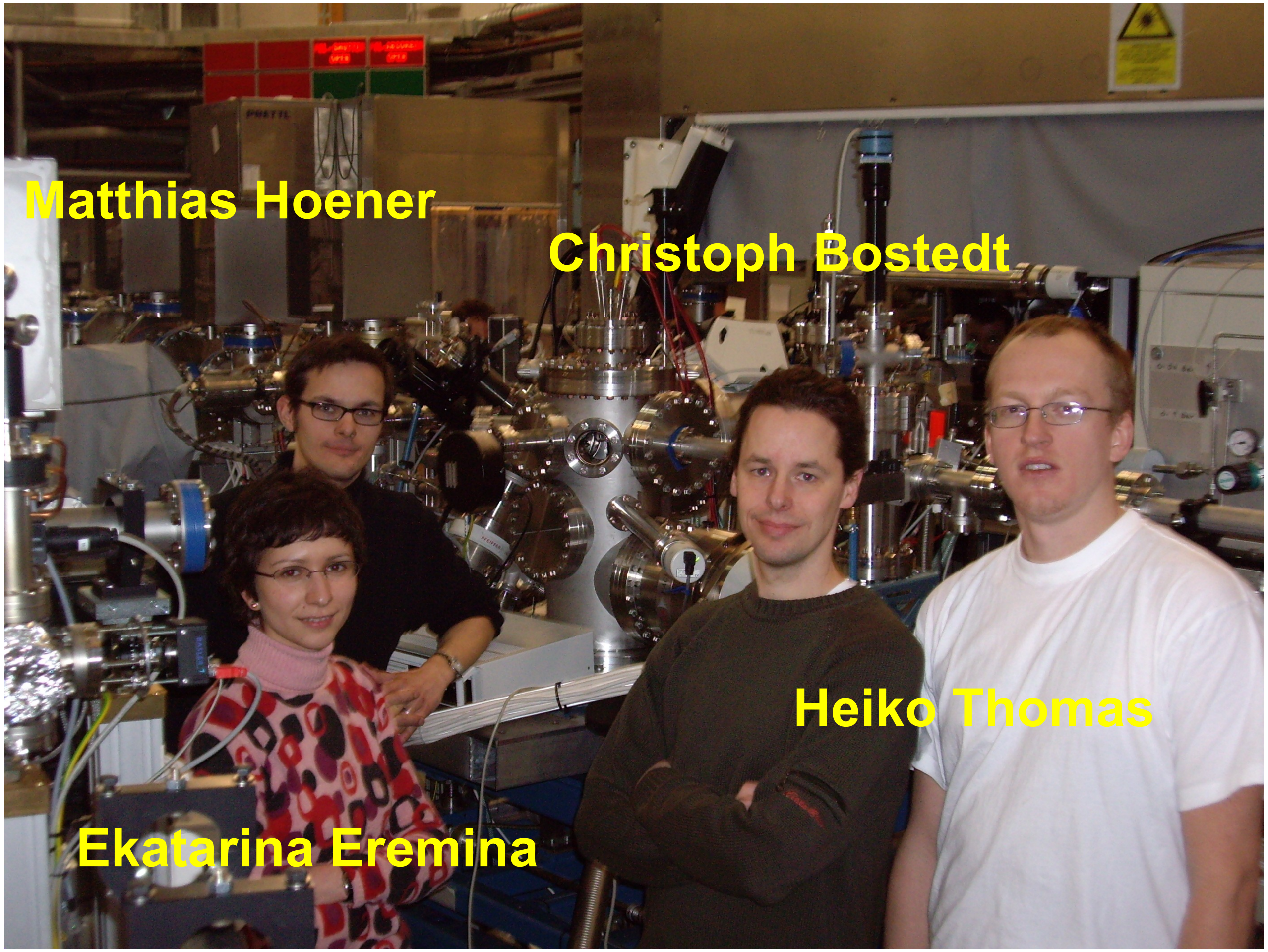


- **Mechanisms of energy absorption and ionization**
- **Non-linear / multi-photon processes observed?**
- **Timescales of electron emission and of ion motion**
- **New processes identified?**

λ= 100nm (2002)	**32 nm/13 nm (2007-2009)**	**future 6 - 0.1 nm**
valence electrons	**valence/innershell electrons**	**atomic resolution**

The experimental group

Technische Universität Berlin



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Collaboration
with the groups of J. Hajdu (Uppsala, Stanford) and H. Chapman (CFEL)
R. Hartmann, C. Reich, L. Strüder, MPG Halbleiterlabor

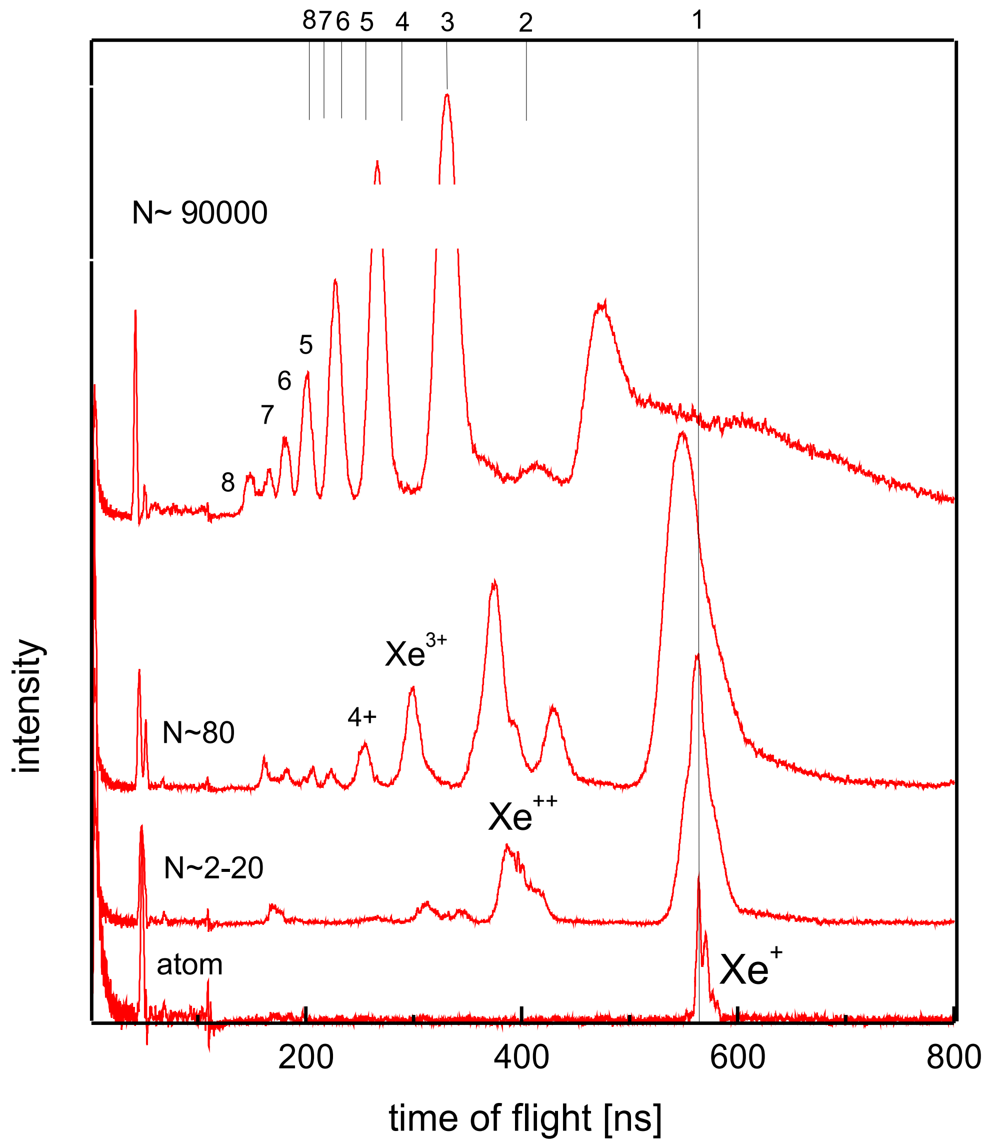
Evolution of irradiated samples

Two phases:

- non-equilibrium ionization phase: starts with the photon irradiation and lasts until thermalization of electrons and saturation of ionizations from ground state is reached

- semi-equilibrium expansion phase: electron-ion plasma in local thermal equilibrium, ions and electrons slowly escaping from outer shells → expansion of the sample

Time of flight mass spectra of Xe atoms and clusters at radiation wavelength 100 nm



10^{12}-10^{14} W/cm²

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

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

- **Multiply charged ions from clusters, keV energy**
- **Only singly charged ions from atoms**

↓↓

Dedicated theoretical study needed to explain the enhanced energy absorption

pulse duration ~ 50 fs

Theoretical models proposed

- Enhanced inverse bremsstrahlung heating of quasi-free electrons within the cluster [Santra, Greene]. Enhanced heating rate obtained with effective atomic potential. High charge states produced during collisional ionizations [R. Santra, Ch. H. Green, PRL 91, 233401 (2003)]
- High charge states within clusters are produced by single photon absorptions due to the suppression of the interatomic potential barriers within the cluster environment [Georgescu, Saalmann, Siedschlag, Rost] [C. Siedschlag, J. M. Rost , PRL 93, 43402 (2004)]
- Heating of quasi-free electrons through many-body recombination [Jungreuthmayer, Ramunno, Zanghellini, Brabec]. High charge states produced during collisional ionizations.

[C. Jungreuthmayer et al. , J. Phys. B 38, 3029 (2005)]

Theoretical modelling

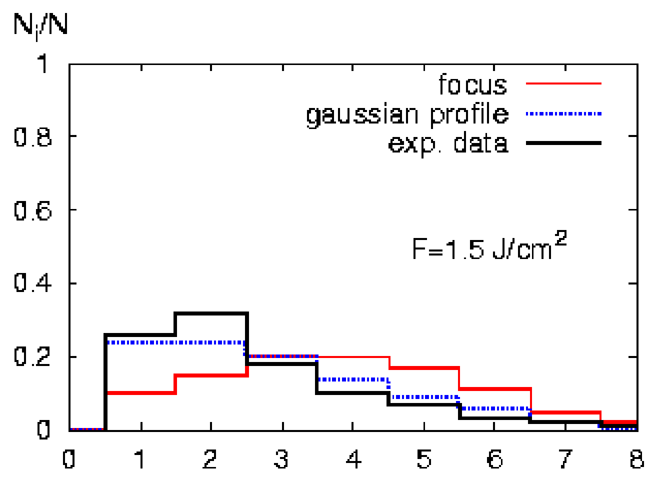
What happens if all enhancement factors are included in one model?

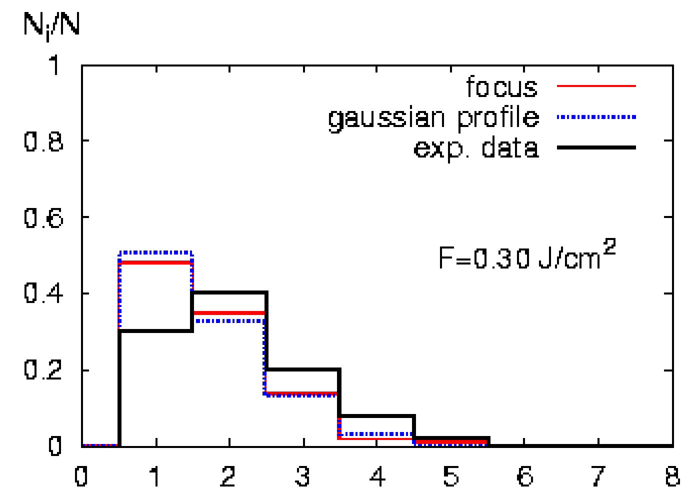
- Creation of high charges through: photoionizations → IB heating of quasi-free electrons as proposed by [Santra,Greene] → collisional ionizations / recombinations;
- Modification of atomic potentials by electron screening and ion environment tested
- Plasma regime tested → possible contribution of many-body effects (many-body recombination)

↑ ↑ ↑ ↑

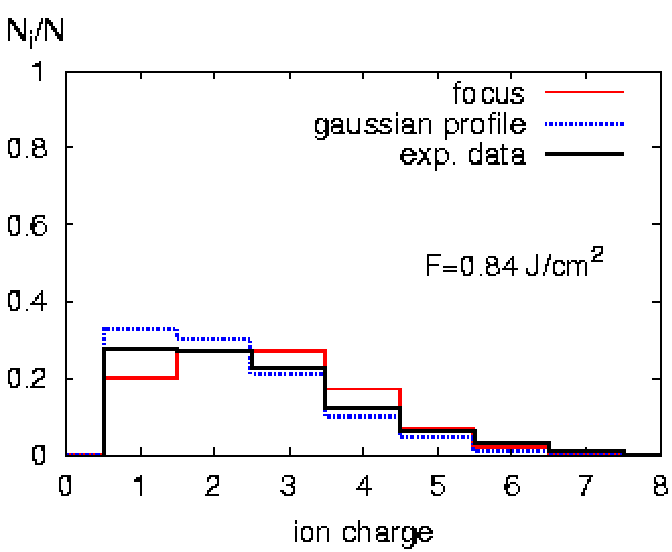
- Independent cross-check with MD simulation successful [F.Wang]

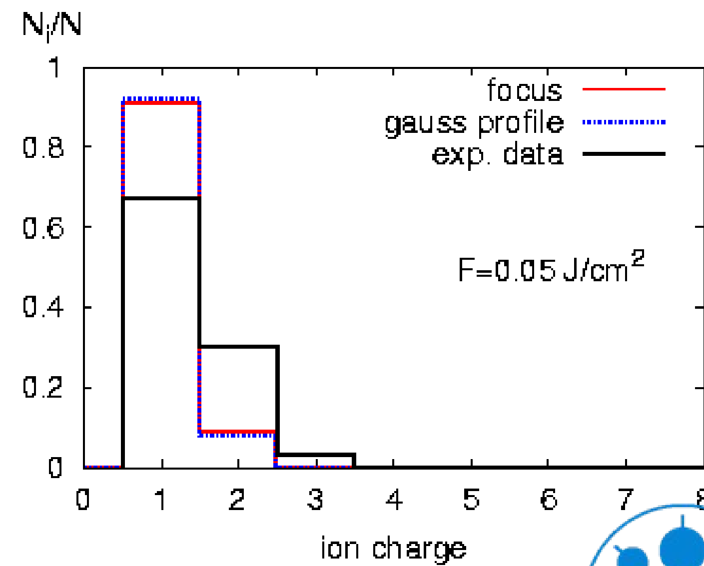
Clusters of 2500 xenon atoms irradiated with 100nm FEL pulses of various energies





Ion fractions at various pulse fluences

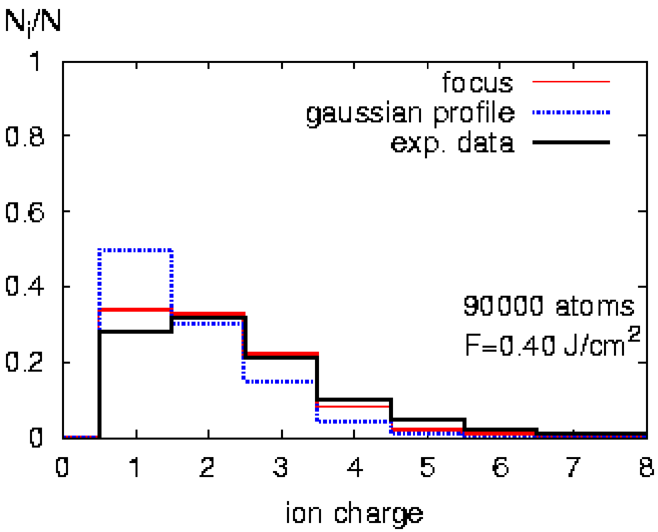


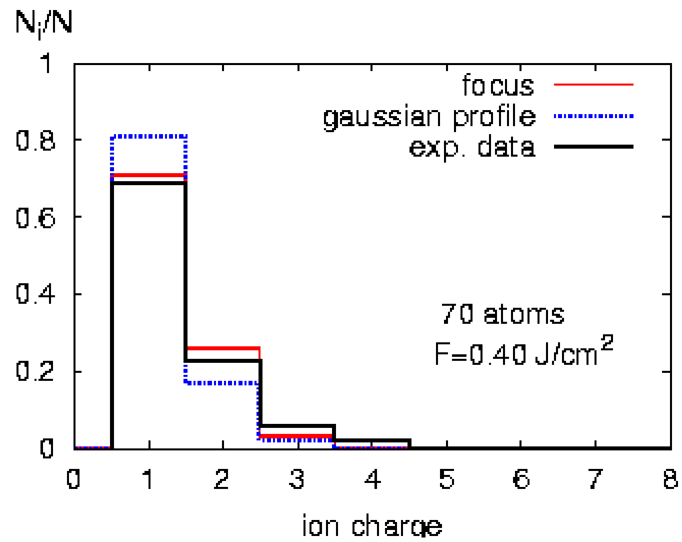


TOF detects only ions !

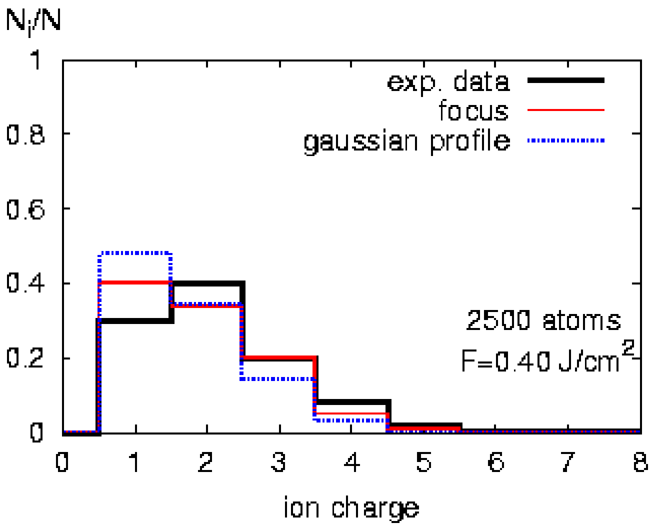
[Ziaja et al., Phys. Rev. Lett. 102 (2009) 205002]

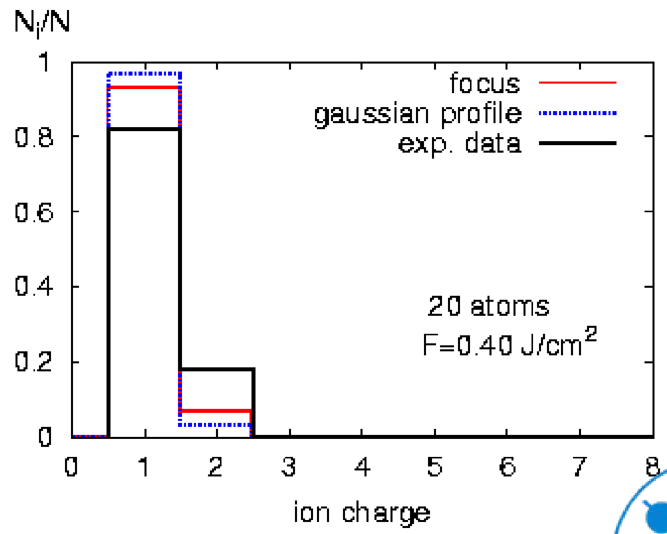
Clusters of various size irradiated with 100nm FEL pulses of a fixed flux, F=0.4 J/cm²





Ion fractions at various cluster size

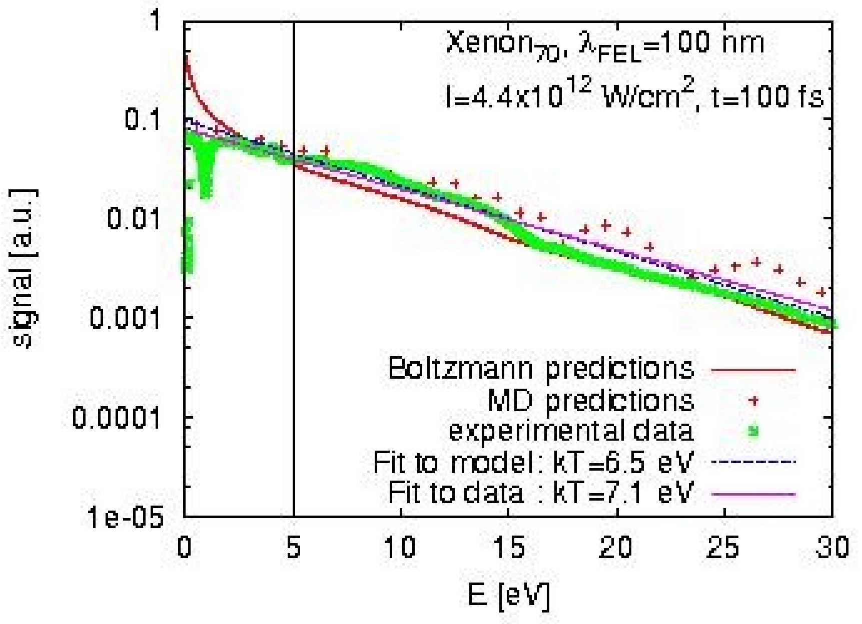


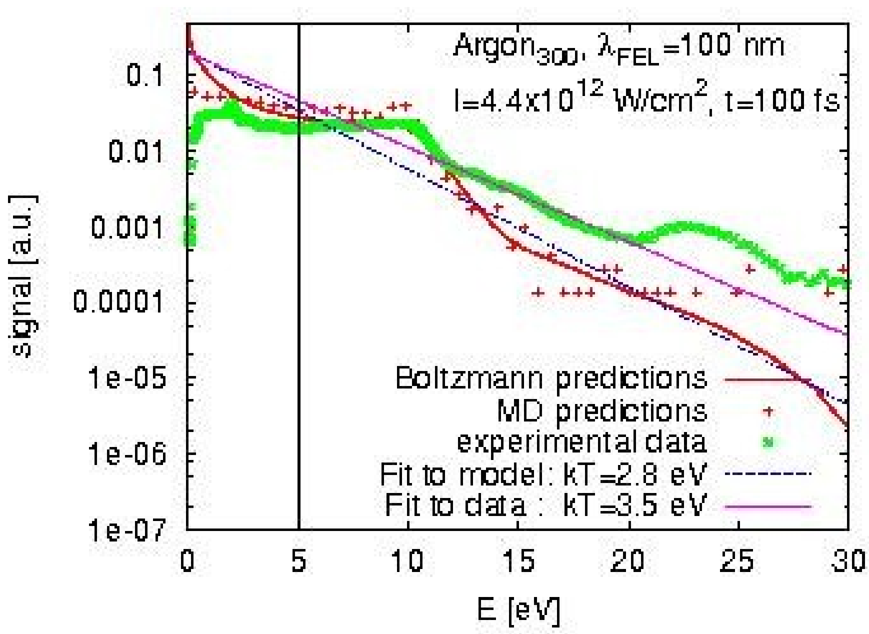


TOF detects only ions !

Electron spectroscopy at 100 nm: indication of energy absorption mechanisms

Electron emission spectra at 100 nm for Xe (70) and Ar (300):

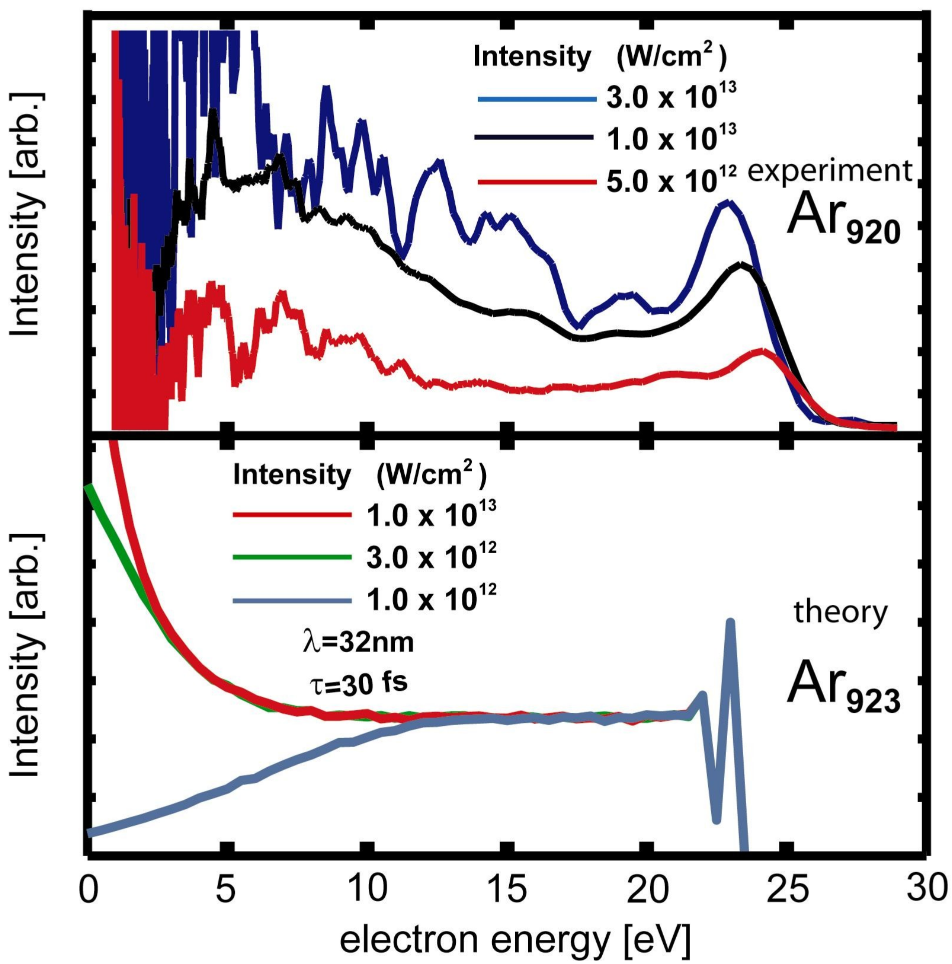


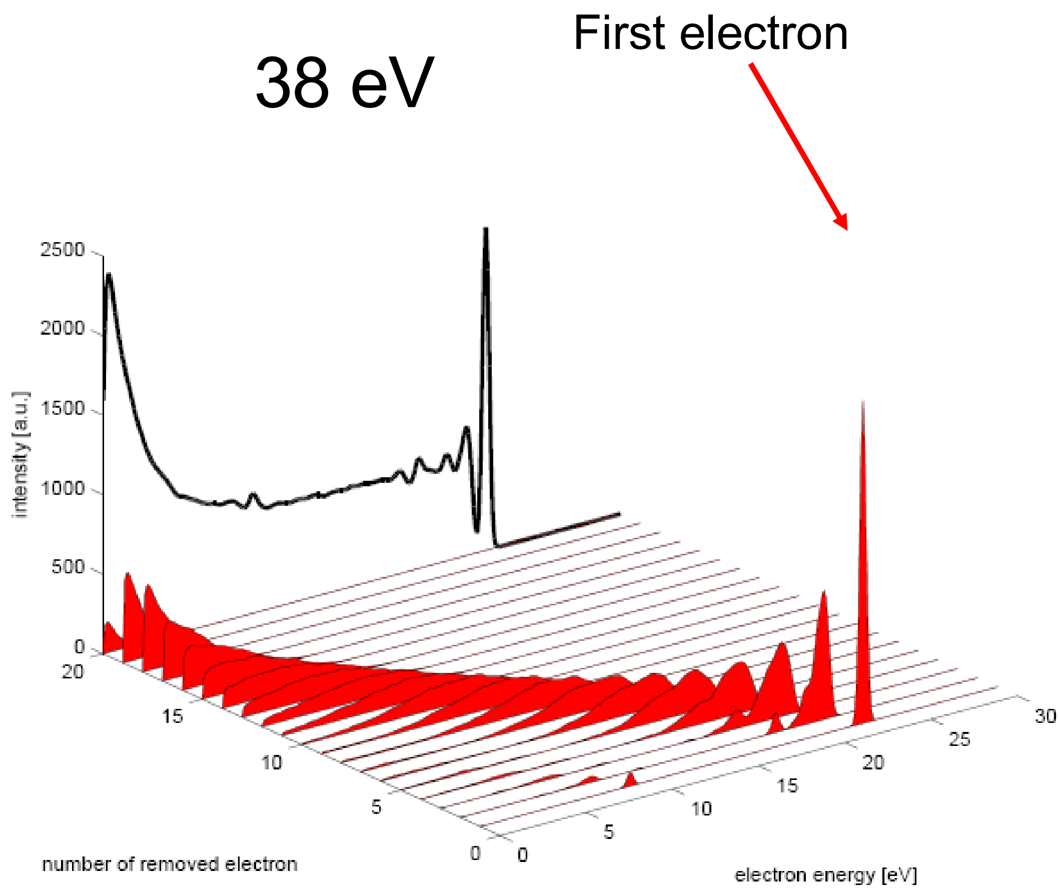


Plasma formation, intense heating of quasi-free electrons

[Ziaja et al., New J. Phys. 11 (2009) 103012]

Electron spectroscopy at 32 nm: indication of energy absorption mechanisms



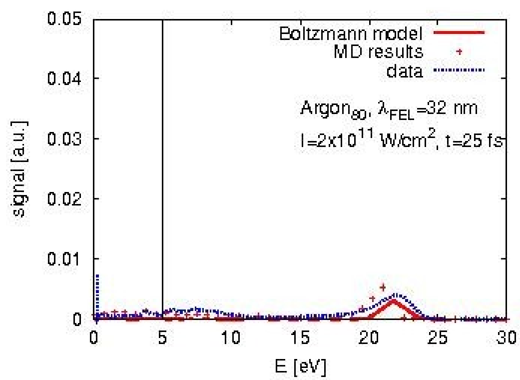


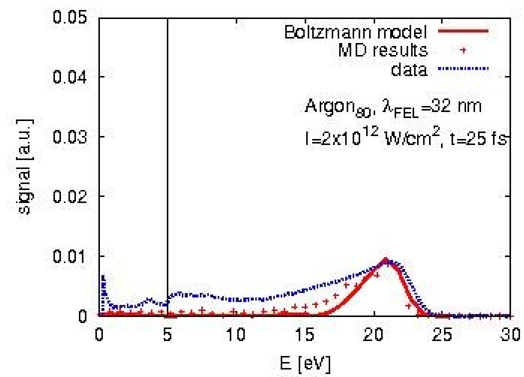
no thermionic electron emission
no plasma absorption [T. Fennel et al.]

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

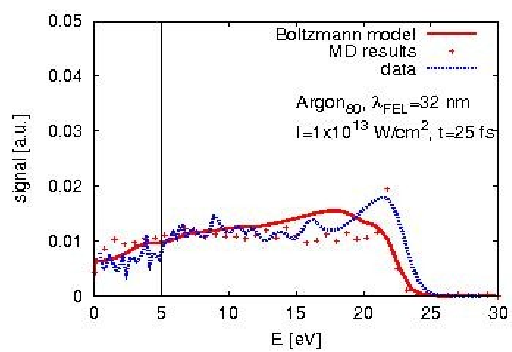
Electron spectroscopy at 32 nm
indication of energy absorption mechanisms

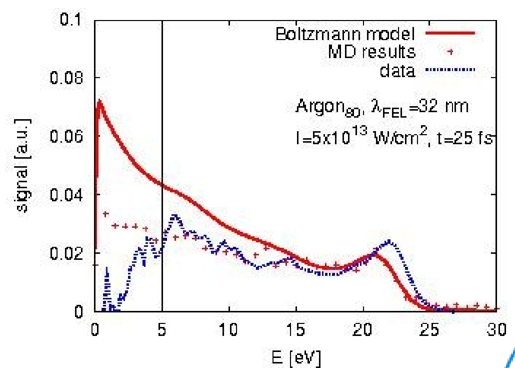
Electron emission spectra at 32 nm for Ar(80) and Ar (150):
sequential ionization
[Ziaja et al., New J. Phys. 11 (2009) 103012]





FEL pulse length=25 fs

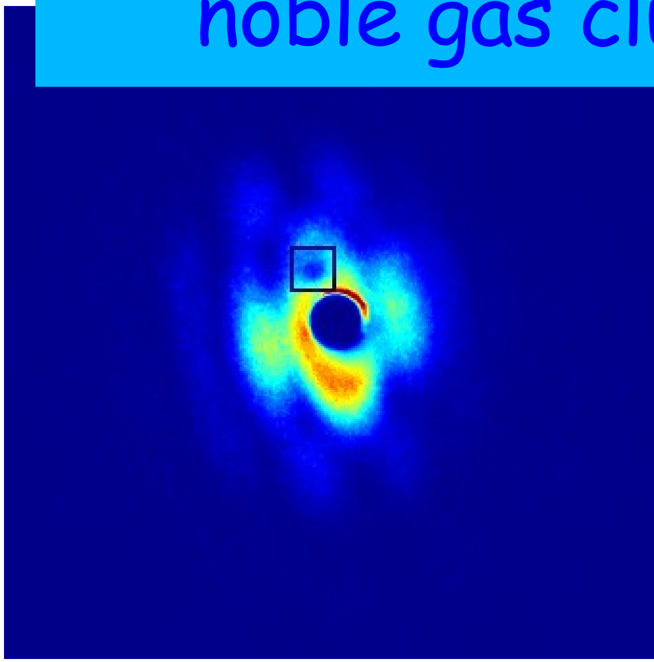




Good agreement of the results with MD simulations by the Rostock group [T. Fennel et al.] and in-house MD simulations [F. Wang]

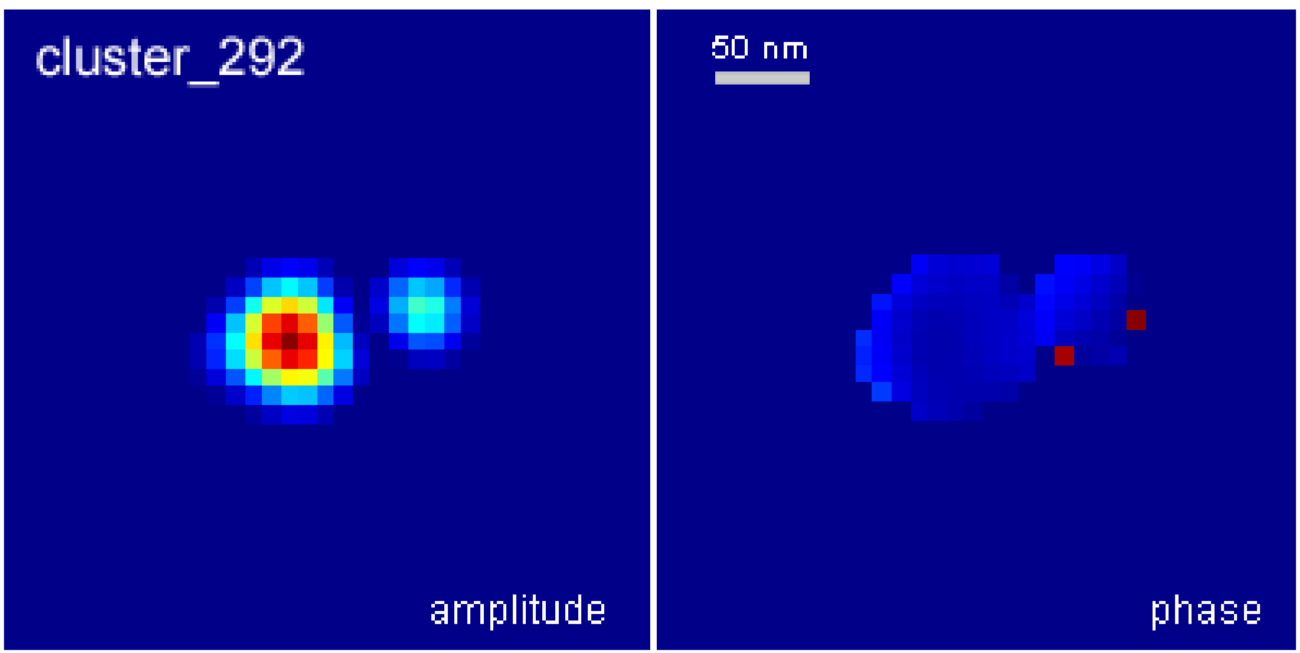
Single shot scattering and imaging of large noble gas clusters at wavelength ~ 13 nm

[C. Bostedt, H. Chapman, F. Wang and T. Möller]



scattering pattern

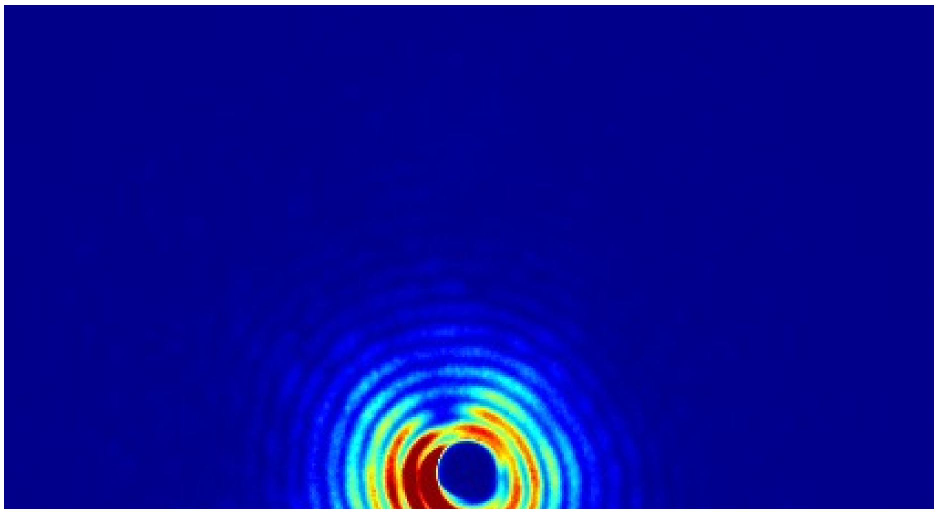
Wavelength 13.7 nm



reconstructed image

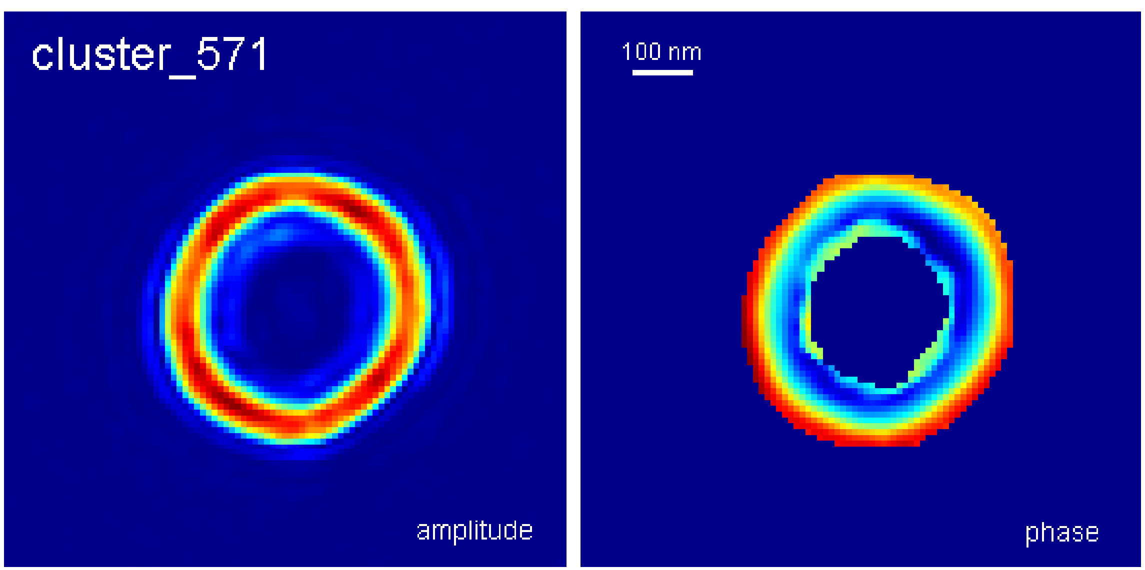
two clusters in
direct contact

Single shot scattering and imaging of large clusters: reconstruction



scattering pattern

Wavelength 13.7 nm



reconstructed image

large cluster

penetration depth of light

Soft X-Ray Thomson Scattering in Warm Dense Hydrogen

R.R. Fäustlin, S. Toleikis et al.

[Phys. Rev. Lett. 104 (2010), 125002]

Collaboration

- **DESY, Hamburg**
  S. Düsterer, R.R. Fäustlin, T. Laarmann, H. Redlin,
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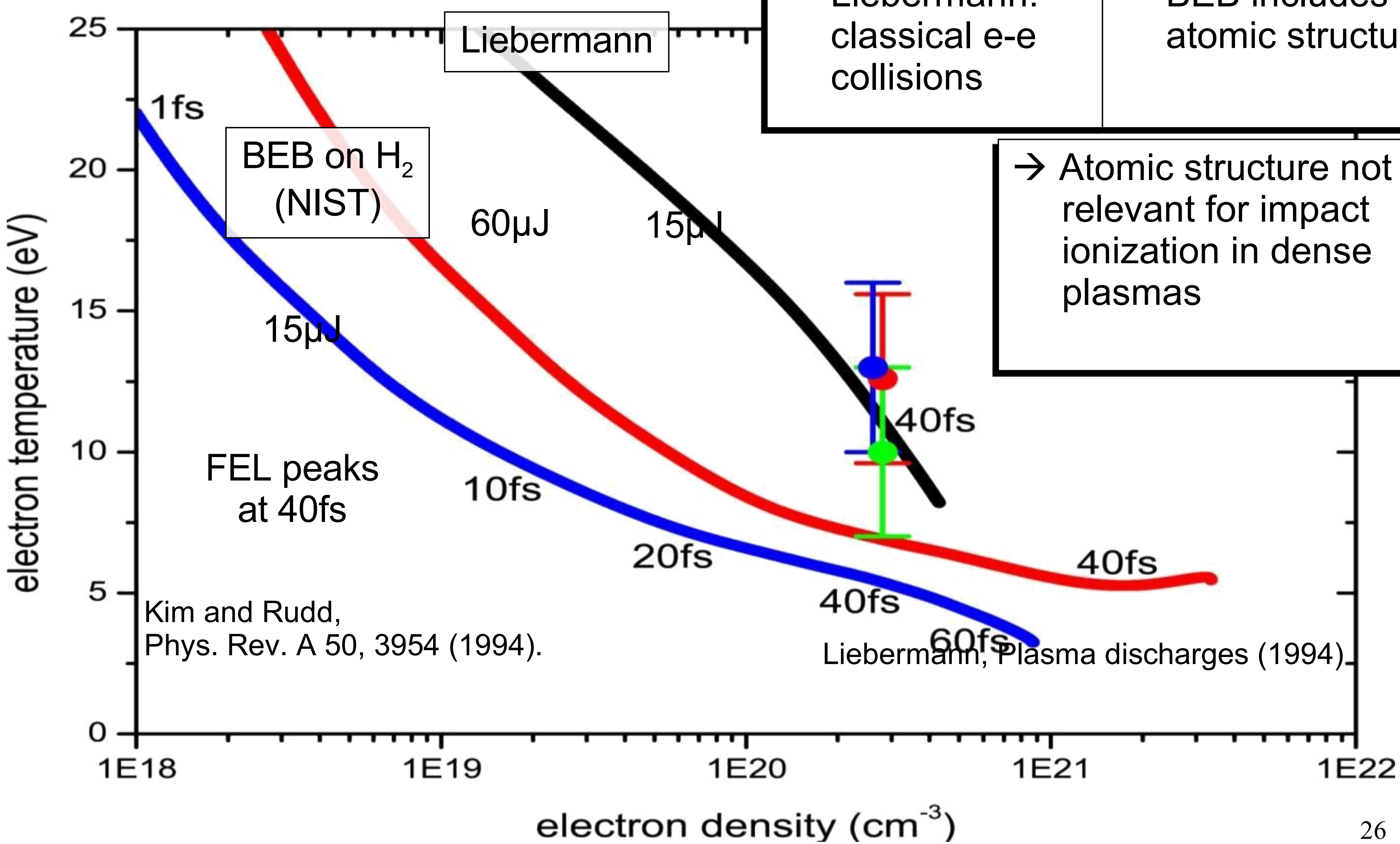
- **University of California, Berkeley**
  H.J. Lee
- **University of Jena**
  E. Förster, I. Uschmann, U. Zastrau
- **LLNL, Livermore**
  T. Döppner, S.H. Glenzer
- **University of Oxford / RAL, Chilton, Didcot**
  G. Gregori, B. Li, J. Mithen, J. Wark
- **University of Rostock**
  T. Bornath, C. Fortmann, S. Göde, R. Irsig,
  K.-H. Meiwes-Broer, A. Przystawik, R. Redmer,
  H. Reinholz, G. Röpke, R. Thiele, J. Tiggesbäumker

Helmholz Association: VH-VI-104; **Science and Technology Facilities Council UK**;
German Research Foundation: GRK 1355, SFB 652, LA 1431/2-1; **DOE**: DE-AC52-07NA27344,
LDRDs 08-ERI-002, 08-LW-004; **German Ministry for Education and Research**: FSP 301-FLASH;
European Community: RII3-CT-2004-506008 (IA-SFS)

We Validate Impact Ionization Models

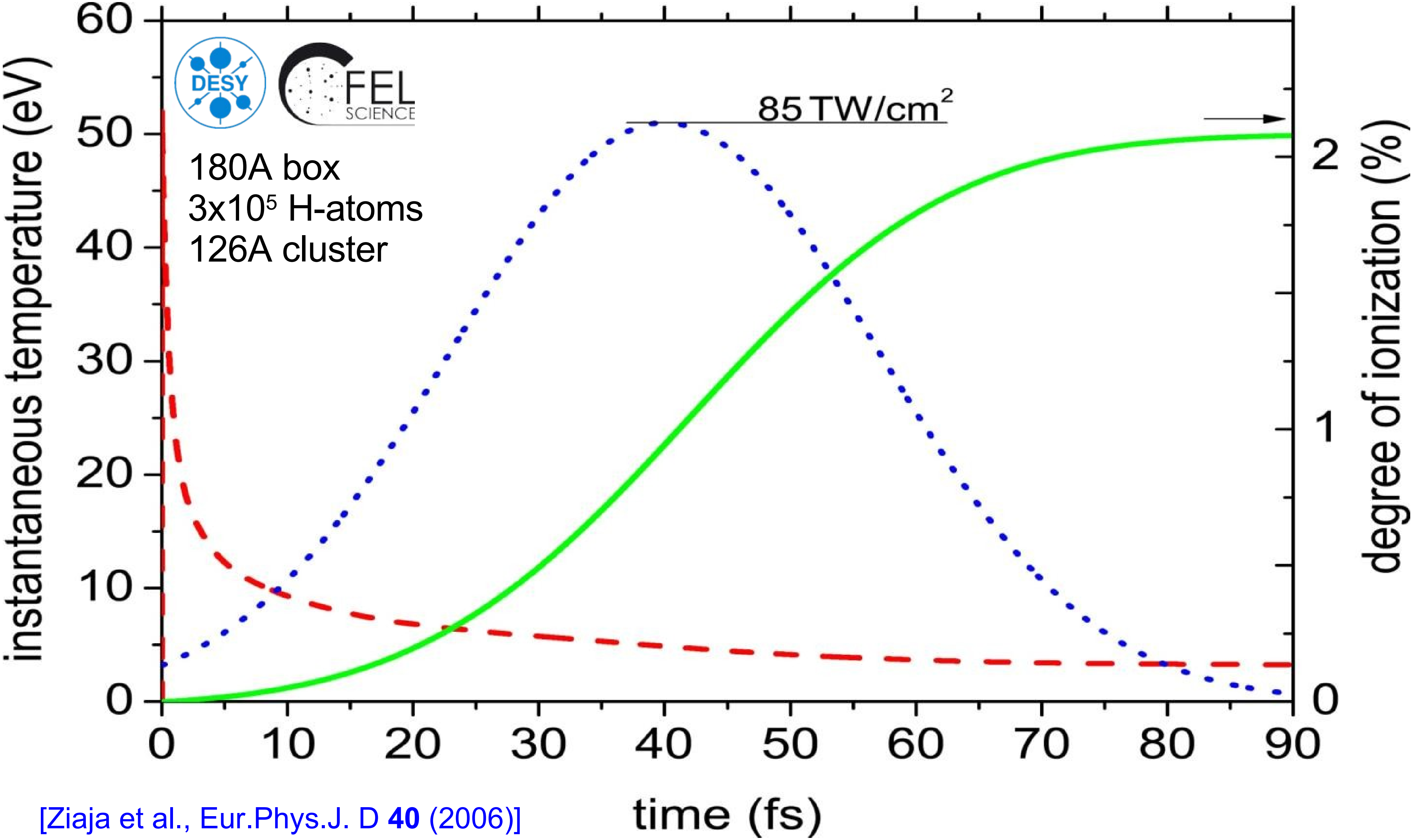
- Liebermann: classical e-e collisions
- BEB includes atomic structure

→ Atomic structure not relevant for impact ionization in dense plasmas



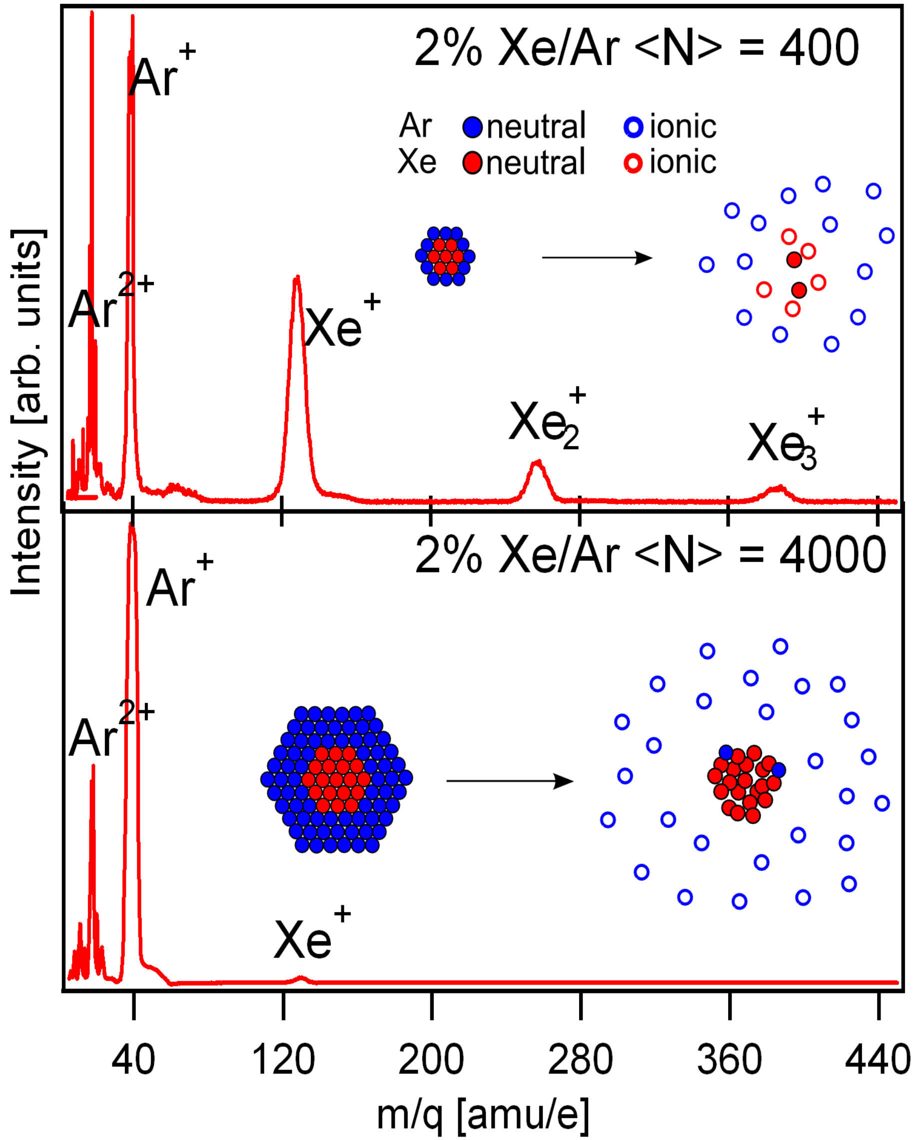
Simulations performed for clusters consisting of 0.5 million atoms

STS Probes fs Electron Thermalization



[Ziaja et al., Eur.Phys.J. D **40** (2006)]

Perspectives for imaging: delayed ionization and expansion through tampering.



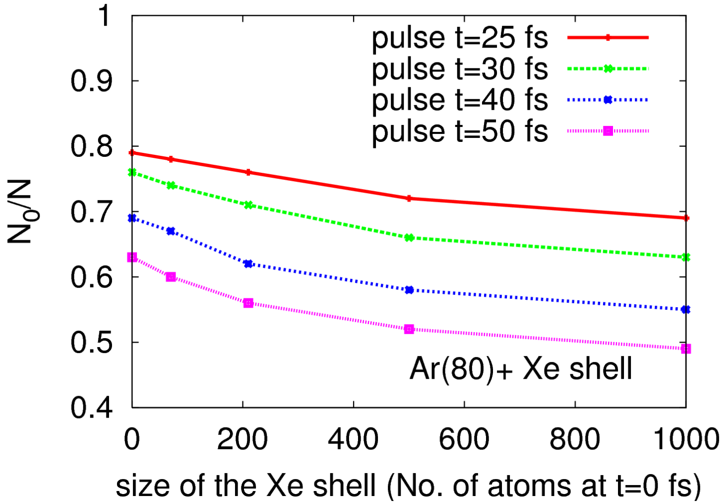
Experiment:
13 nm/ 92 eV, 10^{14} W/cm^2

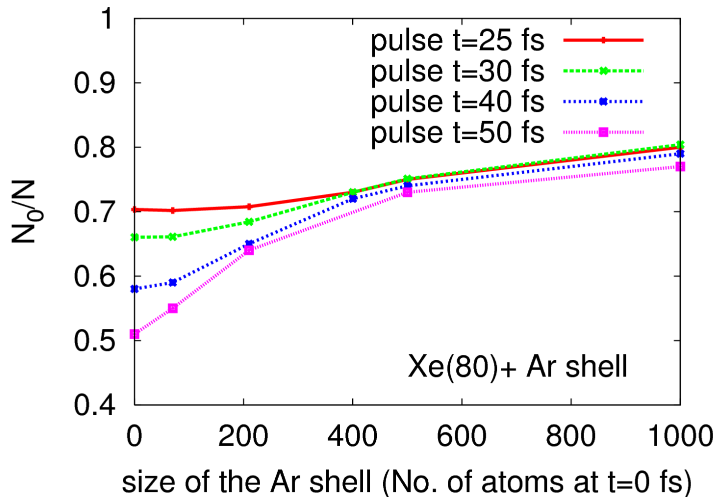
- **singly charged Ar ions from the surface**
- **strong size effect**
- **Xe plasma in the interior recombines, neutral atoms?**
- **delayed cluster expansion**

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

DESY

Perspectives for imaging: increased/decreased ionization in mixed Xe/Ar clusters at 32 nm





32 nm/ 40 eV, 10^{13} W/cm^2

- **Increased core ionization for Ar/Xe**
- **Decreased core ionization for Xe/Ar**

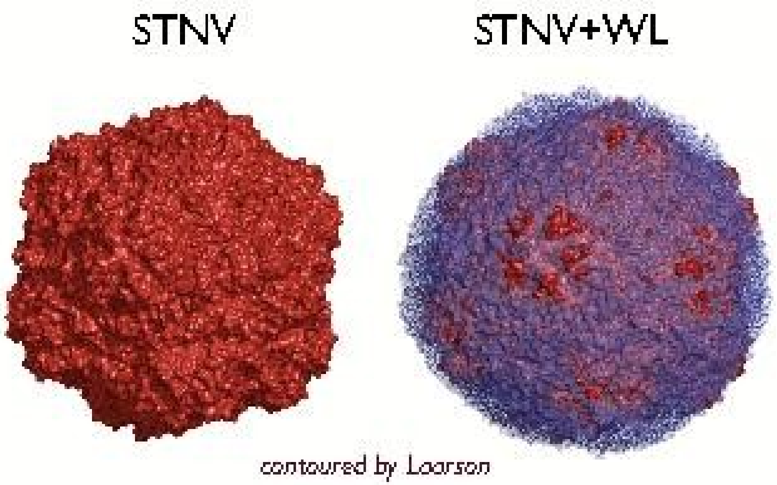
PRELIMINARY!

DESY

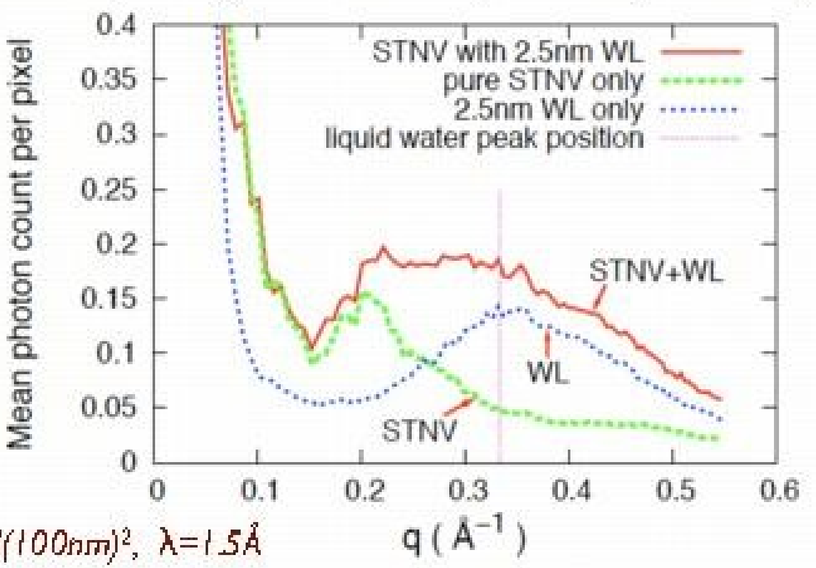
Perspectives for imaging: loss of structural and orientational information due to the water layer around the imaged object

Satellite Tobacco Necrosis virus (STNV, PDB ID: 2BUK)
object size ~17 nm, ~0.18M atoms, icosahedral symmetry.

* dynamic simulations with GROMACS (Larrson & van der Spoel)
    no radiation damage considered
    generate random water layers
* diffractive simulations with obtained atom coordinates (use Moltrans code)



q-averaged diffraction patterns (no noise)



flux=10^{12} ph/(100nm)^2, λ=1.5Å

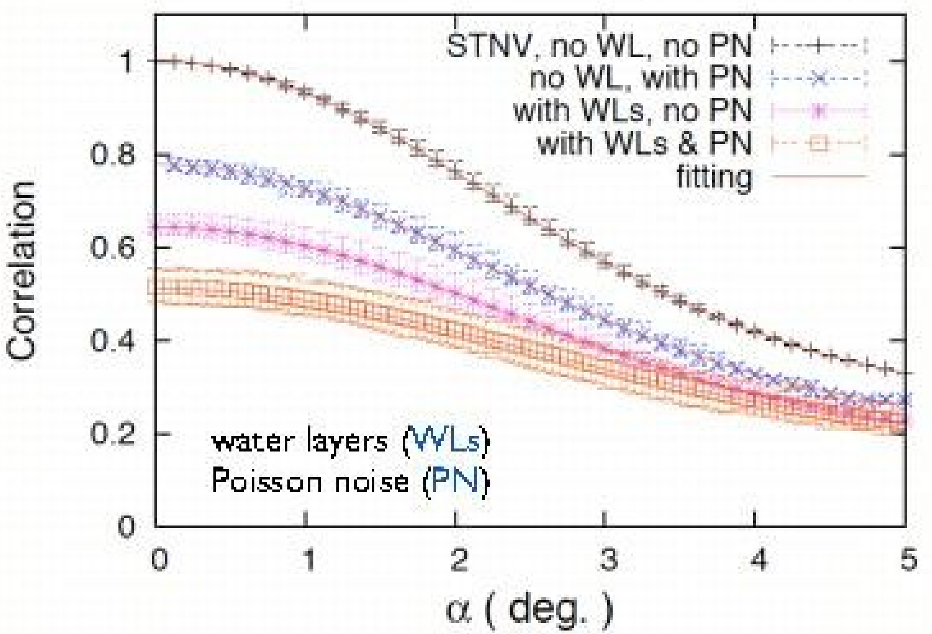
Perspectives for imaging: loss of structural and orientational information due to the water layer around the imaged object

Correlation analysis

examples of correlation curves (given q, between different views)

$$G(\alpha, q) \equiv \frac{\tilde{I}(\alpha, q) \cdot \tilde{I}(0, q)}{\sqrt{\langle \tilde{I}(\alpha, q)^2 \rangle_q} \cdot \sqrt{\langle \tilde{I}(0, q)^2 \rangle_q}}$$

correlations in given q range: $[0.069, 0.104]\ \text{Å}^{-1}$



* Suppression of correlation
* value fluctuation
* averaged curve perfectly fitted by Gaussian-like function:

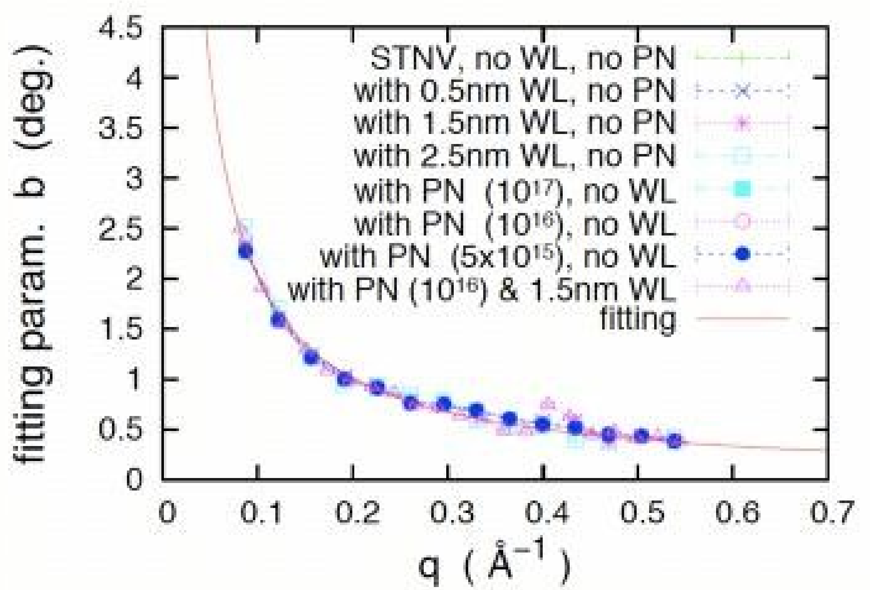
$$G(\alpha, q) = a(q)\, e^{-\alpha^2/2b(q)^2} + c(q)$$

Perspectives for imaging: loss of structural and orientational information due to the water layer around the imaged object

Fitting results of correlation curves (all q ranges)

$$G(\alpha, q) = a(q)\, e^{-\alpha^2/2b(q)^2} + c(q)$$

fitting parameter b(q) & its fitting



* Orientation information remained
* Fitting q-dependence by

$$b(q) = (4\, R_0\, q)^{-1}$$

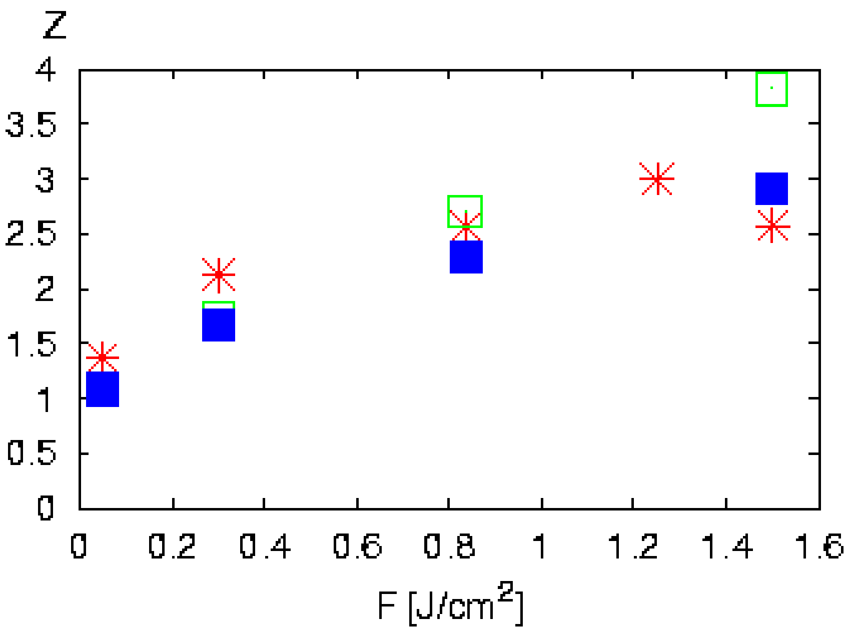
(half-Shannon angle)

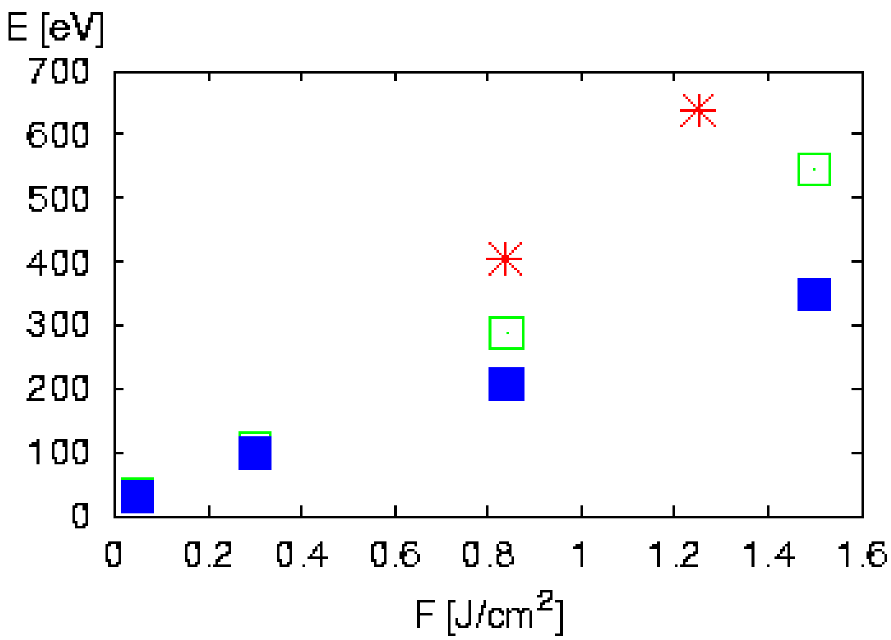
fitting value $R_0 \sim 71$ Å

Summary and outlook

- We constructed a useful tool for studying the evolution of FEL irradiated samples (computationally efficient treatment of large samples)
- Our model is so far the only one that gives an accurate description of all of the experimental data collected from atomic clusters at 100 nm and 32 nm wavelength
- Good agreement with data from warm dense matter hydrogen experiment
- Several problems can be studied in the next future:
- evolution of clusters irradiated with XUV and X-rays
- clusters of various structures
- mechanisms of slowing down the cluster explosion
- samples exposed to ultra short and/or ultra intense pulses

Clusters of 2500 xenon atoms irradiated with 100nm FEL pulses of various energies

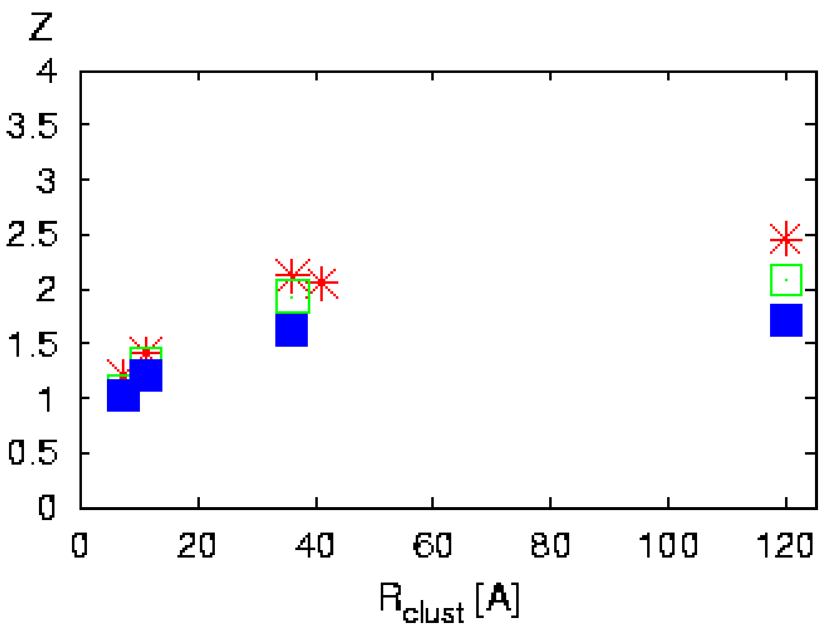


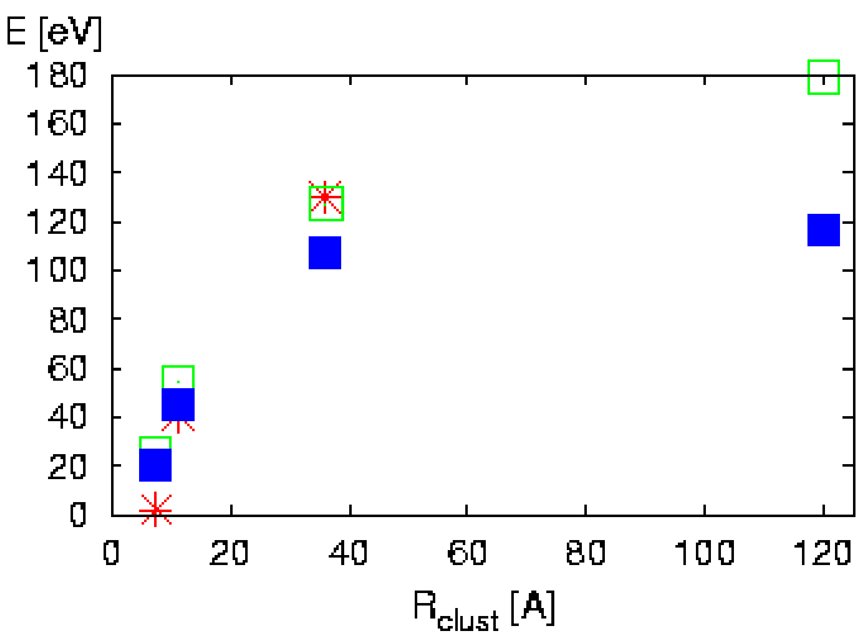


Results from model □ (in focus) Experimental data *

□ (averaged over spatial pulse profile)

Clusters of various size irradiated with 100 nm FEL pulses of a fixed flux, F=0.4 J/cm^2





Results from model □ (in focus) Experimental data *

□ (averaged over spatial pulse profile)