Dynamics of matter induced by high intensity X-rays

23.5.2017 – LII Zakopane School of Physics

Zoltan Jurek, Sang-Kil Son, Beata Ziaja, Robin Santra

Theory Division,

Center for Free-Electron Laser Science, DESY

Deutsches Elektronen-Synchrotron (DESY), Hamburg

CFEL–DESY Theory Division

- **Theoretical** and **computational** tools are developed to predict the behavior of **matter exposed to intense electromagnetic radiation**.
- **Quantum-mechanical** and **classical techniques** are employed to study **ultrafast** processes happen on *ps* (10^{-12} s) down to *as* (10^{-18} s) timescales.
- **Research interest**: Dynamics of excited many-electron systems; Motion of atoms during chemical reactions; X-ray radiation damage in matter; ...

Members of the CFEL-DESY Theory Division: C. Arnold, S. Bazzi, Y.-J. Chen, O. Geffert, D. Gorelova, L. Inhester, A. Hanna, Z. Jurek, A. Karamatskou, M. Krishna, Z. Li, V. Lipp, M. A. Malik, P. K. Mishra, **R. Santra (Division Director)**, V. Saxena, S.-K. Son, V. Tkachenko, K. Toyota, R.Welsch, B. Ziaja

Subgroups:

'Ab-initio X-ray Physics' (S.-K. Son)
'Chemical Dynamics' (R.Welsch)
'Modeling of Complex Systems' (B. Ziaja)

Outline

- Introduction

  Elements of x-ray – matter interaction

- Complex dynamics of matter induced by high intensity X-rays

  via experiments and simulations

  – Sequential single photon absorption
  – Non-equilibrium dynamics
  – Coulomb explosion
  – Nanoplasma formation
  – Radiation damage

Dynamics of matter induced by high intensity X-rays

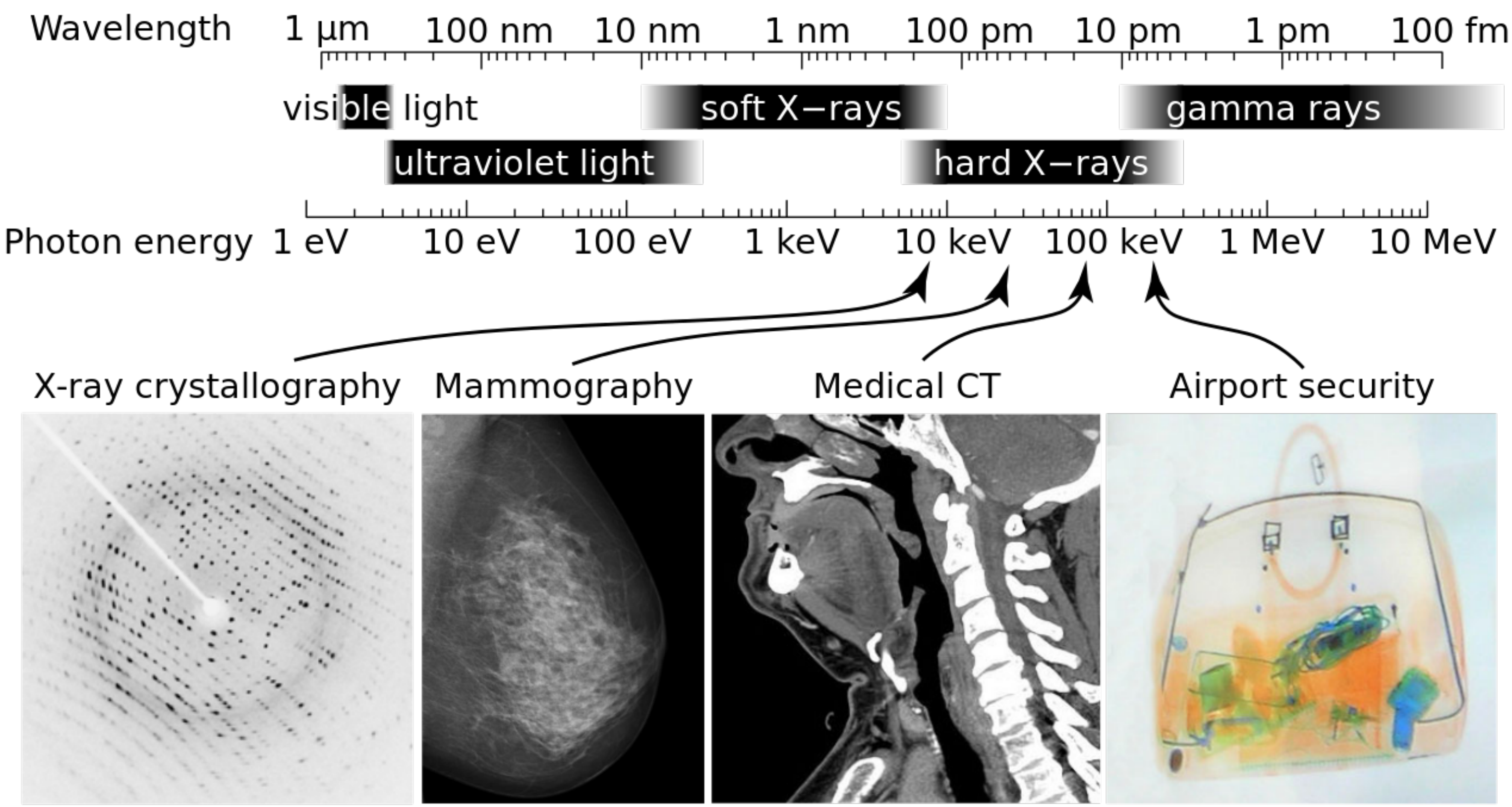
Dynamics of matter induced by high intensity X-rays

X-rays

- Electromagnetic radiation

Photon Energy: 100 eV – 100 keV

Wavelength: 0.01 nm – 10 nm



https://en.wikipedia.org/wiki/X-ray

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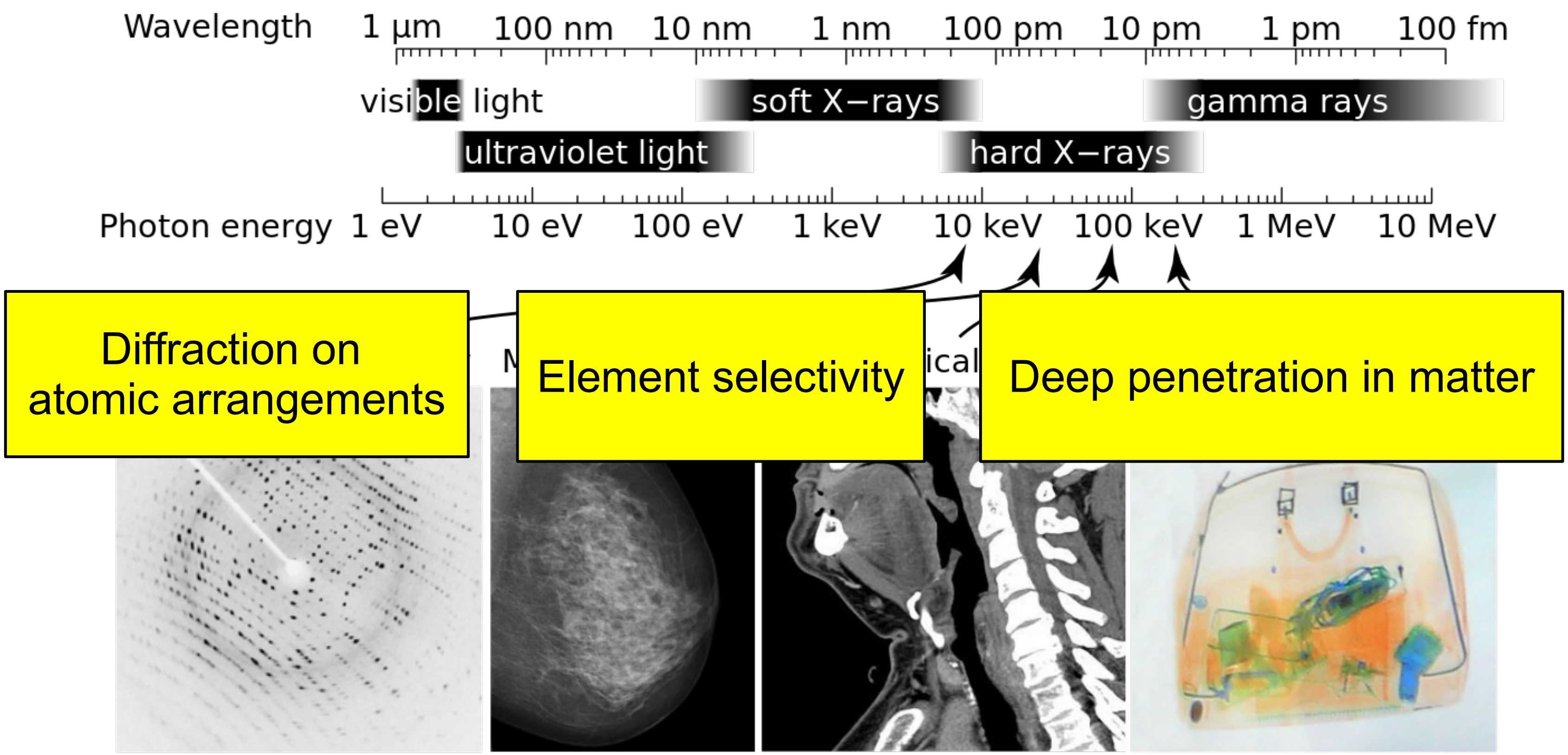

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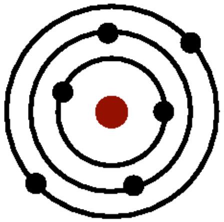
X-ray – Matter interaction

- Photon energy: 100 eV – 100 keV $\ll$ $m_e c^2$ $\rightarrow$ no relevance in particle physics

  $\geq$ electron binding energies in the atoms



Predominant **interaction with atomic electrons**

> **Hamiltonian** in QED formalism

Minimal coupling, Coulomb gauge

$$\hat{H} = \hat{H}_{\mathrm{mol}} + \hat{H}_{\mathrm{EM}} + \hat{H}_{\mathrm{int}}$$

$$\hat{H}_{\mathrm{int}} = \alpha \int \mathrm{d}^3x\, \hat{\psi}^\dagger(\boldsymbol{x}) \left[\hat{\boldsymbol{A}}(\boldsymbol{x}) \cdot \frac{\boldsymbol{\nabla}}{\mathrm{i}} \right] \hat{\psi}(\boldsymbol{x}) + \frac{\alpha^2}{2} \int \mathrm{d}^3x\, \hat{\psi}^\dagger(\boldsymbol{x}) \hat{A}^2(\boldsymbol{x}) \hat{\psi}(\boldsymbol{x}),$$

$$\hat{H}_{\mathrm{EM}} = \sum_{\boldsymbol{k},\lambda} \omega_k \hat{a}^\dagger_{\boldsymbol{k},\lambda} \hat{a}_{\boldsymbol{k},\lambda}$$

$$\hat{H}_{\mathrm{el}} = \int \mathrm{d}^3x\, \hat{\psi}^\dagger(\boldsymbol{x}) \left\{ -\frac{1}{2}\nabla^2 - \sum_n \frac{Z_n}{|\boldsymbol{x} - \boldsymbol{R}_n|} \right\} \hat{\psi}(\boldsymbol{x}) + \frac{1}{2} \int \mathrm{d}^3x \int \mathrm{d}^3x'\, \hat{\psi}^\dagger(\boldsymbol{x}) \hat{\psi}^\dagger(\boldsymbol{x}') \frac{1}{|\boldsymbol{x} - \boldsymbol{x}'|} \hat{\psi}(\boldsymbol{x}') \hat{\psi}(\boldsymbol{x})$$

R. Santra *J. Phys. B* **42** 023001 (2009)

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X-ray – Matter interaction: Theory

- Interaction: perturbation

$$\hat{H} = \underbrace{\hat{H}_{\mathrm{mol}} + \hat{H}_{\mathrm{EM}}}_{\hat{H}_0} + \hat{H}_{\mathrm{int}}$$

Perturbation

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$$\begin{aligned} w_{FI} &= \frac{|S_{FI}|^2}{T} \\ &= 2\pi\delta(E_F - E_I) \\ &\times \left| \langle F|\hat{H}_{\mathrm{int}}|I\rangle + \sum_M \frac{\langle F|\hat{H}_{\mathrm{int}}|M\rangle\langle M|\hat{H}_{\mathrm{int}}|I\rangle}{E_I - E_M + \mathrm{i}\epsilon} + \cdots \right|^2 \end{aligned}$$

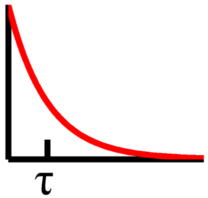
R. Santra *J. Phys. B* **42** 023001 (2009)

X-ray – Matter interaction

- Parameters describing and quantifying processes

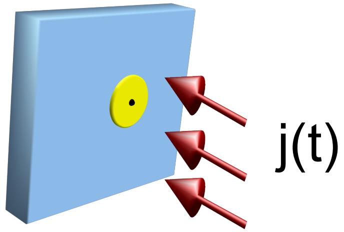
  – **rate** (w) → **lifetime**: $\tau = 1 / w$

  → **probability** $= 1 - \exp\left(- \int_{-\infty}^{T} w(t)\, dt \right)$



  – **cross section** (σ) → $w(t) = \sigma \cdot j(t)$

Flux of
probe particles



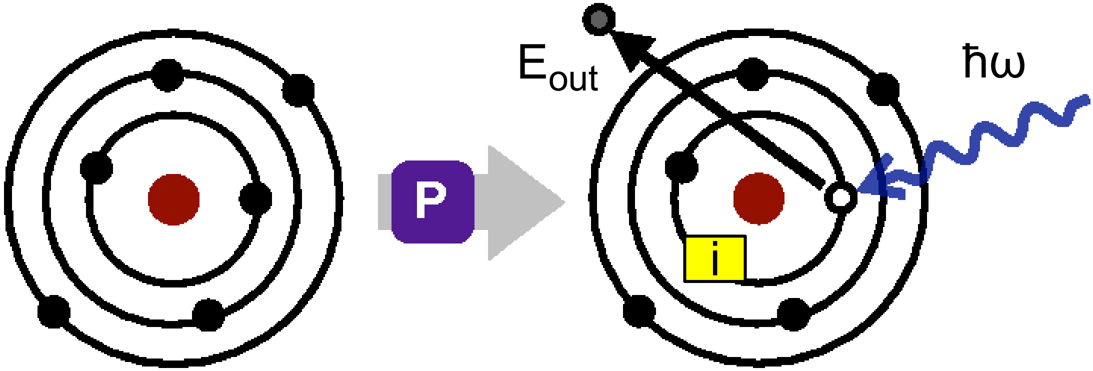
X-ray – atom predominant interaction: Photoionization

- **Photoionization** (photoabsorption)

$$E_{out} = \hbar\omega - (E^{(f)}_{atom} - E^{(i)}_{atom})$$

$$\sim \hbar\omega - B_i$$

Binding energy: $B_i > 0$



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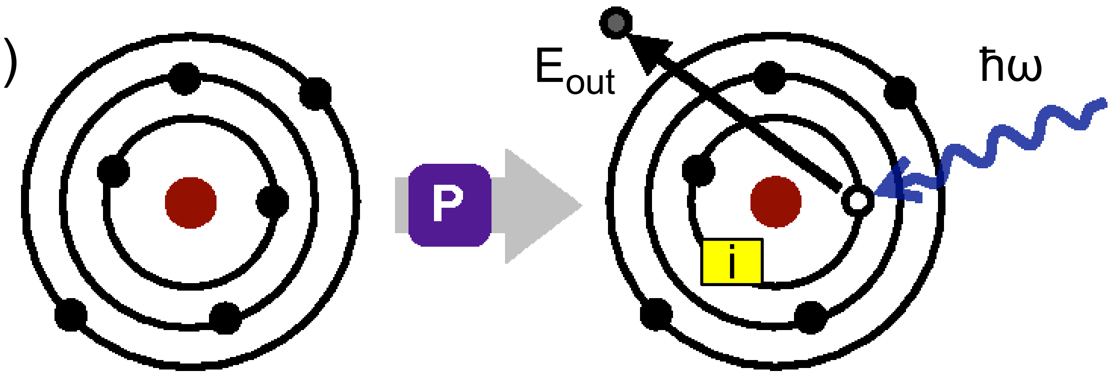

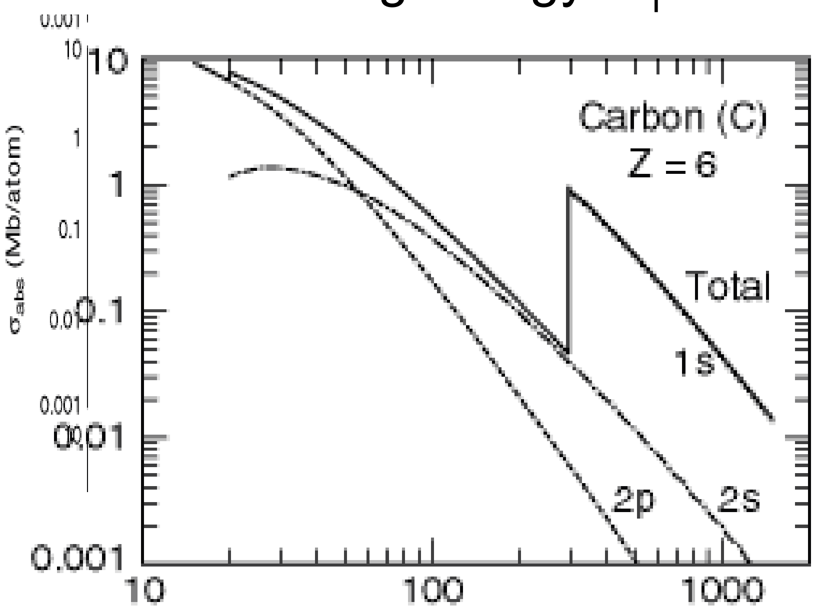

http://xdb.lbl.gov/

- **Element selectivity**
  element dependent absorption
  e.g. H-like ion: $\sigma \sim Z^5$
- **Core electrons first** $B_i < \hbar\omega$
  subshell dependent cross sections
- **Deep penetration**
  small cross sections
  $\rightarrow$ long mean free path $\lambda=1/(\rho\sigma)$

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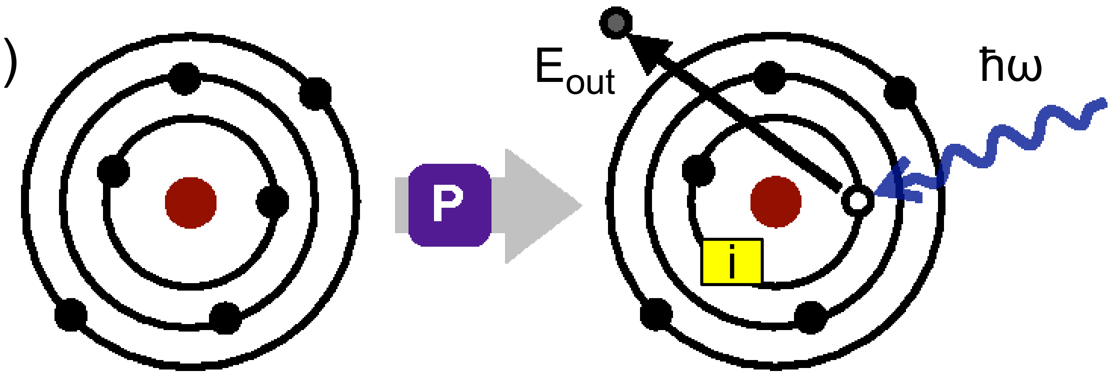

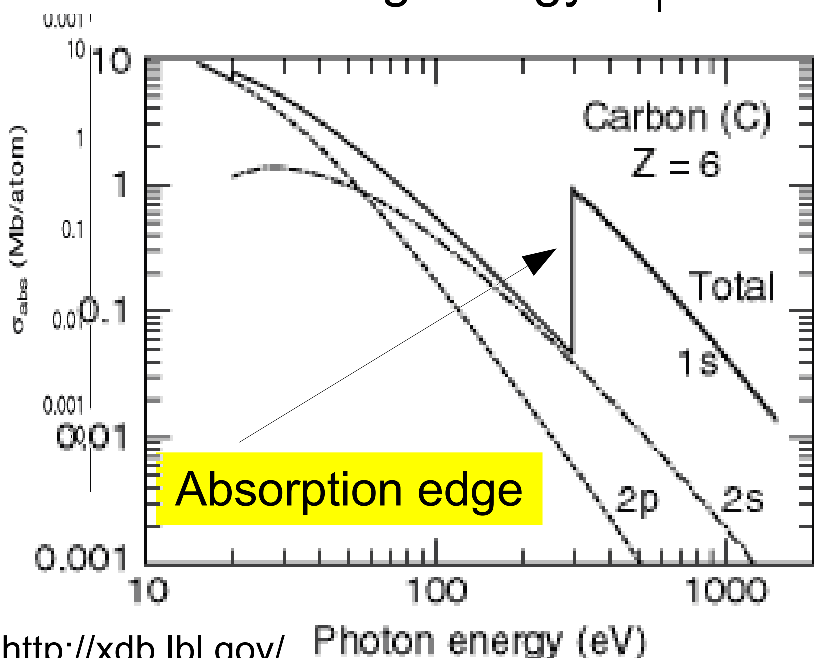

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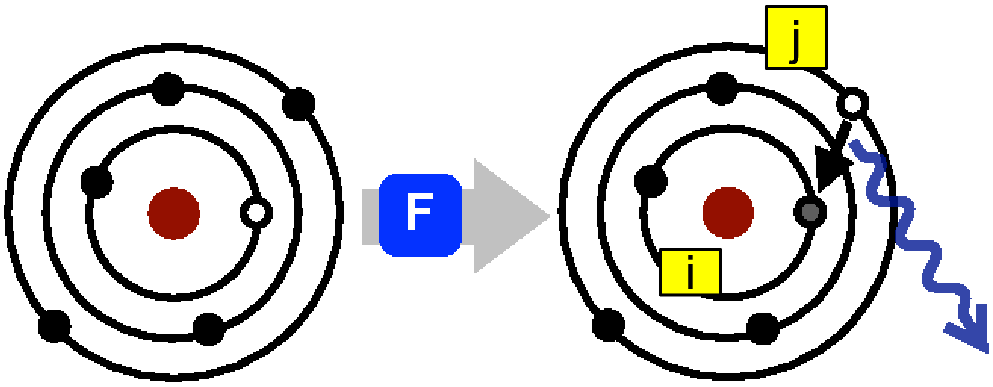
X-ray induced processes in an atom: Inner shell decays

Fluorescent decay

$$\hbar\omega = E^{(f)}_{atom} - E^{(i)}_{atom}$$

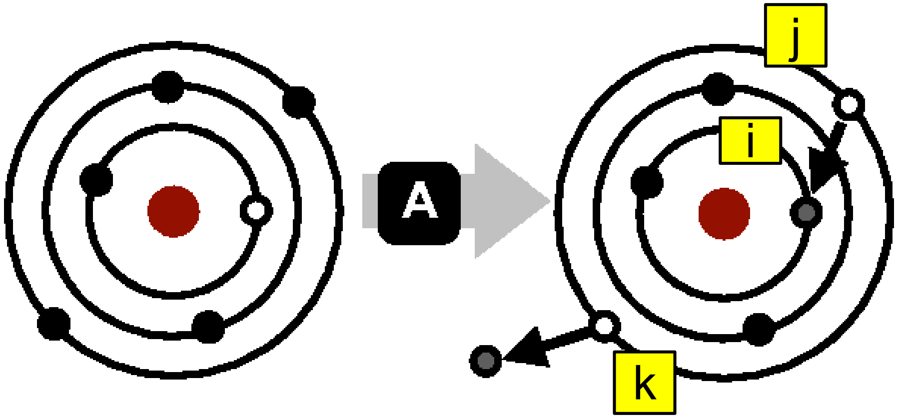
$$\sim B_i - B_j$$



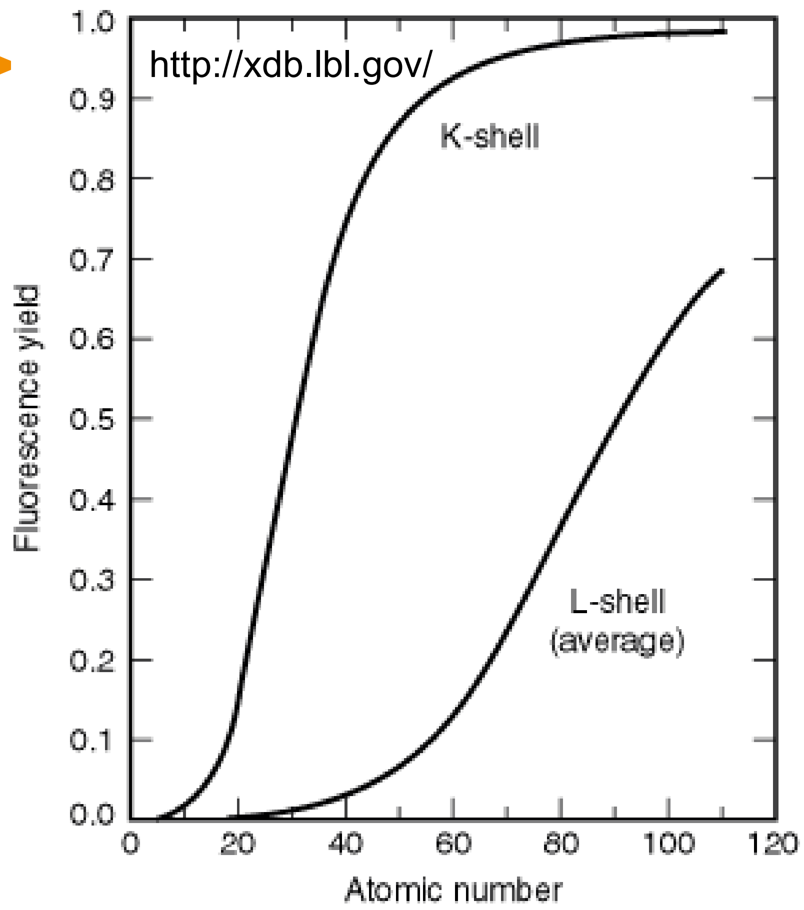
Auger decay

$$E_{Auger} = E^{(f)}_{atom} - E^{(i)}_{atom}$$

$$\sim (B_i - B_j) - B_k$$



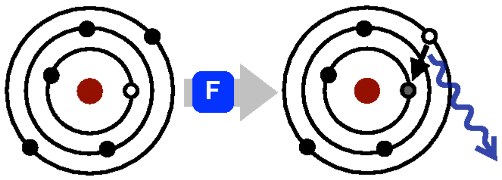
X-ray induced processes in an atom: Competition of decays



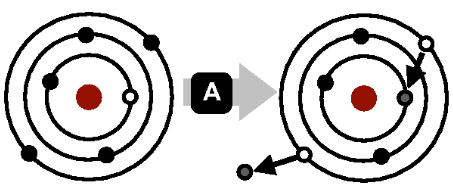
Fluorescence yields for K and L shells:
probability of a core hole being filled by a
radiative process, **in competition with non-radiative processes**

From simple model
(with H-like and planewave):

– **Fluorescent rate** $w_{fluor} \sim Z^4$
lifetime: < 1fs … ps

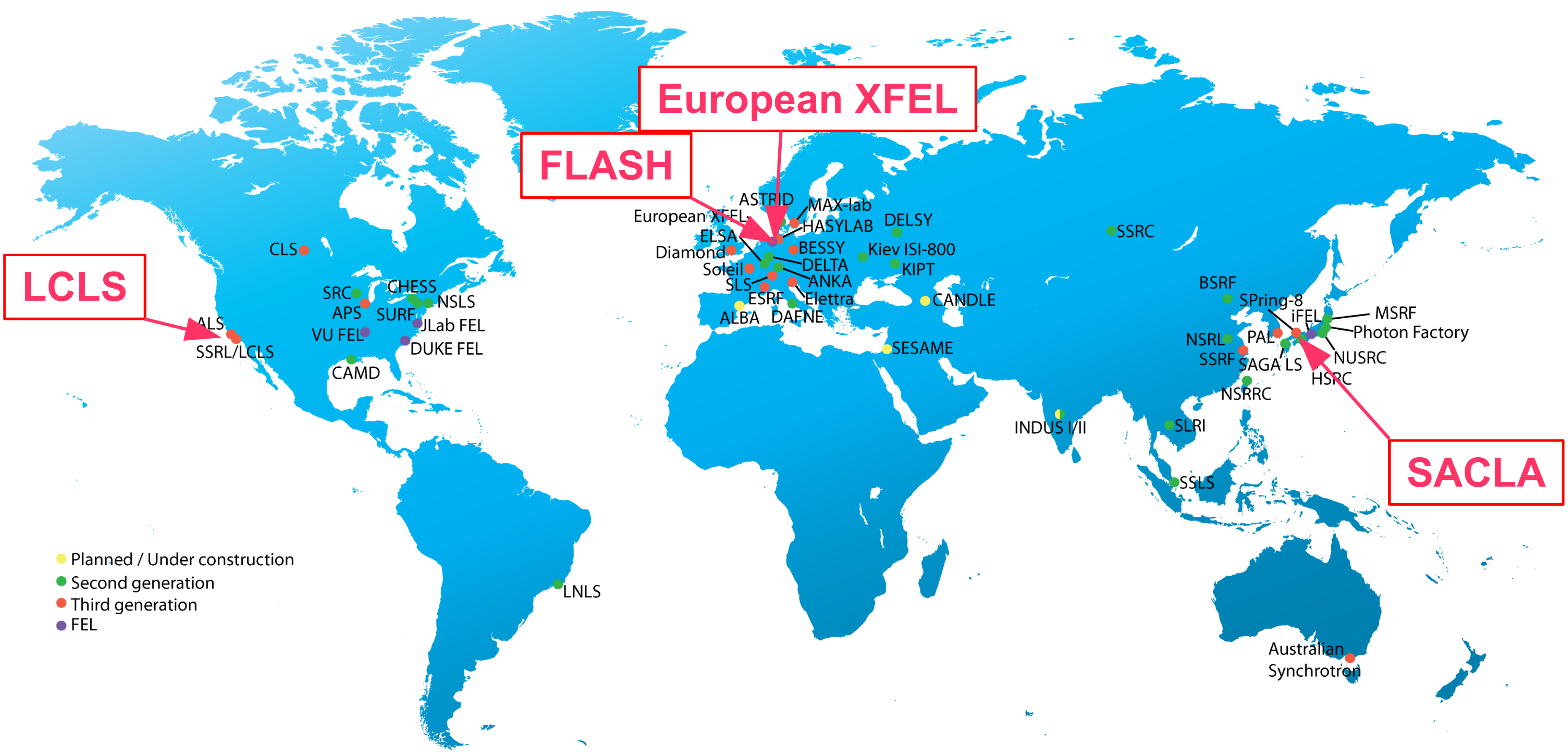


– **Auger** decay **rate** $w_{Auger} \sim Z^0$
lifetime: ~1 … 10fs



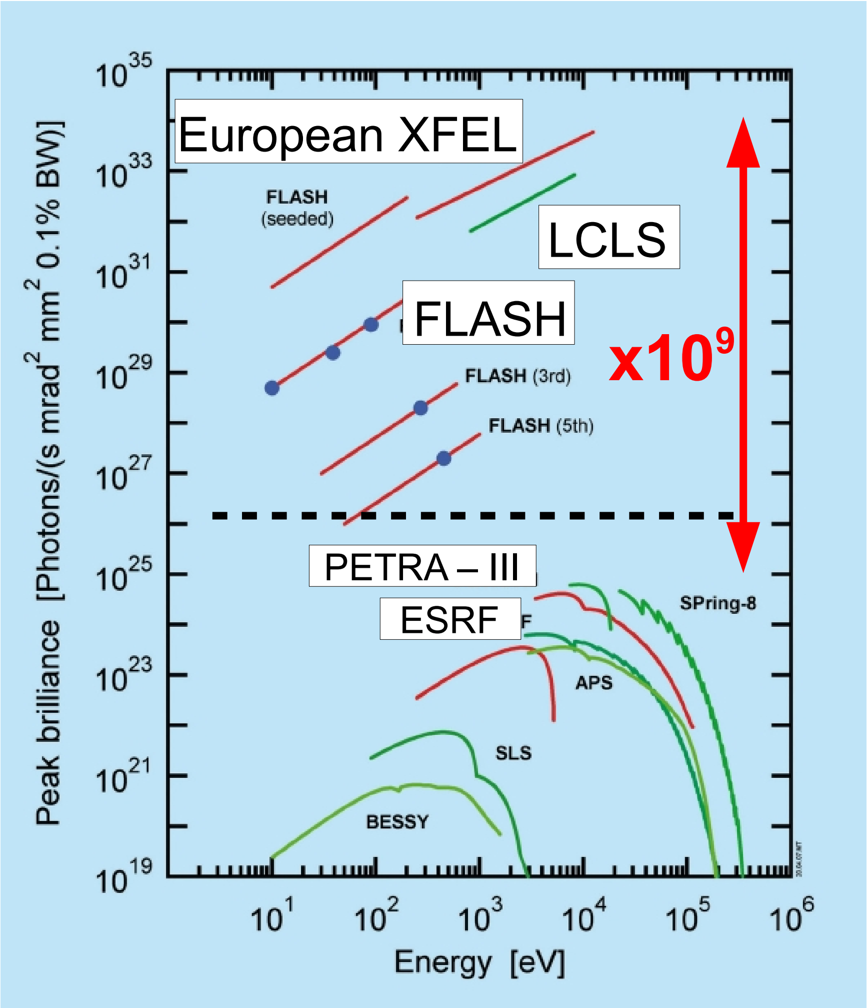
Dynamics of matter induced by high intensity X-rays

Synchrotrons and Free-Electron Lasers in the world



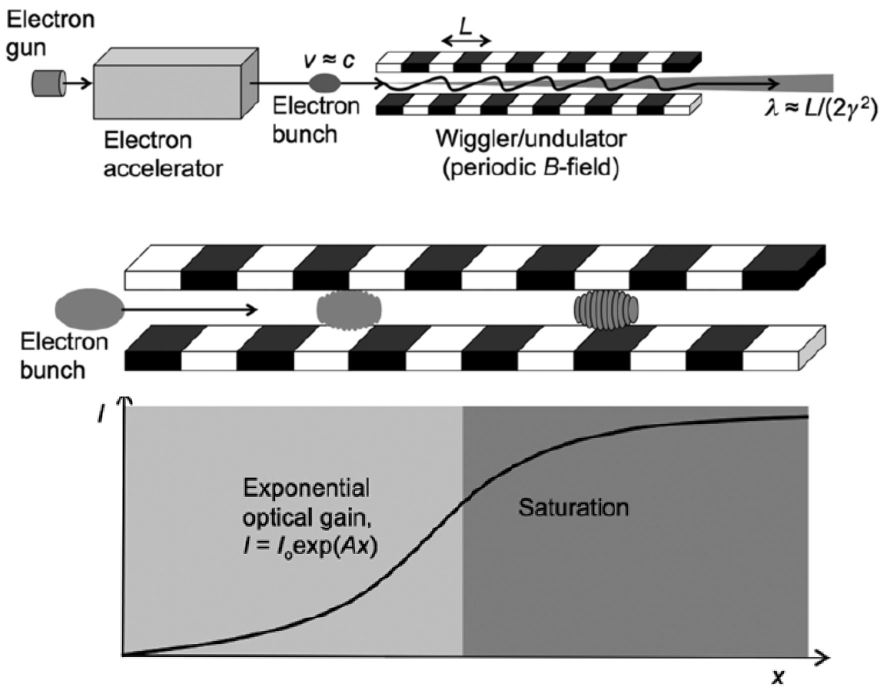
diamond.ac.uk

High x-ray intensity → how high is it?



photon-science.desy.de

X-ray Free-Electron Laser (XFEL)



T_{pulse} ~ 10 – 100 fs

Ribic, Margaritondo, *J. Phys. D* **45** 213001 (2012)

Pellegrini, *Rev. Mod. Phys.* **88** 015006 (2016)

High x-ray intensity → how high is it?

- **Probability of photoionization** during a single pulse (disregarding all other processes)

$$p = 1 - \exp\left(-\int_{-\infty}^{T=+\infty} w(t)\,dt\right) = 1 - \exp\left(-\int_{-\infty}^{T=+\infty} \sigma\, j(t)\,dt\right)$$

$$\sim 1 - \exp(-\sigma N_{photon} / A_{focus}) = 1 - \exp(-\sigma F) \qquad \text{(F: Fluence)}$$

Cross section for Carbon at 1 keV: $\sigma_{Carbon} \sim 0.044$ Mb ($= 4.4 \cdot 10^{-24}$ m^2)

	Synchrotron	XFEL
N_{photon}/ pulse	10^6	10^{12}
A_{focus}	1μm^2	1μm^2
T_{pulse}	~ 20 ps	~ 10 … 100 fs
p_{Carbon}	$4.4 \cdot 10^{-5}$	**0.988**
Signal vs. Fluence	linear	**non-linear**

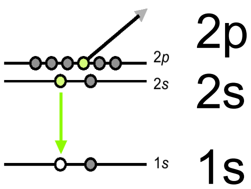
Dynamics of matter induced by high intensity X-rays

Single atoms in intense x-rays

- Theory: **sequential single photon absorption** dominates

- Proof of principle measurement at Linac Coherent Light Source on Neon

Synchrotron: up to Ne^{2+}



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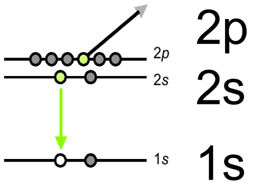
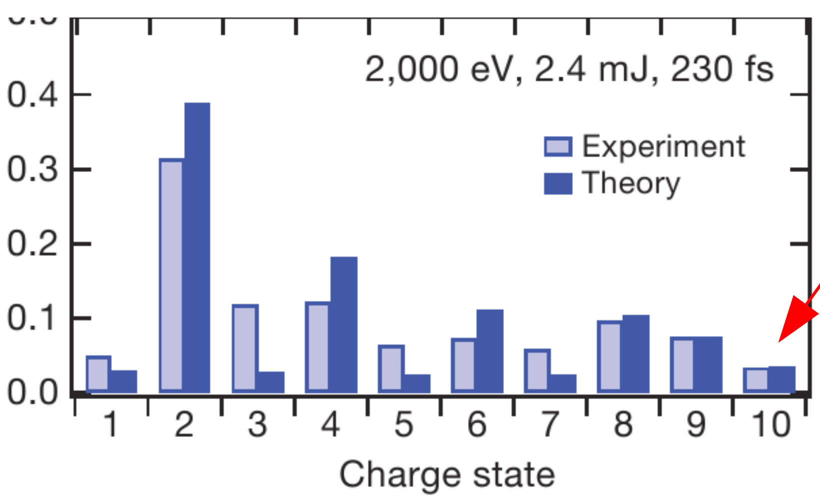
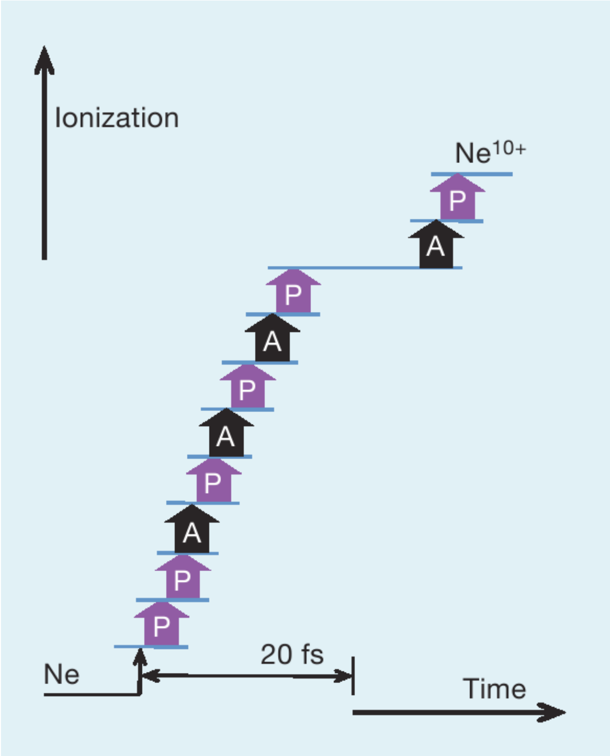


LCLS:

Ne^{10+} : bare core!





L. Young et al *Nature* **466** 56 (2010)

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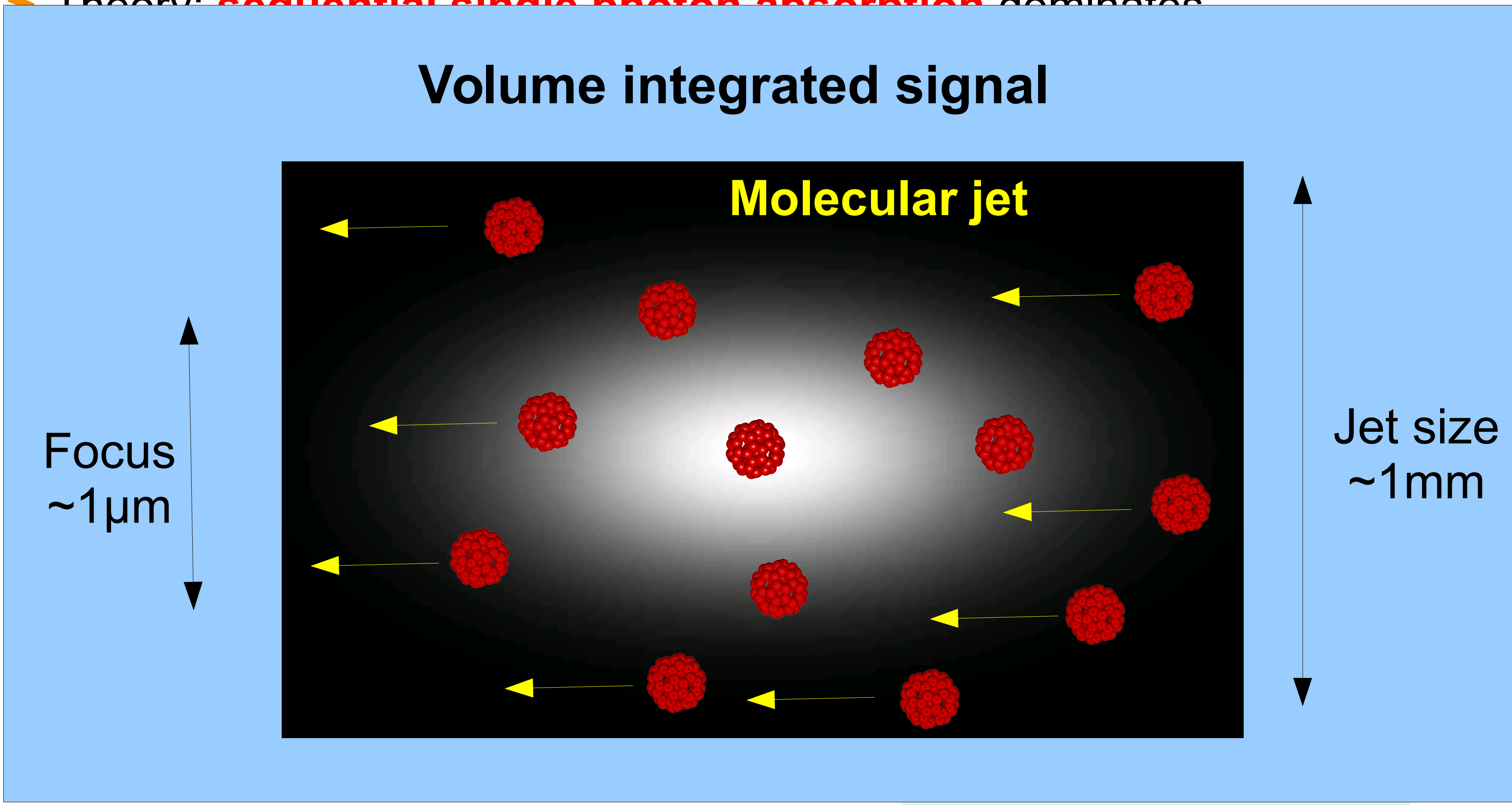

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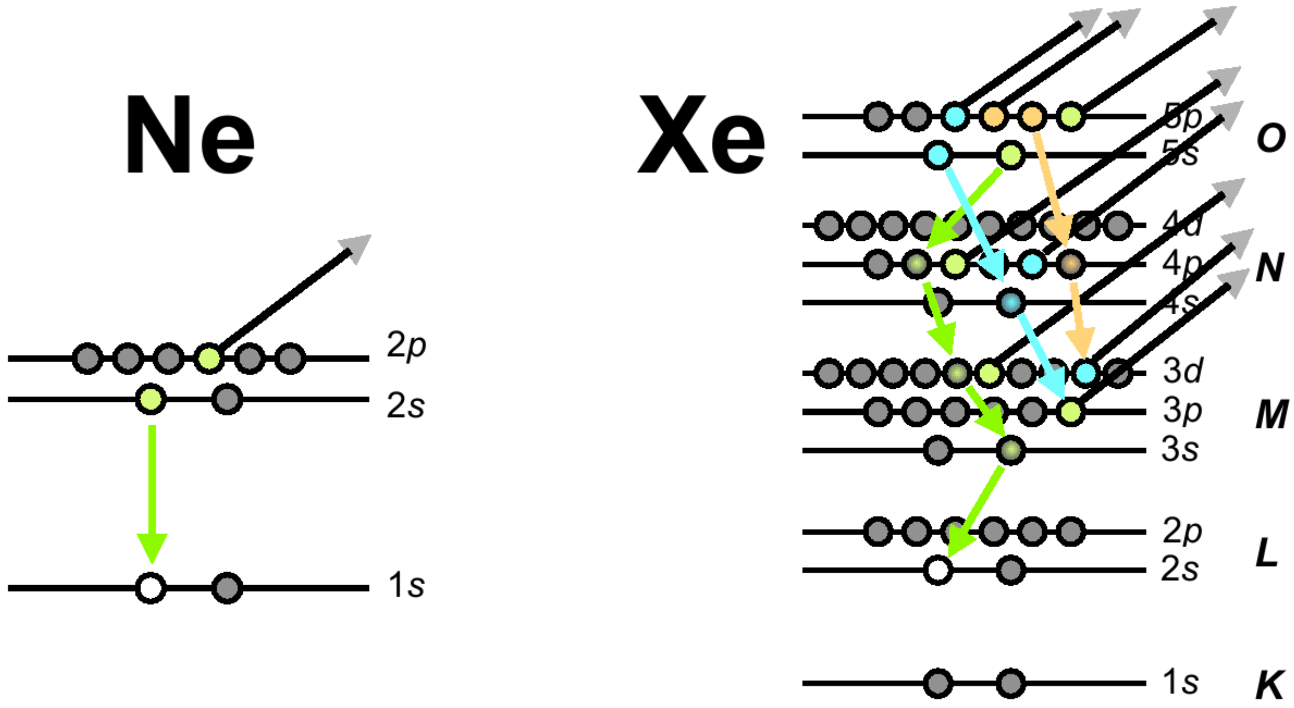
High intensity x-ray induced dynamics: challenge for theory

> **Various** different **electronic configurations** may appear transiently



Multiphoton absorption after/during decay cascade

→ More than 20 million multiple-hole configurations

→ More than 2 billion x-ray-induced processes

High intensity x-ray induced dynamics: challenge for theory

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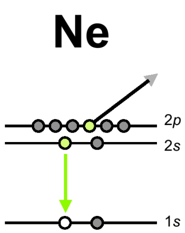
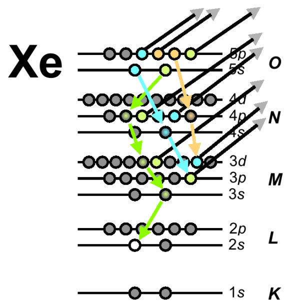
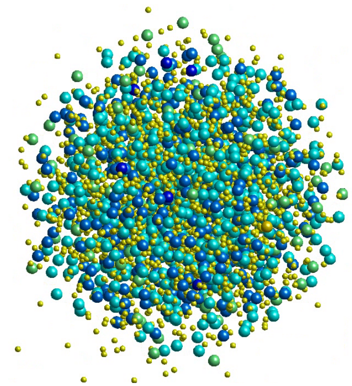
  In many atom systems: **environmental effects**







Non-equilibrium dynamics

- **Highly excited matter** → how to capture **theoretically**?

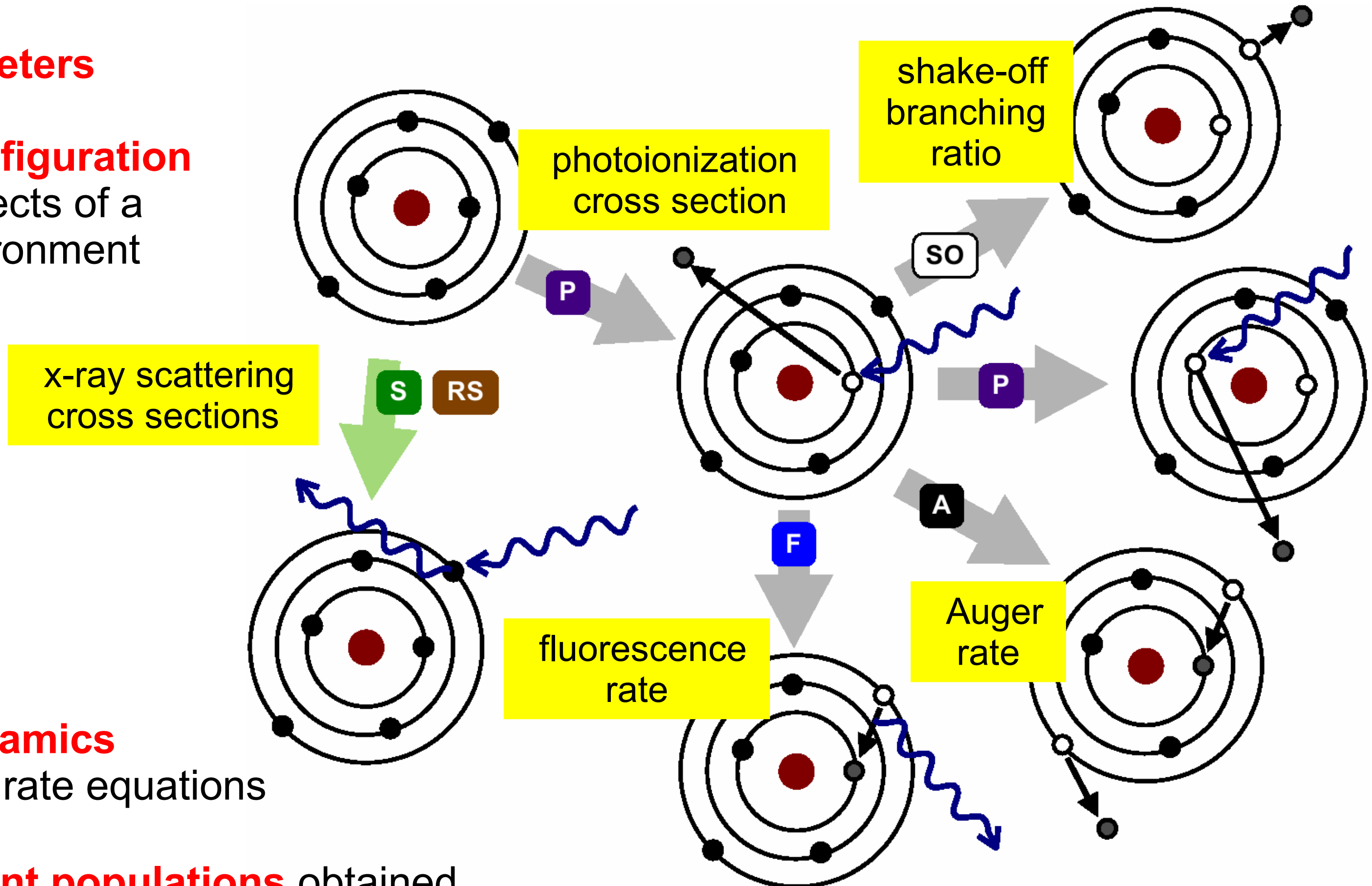
- For single atoms: **XATOM** (ab initio code)

- For atomic clusters, many-atom systems: **XMDYN**

  (Monte Carlo / Molecular Dynamics code)

Computational tool: XATOM

- Ab initio code based on the Hartree-Fock-Slater approach
  - **Atomic parameters** for **arbitrary electronic configuration**
    > also with effects of a plasma environment



  - **Ionization dynamics**
    > described by rate equations
  - **Time-dependent populations** obtained
    > e.g. charge state distribution

Son, Young & Santra, *Phys. Rev. A* **83**, 033402 (2011)
Jurek, Son, Ziaja & Santra, *J. Appl. Cryst*. **49**, 1048 (2016)

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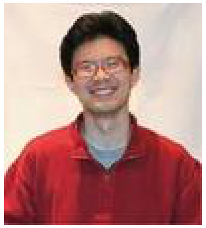
Computational tool: XMDYN

Atomistic Model + Molecular Dynamics (MD) in-house code

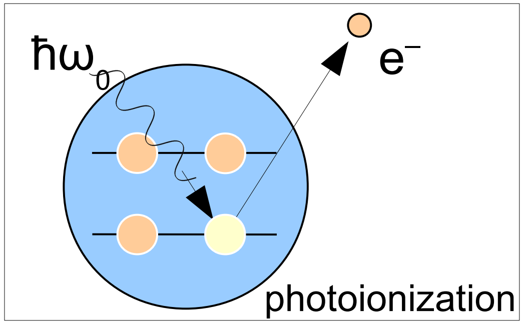
- Bound electrons → **Occupation numbers**

  Inner-shell processes (ph.eff./Auger/fluor.): Monte Carlo

  Rates by **XATOM** package (Sang-Kil Son, Robin Santra)




- **Real space dynamics**: MD
  - atoms/ions and (quasi-) free electrons: classical particles
  - classical force fields: Coulomb ; Newton's equations
- Phenomena due to the **molecular environment**
  - chemical bonds (force fields)
  - secondary ionization
  - recombination



Jurek, Son, Ziaja & Santra, *J. Appl. Cryst*. **49**, 1048 (2016)
B. Murphy *et al.*, *Nat. Commun*. **5** 4281 (2014)

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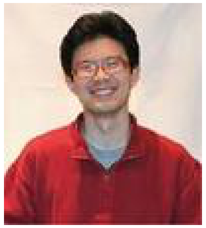

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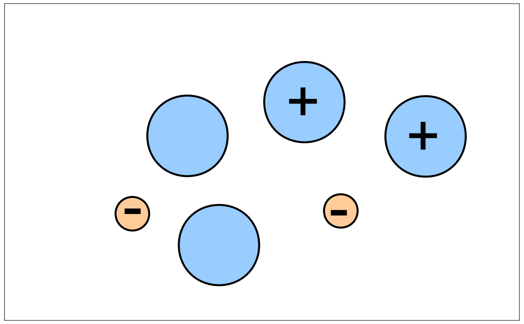

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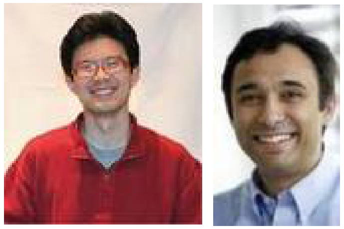

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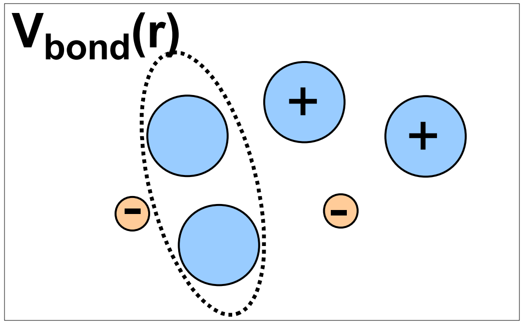

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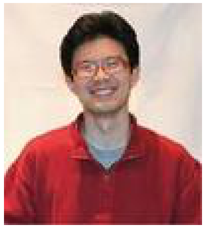

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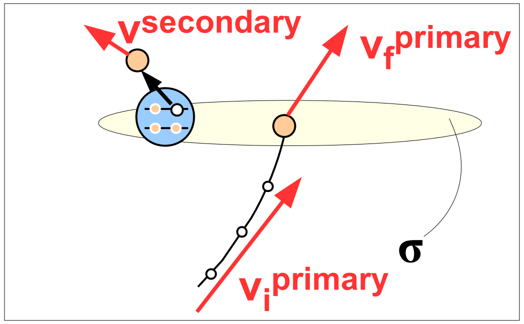

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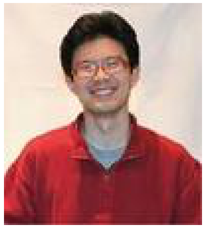
Atomistic Model + Molecular Dynamics (MD) in-house code

- Bound electrons → **Occupation numbers**

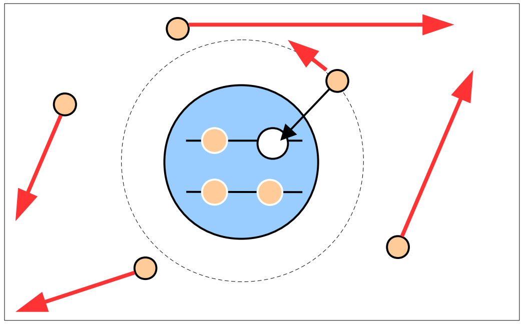
  Inner-shell processes (ph.eff./Auger/fluor.): Monte Carlo

  Rates by **XATOM** package (Sang-Kil Son, Robin Santra)



- **Real space dynamics**: MD
  - – atoms/ions and (quasi-) free electrons: classical particles
  - – classical force fields: Coulomb ; Newton's equations

- Phenomena due to the **molecular environment**
  - – chemical bonds (force fields)
  - – secondary ionization
  - – **recombination**



Jurek, Son, Ziaja & Santra, *J. Appl. Cryst*. **49**, 1048 (2016)
B. Murphy *et al.*, *Nat. Commun*. **5** 4281 (2014)

Computational tool: XMDYN

Atomistic Model + Molecular Dynamics (MD) in-house code

> Bound electrons → **Occupation numbers**
>
> **Inner-shell processes** (ph.eff./Auger/fluor.): Monte Carlo


– **A microscopic description of the dynamics**

– **One XMDYN run** → **One realization of the stochastic dynamics**

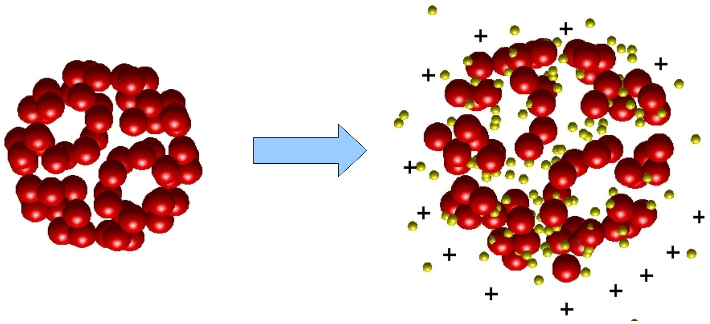
Jurek, Son, Ziaja & Santra, *J. Appl. Cryst*. **49**, 1048 (2016)
B. Murphy *et al.*, *Nat. Commun*. **5** 4281 (2014)

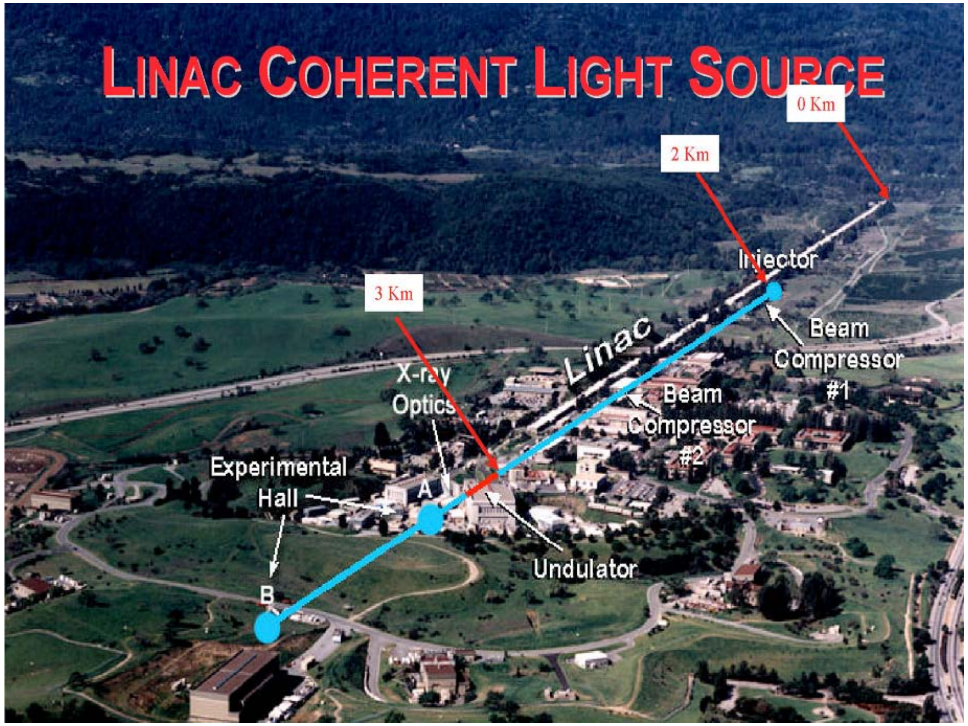
Fullerenes at high x-ray intensity

C_{60} @ LCLS – The Experiment

- **Nora Berrah** (WMU) *et al.*

  $\mathbf{C_{60}}$ **molecules** irradiated **at LCLS**

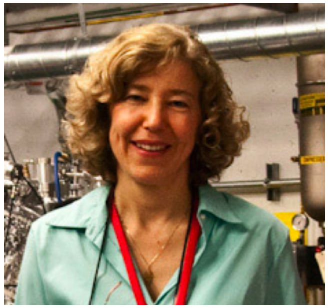




- The Goal:

  to learn about the XFEL-induced dynamics of a highly ionized complex system via **spectroscopy**

C_{60} @ LCLS – The Project & Collaboration

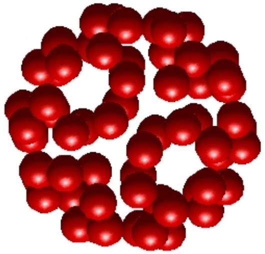


> **Experiment: Nora Berrah**

B. F. Murphy, T. Osipov, L. Fang, M. Mucke, J.H.D. Eland, V. Zhaunerchyk, R. Feifel, L. Avaldi, P. Bolognesi, C. Bostedt, J. D. Bozek, J. Grilj, M. Guehr, L. J. Frasinski, J. Glownia, D. T. Ha, K. Hoffmann, E. Kukk, B. K. McFarland, C. Miron, E. Sistrunk, R. J. Squibb, K. Ueda

> **Theory: CFEL-DESY Theory Division**

Z. Jurek, S.-K. Son, R. Santra



Irradiation conditions:

– $\hbar\omega$ = 485 – 800 eV

– T = 4 – 90 fs

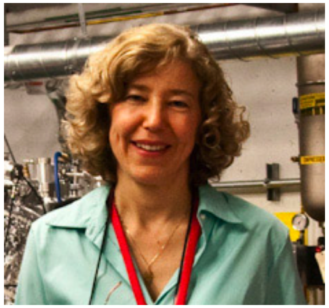
– ε = 0.3 – 0.9 mJ

Ion data measured

B. Murphy *et al.*, Nat. Commun. **5** 4281 (2014)

,

C_{60} @ LCLS – The Project & Collaboration



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- **Theory: CFEL-DESY Theory Division**

  Z. Jurek, S.-K. Son, R. Santra

Irradiation conditions:

High Intensity:

Fluence ~ 15 × (1 / $\sigma^{C}_{ph.ion.}$)

ion data measured

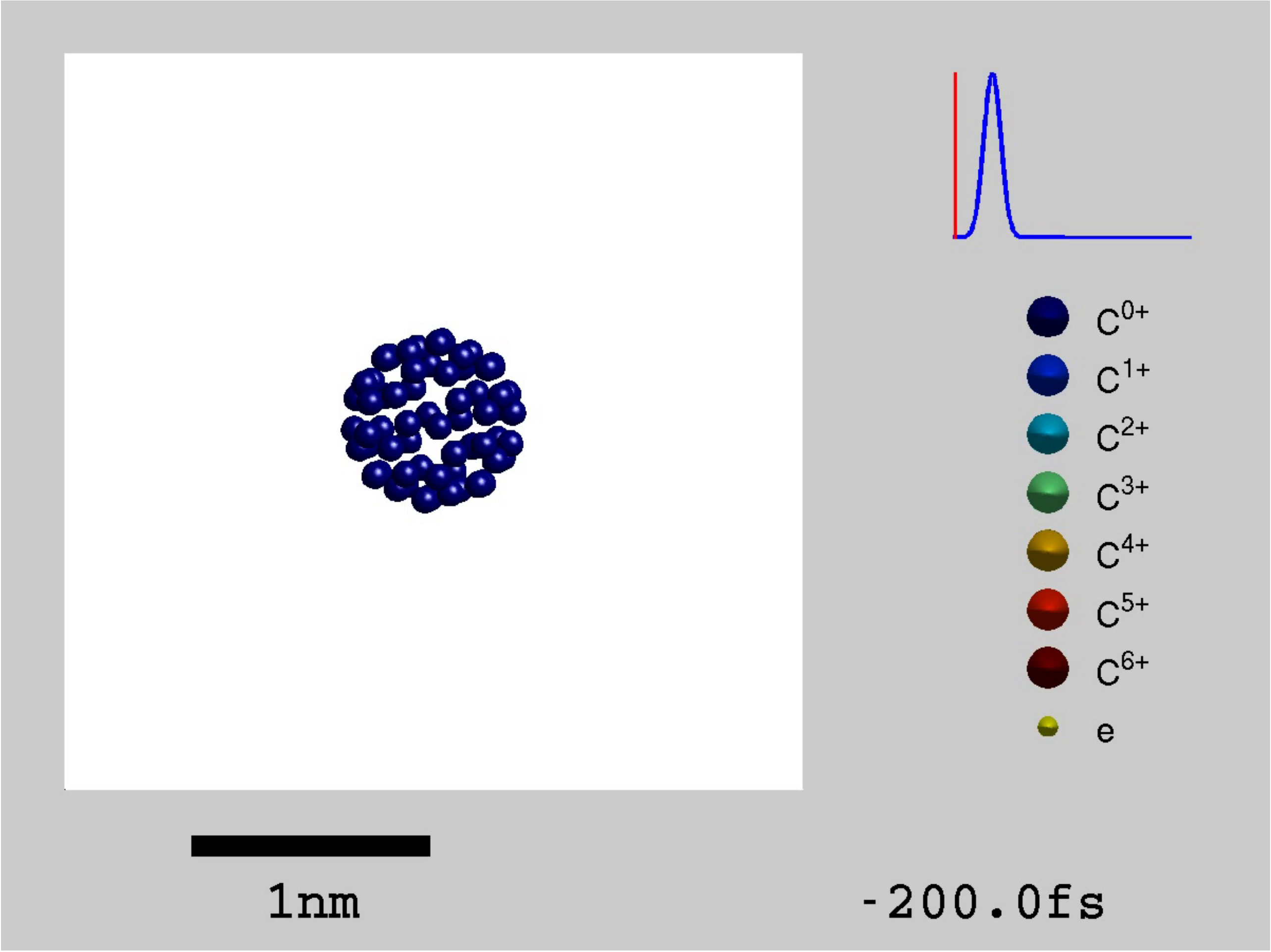
B. Murphy *et al.*, Nat. Commun. **5** 4281 (2014)

CFEL SCIENCE

DESY

C_{60} @ LCLS: Simulation

> Explosion in the focus

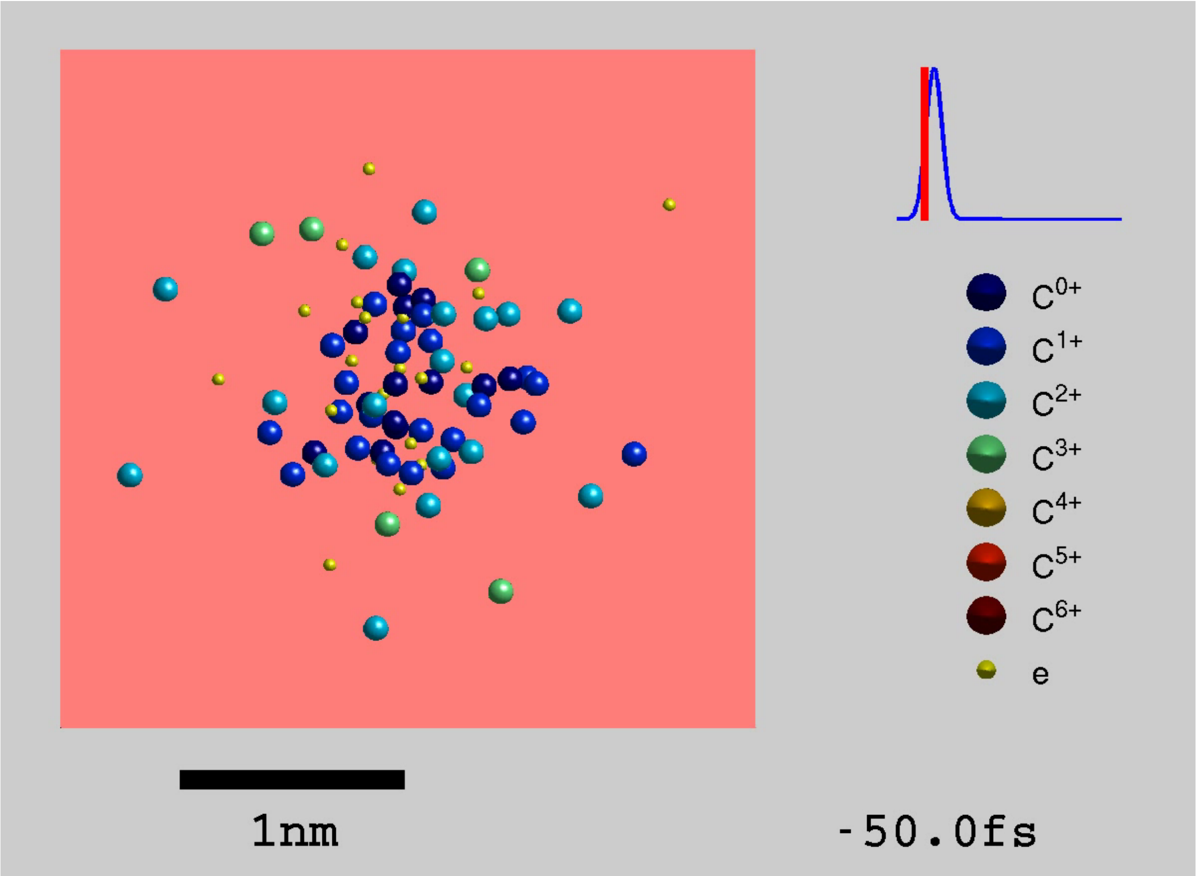


E_{photon} = 485 eV

T_{pulse} = 100 fs

C_{60} @ LCLS: Simulation

> Explosion in the focus



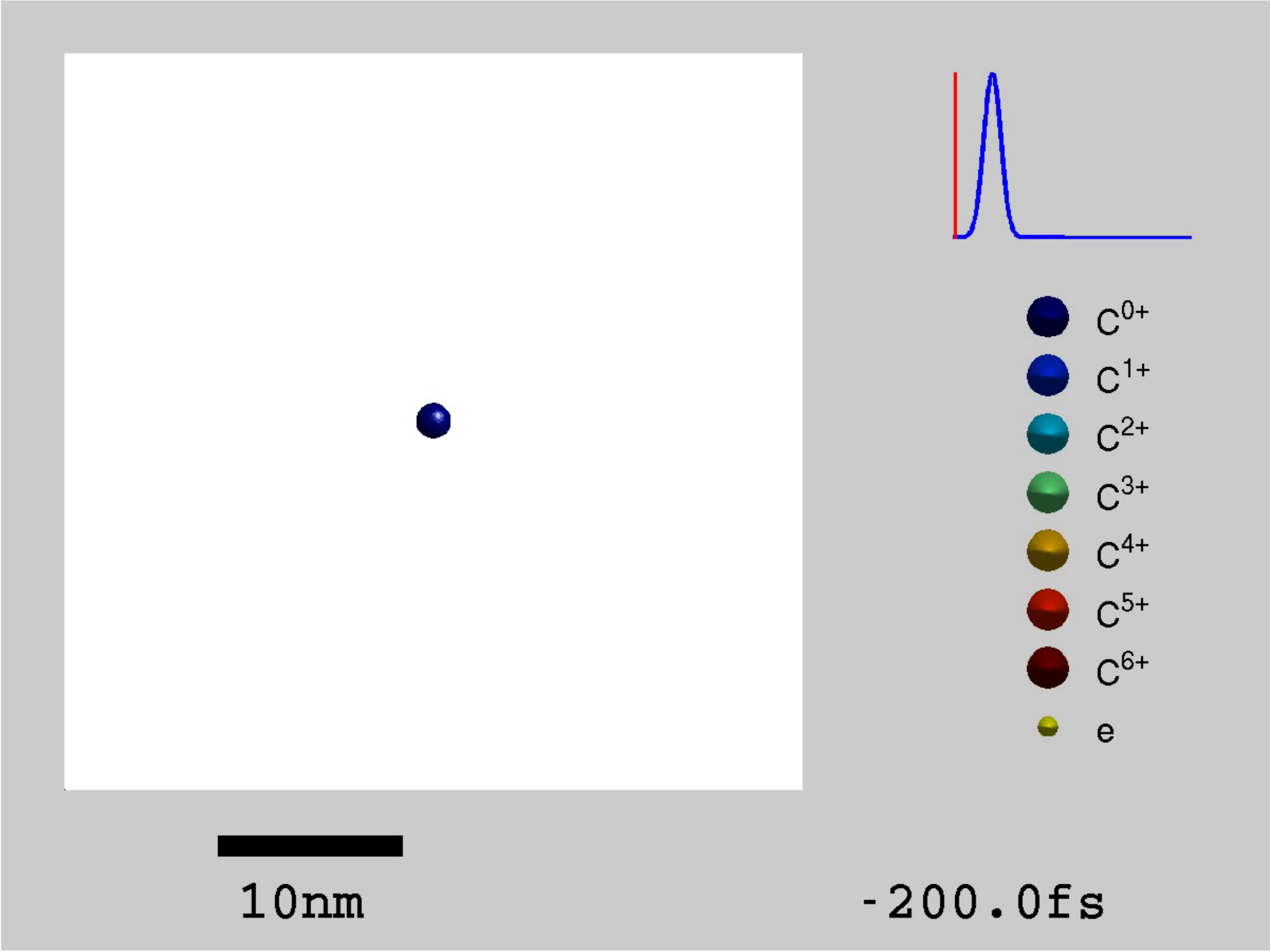
E_{photon} = 485 eV

T_{pulse} = 100 fs

Coulomb explosion

C_{60} @ LCLS: Simulation

> Explosion in the focus



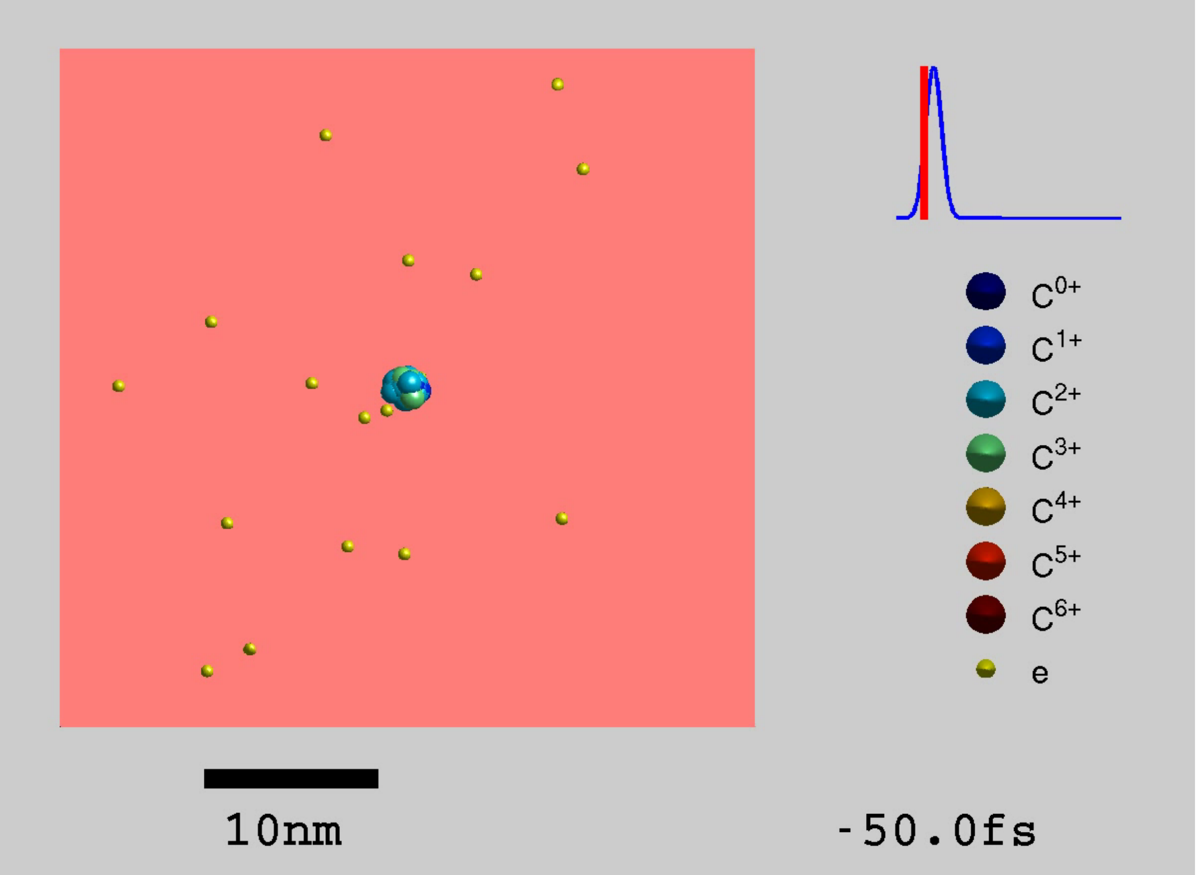
E_{photon} = 485 eV

T_{pulse} = 100 fs

Coulomb explosion

C_{60} @ LCLS: Simulation

> Explosion in the focus



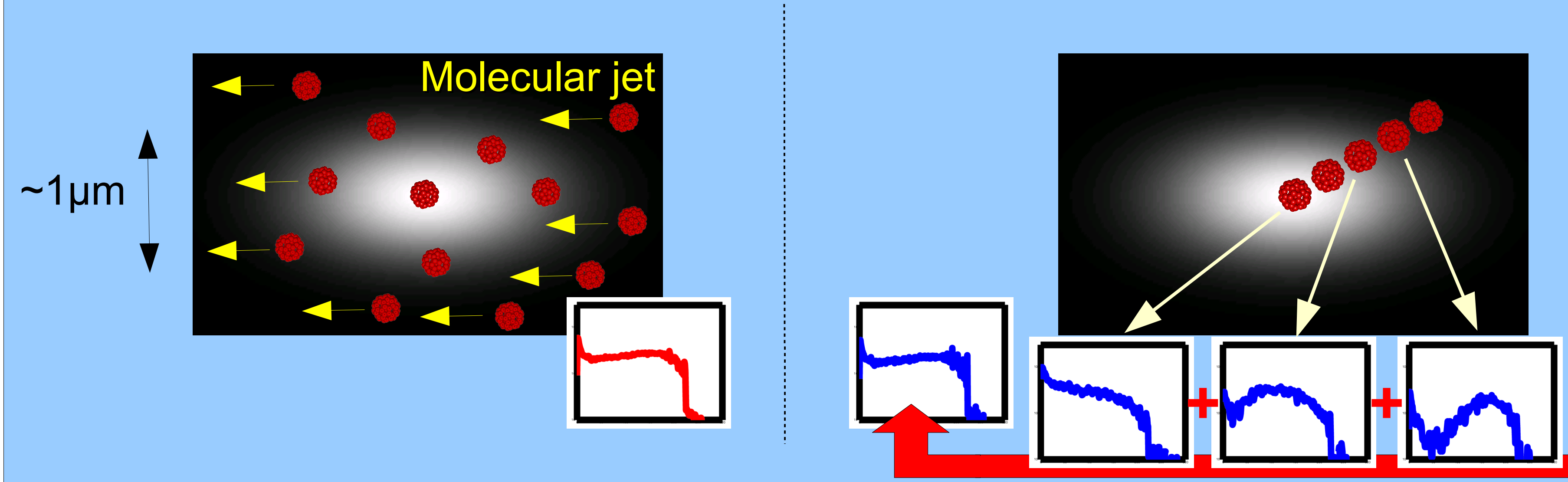
E_{photon} = 485 eV

T_{pulse} = 100 fs

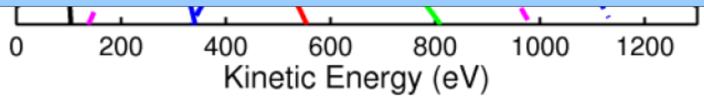
Coulomb explosion

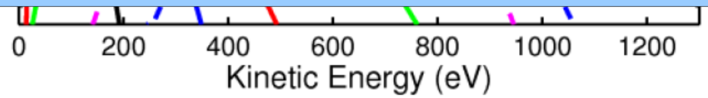
C_{60} @ LCLS – The Observables. Experiment vs. Theory

Volume integrated signal



Experiment vs. Theory





C_{60} @ LCLS – The Observables. Experiment vs. Theory

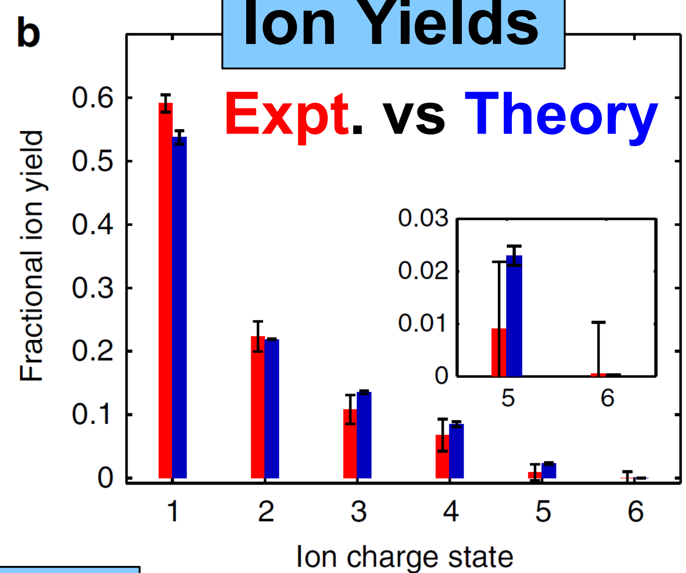
> **Atomic ions**

Theory: **No parameter fitting**!

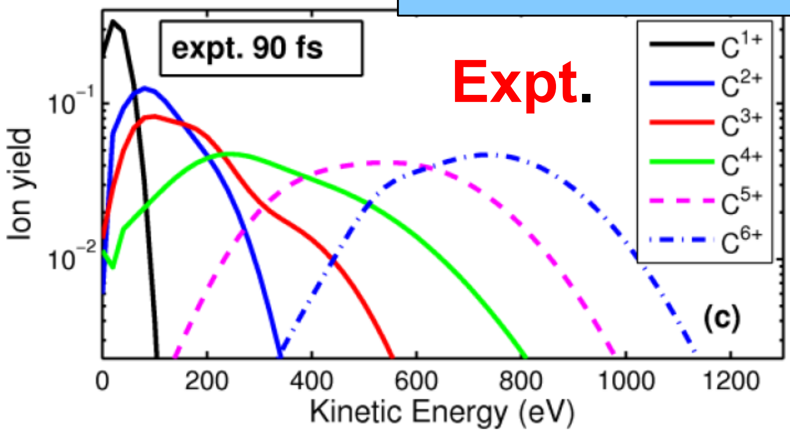
B. Murphy *et al.*, Nat. Commun. **5** 4281 (2014)

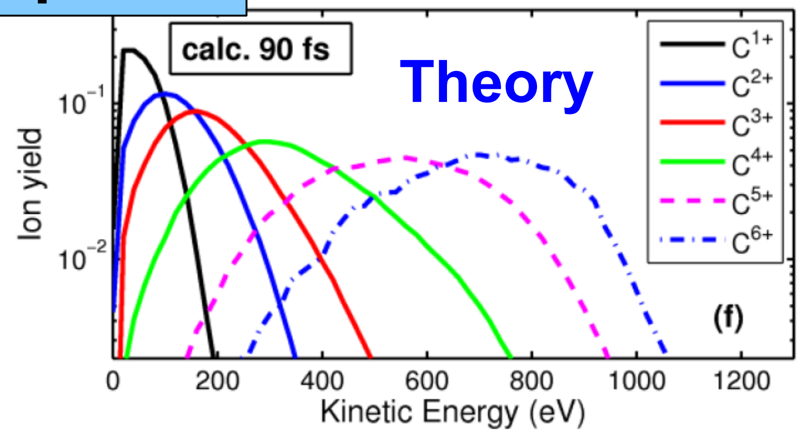
N. Berrah *et al.*, Faraday Discuss. **171** 471 (2014)

Ion Yields



Kinetic Energy spectra





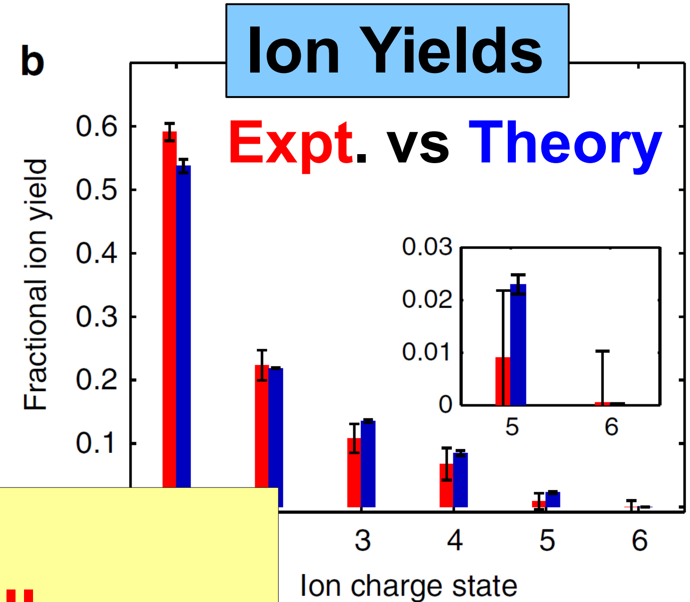
C_{60} @ LCLS – The Observables. Experiment vs. Theory

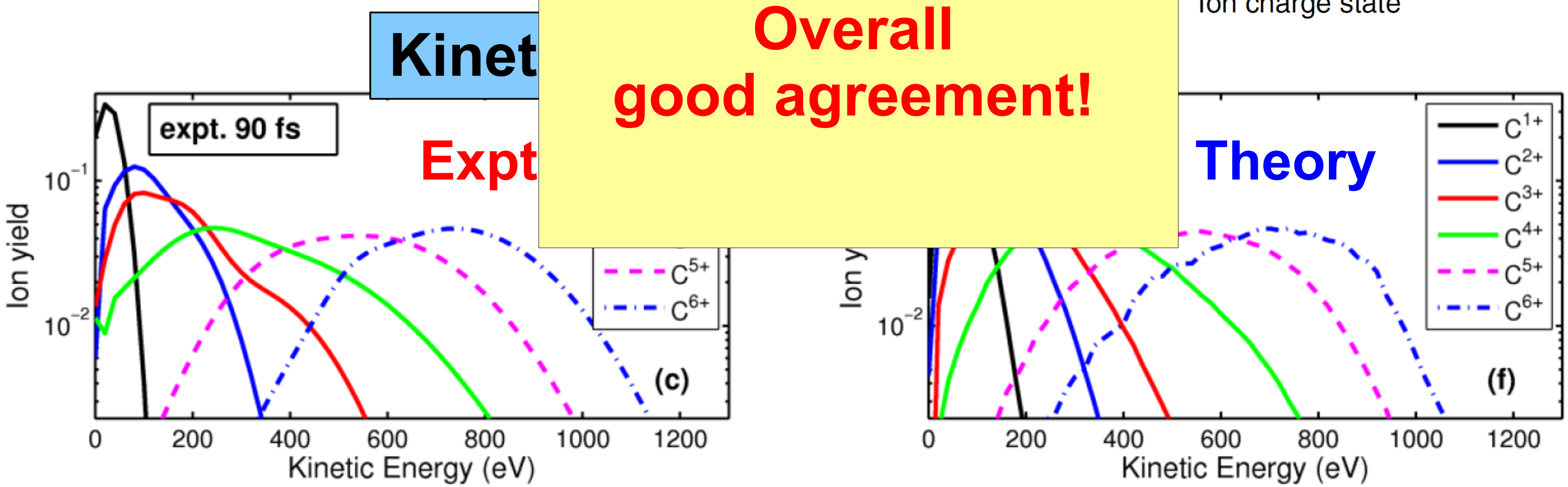
> **Atomic ions**

Theory: **No parameter fitting**!

B. Murphy *et al.*, Nat. Commun. **5** 4281 (2014)

N. Berrah *et al.*, Faraday Discuss. **171** 471 (2014)





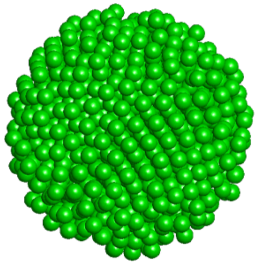
Overall good agreement!

Rare gas atomic clusters at high x-ray intensity

Rare gas clusters @ SACLA – The Experiment

- **Kiyoshi Ueda** (Tohoku Univ.) *et al.*

  Ar, Xe clusters irradiated **at SACLA**



- The Goal:

  to learn about the properties of nanoplasma formed due to XFEL irradiation via **spectroscopy**

Rare gas clusters @ SACLA – The Collaboration

- **Experiment: Kiyoshi Ueda**

  T. Tachibana, H. Fukuzawa, K. Motomura, K. Nagaya,

  S. Wada, P. Johnsson, M. Siano, S. Mondal, Y. Ito, M. Kimura, T. Sakai,

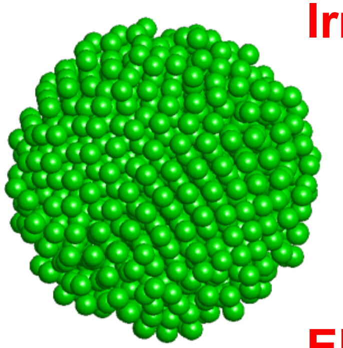
  K. Matsunami, H. Hayashita, J. Kajikawa, X.-J. Liu, E. Robert, C. Miron,

  R. Feifel, J. P. Marangos, K. Tono,

  Y. Inubushi, M. Yabashi, M. Yao

- **Theory: CFEL-DESY Theory Division**

  Z. Jurek, S.-K. Son, B. Ziaja, R. Santra



Irradiation conditions:

– $\hbar\omega = 5 – 5.5$ keV

– $T = 10$ fs

– $\varepsilon \sim 0.24$ mJ

Electron data measured

T. Tachibana, Sci. Rep. **5** 10977 (2015)

Rare gas clusters @ SACLA – The Collaboration

- **Experiment: Kiyoshi Ueda**

  T. Tachibana, H. Fukuzawa, K. Motomura, K. Nagaya, S. Wada, P. Johnsson, M. Siano, S. Mondal, Y. Ito, M. Kimura, T. Sakai, K. Matsunami, H. Hayashita, J. Kajikawa, X.-J. Liu, E. Robert, C. Miron, R. Feifel, J. P. Marangos, K. Tono, Y. Inubushi, M. Yabashi, M. Yao

- **Theory: CFEL-DESY Theory Division**

  Z. Jurek, S.-K. Son, B. Ziaja, R. Santra

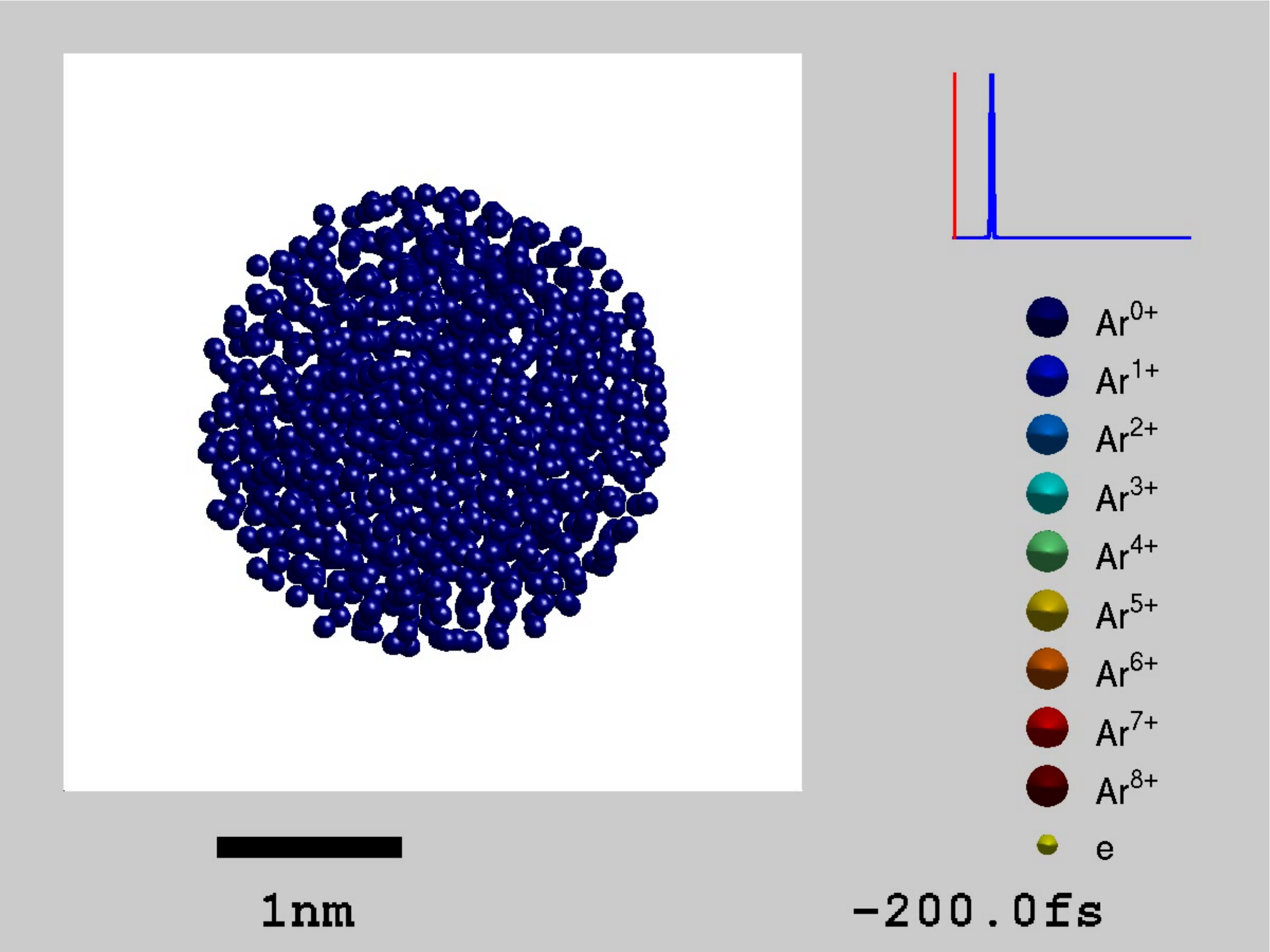
High Intensity:

Fluence ~ 0.16 × (1 / $\sigma^{Ar}_{ph.ion.}$)

T. Tachibana, Sci. Rep. **5** 10977 (2015)

Rare gas clusters @ SACLA: Simulation

> Ar_{1000} explosion in the focus

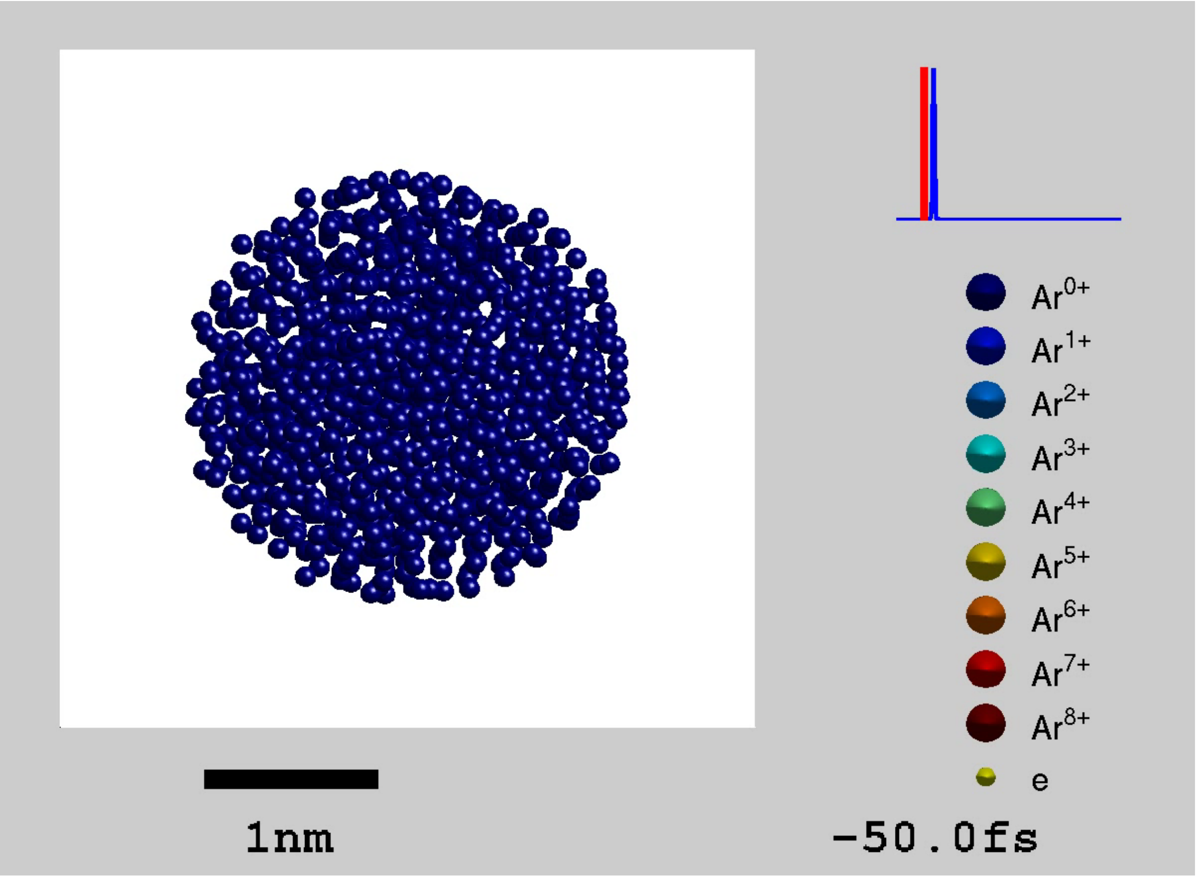


E_{photon} = 5 keV

T_{pulse} = 10 fs

Rare gas clusters @ SACLA: Simulation

> Ar_{1000} explosion in the focus



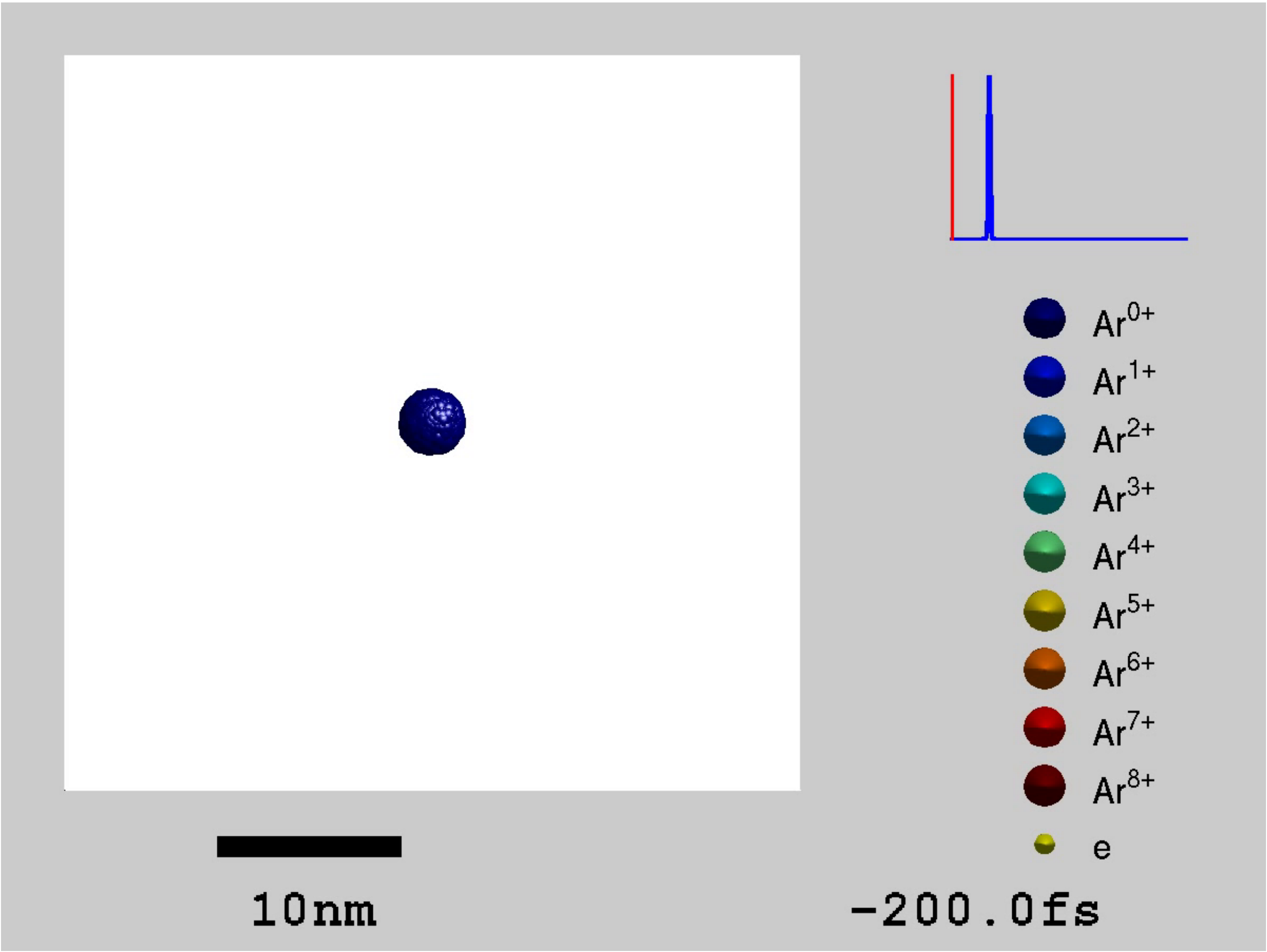
E_{photon} = 5 keV

T_{pulse} = 10 fs

Nanoplasma formation

Rare gas clusters @ SACLA: Simulation

> Ar_{1000} explosion in the focus



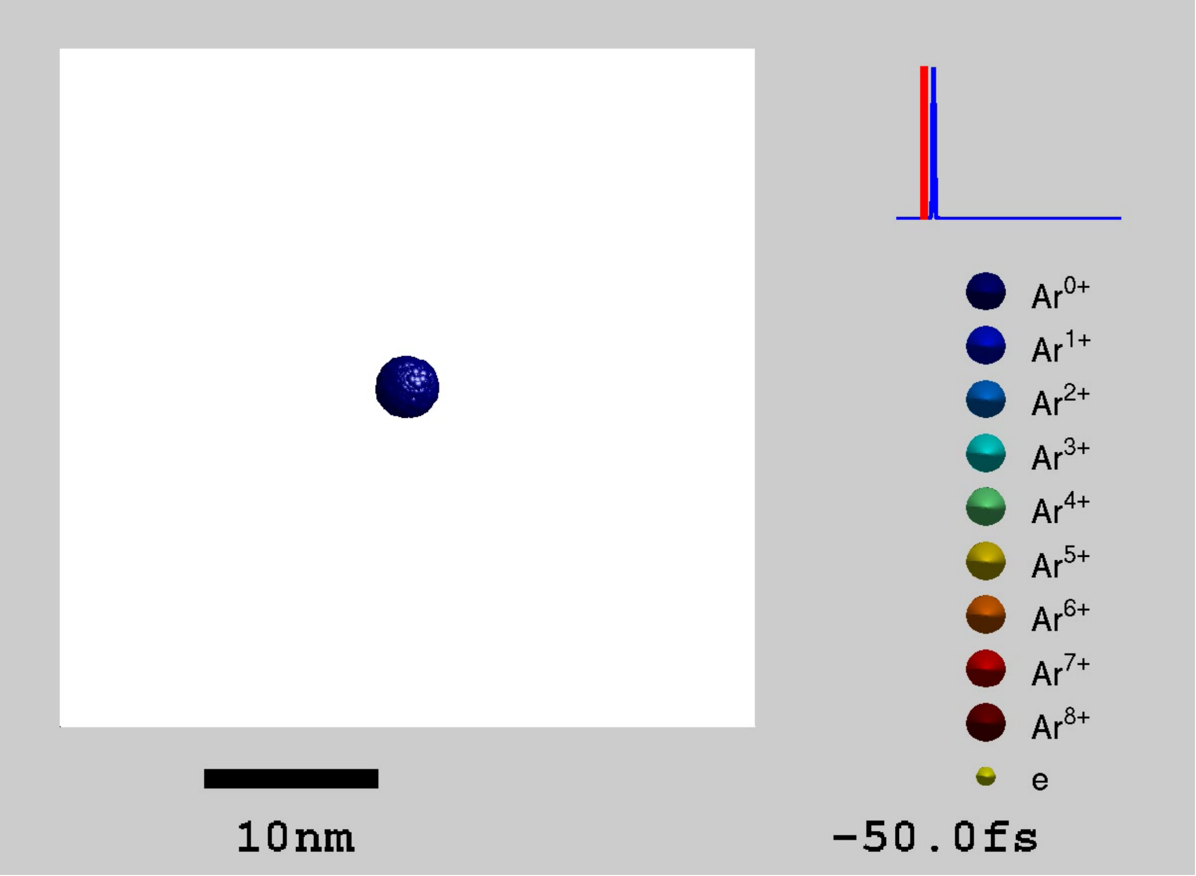
E_{photon} = 5 keV

T_{pulse} = 10 fs

Nanoplasma formation

Rare gas clusters @ SACLA: Simulation

> Ar_{1000} explosion in the focus



E_{photon} = 5 keV

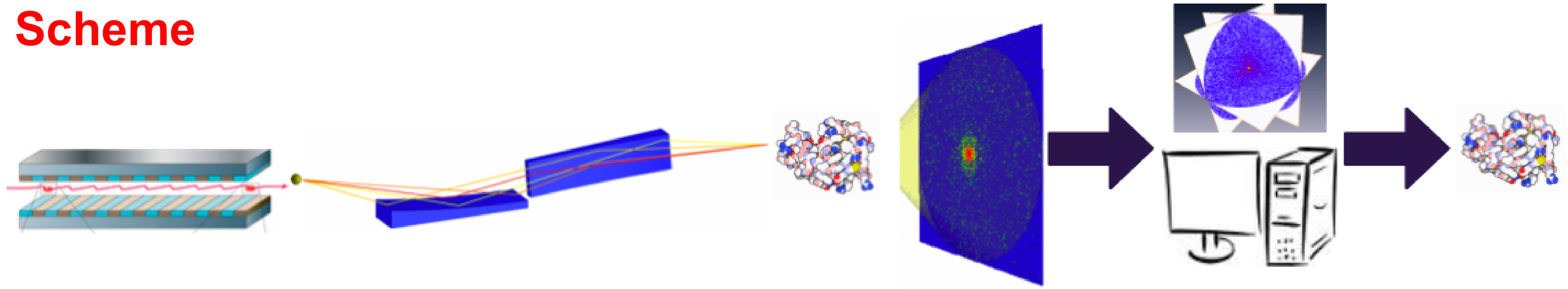
T_{pulse} = 10 fs

Nanoplasma formation

Single Molecule Imaging Start-To-End (S2E) Simulations at the European XFEL

Single Molecule Imaging S2E Simulations

> Scheme

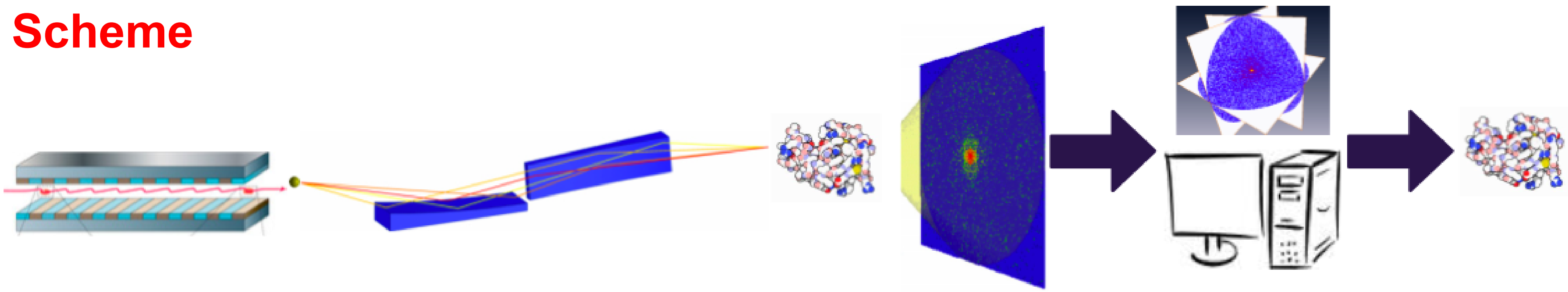


Images: *Nature Photonics* **4**, 814–821 (2010), x-ray-optics.de, pdb.org, J. Phys. B: At. Mol. Opt. Phys. 43 (2010) 194016, SPB CDR

XFEL Pulse generated in the undulator	Focusing **Optics**	**Target**	**Single shot** 2D Diffraction **Patterns** (Detectors)	**Orienting** patterns and **Phasing**: **Reconstruction**

Single Molecule Imaging S2E Simulations

> Scheme



Images: *Nature Photonics* **4**, 814–821 (2010), x-ray-optics.de, pdb.org, J. Phys. B: At. Mol. Opt. Phys. 43 (2010) 194016, SPB CDR

> S2E simulations @ European XFEL

http://www.xfel.eu/sims2e

Goal: realistic simulations

Potential:
- explore possible parameter spaces
- plan experiments, optimize setup, save beamtime
- feedback to data post-processing

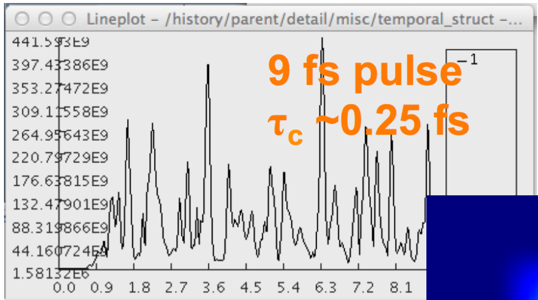
Requirement: framework defined, simulation tools

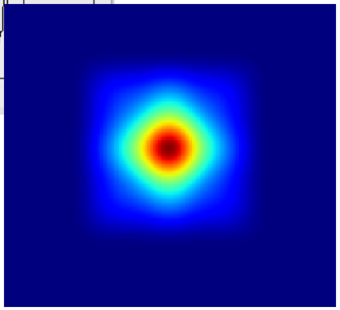
Single Molecule Imaging S2E Simulations

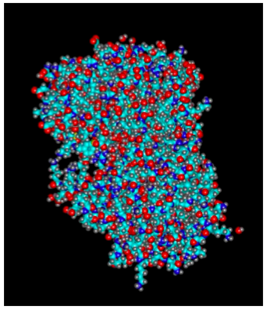
> S2E simulations @ European XFEL

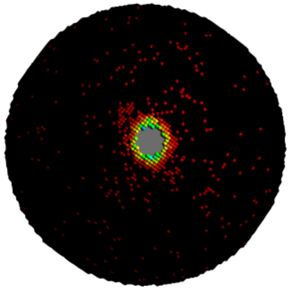
http://www.xfel.eu/sims2e

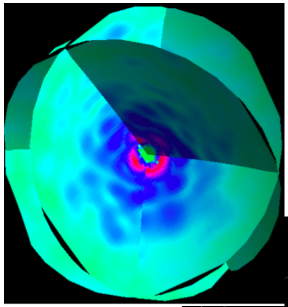
FEL source → Optics → Photon/Matter Interaction → Diffr. Patterns → Analysis

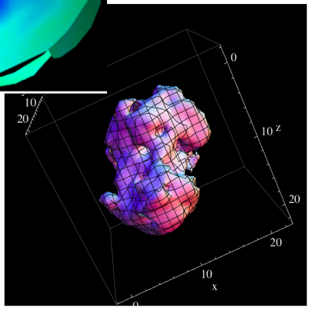












Images:
courtesy of A. Mancuso
and collaborators

> Codes:

FAST | **WPG** | **XMDYN** | **SingFEL** | **EMC & DM**

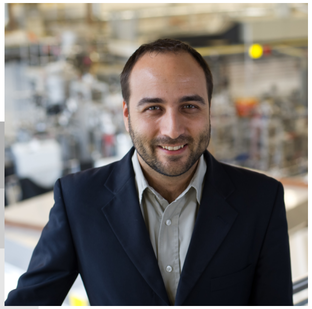
Yoon et al, Sci. Rep. **6**, 24791 (2016)

CFEL SCIENCE

DESY

Single Molecule Imaging S2E Simulations – Collaboration

> Project Leader: Adrian P. Mancuso



Name	Organization	Role
Chun Hong Yoon	European XFEL & CFEL	Fast diffraction calculation, interfaces, more
Liubov Samoylova	European XFEL X-ray	X-ray optics, propagation code
Alexey Buzmakov	Institute of Crystallography	WPG propagation framework, interfaces
Oleg Chubar	Brookhaven National Lab	SRW wave optics core library for propagation
Zoltan Jurek	CFEL	Photon–Matter Interaction Simulation
Robin Santra	CFEL	Photon–Matter Interaction Simulation
Beata Ziaja	CFEL	Photon–Matter Interaction Simulation
Mikhail Yurkov	DESY	Source photon field simulations
Evgeny Schneidmiller	DESY	Source photon field simulations
Duane Loh	NUS	Orientation Algorithms, Image Reconstruction
Carsten Fortmann-Grote	European XFEL	Interfaces, SIMEX platform
Adrian Mancuso	European XFEL	Coordinator, Image Reconstruction
Thomas Tschentscher	European XFEL	European XFEL Director for optics and SPB

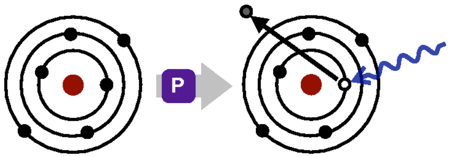
http://www.xfel.eu/sims2e

CFEL SCIENCE

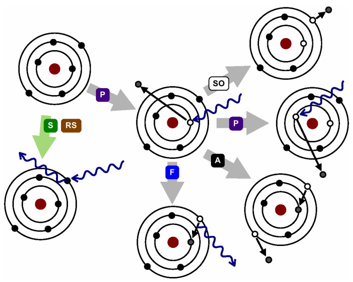
DESY

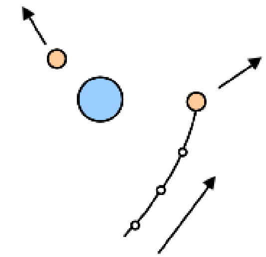
Summary

- **Elements of x-ray physics**



- Simulation tools **XATOM** and **XMDYN**

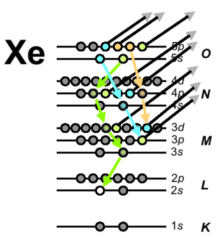


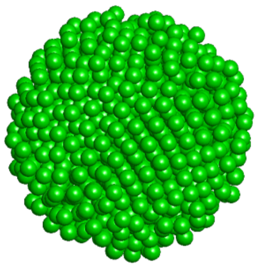


- Ionization dynamics of **single atoms**

  Ionization and real space dynamics

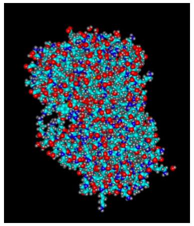
  of finite **many atom systems**

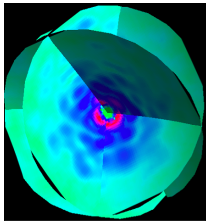


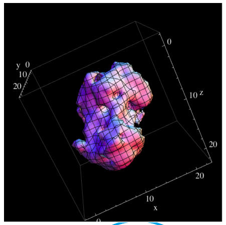


- Realistic **simulations of**

  single molecule imaging experiments

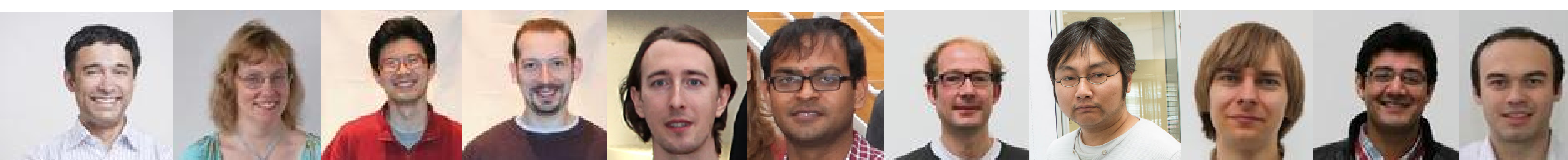






XTOOLS of CFEL–DESY Theory Division

- **XATOM**[1]: an ab initio integrated toolkit for x-ray atomic physics
- **XMOLECULE**[2]: an ab initio integrated toolkit for x-ray molecular physics
- **XMDYN**[3]: an MD/MC tool for modeling matter at high intensity x-rays
- **XHYDRO**[4]: a hydrodynamic tool for simulating plasma in local equilibrium
- **XSINC**[5]: a tool for calculating x-ray diffraction patterns for nano-crystals
- **XTANT**[6]: a hybrid tight-binding/MD/MC tool to study phase transitions
- **XCASCADE**[7]: MC tool to treat electron cascades at low x-ray excitation
- **XCALIB**[8]: an XFEL pulse profile calibration tool based on ion yields
- **BOLTZMANN**[9]: a kinetic approach for XFEL–matter interaction



R. Santra	B. Ziaja	S.-K. Son	Z. Jurek	N. Medvedev	V. Saxena	L. Inhester	K. Toyota	V. Lipp	M.M. Abdullah	V. Tkachenko
1-5,8	3,4,6,7,9	1,2,3,8	3,5,8	6,7 (now in Prague)	4 (now in India)	1,2	1,8	6,7	3,5	6,7

http://www.desy.de/~xraypac

X-ray physics: useful references

Theory

IOP Publishing — Journal of Physics B: Atomic, Molecular and Optical Physics

J. Phys. B: At. Mol. Opt. Phys. **42** (2009) 023001 (16pp) — doi:10.1088/0953-4075/42/2/023001

PHD TUTORIAL

Concepts in x-ray physics

Robin Santra

Argonne National Laboratory, Argonne, IL 60439, USA
and
Department of Physics, University of Chicago, Chicago, IL 60637, USA

E-mail: rsantra@anl.gov

Received 26 November 2008
Published 29 December 2008
Online at stacks.iop.org/JPhysB/42/023001

Abstract
A basic introduction to the theory underlying x-ray processes is provided. After general remarks on the practical advantages of using x-rays for probing matter, the derivation of the minimal-coupling Hamiltonian within nonrelativistic quantum electrodynamics is outlined. Perturbation theory is reviewed and applied to describe x-ray-induced processes. In connection with x-ray absorption, inner-shell binding energies and the photon energy dependence of the x-ray absorption cross section are discussed. In the context of x-ray scattering, atomic and molecular scattering factors are introduced, the complex index of refraction is derived, and the nonrelativistic theory of Compton scattering is described. The final topic is x-ray fluorescence and Auger decay of inner-shell-excited systems.

1. Introductory remarks

Since their discovery in the year 1895 by Wilhelm Conrad Röntgen [1], x-rays have become an indispensable tool for studying the structure and electronic properties of matter. Equally important is the role x-rays have come to play in medicine, archaeology, art, security, astronomy and other applications. To date, 19 Nobel prizes have been awarded for x-ray-related research (W C Röntgen 1901, M von Laue 1914, W H Bragg and W L Bragg 1915, C G Barkla 1917, K M G Siegbahn 1924, A H Compton 1927, P J W Debye 1936, H J Muller 1946, M F Perutz and J C Kendrew 1962, F H C Crick, J D Watson, and M H F Wilkins 1962, D Crowfoot Hodgkin 1964, W N Lipscomb 1976, A M Cormack and G N Hounsfield 1979, K M Siegbahn 1981, H A Hauptman and J Karle 1985, J Deisenhofer, R Huber and H Michel 1988, P D Boyer and J E Walker 1997, P Agre and R MacKinnon 2003, R Kornberg 2006).

As a new generation of x-ray sources—so-called x-ray free-electron lasers [2, 3]—is about to come online [4–6], it is timely to familiarize newcomers, experimentalists and theorists alike, with some of the basic properties that make x-rays such a powerful tool. This is attempted in this tutorial. The emphasis is on the development of a consistent theoretical framework that may be employed to describe a variety of x-ray processes. Throughout, the x-rays are assumed to be used as a weak, essentially nonperturbative *probe*. Basic applications of x-rays will be covered. However, since this is a tutorial, there is not a single topic that is discussed in great depth, and some topics, e.g., x-ray sources and x-ray optics [7, 8], are not discussed at all.

This tutorial is structured as follows. In section 2, the derivation of the Hamiltonian underlying nonrelativistic quantum electrodynamics is sketched. This Hamiltonian, in combination with perturbation theory (section 3), allows one to describe all basic x-ray processes. X-ray absorption is the topic of section 4. In section 5, x-ray scattering processes are discussed. Finally, in section 6, relaxation processes following the excitation of an inner-shell electron are treated.

Atomic units are employed, i.e., $m_e = 1, |e| = 1, \hbar = 1$ and $c = 1/\alpha$, where m_e is the electron mass, e is the electron charge, $\hbar$ is Planck's constant divided by 2π, c is the speed of light in vacuum and $\alpha = \frac{e^2}{\hbar c} \approx 1/137$ is the fine-structure constant. The atomic unit of length is the bohr, $a_0 = \frac{1}{\alpha}\frac{\hbar}{m_e c} \approx 0.529$ Å. The atomic unit of cross section is $a_0^2 \approx 28.0$ Mb, where a *barn* (b) equals 10^{-28} m^2. The atomic unit of energy is the hartree, $E_h = m_e c^2 \alpha^2 \approx 27.2$ eV.

Depending on the photon energy, we typically distinguish between the following two x-ray regimes [7, 8]. Photons with an energy between ~10 E_h (~300 eV) and ~100 E_h (a few keV) are called *soft x-rays*. Soft x-rays cover, roughly, the 1s binding energies for elements ranging from carbon

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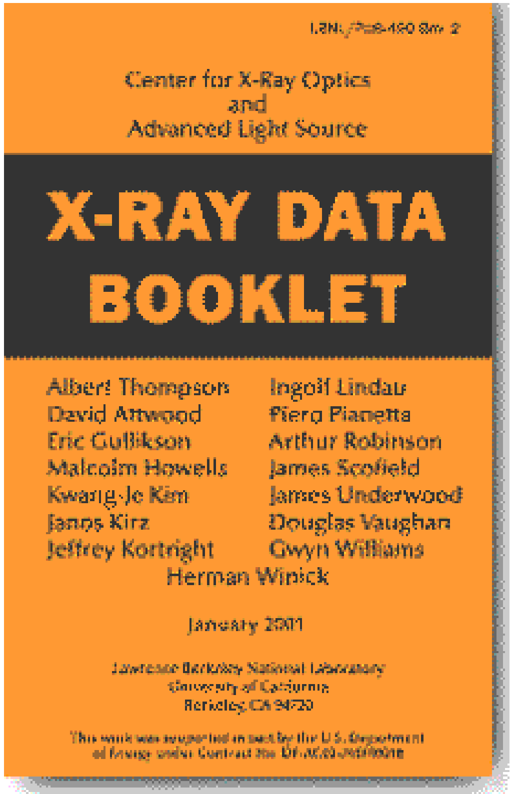
Practical info



http://xdb.lbl.gov/

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