Phenomena at High X-ray Intensity: Part 2

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Hamburg, Germany

XMOLECULE


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Ludger Inhester


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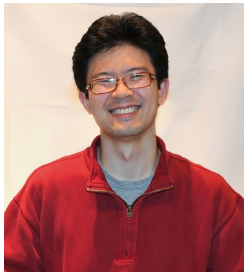
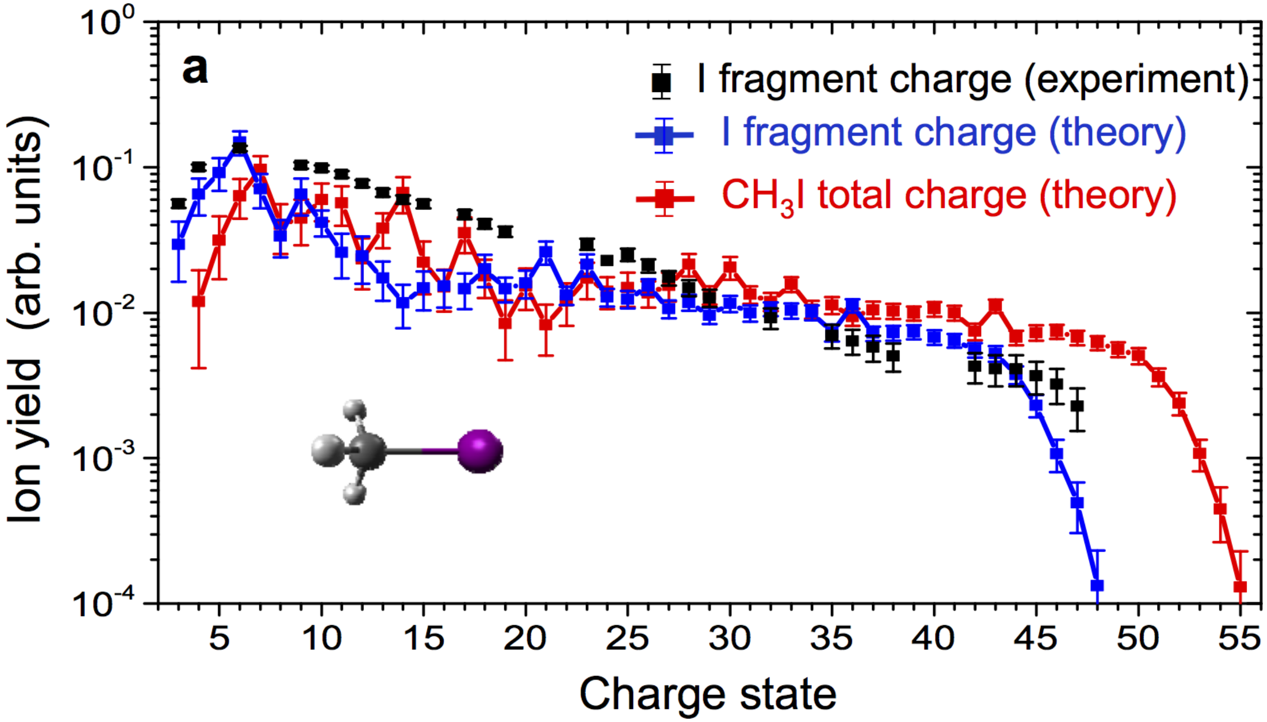

Sang-Kil Son

- 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!



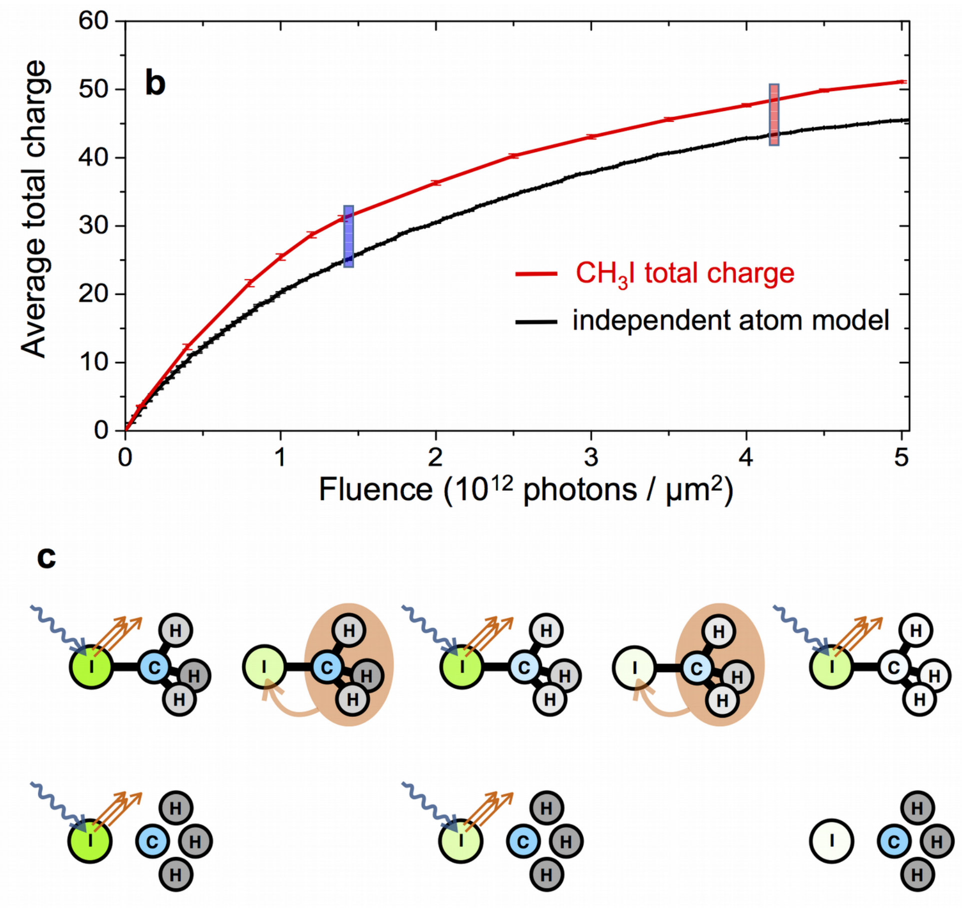
Photon energy: 8.3 keV

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

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

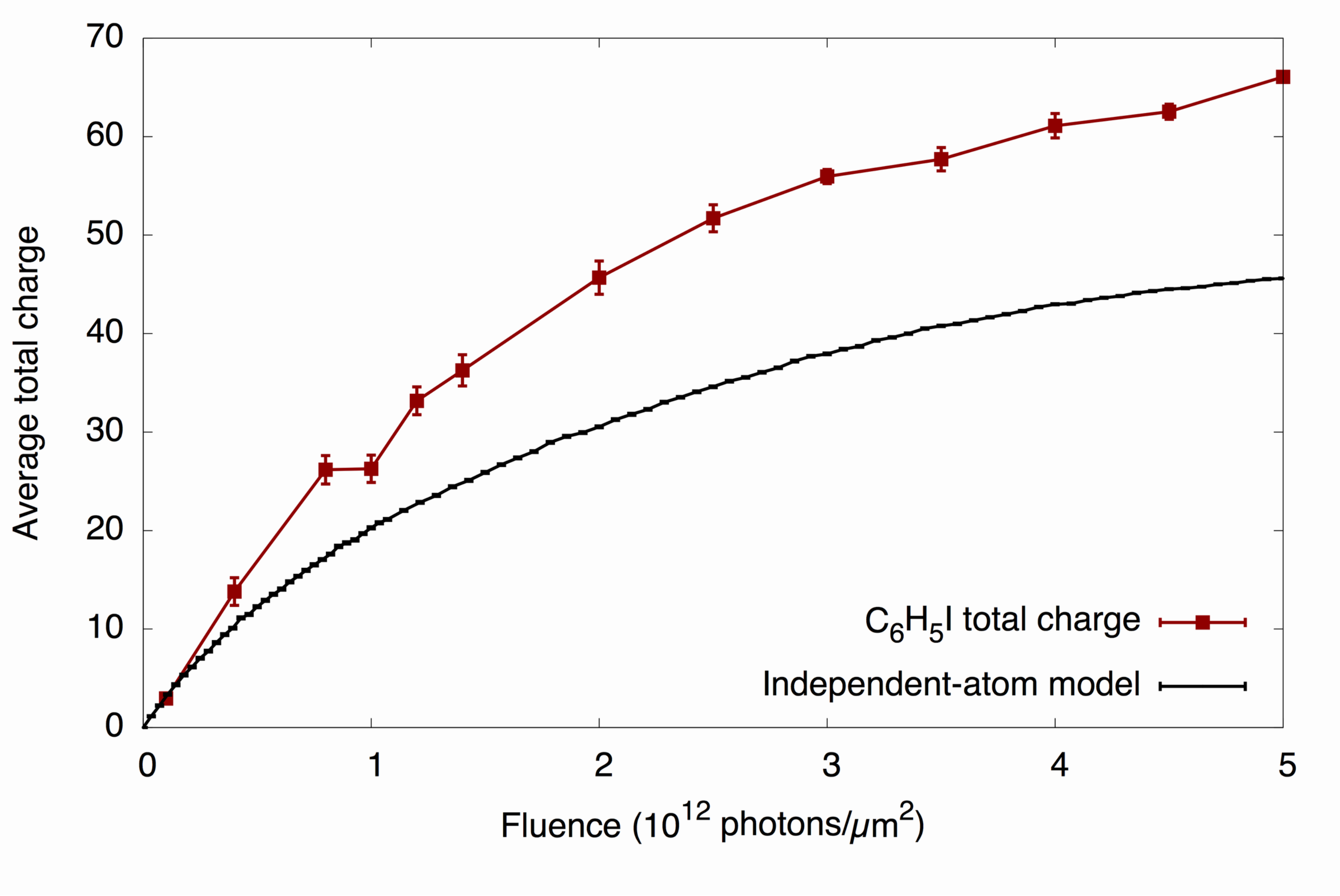
A. Rudenko *et al.*, Nature **546**, 129 (2017).

New ionization enhancement mechanism (molecular effect!)



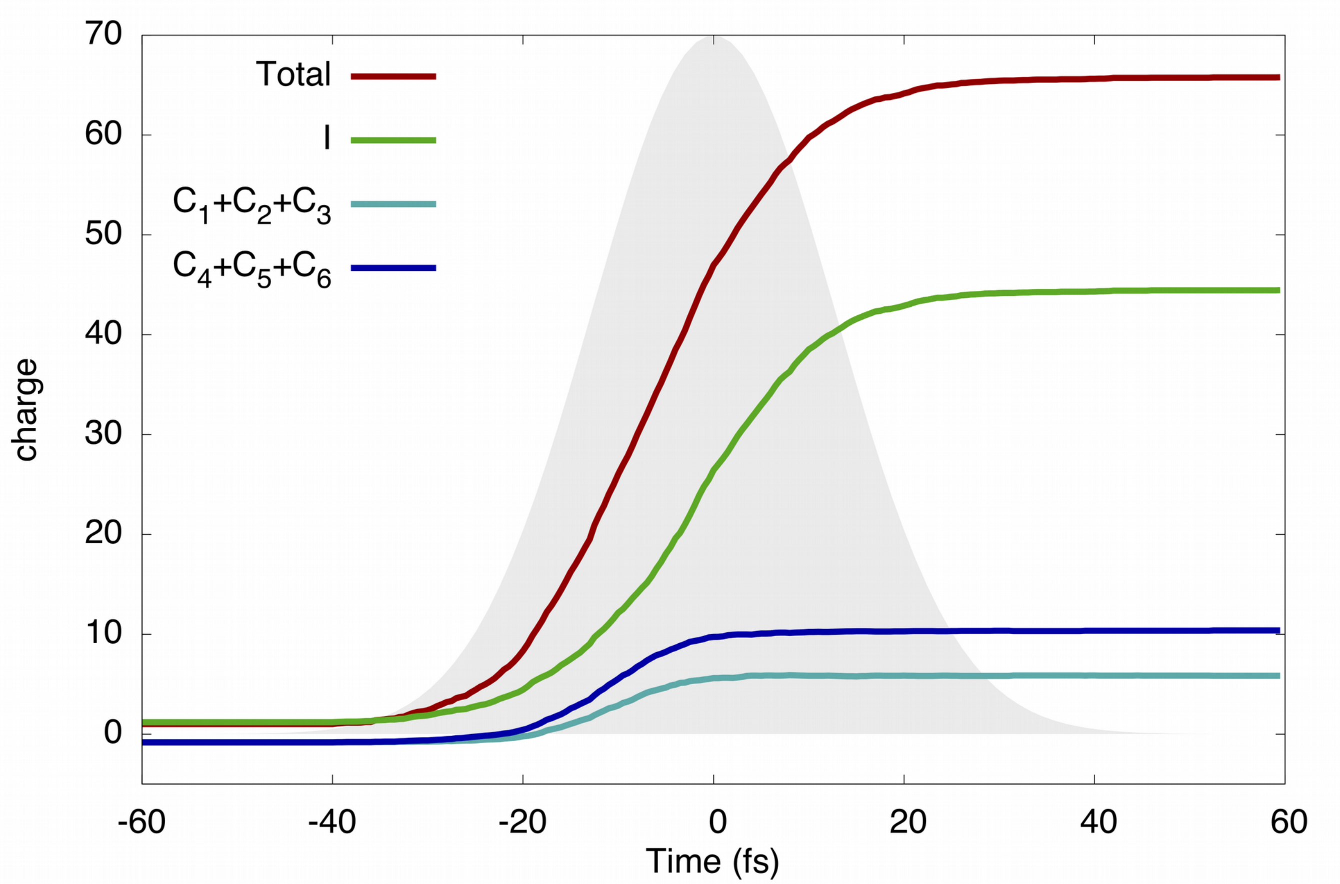
A. Rudenko *et al.*, Nature **546**, 129 (2017).

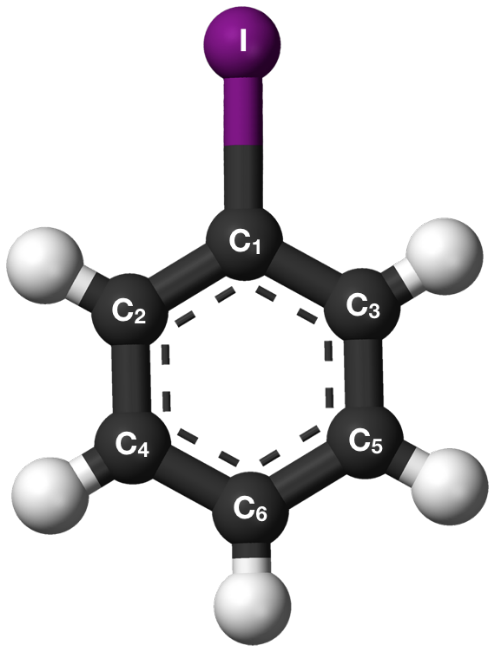
Iodobenzene (photon energy 8.3 keV)



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

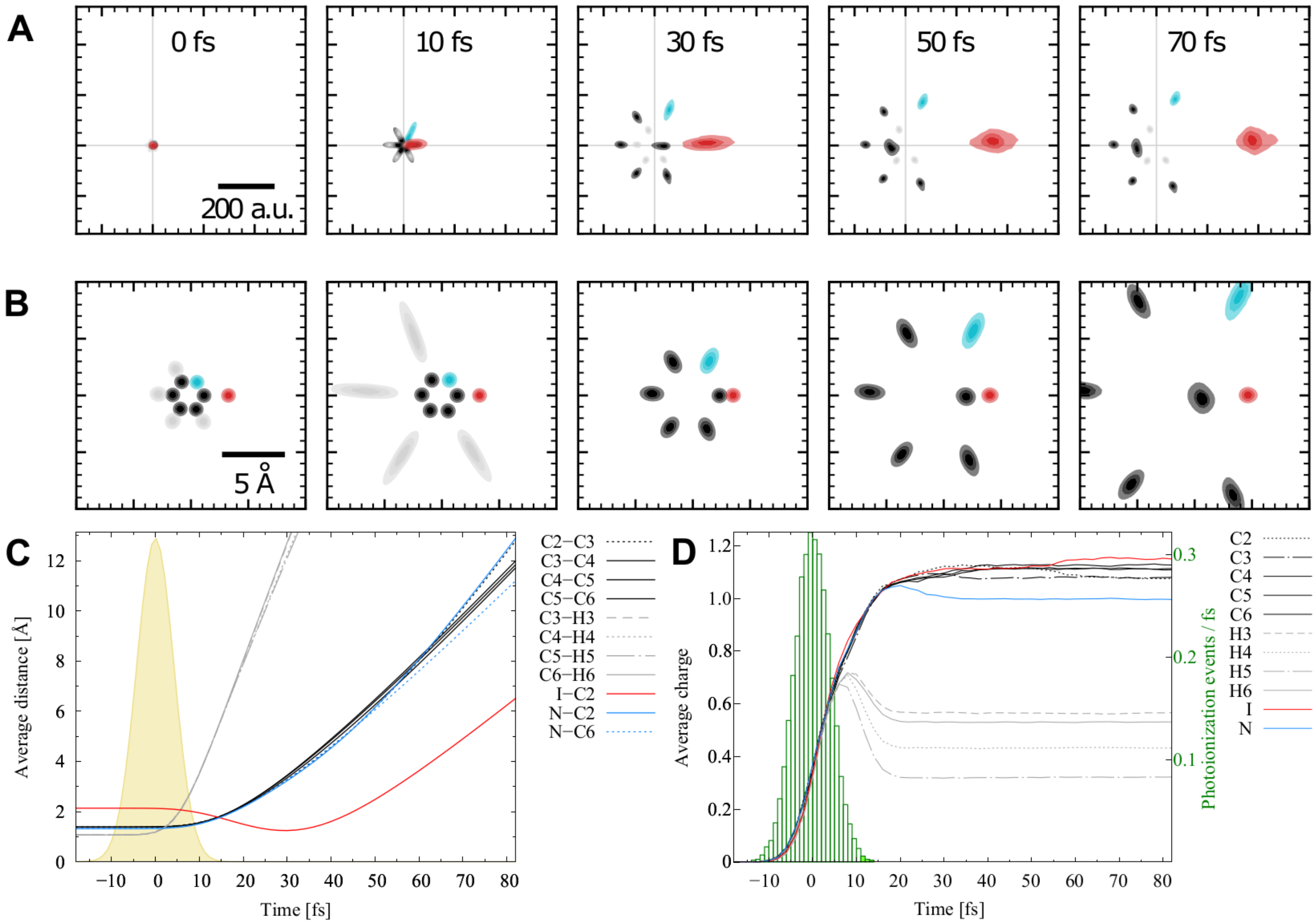
Ionization dynamics in iodobenzene (photon energy 8.3 keV, fluence 5×10^{12} photons/μm^2)





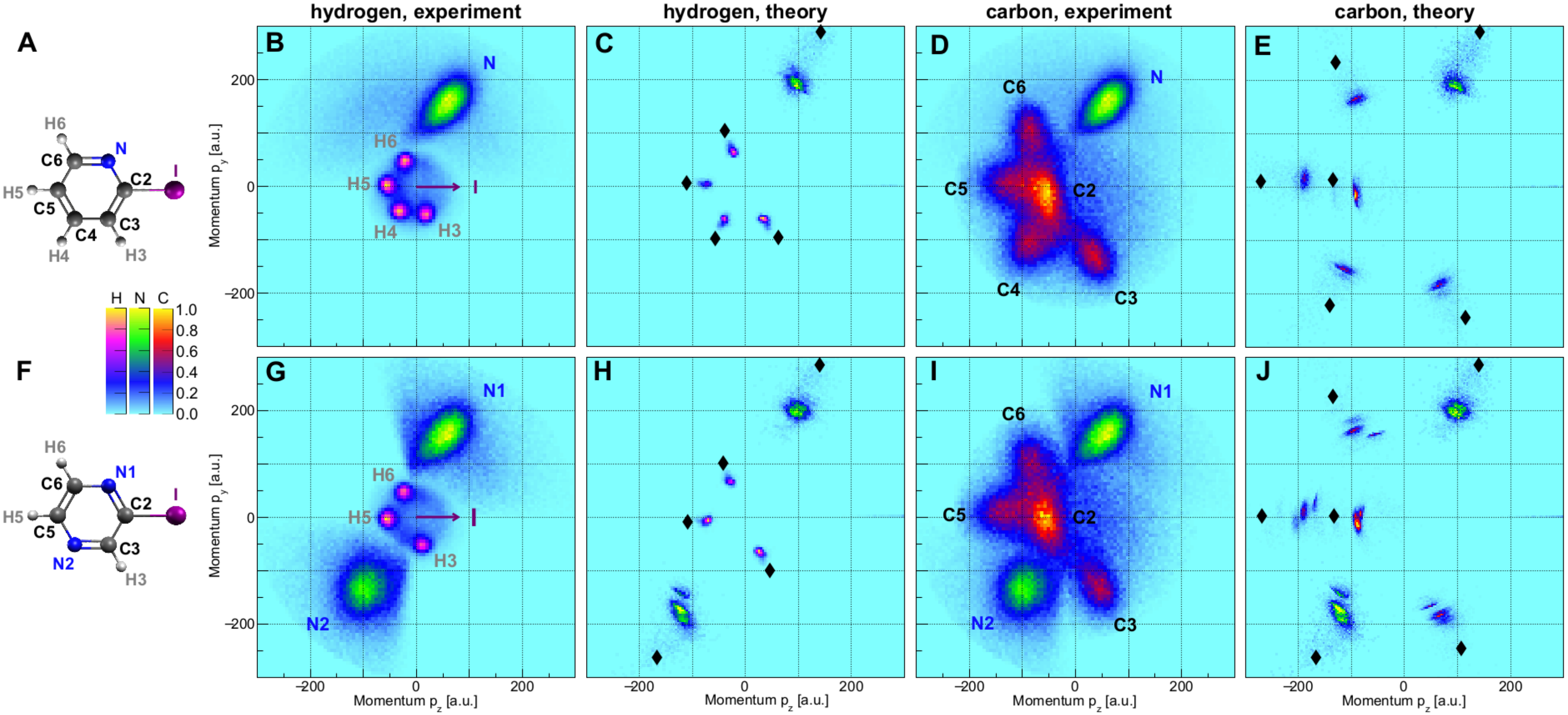
Y. Hao *et al.*, Phys. Rev. A **100**, 013402 (2019).

Single-molecule imaging via XFEL-driven Coulomb explosion: theory for 2-iodopyridine



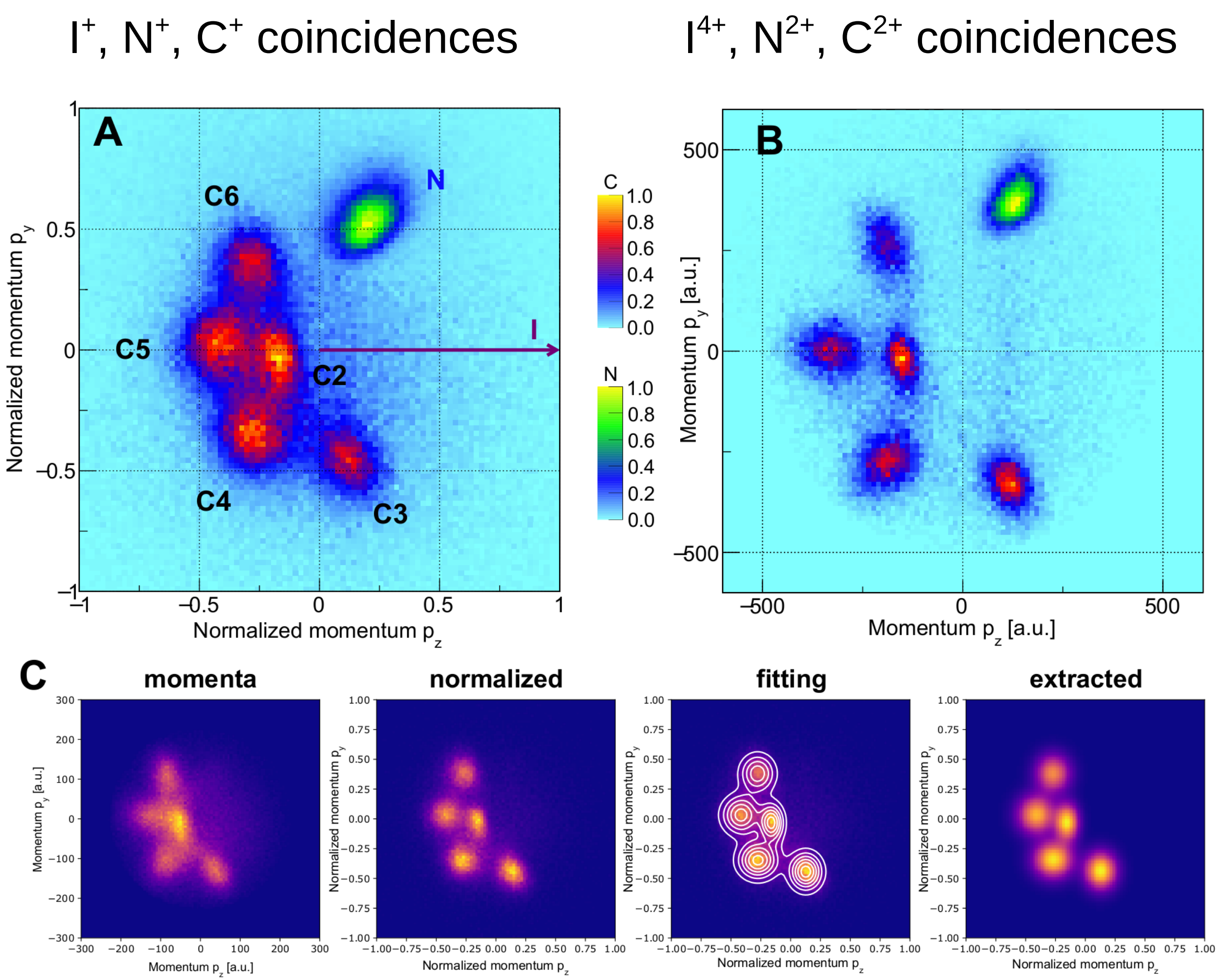
R. Boll *et al.*, Nature Phys. **18**, 423 (2022).

High-quality Coulomb explosion imaging at the European XFEL



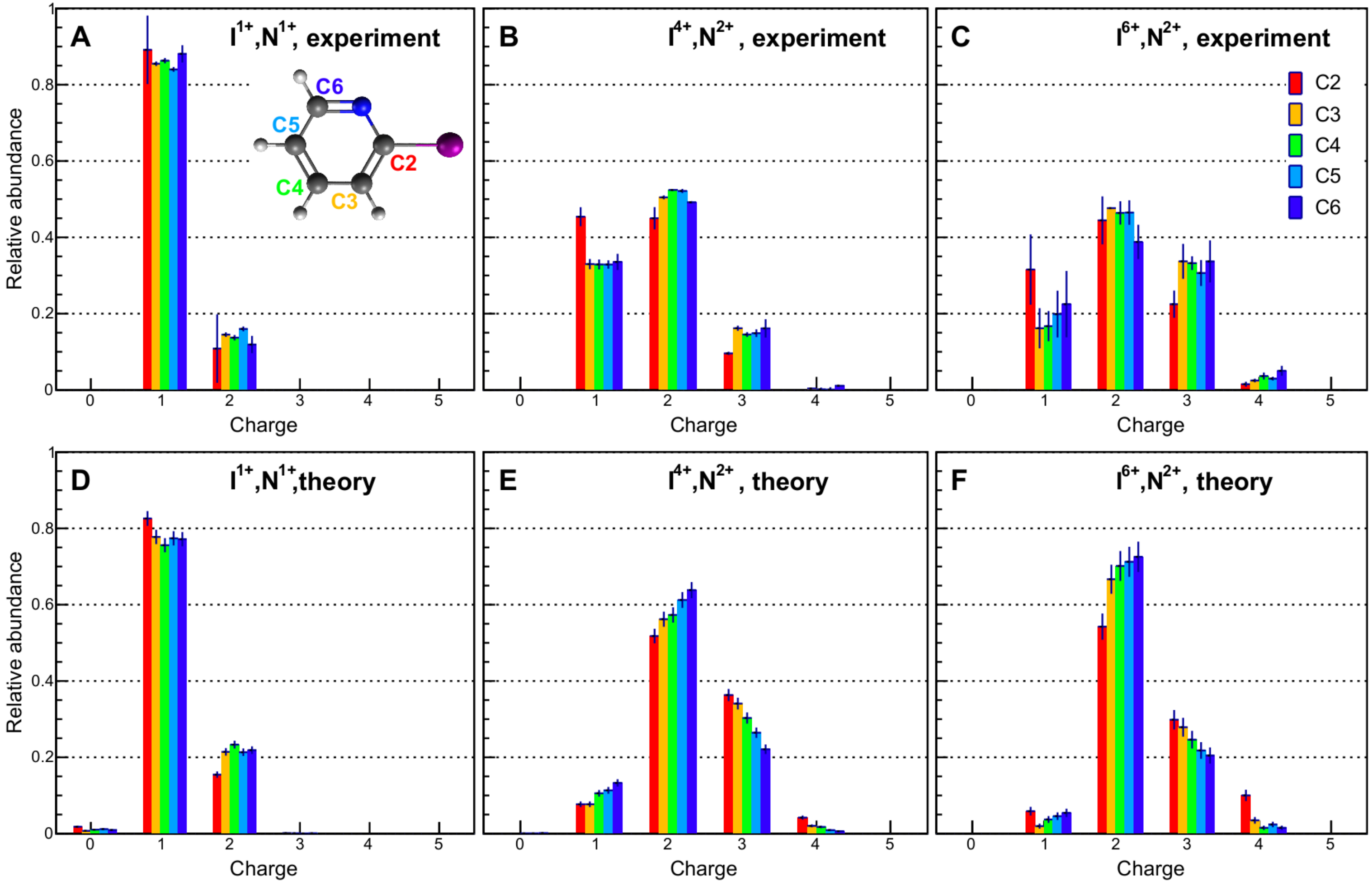
R. Boll *et al.*, Nature Phys. **18**, 423 (2022).

Momentum-space normalization and extraction of site-specific charge abundances



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

Charge state distributions of carbon atoms



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

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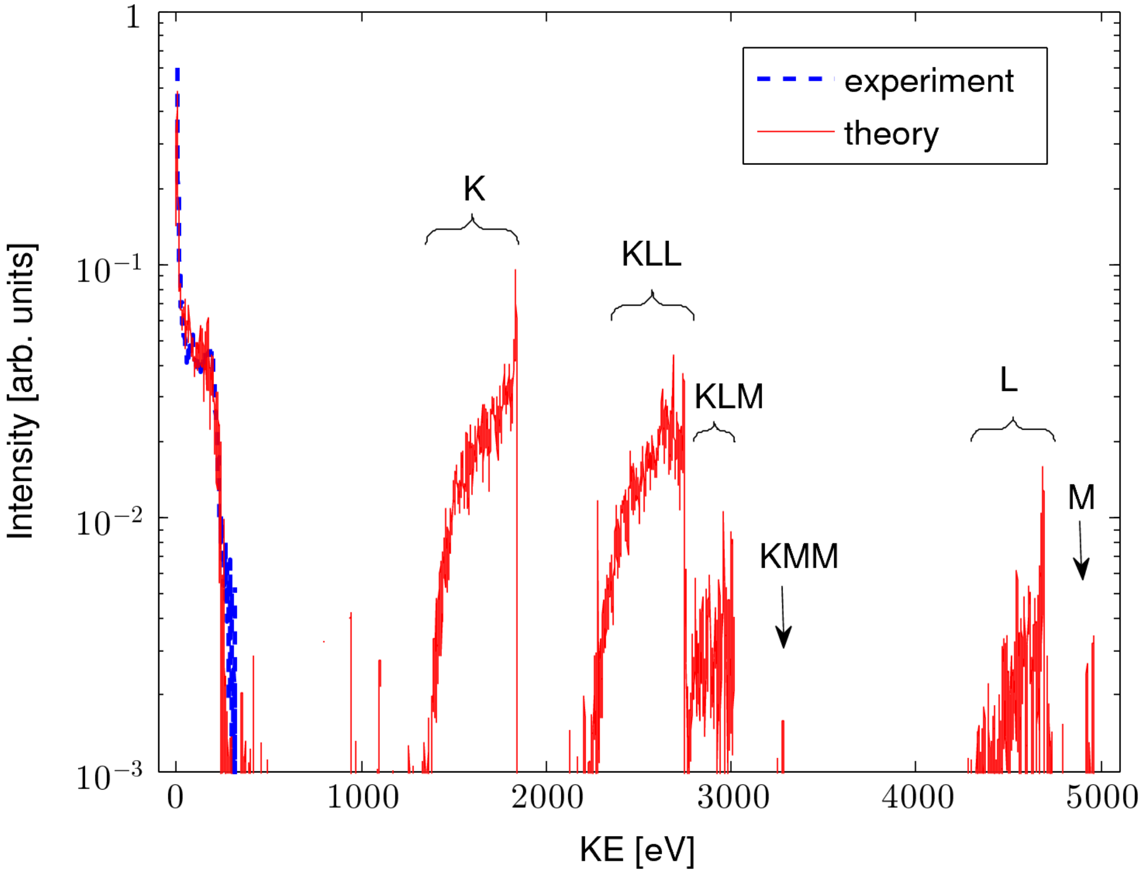
1: European XFEL
2: CFEL, DESY
...

Argon clusters @ SACLA (Kiyoshi Ueda *et al.*)

> **Theoretical and experimental electron kinetic energy spectra**,

5 keV, 30 fs

N=1000 atoms



– Experiment

– Theory

no parameter fitting!

T. Tachibana *et al.*, Scientific Reports **5**, 10977 (2015).

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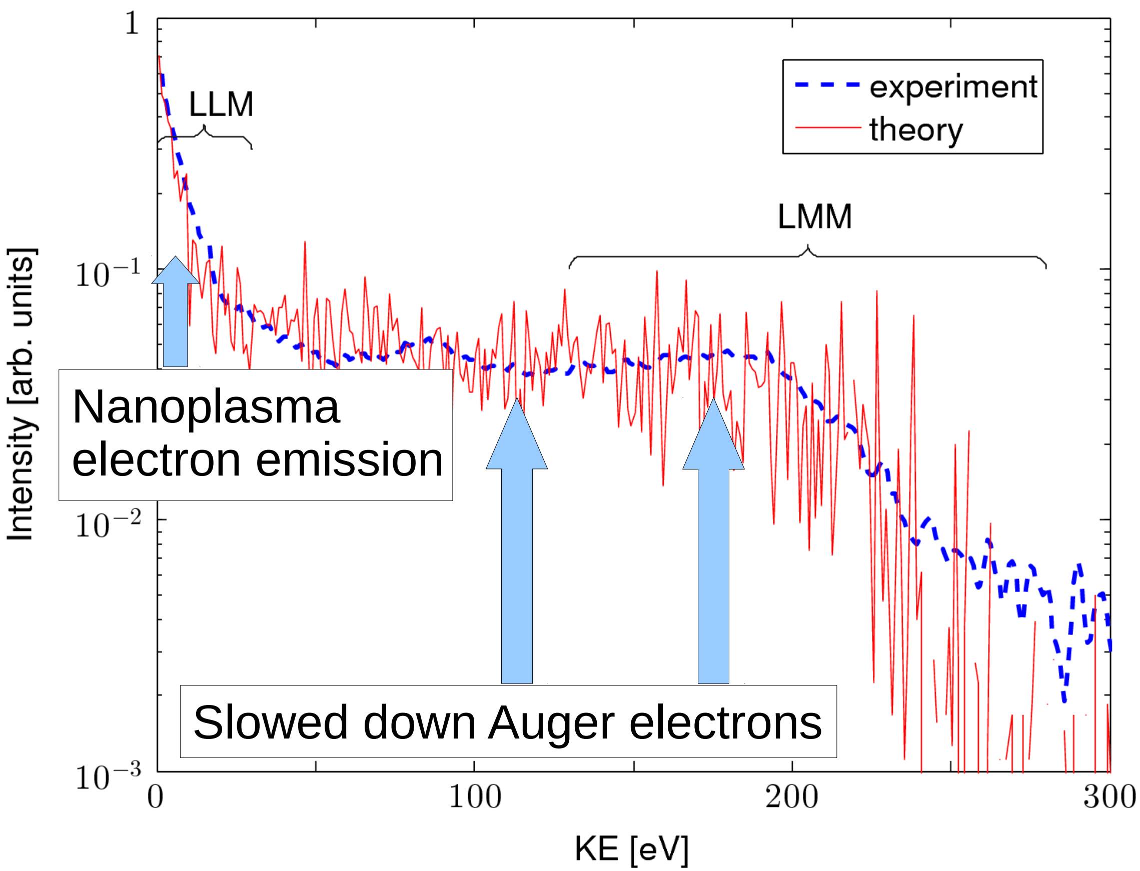

– Experiment

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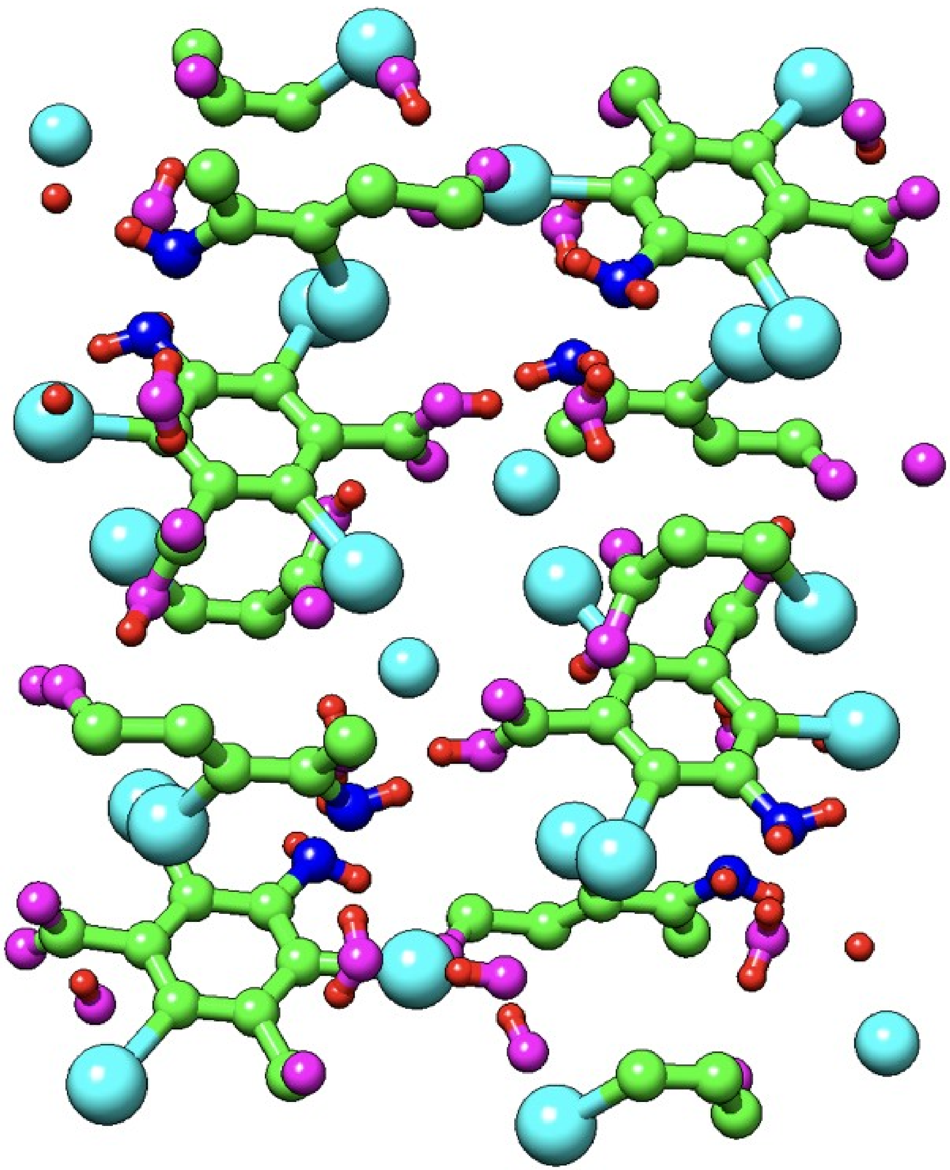
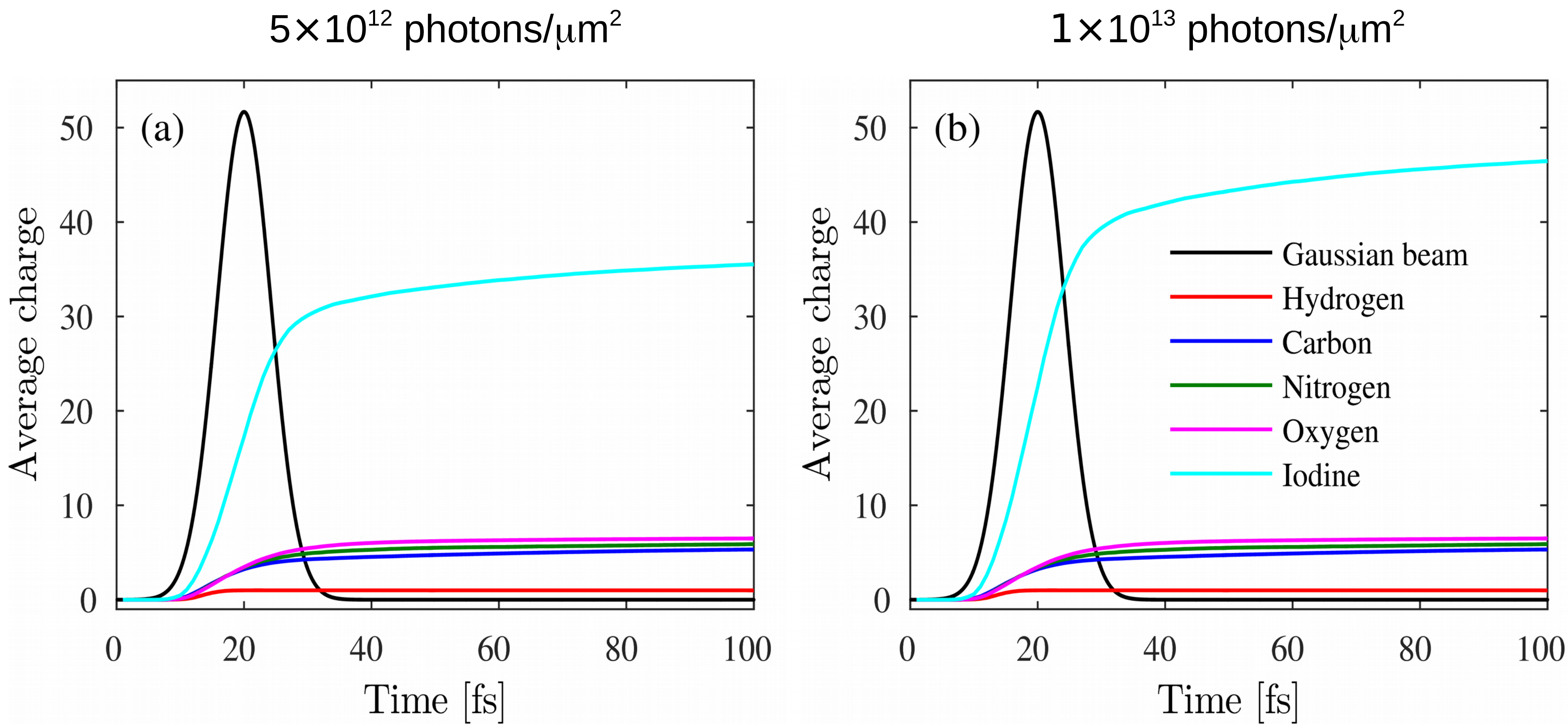
XMDYN using periodic boundary conditions



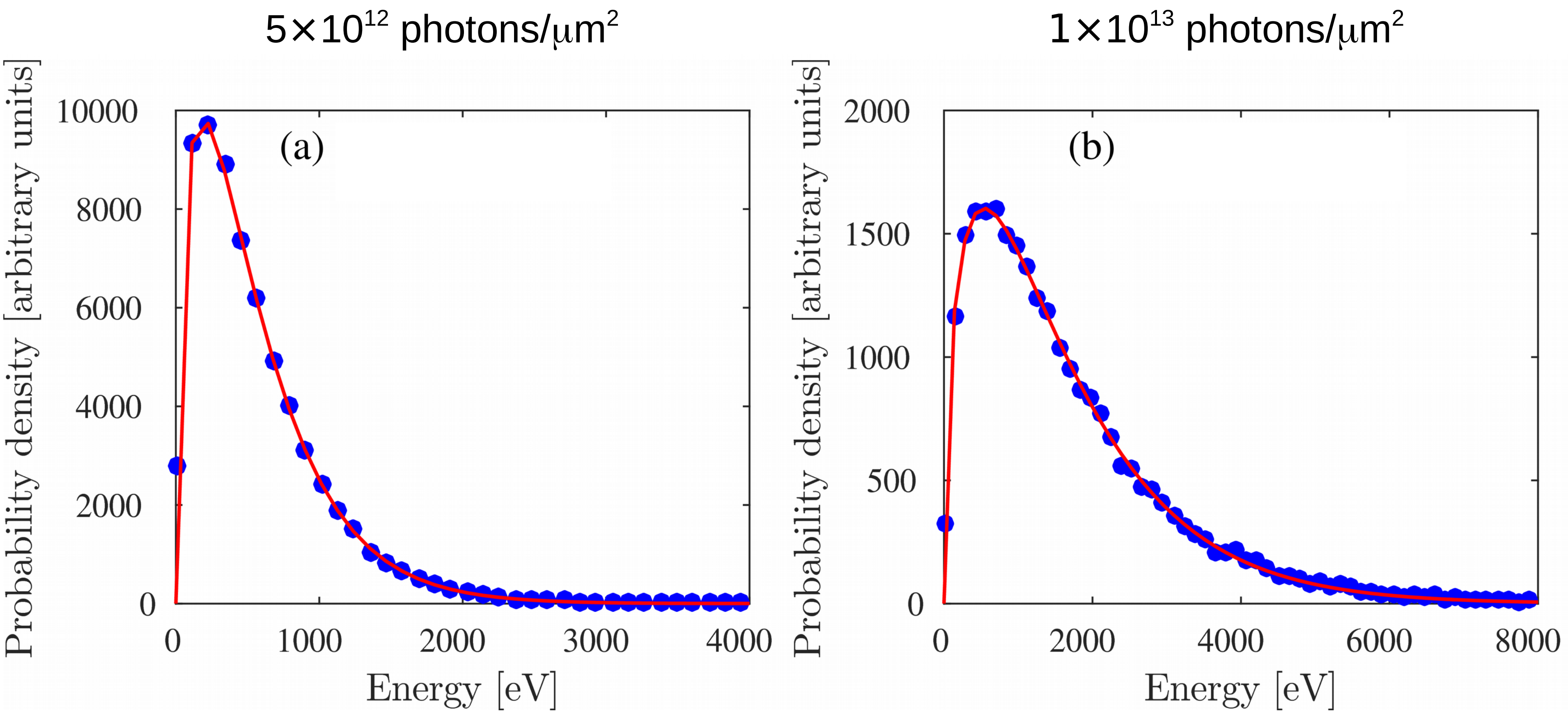
I3C crystal
(5-amino-2,4,6-triiodo-isophthalic acid,
$C_8H_4I_3NO_4$)

Ionization dynamics in I3C crystal (photon energy 9.7 keV)



M. M. Abdullah *et al.*, Phys. Rev. E **96**, 023205 (2017).

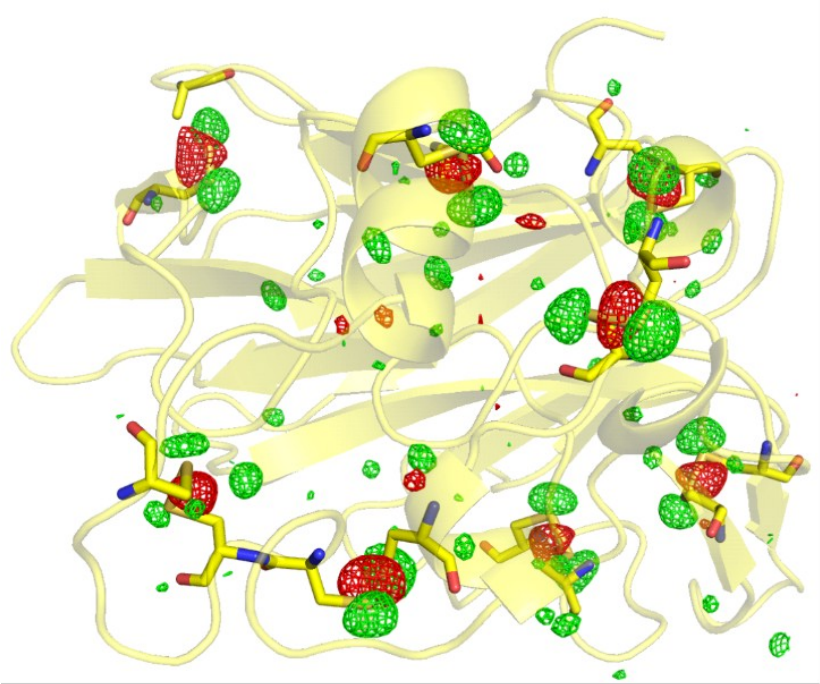
Electron thermalization in I3C crystal (250 fs after a 9.7-keV x-ray pulse)

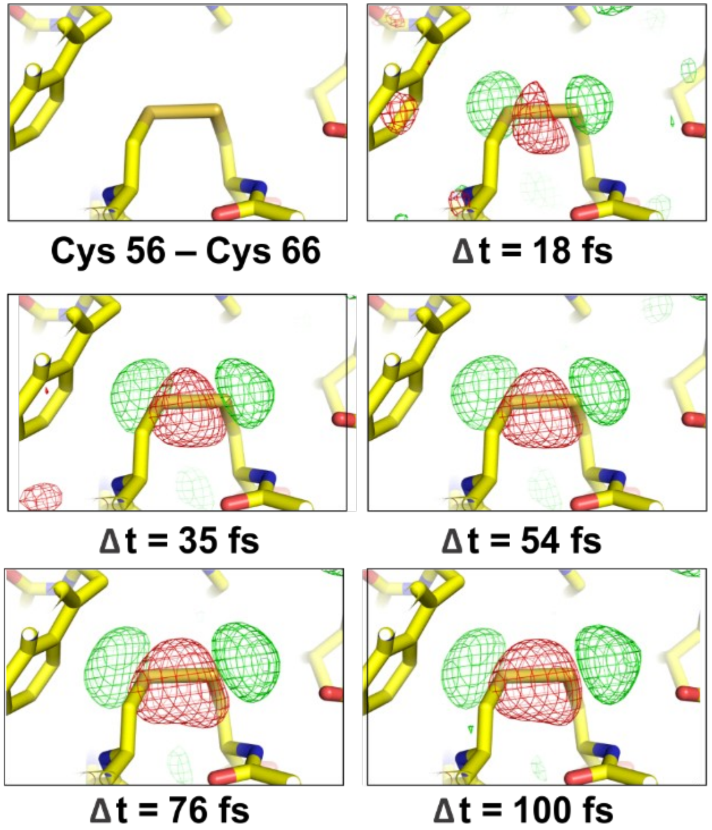


M. M. Abdullah *et al.*, Phys. Rev. E **96**, 023205 (2017).

X-ray pump / x-ray probe SFX study of thaumatin

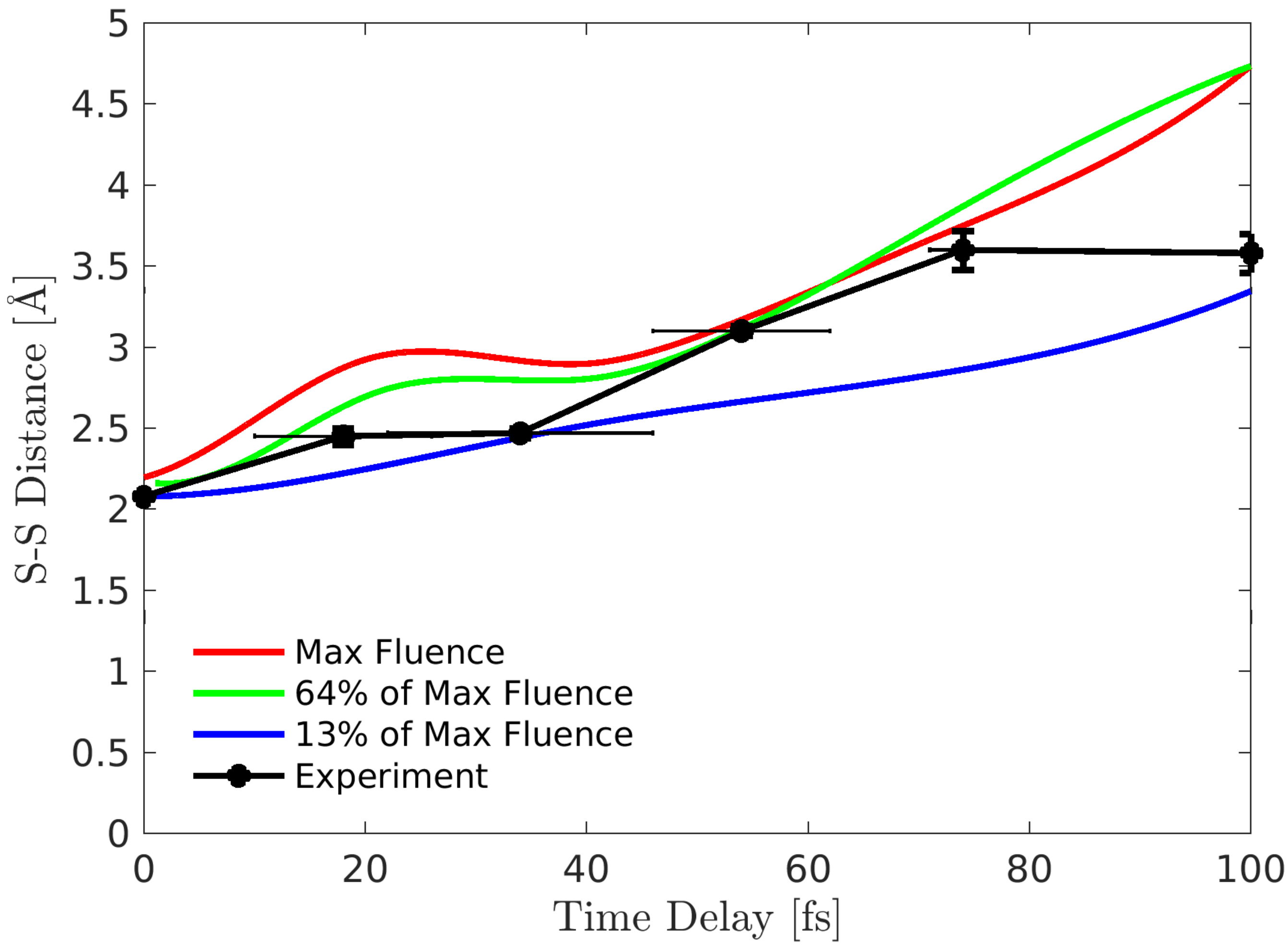
Collaboration with **Ilme Schlichting** et al.
Experiment carried out at LCLS





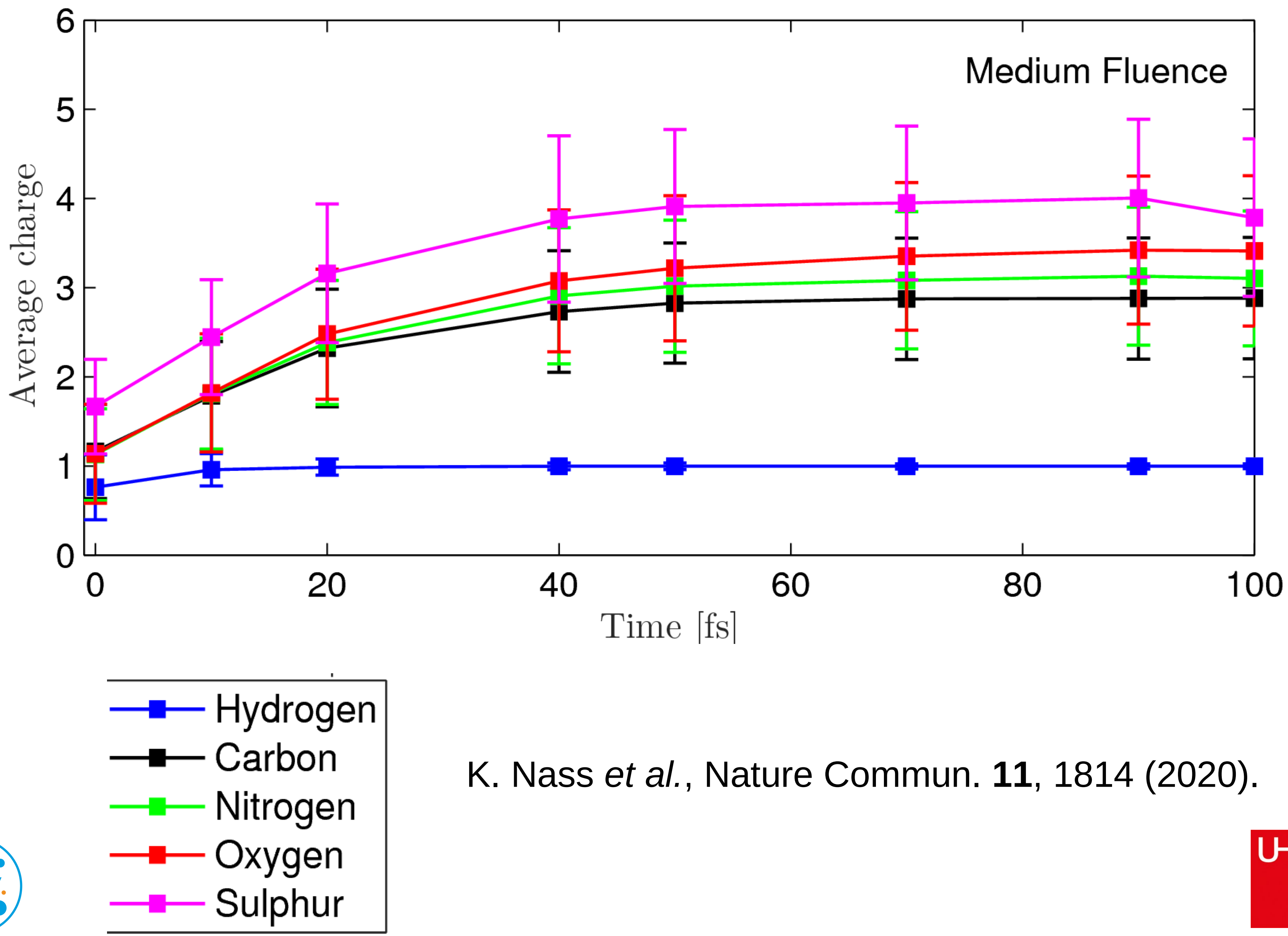
Photon energy: 7.1 keV
Pump (probe) pulse duration: 15 fs (15 fs)
Combined pulse energy: 1 mJ, shared 50%/50% (pump/probe)
Intensity in the focus: 3×10^{19} W/cm^2

Disulfide bond length in thaumatin



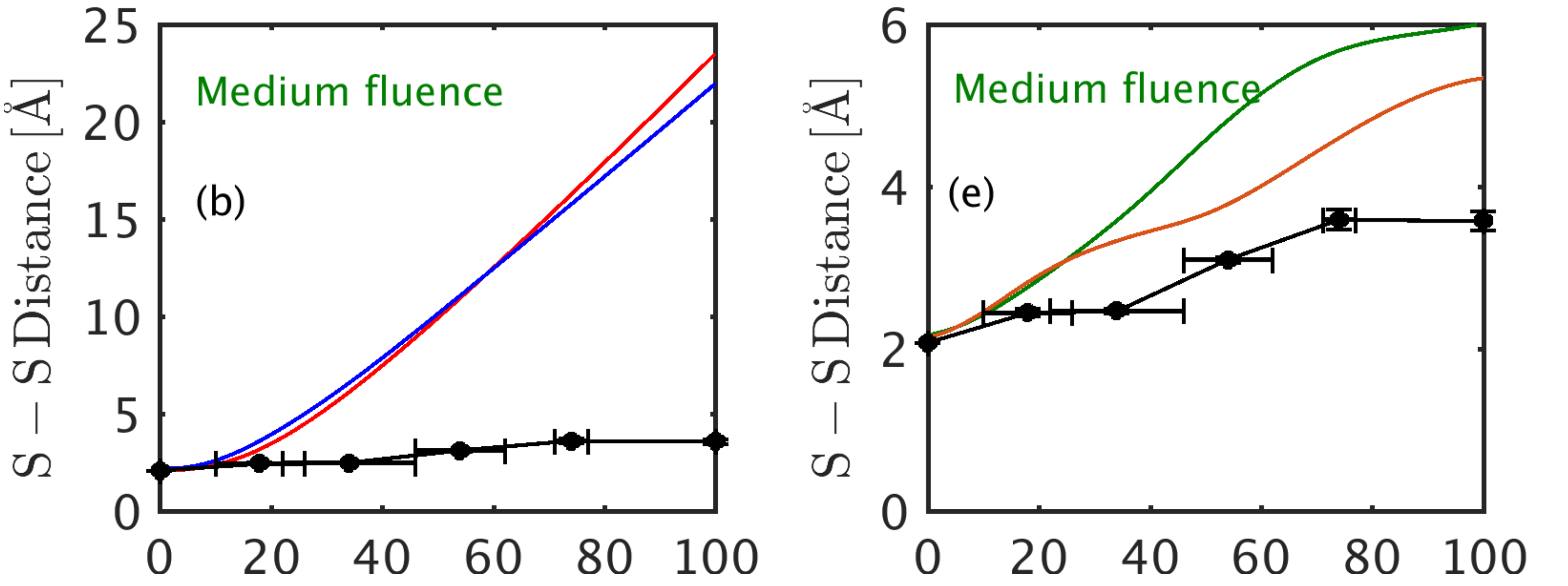
K. Nass *et al.*, Nature Commun. **11**, 1814 (2020).

Average charge as a function of pump-probe delay



K. Nass *et al.*, Nature Commun. **11**, 1814 (2020).

Ion caging and plasma screening

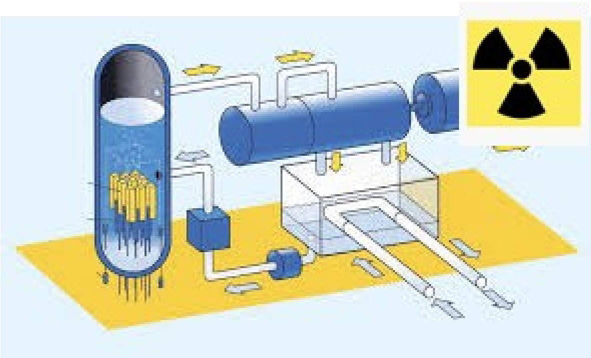


Red: Isolated S-S pair in vacuum
Blue: Isolated S-S pair using charges in crystal environment

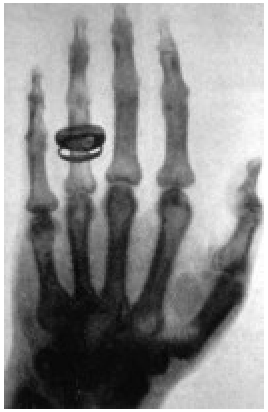
Brown: No Coulomb interaction between S atoms and plasma electrons
Green: No Coulomb interaction between S atoms and non-S atoms

K. Nass *et al.*, Nature Commun. **11**, 1814 (2020).

Effects of radiation

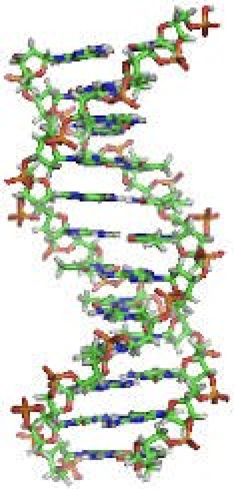


Corrosion in nuclear power plants



Medical x-ray imaging

Air travel and spaceflight

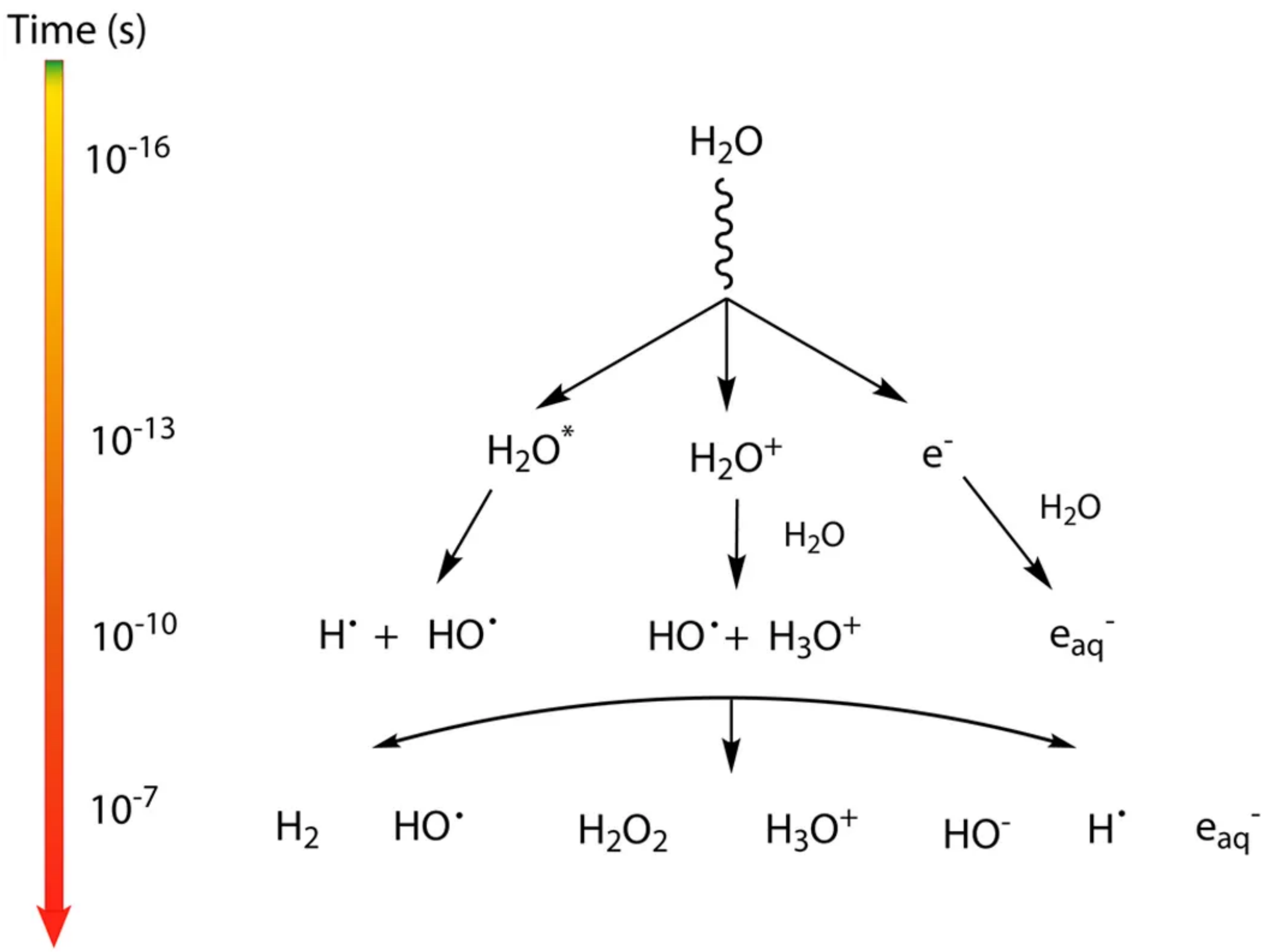


Radiotherapy

What happens microscopically?

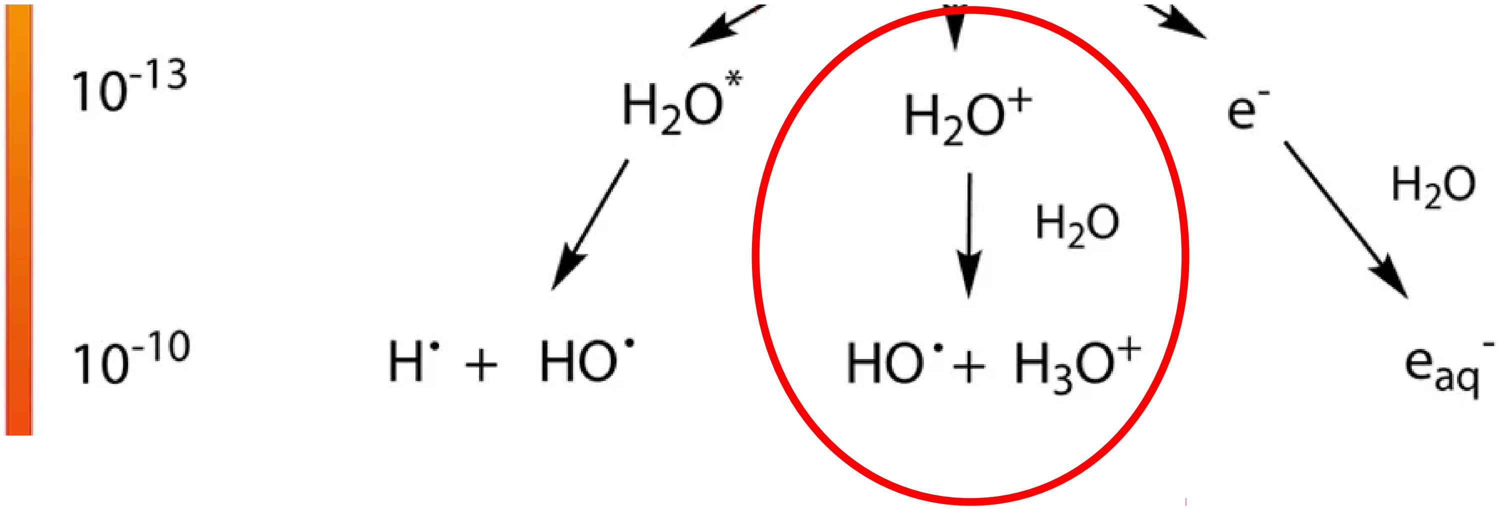
Ionizing radiation (x rays, γ rays, charged particles) gives rise to the **formation of highly reactive radicals**. Particularly, through the **ionization of water**, highly reactive **hydroxyl (OH) radicals** are formed.

How does ionization of water produce OH?



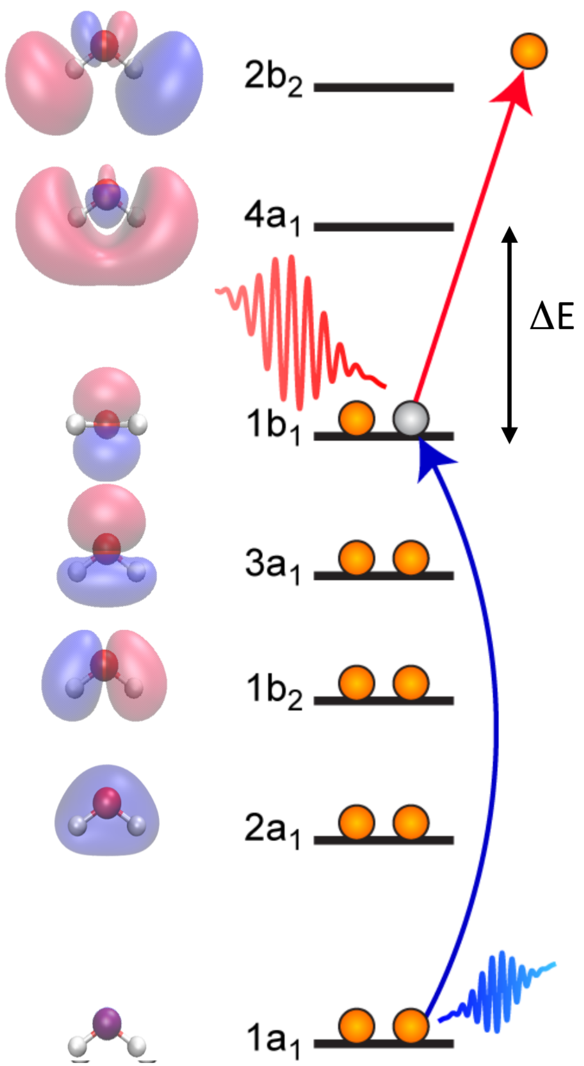
C. M. Lousada *et al.*, Scientific Reports **6**, 24234 (2016).

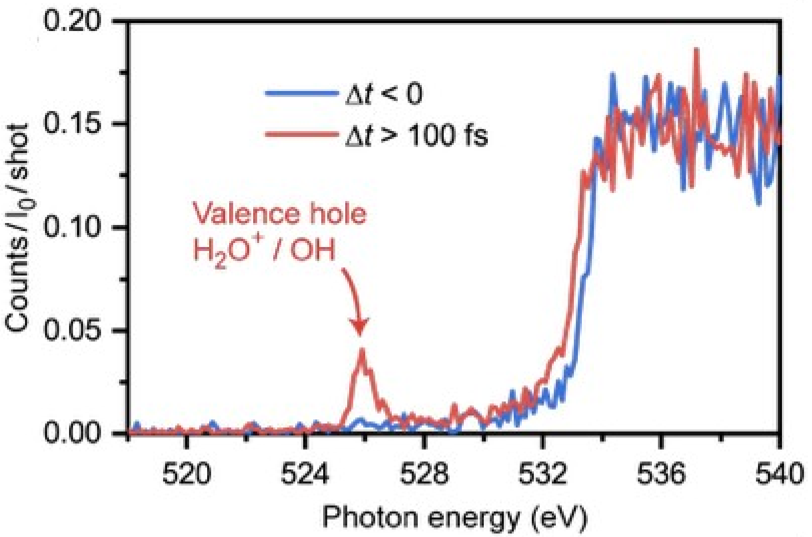
Is it true that the key reaction step takes somewhere between 100 fs and 100 ps?



C. M. Lousada *et al.*, Scientific Reports **6**, 24234 (2016).

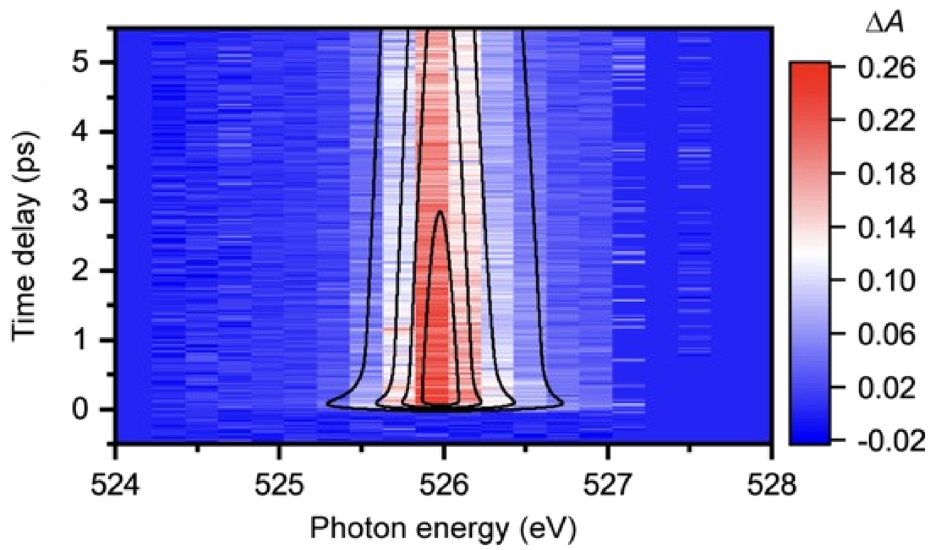
Probing the hole through transient x-ray absorption

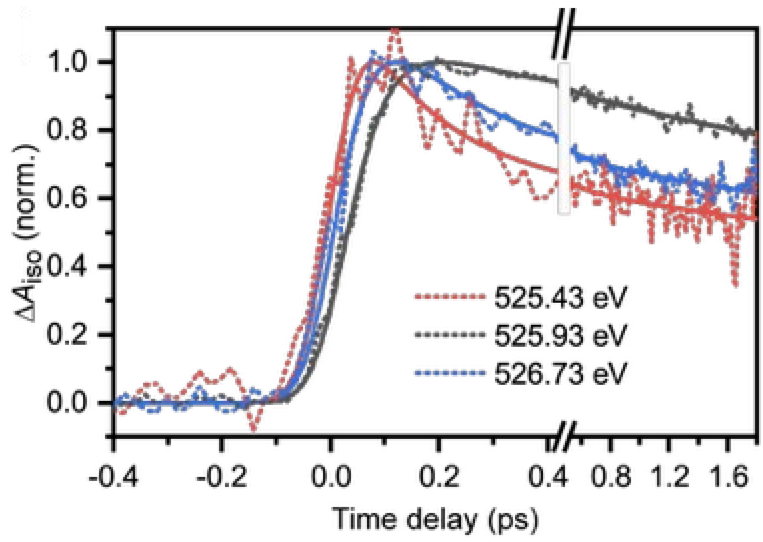




Z.-H. Loh *et al.*,
Science **367**, 179 (2020).

Observed delay dependence of x-ray absorption resonance





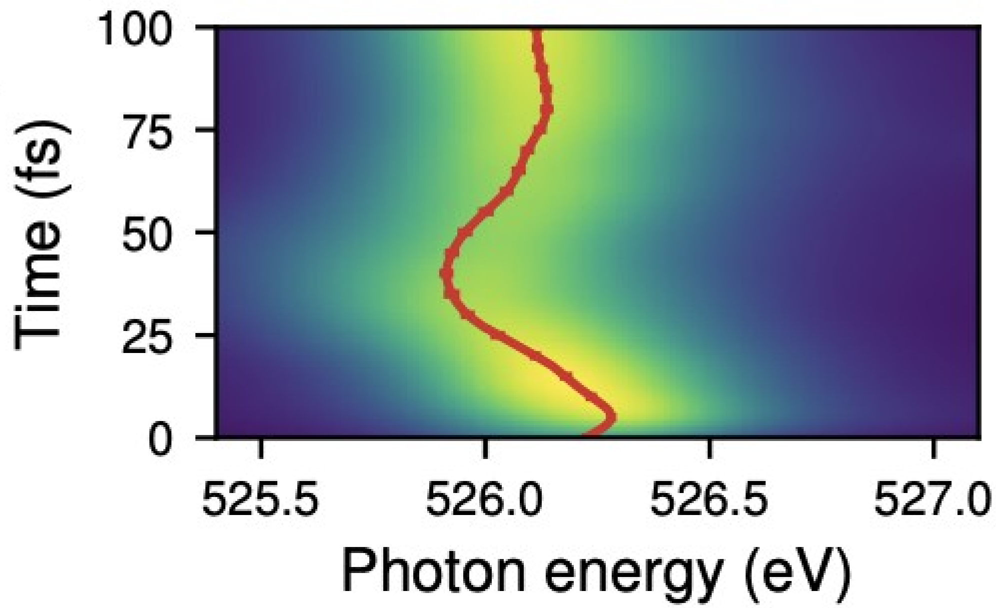
Sequential kinetics

$$A_0 \xrightarrow{\tau_0} A_1 \xrightarrow{\tau_1} A_2 \xrightarrow{\tau_2} A_3$$

$\tau_0 = 46 \pm 10\,\mathrm{fs}$	$\tau_1 = 180 \pm 20\,\mathrm{fs}$	$\tau_2 = 14.2 \pm 0.4\,\mathrm{ps}$
OH formation	Vibrationally hot OH cools	OH + e recombine

Z.-H. Loh *et al.*, Science **367**, 179 (2020).

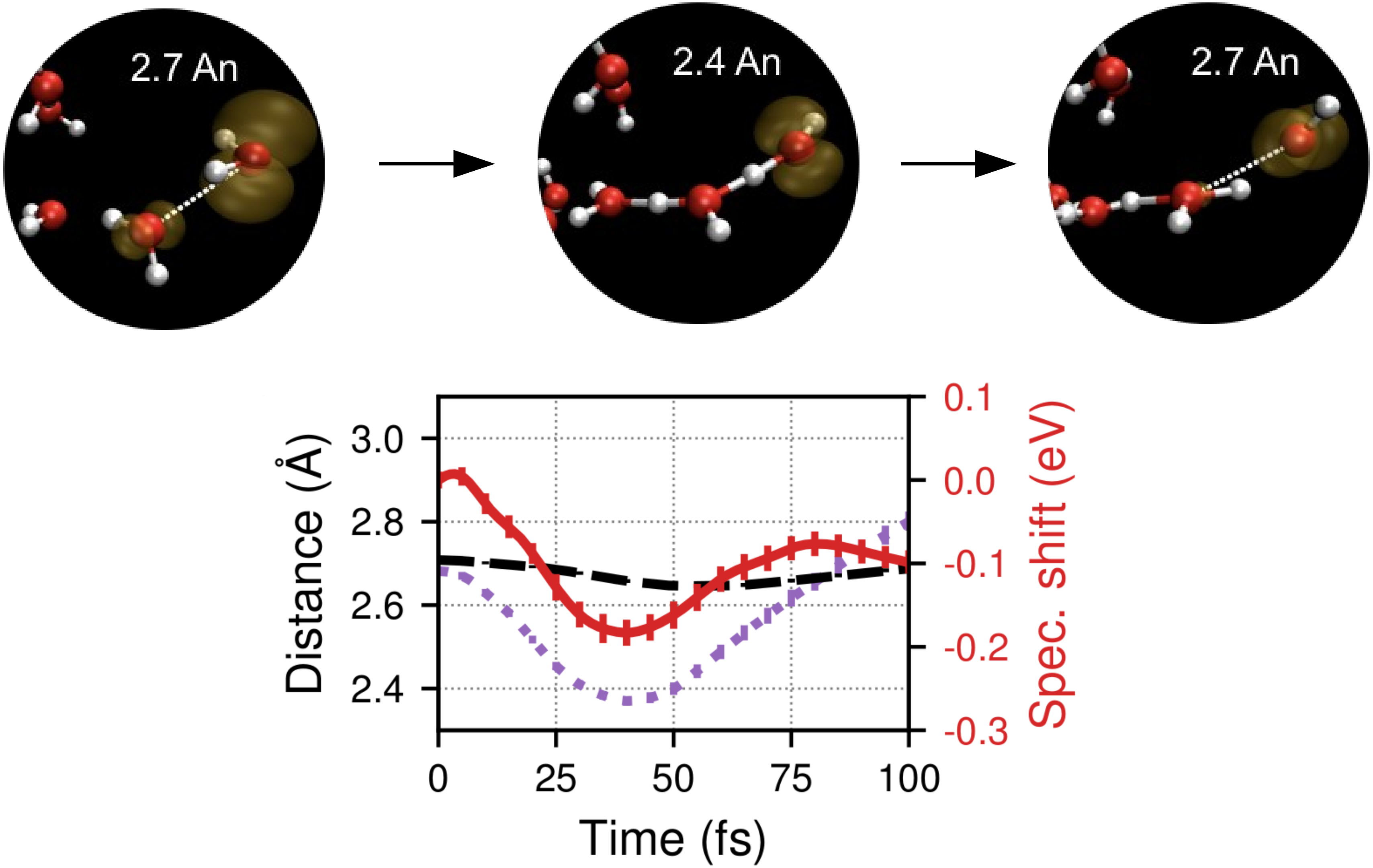
XMOLECULE simulation



How are these spectral dynamics connected to the decay of H_2O^+ and the formation of OH?

Z.-H. Loh *et al.*, Science **367**, 179 (2020).

Impact of chemical environment of H_2O^+/OH



Z.-H. Loh *et al.*, Science **367**, 179 (2020).

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- Sorbonne Université and CNRS: M. Simon
- University of Southern California: K. Nanda, A. I. Krylov
- SLAC National Accelerator Laboratory: S. Moeller, G. Coslovich, J. Koralek, M. P. Minitti, W. F. Schlotter

Conclusions

- Electron transfer in polyatomic systems can lead to significantly enhanced ionization in comparison to independent-atom models.
- Efficient ionization enables Coulomb explosion imaging.
- In spatially extended systems, transient plasmas are formed.
- In such plasmas, atomic displacements are smaller than one might naively expect.
- Ultrafast x-ray absorption enabled the first observation of the proton transfer reaction following ionization of liquid water, giving rise to the chemically aggressive OH radical.