Modelling the interaction of matter with ultrafast and ultraintense x-ray pulses

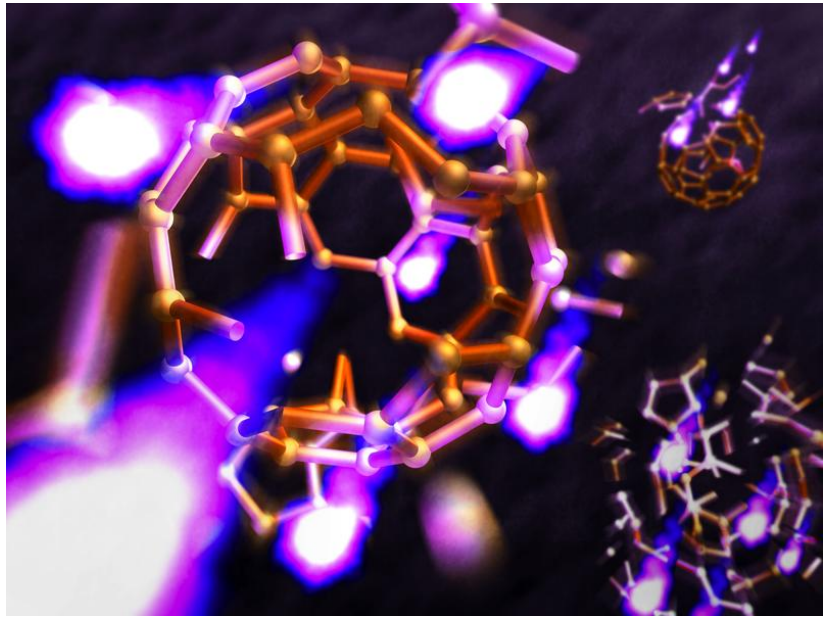
Ludger Inhester, Sang-Kil Son, Zoltan Jurek
FS-CFEL-3
Center for Freel-Electron Laser Science CFEL,
Deutsches Elektronen-Synchrotron DESY
FS computing meeting, 10.11.2023

HELMHOLTZ

CFEL SCIENCE

DESY.

XRAYPAC - a software package for modeling x-ray-induced dynamics of matter

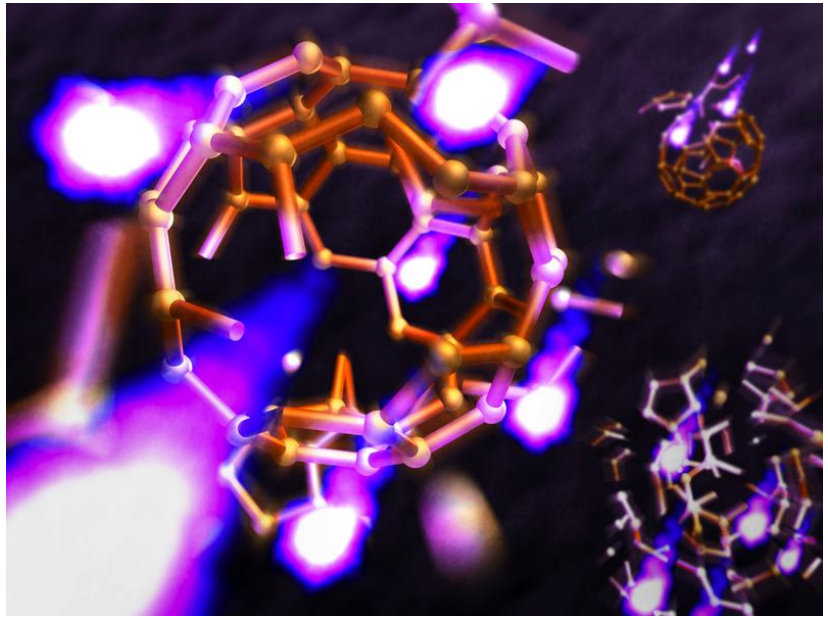


Artist's impression of a buckyball in an X-ray laser flash.
Credit: Greg Stewart/SLAC

- **XATOM**: Atomic electronic structure calculations, interaction of atoms with intense x-ray pulses
- **XMOLECULE**: Molecular electronic structure calculations, ab-initio molecular dynamics
- **XMDYN**: Molecular dynamics based on force-fields including ionization dynamics (photoinduced and collisional ionization)

XRAYPAC is currently distributed under a DESY licence, but is soon to be published as open source https://www.desy.de/~xraypac/

XRAYPAC - a software package for modeling x-ray-induced dynamics of matter



Artist's impression of a buckyball in an X-ray laser flash.
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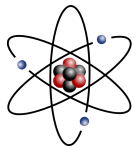
single atom



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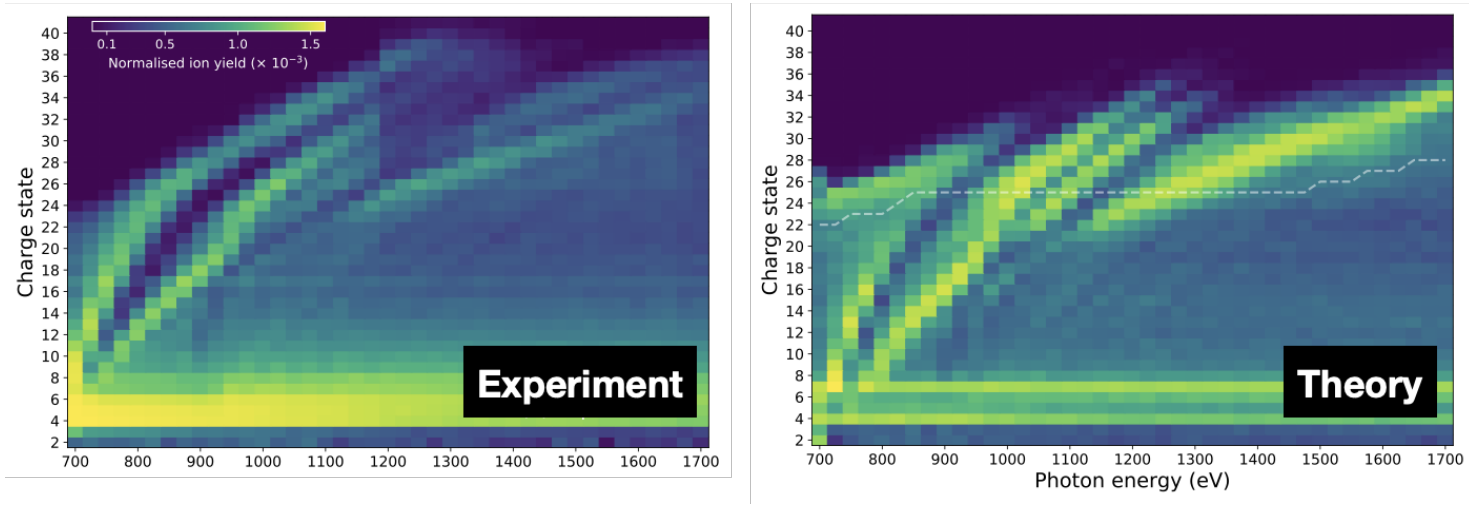
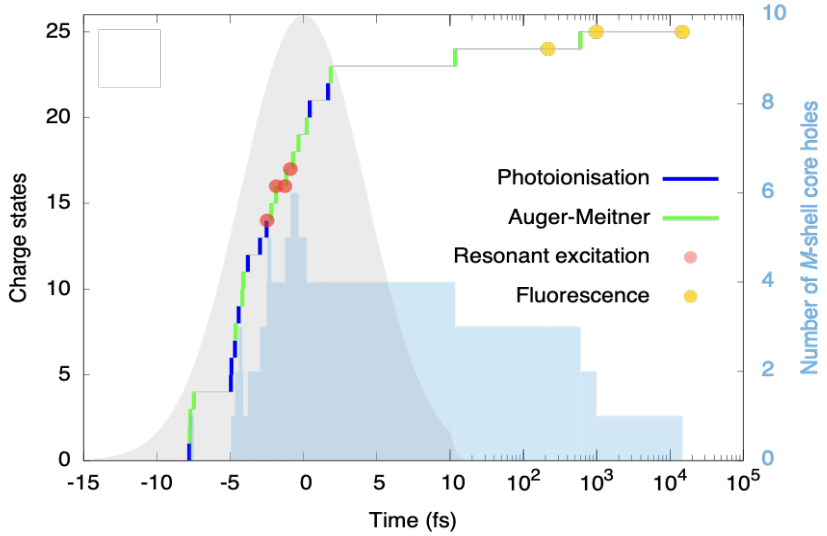
XATOM: Multiple-core-hole resonance spectroscopy of heavy atoms with ultraintense X-ray pulses

- Fundamental interaction between atoms and intense XFEL pulses: **X-ray multiphoton multiple ionization dynamics**
- Xe ion yields measured at the European XFEL as a function of charge state and photon energy
- Ionization dynamics simulation of Xe including resonances: requires to handle ~4.2×10^{60} rate equations ➔ Monte Carlo on the fly
- Quantitative comparison between theory and experiment
- Theory demonstrates that **resonance peaks are formed via massively hollow atoms** featuring as many as six simultaneous core holes

Aljoscha Rörig *et al.*, *Nat. Commun.* **14,** 5738 (2023)

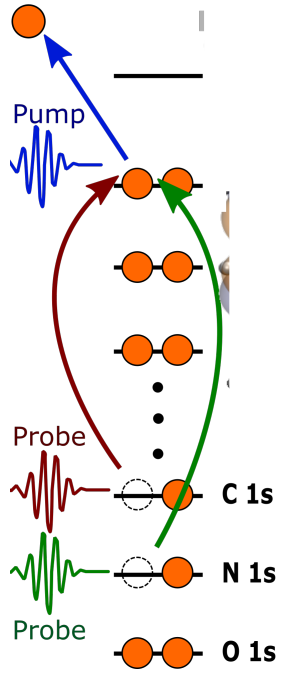


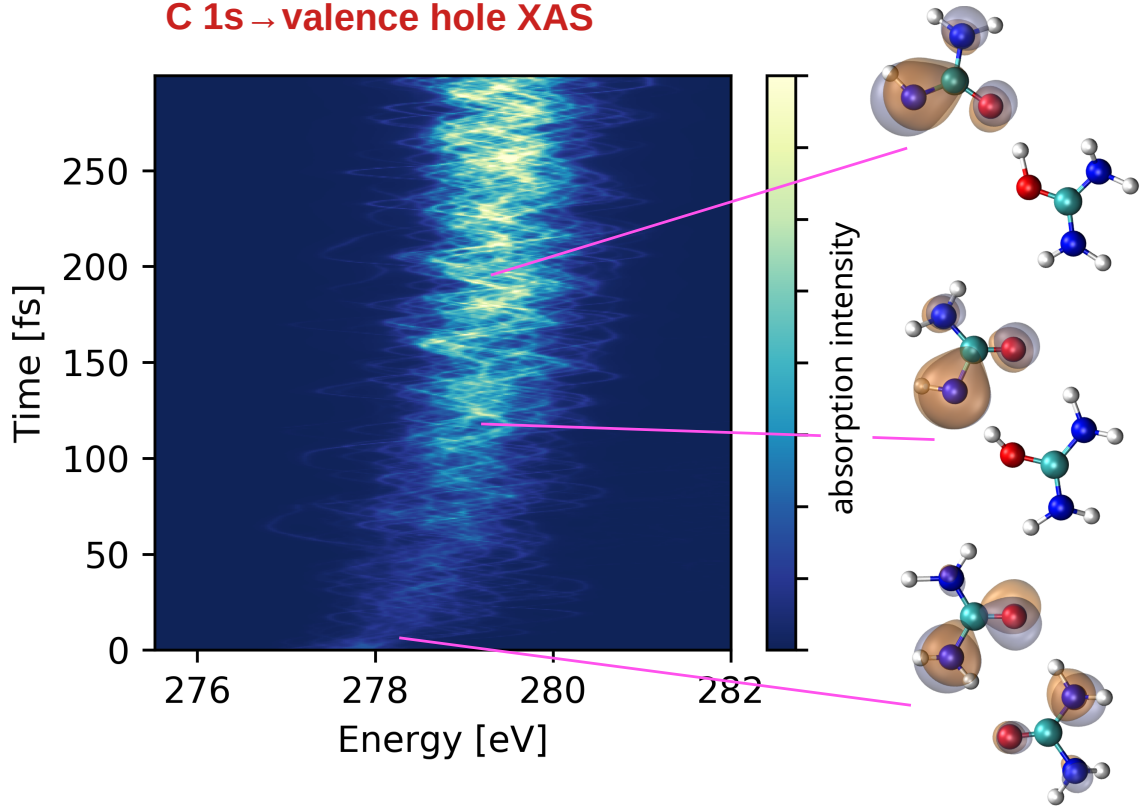


Exemplary ionization pathway of Xe^{25+} at 1250 eV

XMOLECULE: Femtosecond proton transfer in urea solutions probed by X-ray spectroscopy

- **Response of urea to ionizing radiation** is a cornerstone in theories for the development of life on earth (within the “warm little pond scenario”)
- Liquid jet with aqueous urea solution, 400nm pump, HHG-based soft-X-ray probe
- Femtosecond-resolved X-ray absorption spectroscopy reveals: **ionization induces ultrafast proton transfer reaction** between neighboring urea molecules in the solution, thus gives rise to reactive molecular species

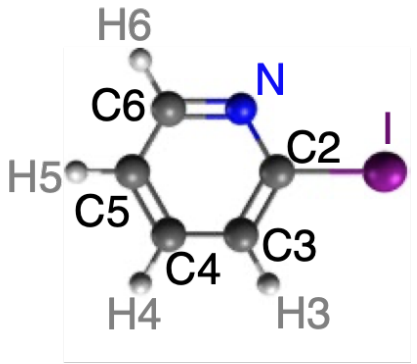


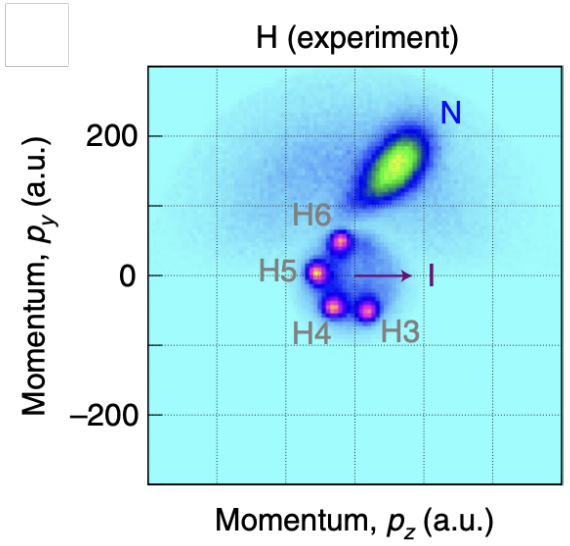


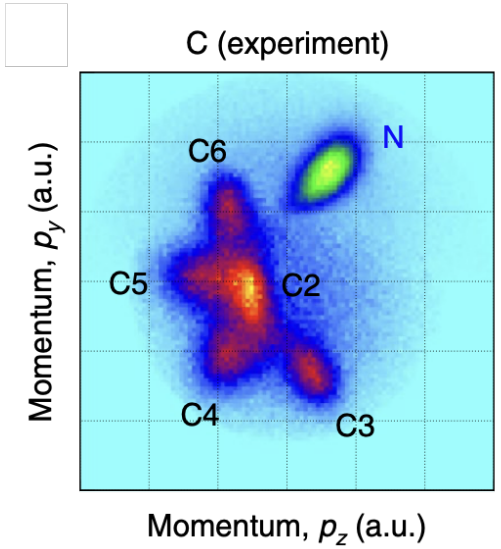
Zhong Yin *et al.*, *Nature* **619**, 749 (2023)

XMDYN: X-ray multiphoton-induced Coulomb explosion imaging of complex single molecules

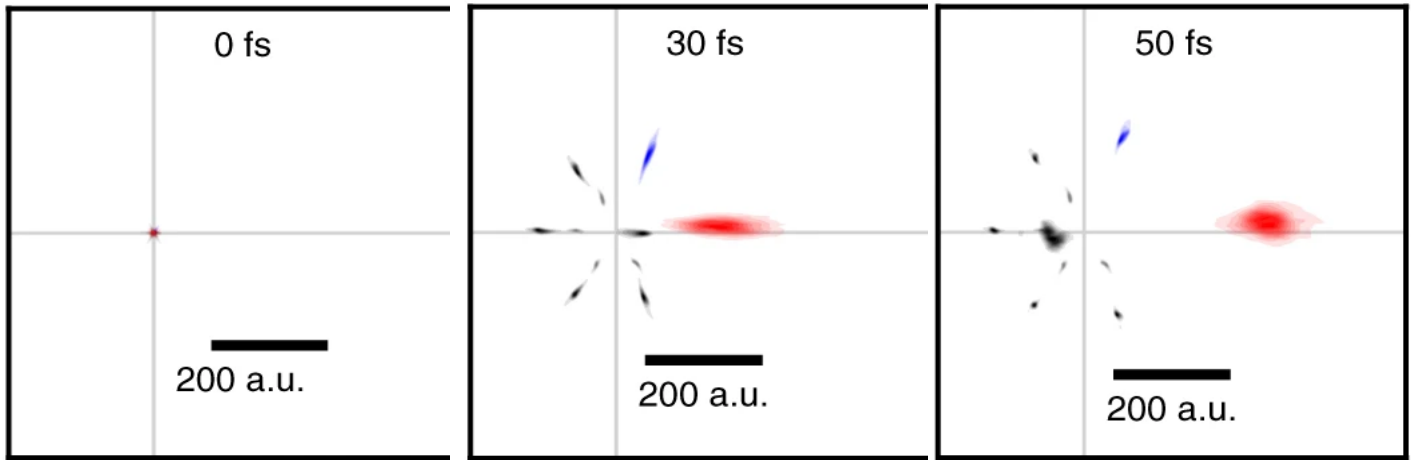
- **Following structural dynamics in real time** is a fundamental goal towards a better understanding of chemical reactions
- Intense femtosecond XFEL pulses trigger rapid and complete **Coulomb explosions** of 2-iodopyridine and 2-iodopyrazine molecules
- Multi-coincidence techniques and theoretical modelling of XMDYN: traces ultrafast hydrogen emission and intramolecular electron rearrangement







Simulation:

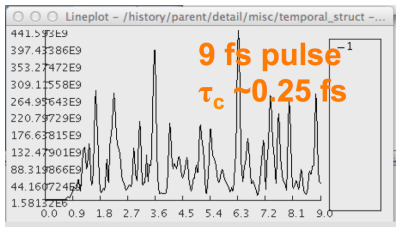


Rebecca Boll *et al.*, *Nat. Phys.* **18**, 423 (2022)

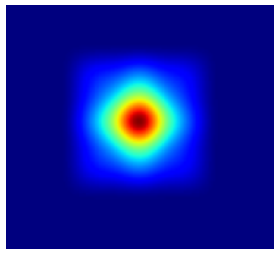
Computational challenges

Running and analyzing simulations often involves complex data analysis and processing:

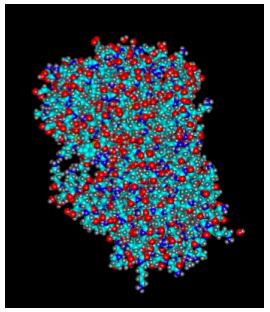
- Multiphoton multiple ionization via intense x-ray radiation involves **trillions of electronic configurations** in a single atom (e.g. xenon). Computing rates and cross section is accelerated using **neural-networks** (work in progress)
- MD simulations involve a **high-dimensional space** data with challenging data analysis tasks: E.g., finding link between motion patterns and x-ray spectroscopic features (e.g. urea). **Partial least square regression** analysis to extract dominant motion pattern in high dimensional data.
- Larger systems (proteins, Natoms ~ 10^5) to address diffractive imaging experiments require the **processing of a large amount of data**. (efficient **data storage**, i.e., hdf)
- Incorporation into **comprehensive simulation framework** of XFEL experiments (**start-to-end simulation**) involves integrating several simulation tools.



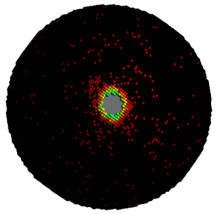
FAST



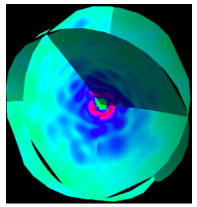
WPG



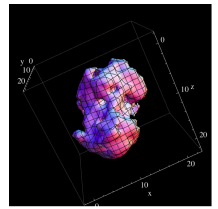
XMDYN



SingFEL



EMC & DM



http://www.xfel.eu/sims2e

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

Thank you

Contact

Deutsches Elektronen-
Synchrotron DESY

www.desy.de

Ludger Inhester
CFEL-DESY-Theory
ludger.inhester@cfel.de