Ultrafast Phenomena: Part 1

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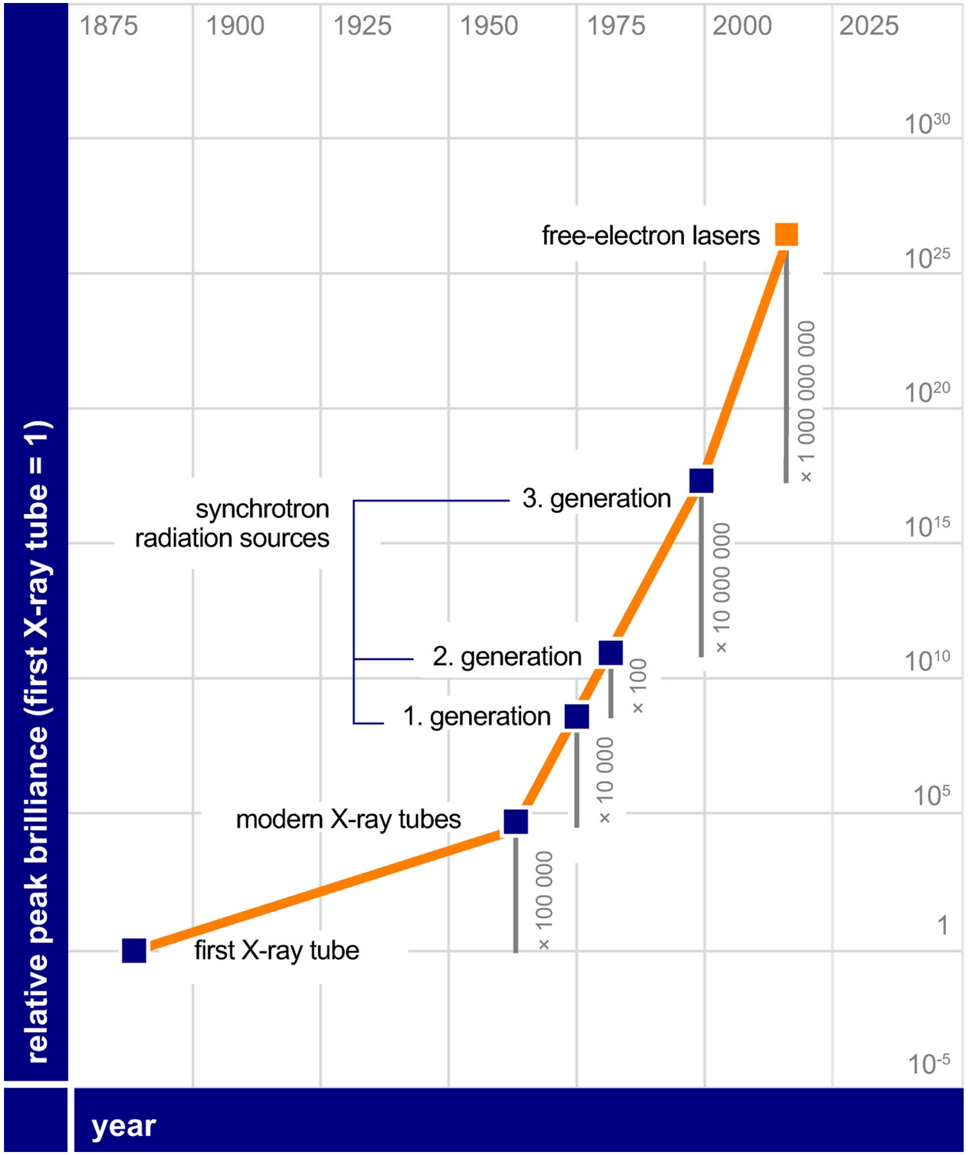
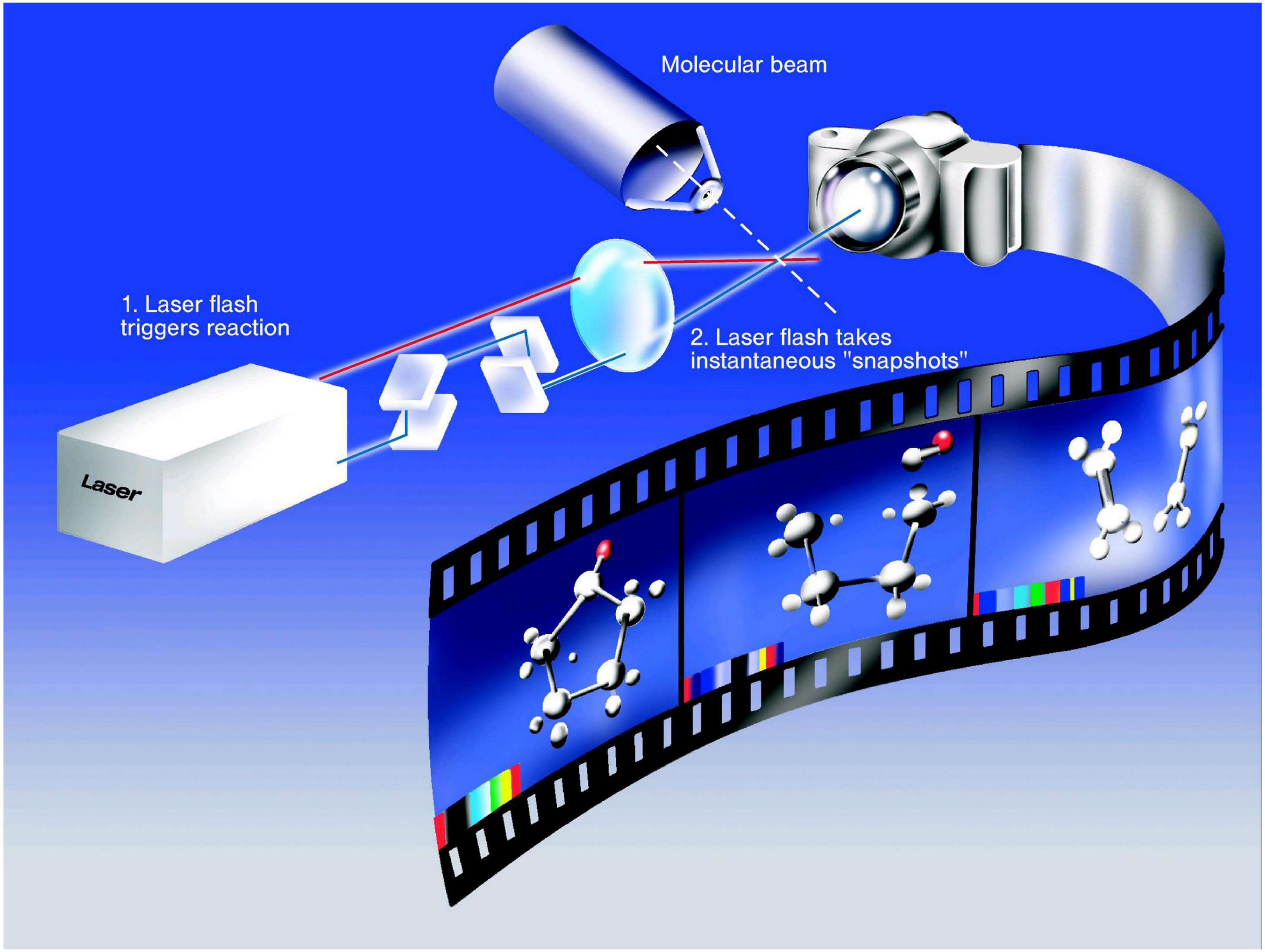
Hamburg, Germany

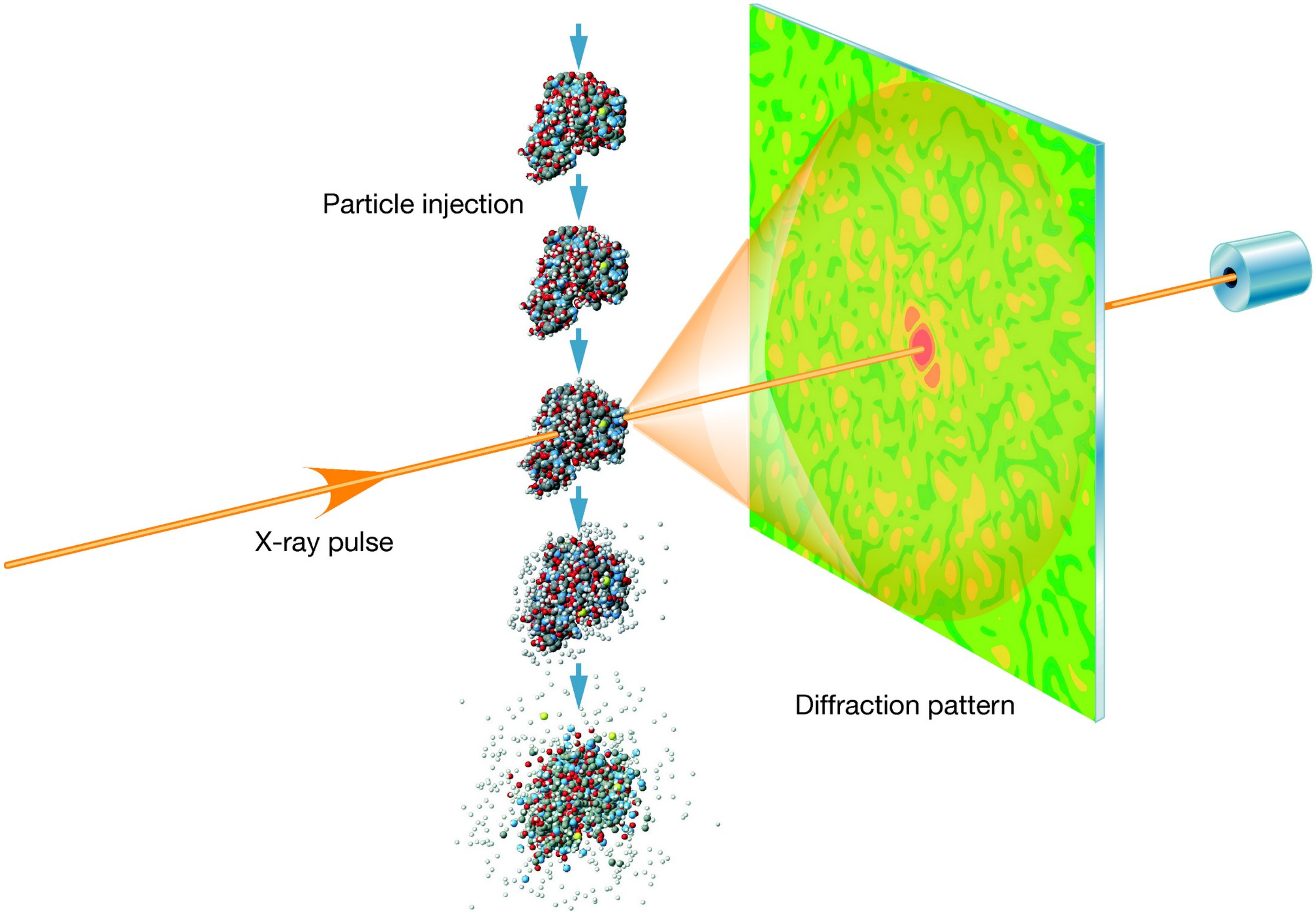
A brief history of x-ray intensity



Making molecular movies: a new tool for femtochemistry

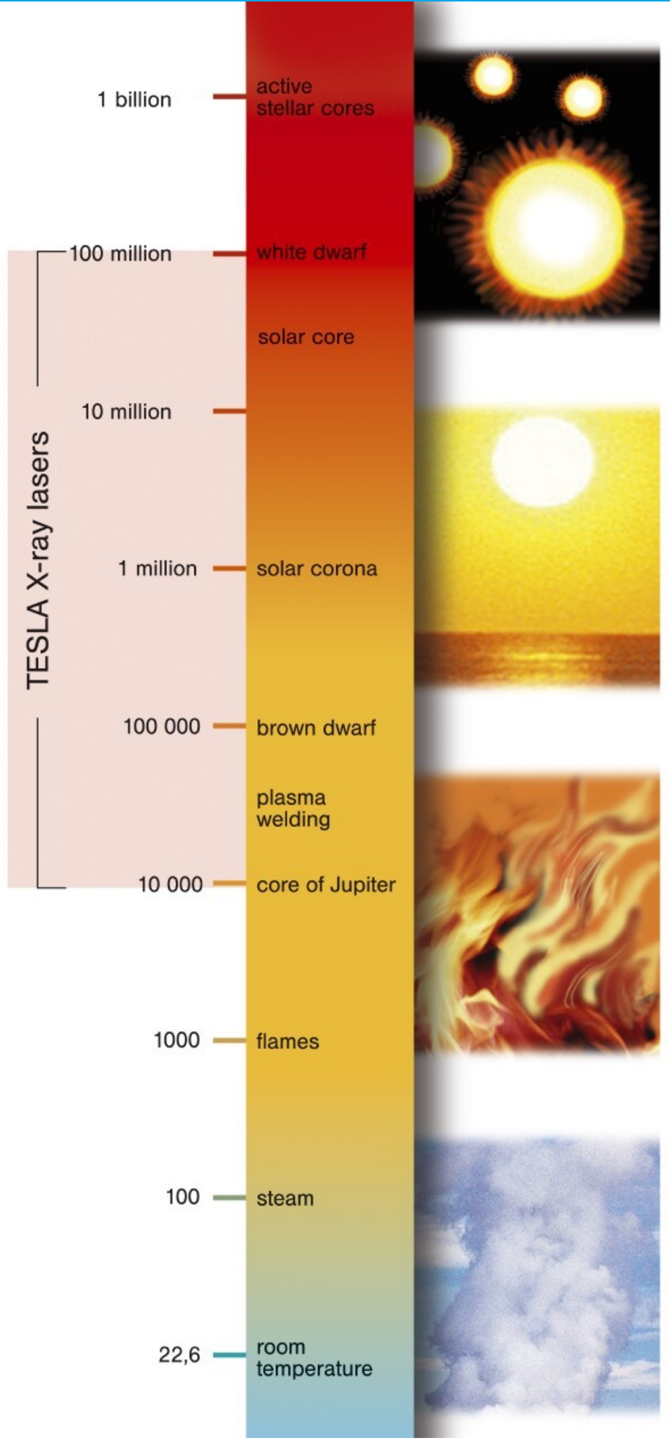


Single-shot structure determination of biomolecules

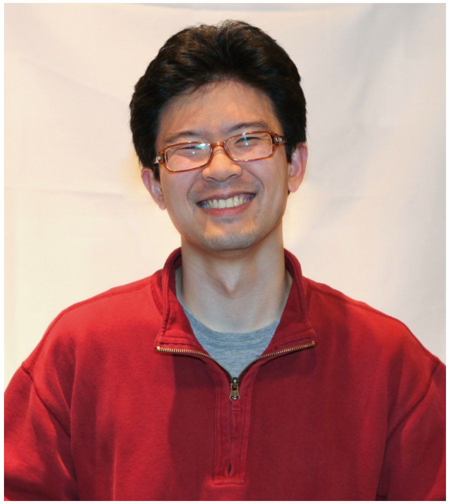


Neutze *et al.*, Nature **406**, 752 (2000).

Generating and probing extreme states of matter



Sang-Kil Son

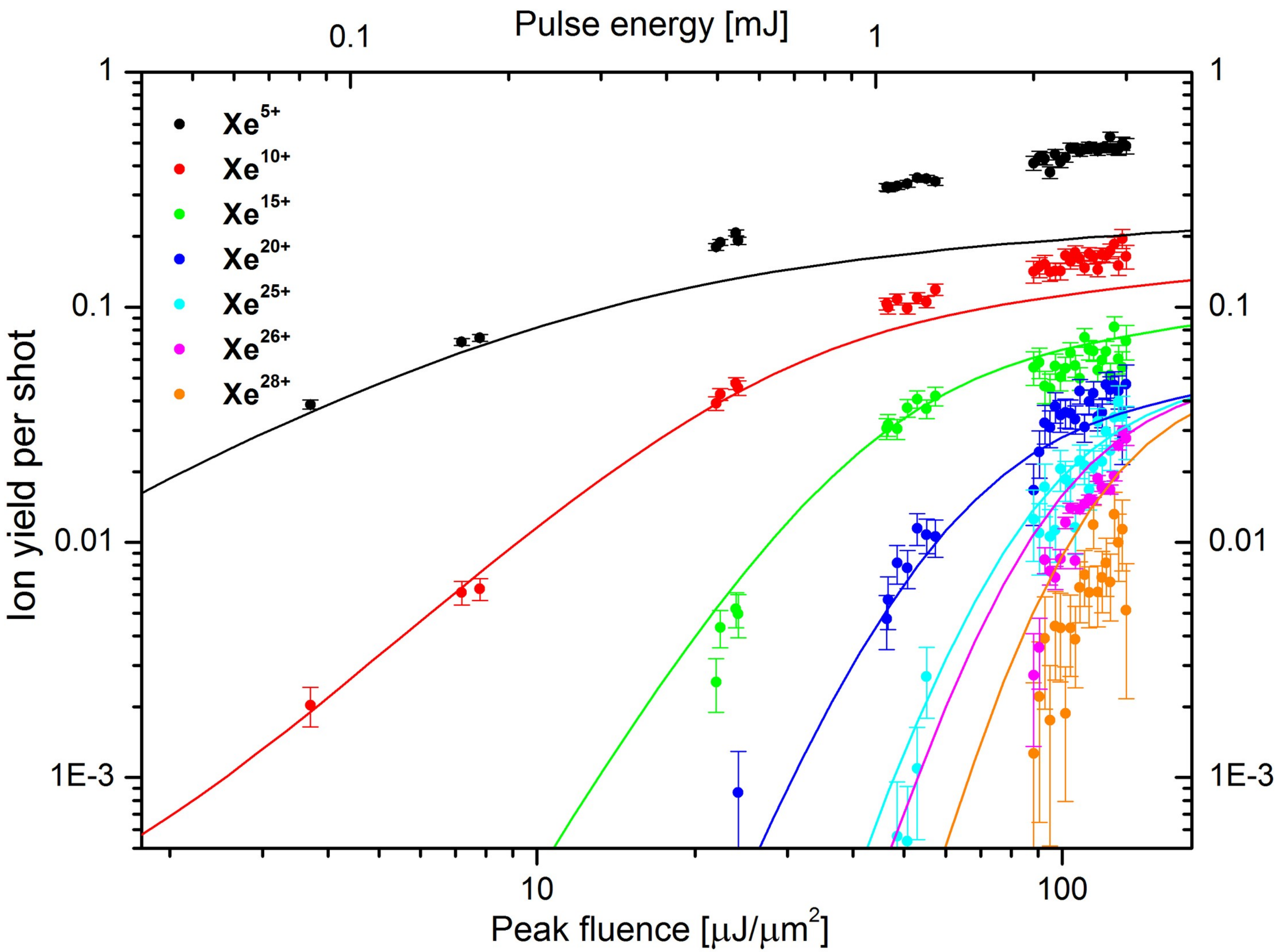


XATOM:
an integrated toolkit for x-ray atomic physics at high intensity

→ ab initio calculation of atomic parameters (subshell photoionization cross sections, electronic decay rates, x-ray scattering cross sections) for arbitrary electronic configurations

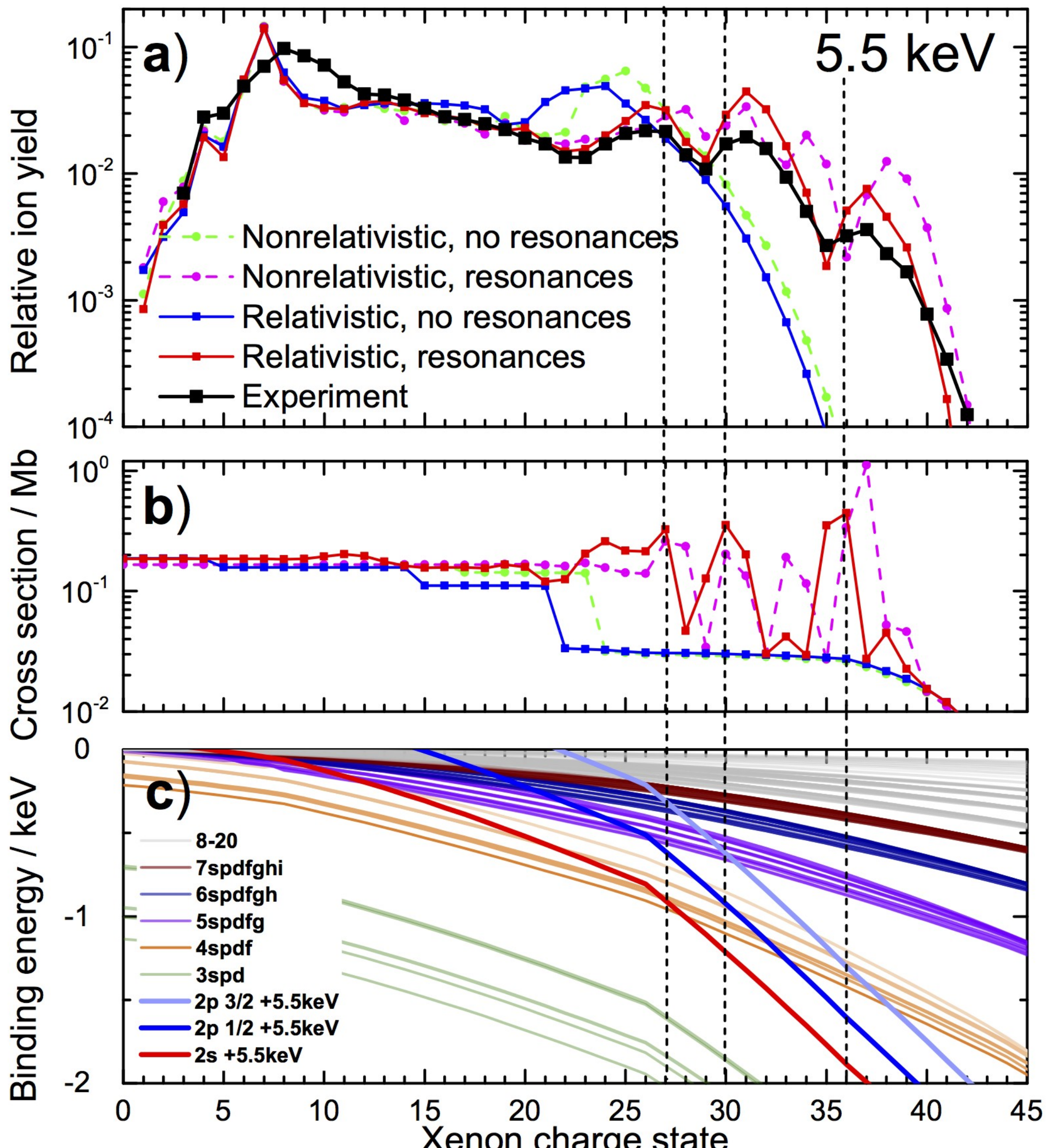
→ description of electronic population dynamics via numerical solution of system of coupled rate equations (one rate equation per electronic configuration)

Comparison between experiment and theory for Xe at 2 keV



B. Rudek *et al.*, Nature Photonics **6**, 858 (2012).

Relativistic and resonant effects in the ionization of heavy atoms by ultra-intense hard x rays



Xe at an x-ray peak intensity exceeding 10^{19} W/cm^2

B. Rudek *et al.*, Nature Commun. **9**, 4200 (2018).

Dramatic increase in the number of coupled rate equations

- Nonrelativistic, no resonances

  → ***23,532,201*** *configurations*

- Relativistic, no resonances

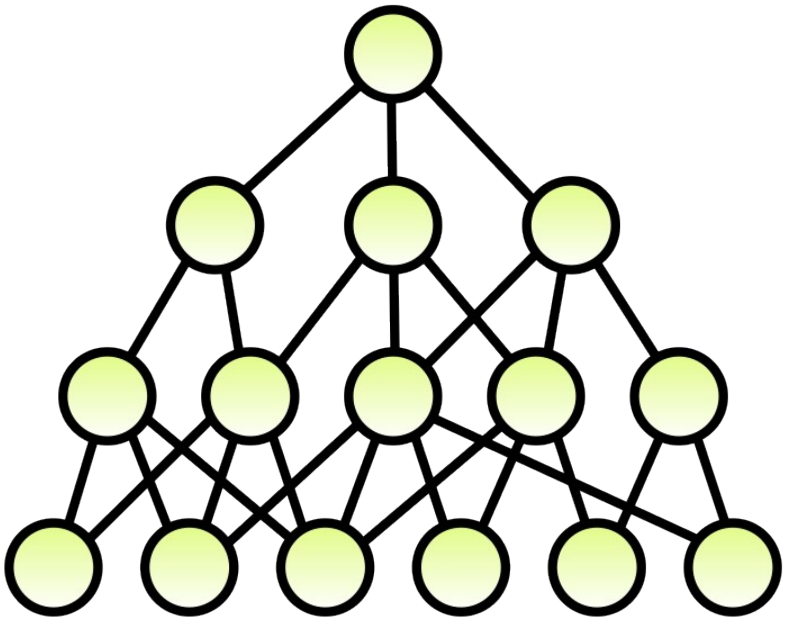
  → ***5,023,265,625*** *configurations*

- Relativistic, including resonances (n_{max} = 30, l_{max} = 7)

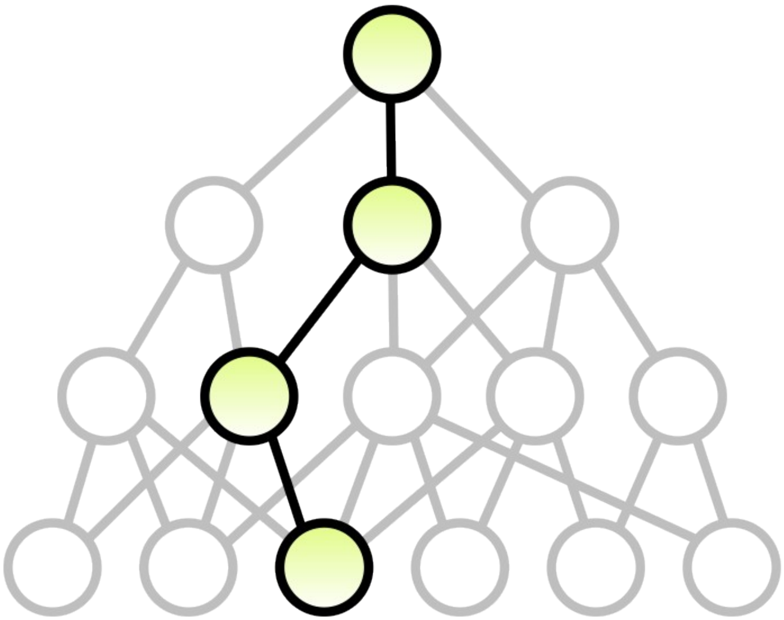
  → ***2.6 × 10⁶⁸*** *configurations*

(ionization from the K shell is excluded in all three cases listed)

Direct integration of rate equations vs. Monte Carlo



solved only once
with all possible pathways



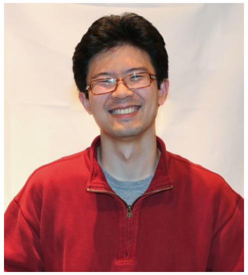
repeated many times

XMOLECULE

Yajiang Hao

Ludger Inhester

Kota Hanasaki



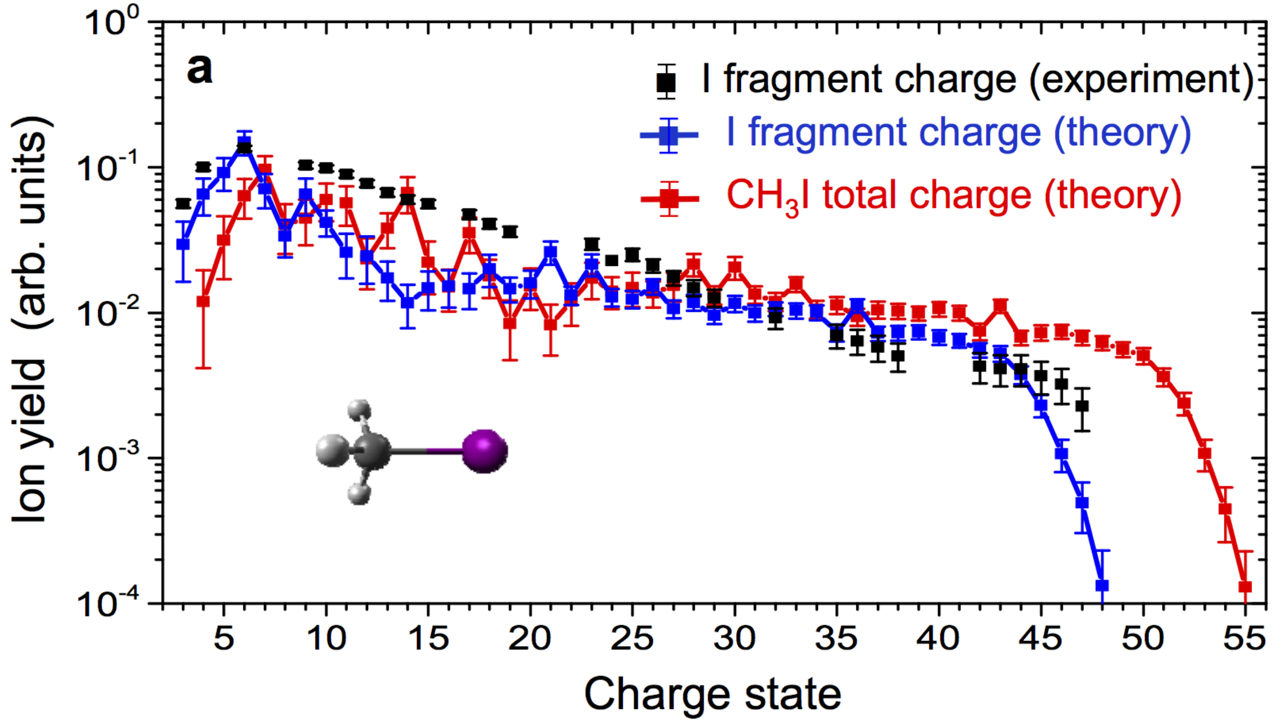
Sang-Kil Son

> An ab-initio electronic-structure approach dedicated to ionization dynamics of molecules

> Self-consistent-field calculation for every electronic configuration formed during interaction with intense XFEL pulse

> Demonstration of a new ionization enhancement mechanism

The highest charge states ever produced using light!



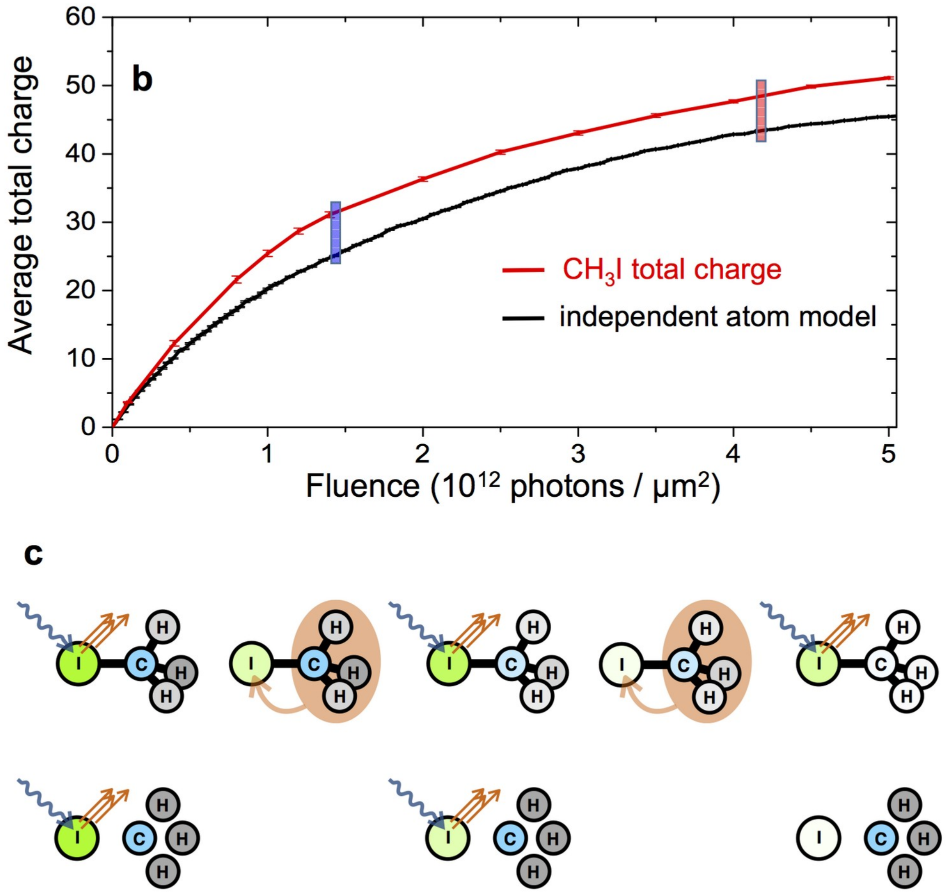
Photon energy: 8.3 keV

X-ray peak intensity:
> 10^{19} W/cm^2

Experimental data taken by Artem Rudenko, Daniel Rolles, and collaborators

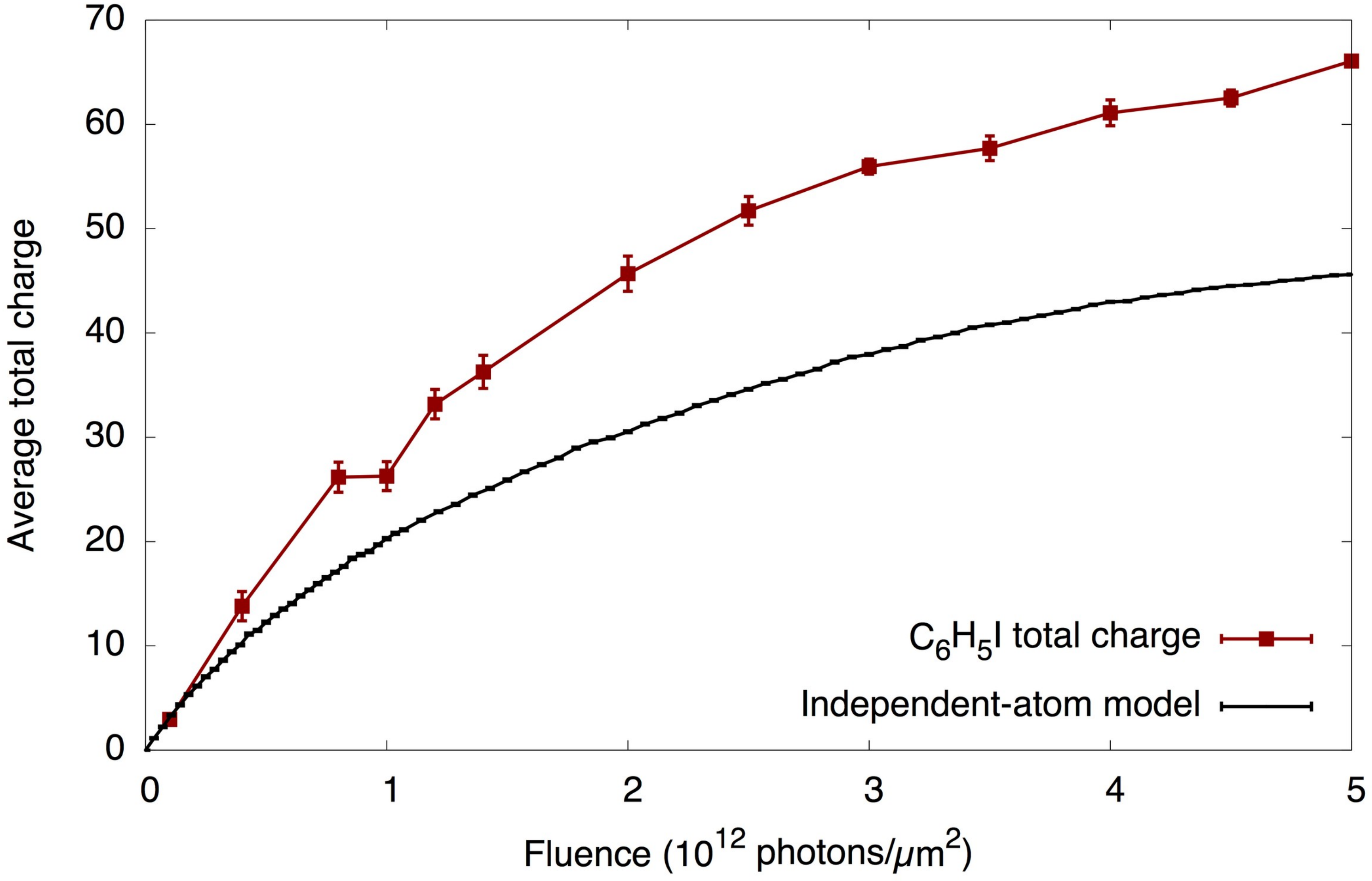
A. Rudenko *et al.*, Nature **546**, 129 (2017).

New ionization enhancement mechanism (molecular effect!)



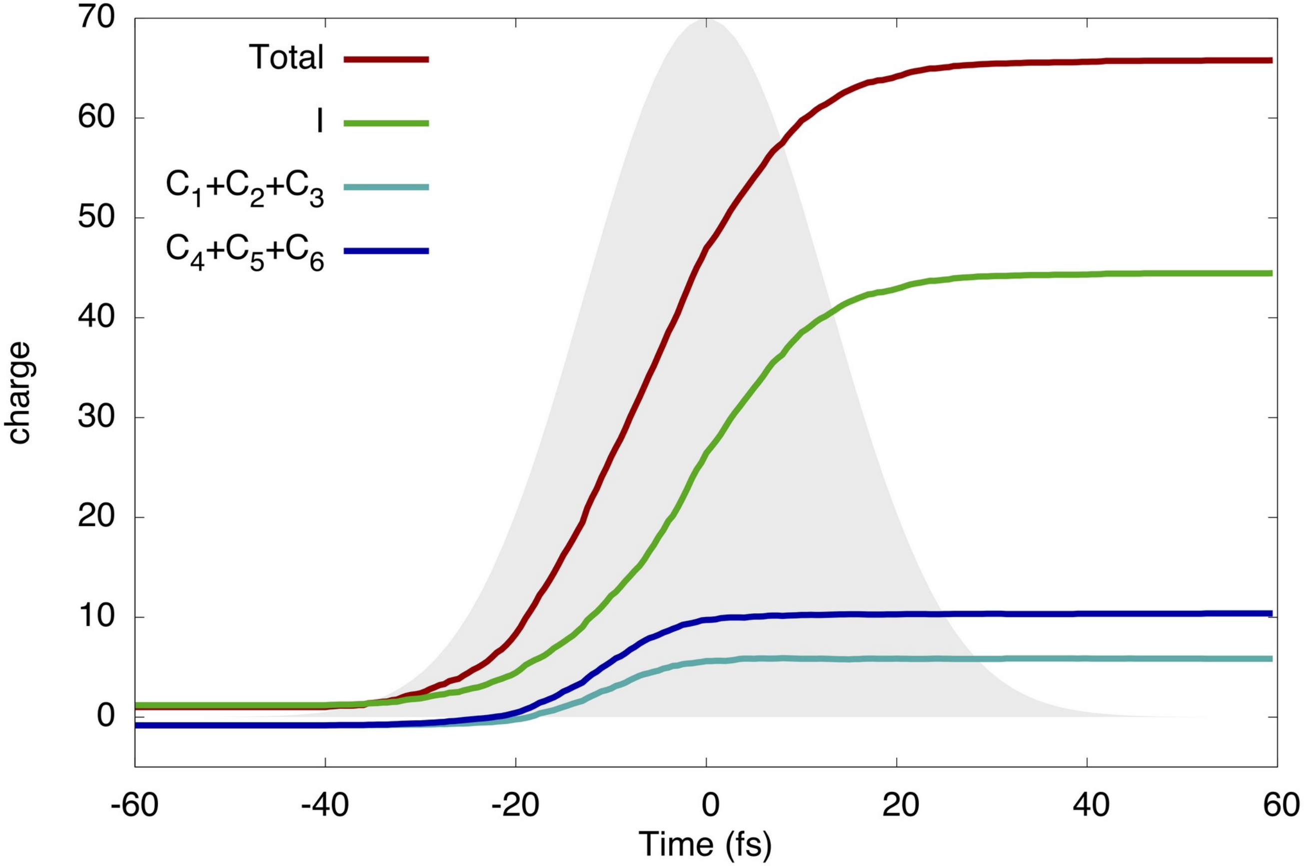
A. Rudenko *et al.*, Nature **546**, 129 (2017).

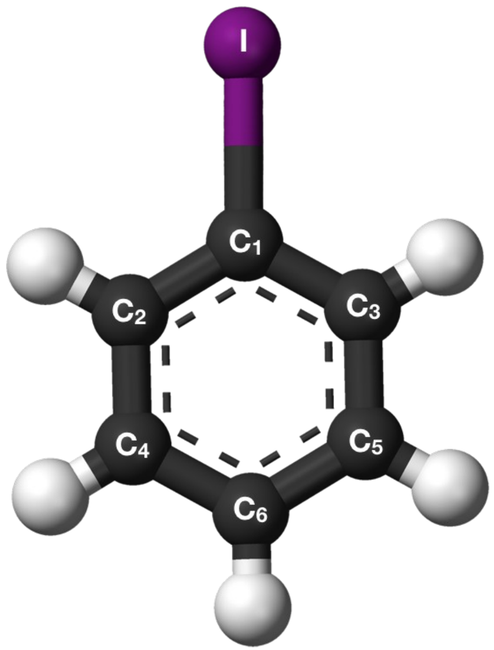
Iodobenzene (photon energy 8.3 keV)



Y. Hao *et al.*, Phys. Rev. A **100**, 013402 (2019).

Ionization dynamics in iodobenzene
(photon energy 8.3 keV, fluence 5×10^{12} photons/μm^2)

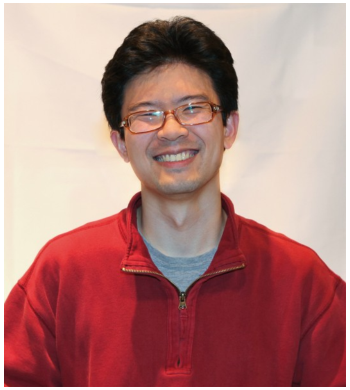




Y. Hao *et al.*, Phys. Rev. A **100**, 013402 (2019).

XMDYN

Zoltan Jurek



Sang-Kil Son

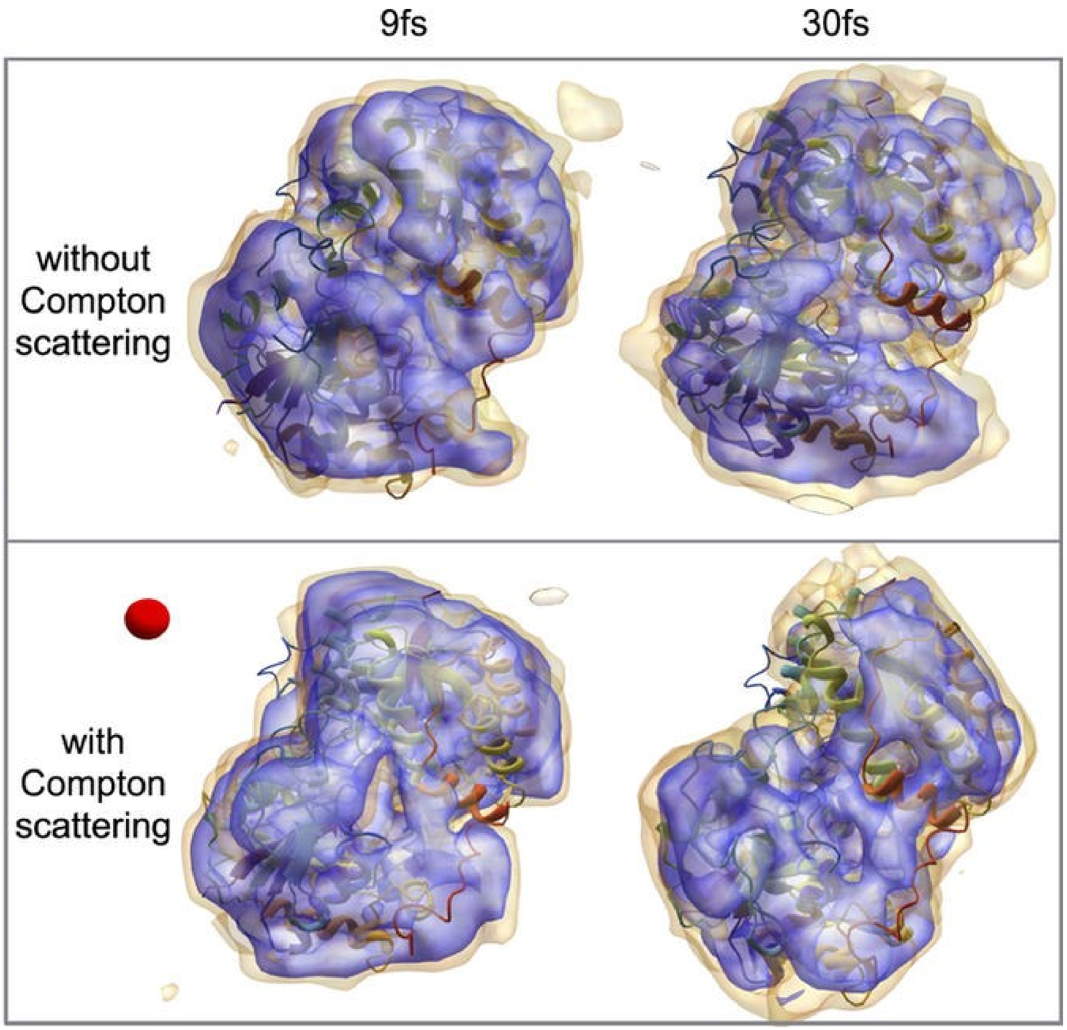
→ ab-initio calculation of atomic parameters (subshell photoionization cross sections, electronic decay rates, x-ray scattering cross sections) for arbitrary electronic configurations → uses XATOM

→ description of electronic population dynamics via Monte Carlo

→ classical molecular dynamics for nuclei and ionized electrons

XMDYN is part of a start-to-end simulation framework for single-particle imaging at the European XFEL

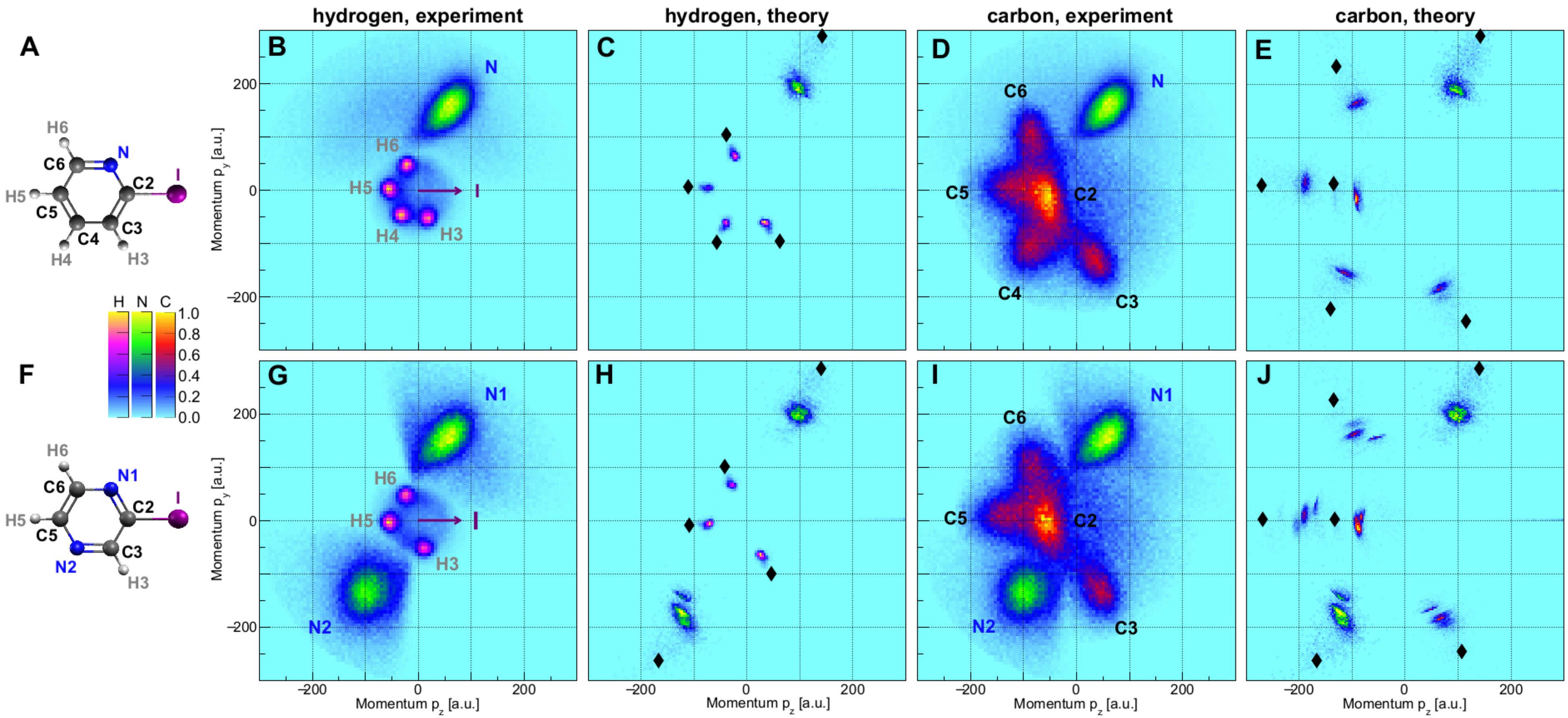
Red reference sphere has a diameter of 7 Å



nitrogenase iron protein

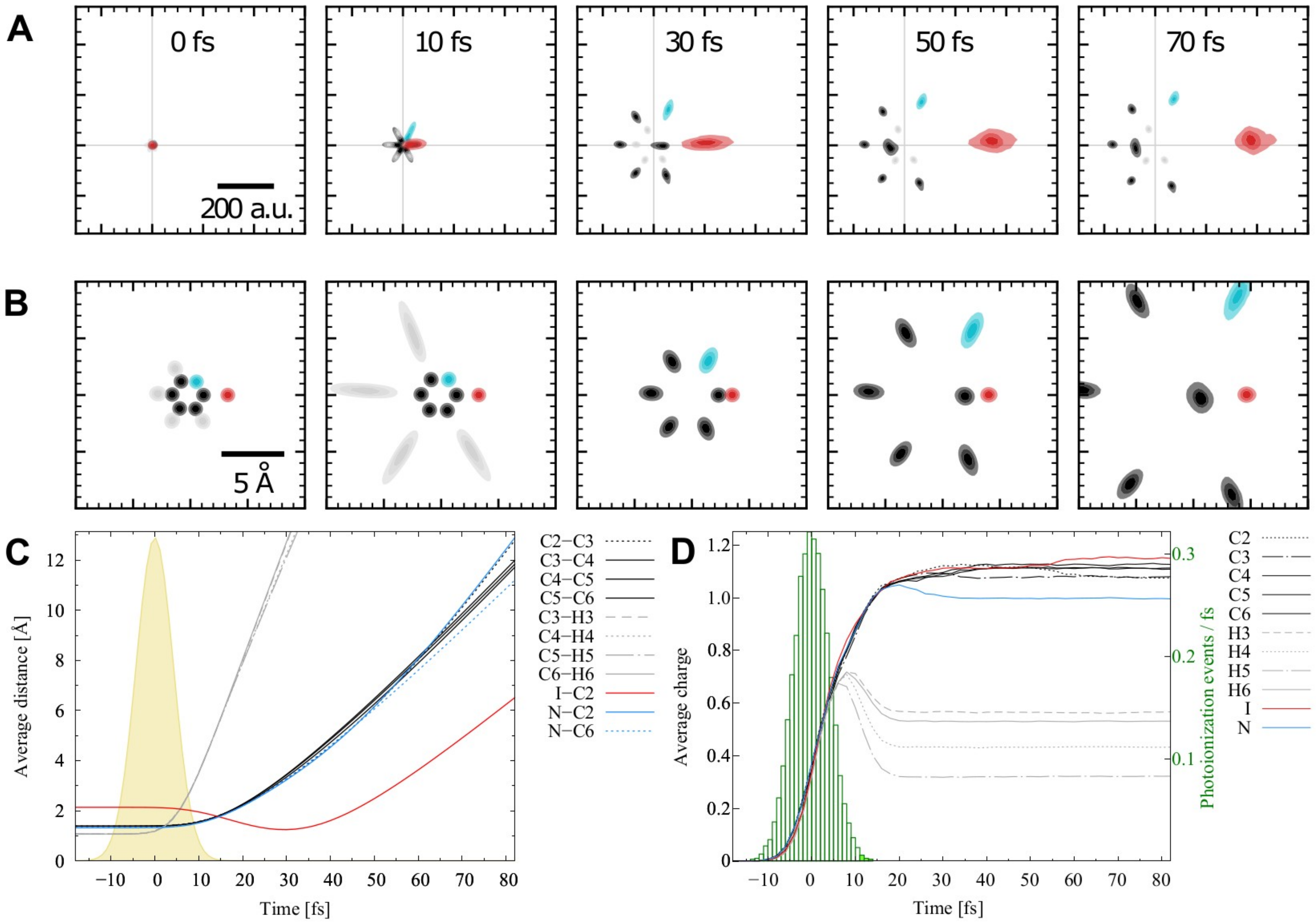
C. H. Yoon *et al.*, Sci. Rep. **6**, 24791 (2016).
C. Fortmann-Grote *et al.*, IUCrJ **4**, 560 (2017).

Demonstration of XFEL-CEI at the European XFEL



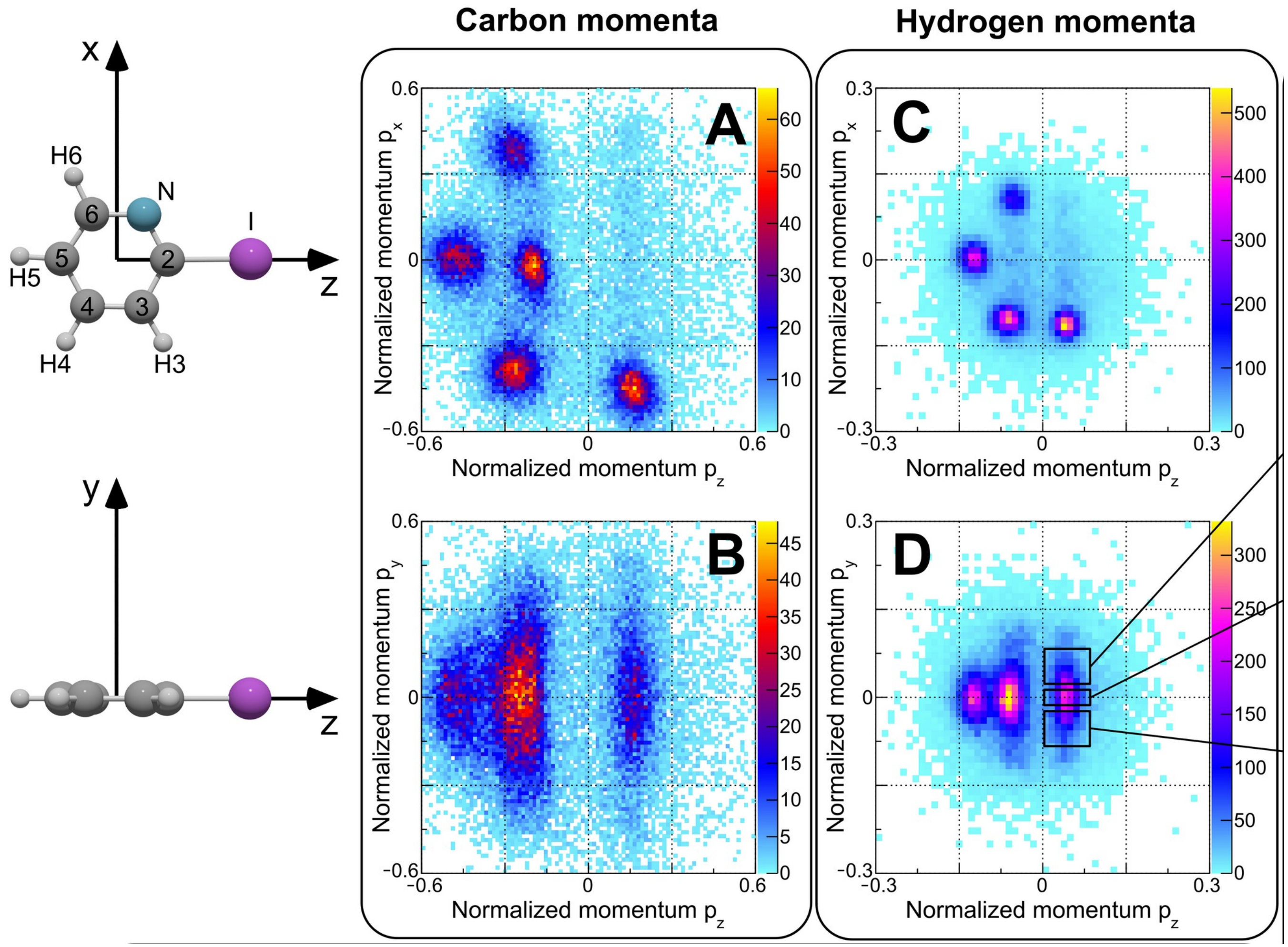
R. Boll *et al.*, Nature Phys. **18**, 423 (2022).
See also Physics Today **75**, 5, 12 (2022).

Theory provides mapping between asymptotic ion momenta and initial real-space structure



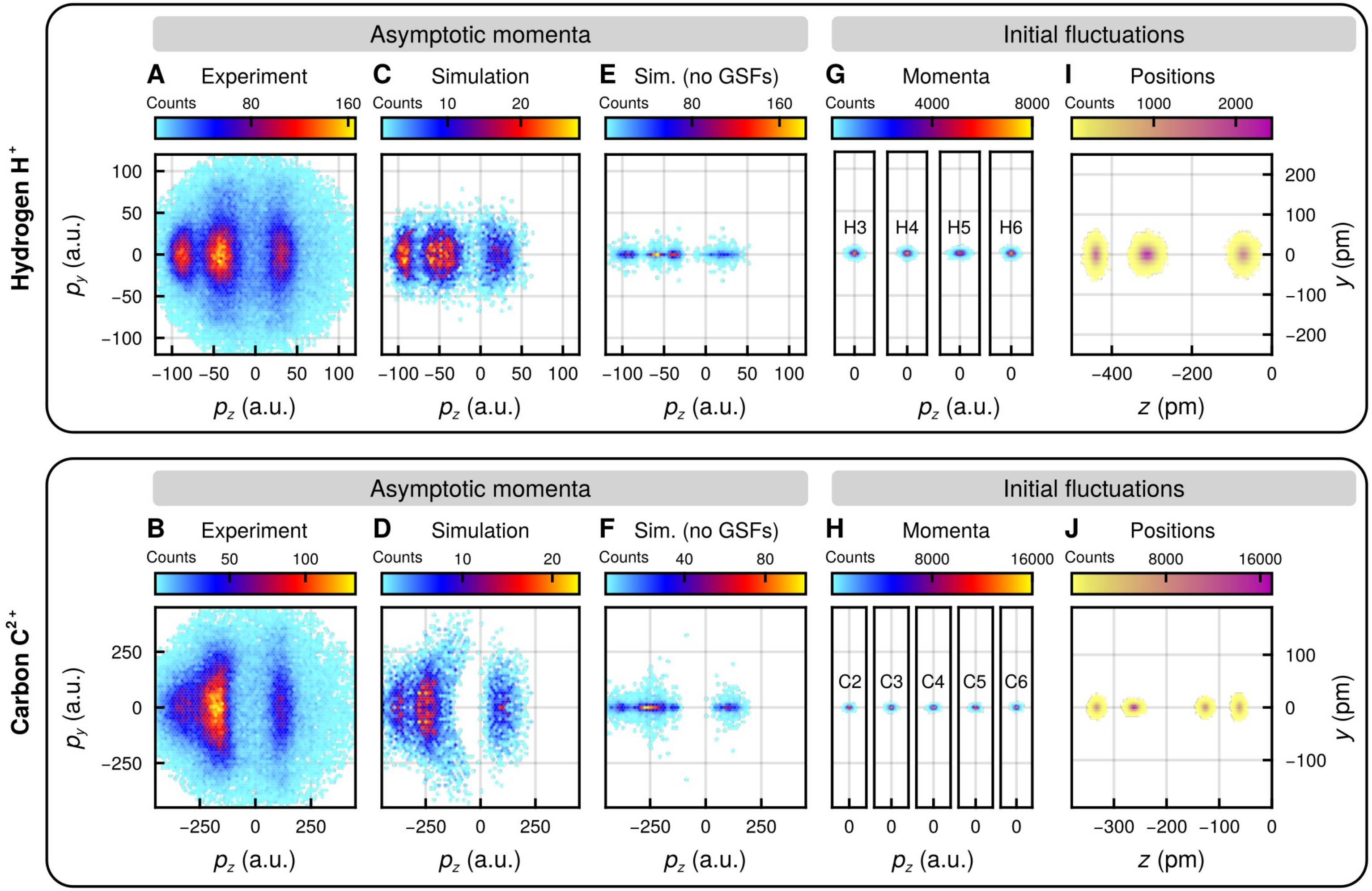
R. Boll *et al.*, Nature Phys. **18**, 423 (2022).

4-fold ion coincidence events (I^{4+}, N^{2+}, C^{2+}, H^{+}): in plane and out of plane



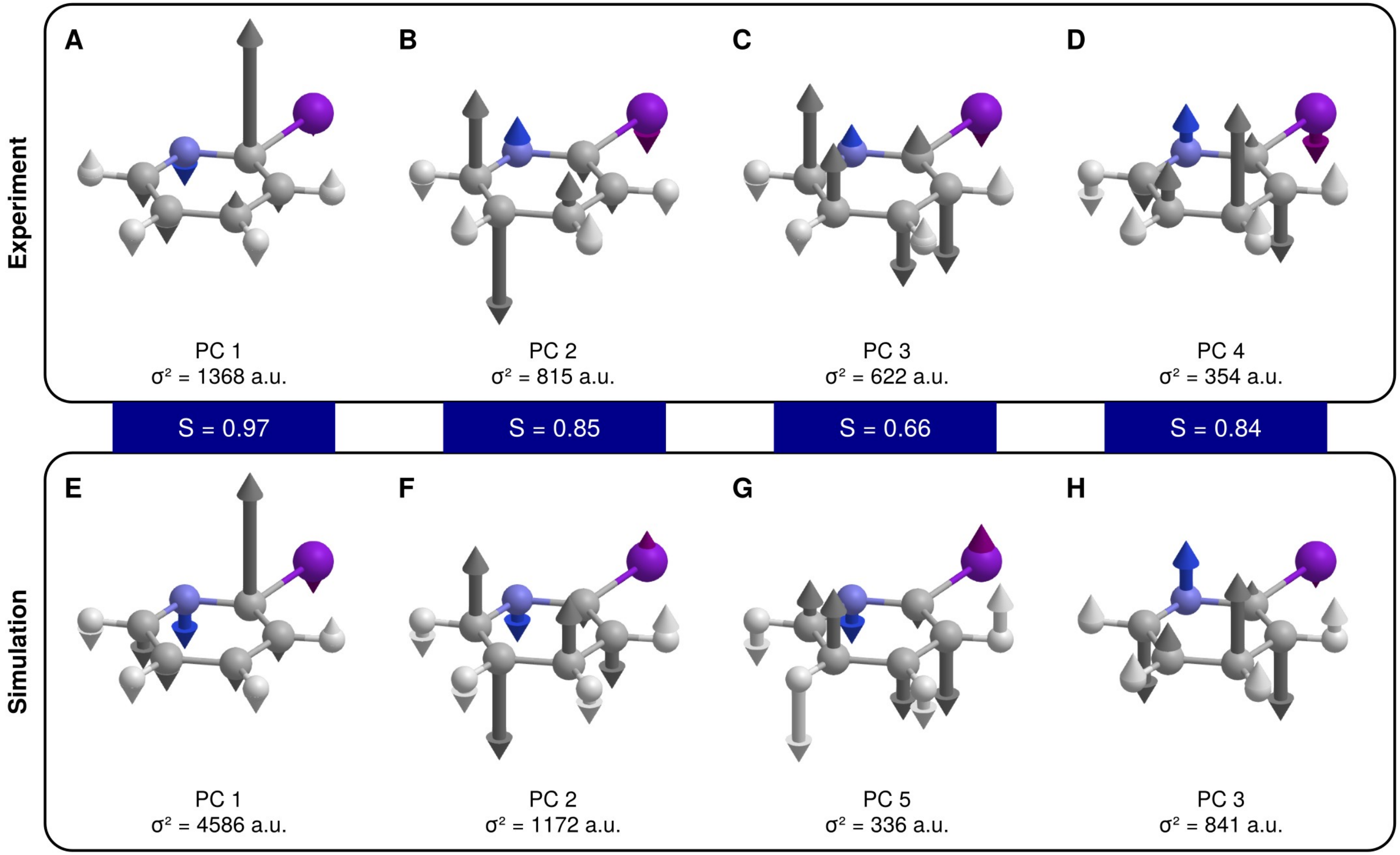
B. Richard *et al.*, Science **389**, 650 (2025).

Impact of ground-state quantum fluctuations



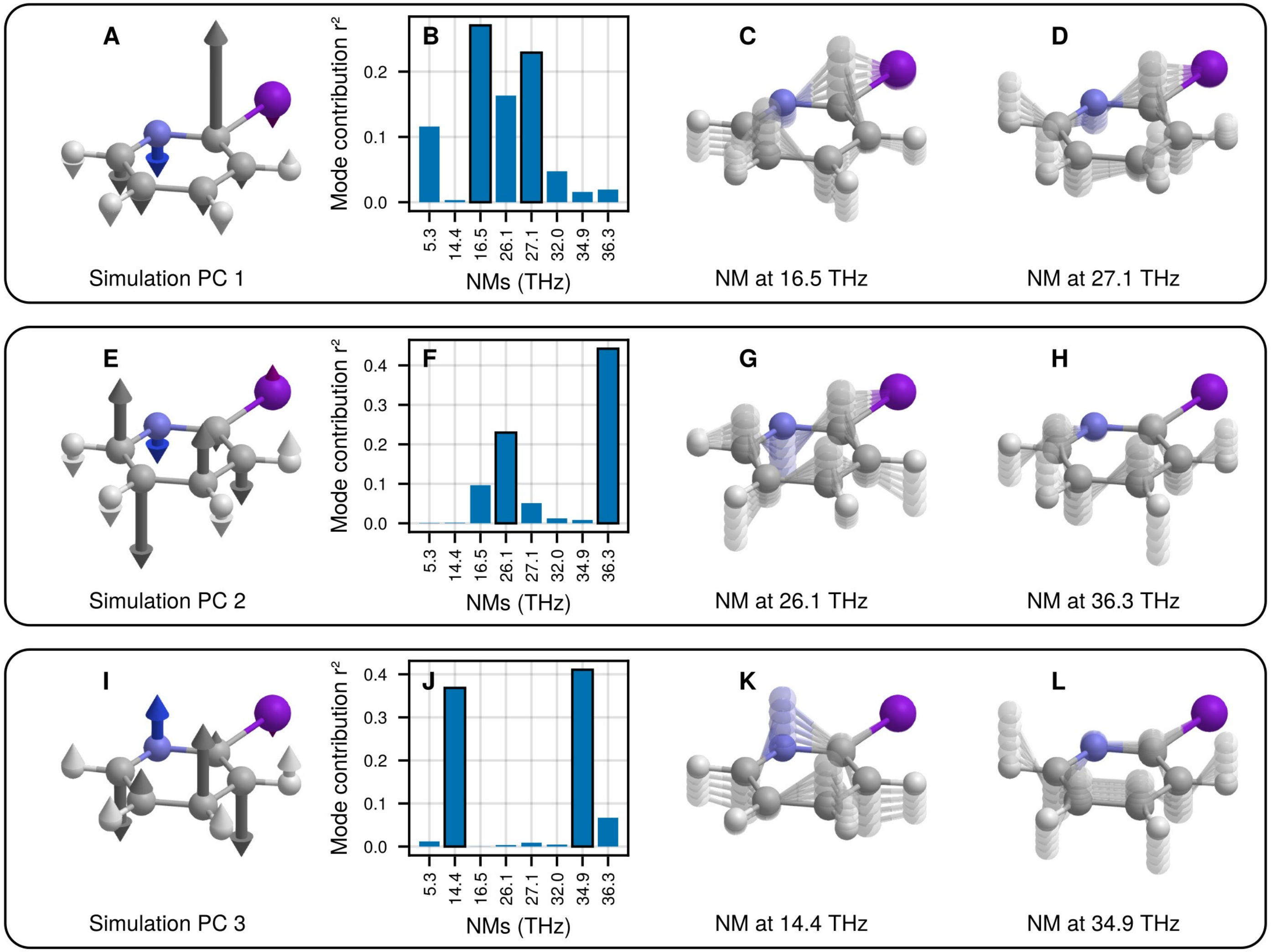
B. Richard *et al.*, Science **389**, 650 (2025).

Capturing the collective nature of the quantum fluctuations



B. Richard *et al.*, Science **389**, 650 (2025).

Mapping between the asymptotic-momentum fluctuations and the initial fluctuations with respect to the normal modes



B. Richard *et al.*, Science **389**, 650 (2025).

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1: CFEL, DESY
4: European XFEL

Conclusions

- Radiation damage at high x-ray intensity is of relevance to applications of XFELs.
- Systematic development of software for quantitative calculation of x-ray-driven effects in matter: XATOM, XMOLECULE, XMDYN.
- Very high charge states are formed as a consequence of the sequential absorption of multiple photons, combined with electronic decay cascades associated with hole formation in deep inner shells.
- Impact of relativistic and resonant effects.

Conclusions

> Electron transfer in polyatomic systems can lead to significantly enhanced ionization.

> Efficient ionization enables Coulomb explosion imaging (XFEL-CEI).

> Direct demonstration of the collective nature of structural quantum fluctuations in a complex molecule has been achieved.

> Advantage of XFEL-CEI is that such high molecular charge states can be reached that the impact of chemical bonds on the dynamics underlying CEI can be significantly reduced.