Phenomena at High X-ray Intensity: Part 1

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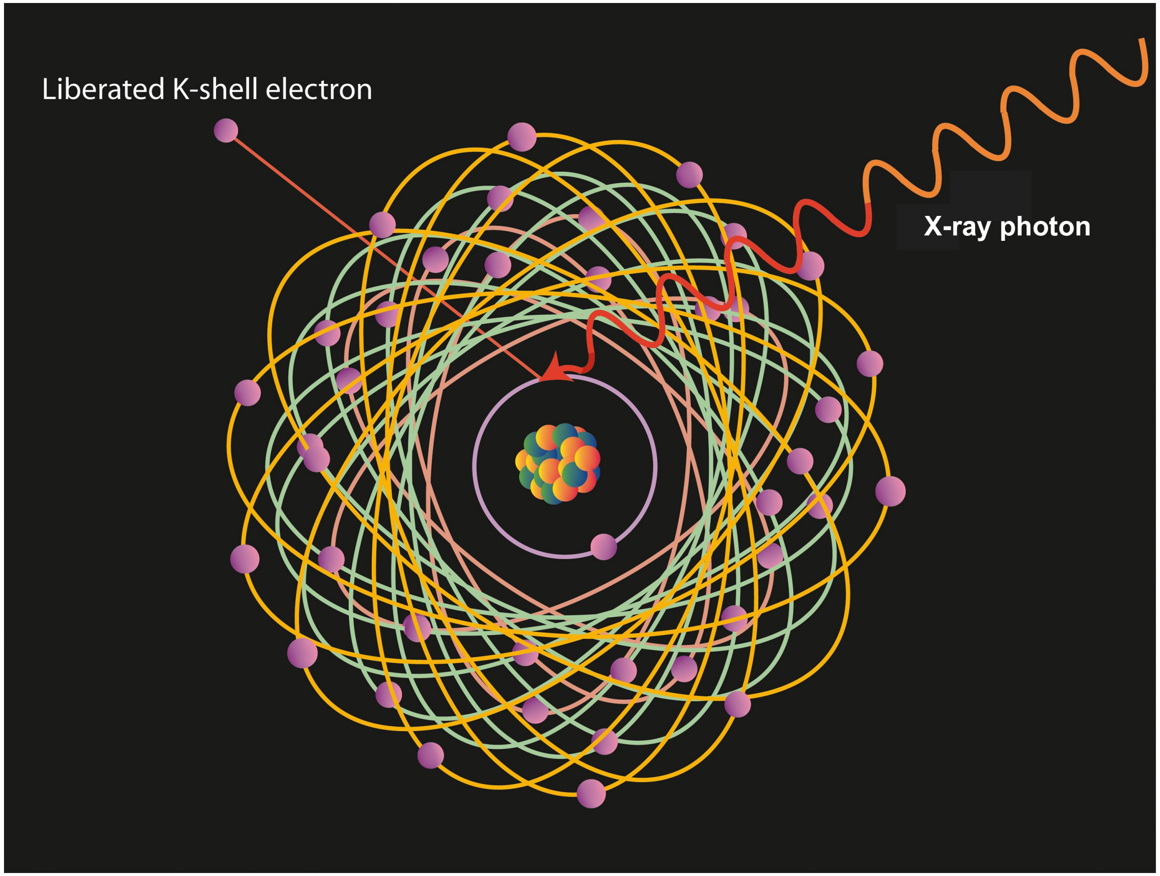
Discovery of x rays by Wilhelm Röntgen in 1895

Mechanism of image formation:

• X-ray photoelectric effect, which depends on the atomic species encountered by the x rays

• Photoelectrons are produced, but they get stuck in the tissue

Dominant x-ray–atom interaction process: photoabsorption



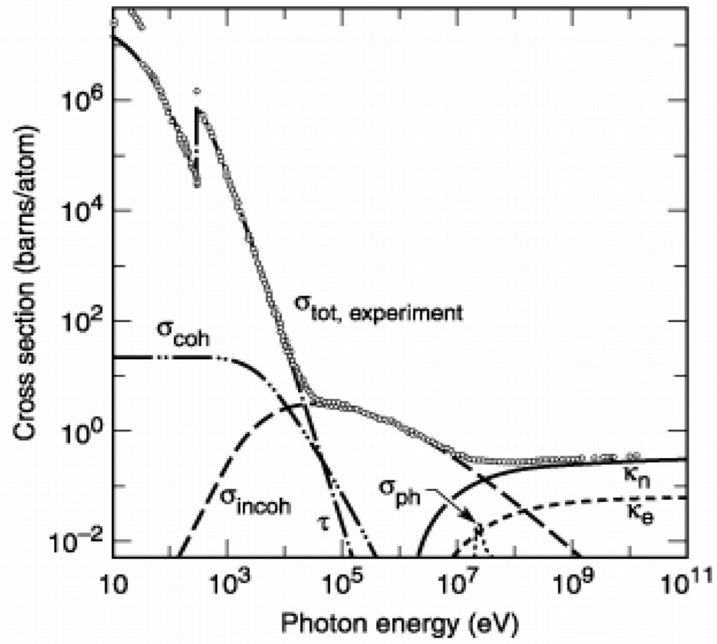


Fig. 3-1. *Total photon cross section* σ_{tot} *in carbon, as a function of energy, showing the contributions of different processes:* τ, *atomic photo-effect (electron ejection, photon absorption);* σ_{coh}, *coherent scat-tering (Rayleigh scattering—atom neither ionized nor excited);* σ_{incoh}, *incoherent scattering (Comp- ton scattering off an electron);* κ_n, *pair production, nuclear field;* κ_e, *pair production, electron field;* σ_{ph}, *photonuclear absorption (nuclear absorption, usually followed by emission of a neutron or other particle). (From Ref. 3; figure courtesy of J. H. Hubbell.)*

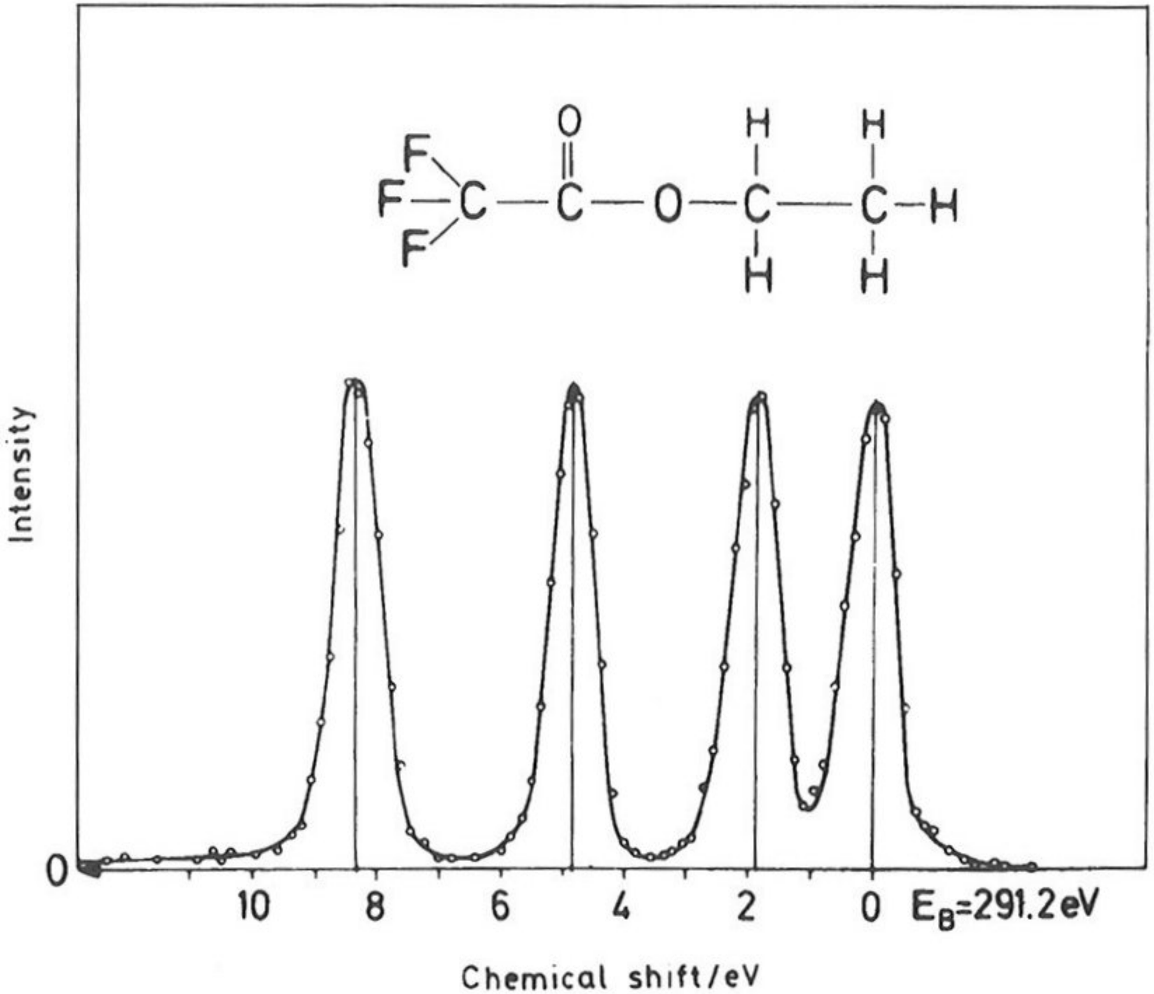
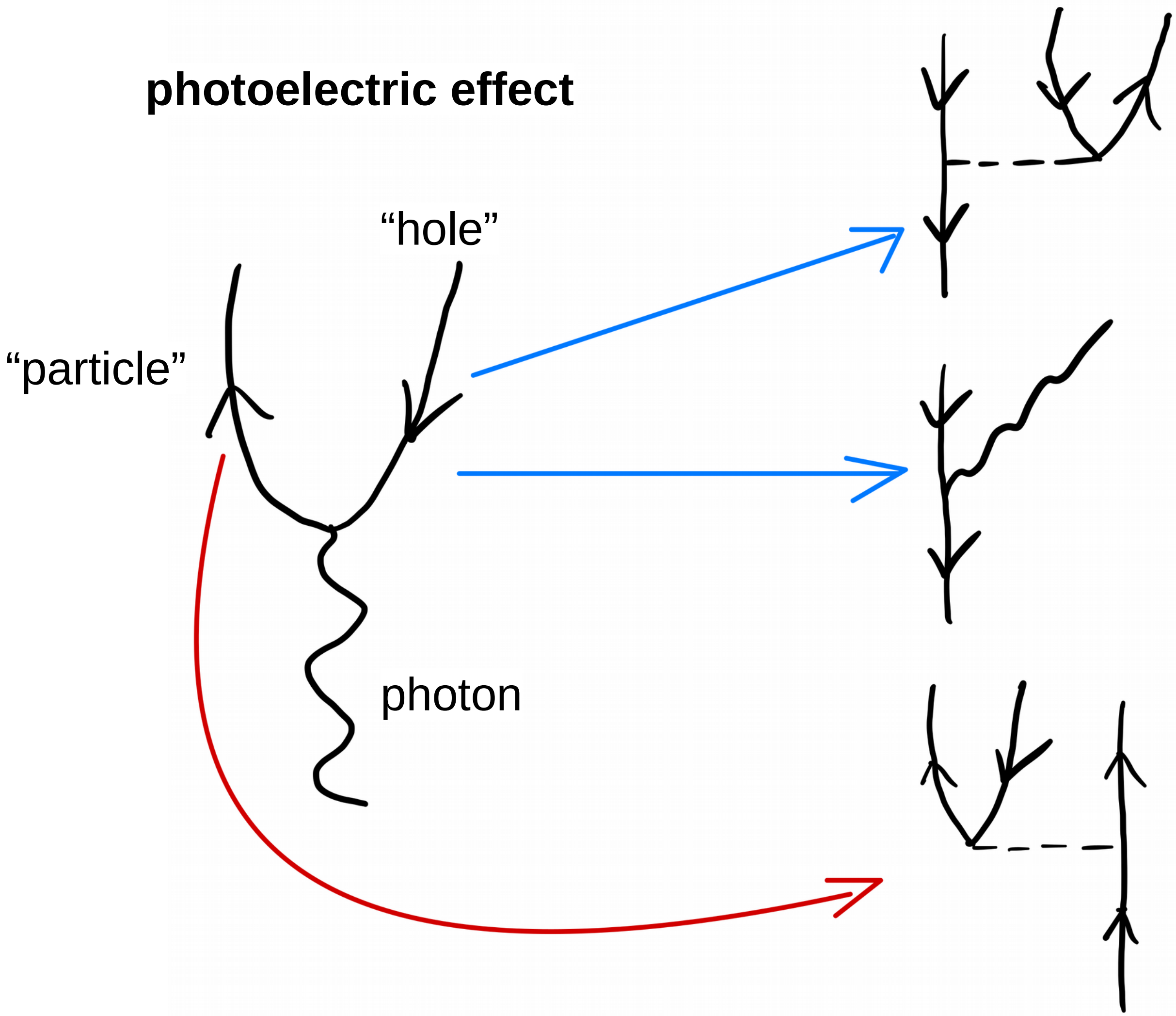


Figure 8.14 The monochromatized Al$K\alpha$ carbon 1s XPS spectrum of ethyltrifluoroacetate showing the chemical shifts relative to an ionization energy of 291.2 eV. (Reproduced, with permission, from Gelius, U., Basilier, E., Svensson, S., Bergmark, T., and Siegbahn, K., *J. Electron Spectrosc.*, **2**, 405, 1974)

photoelectric effect

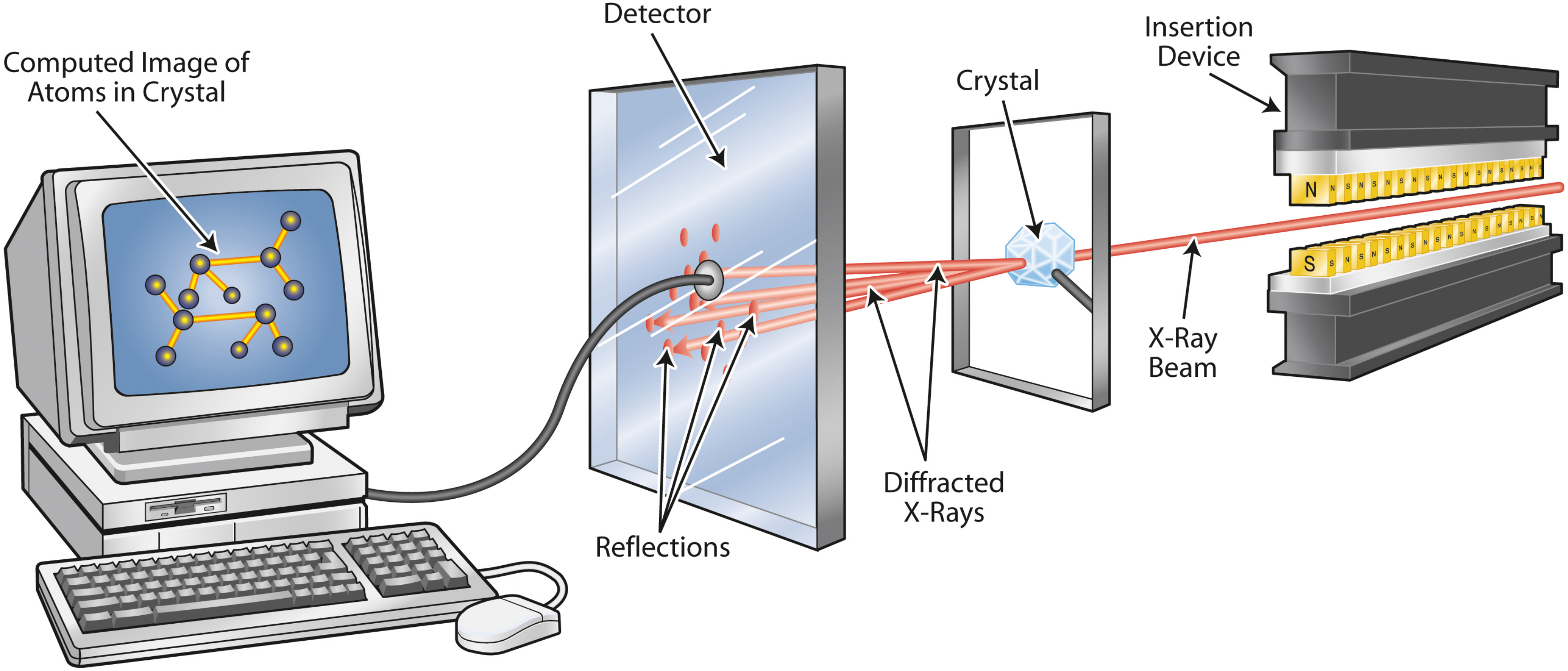


Auger decay
=
Auger-Meitner decay

x-ray fluorescence

electron impact ionization
=
collisional ionization

X-ray crystallography: principle

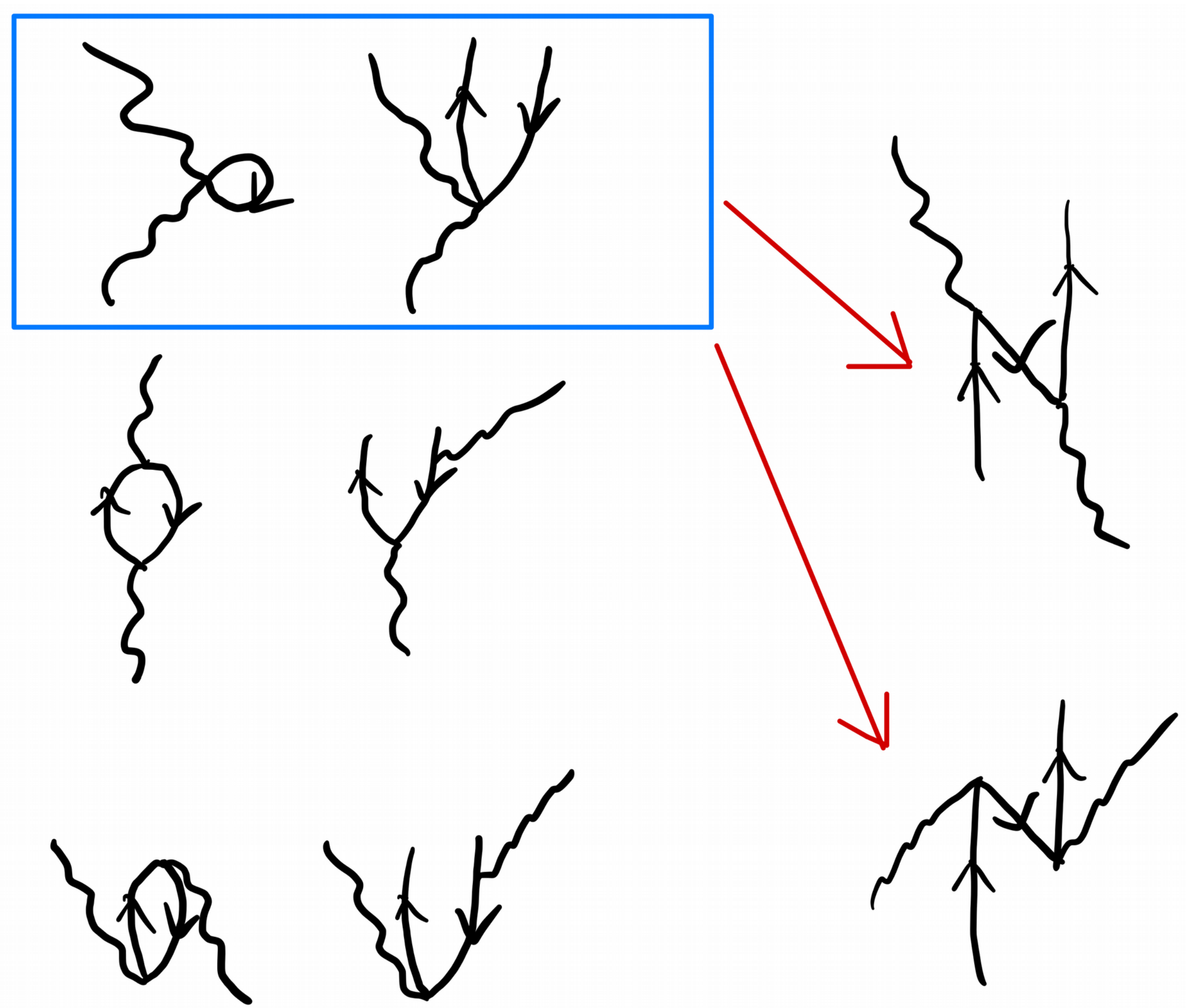


nonrelativistic QED and many-body theory

“A^2” mechanism

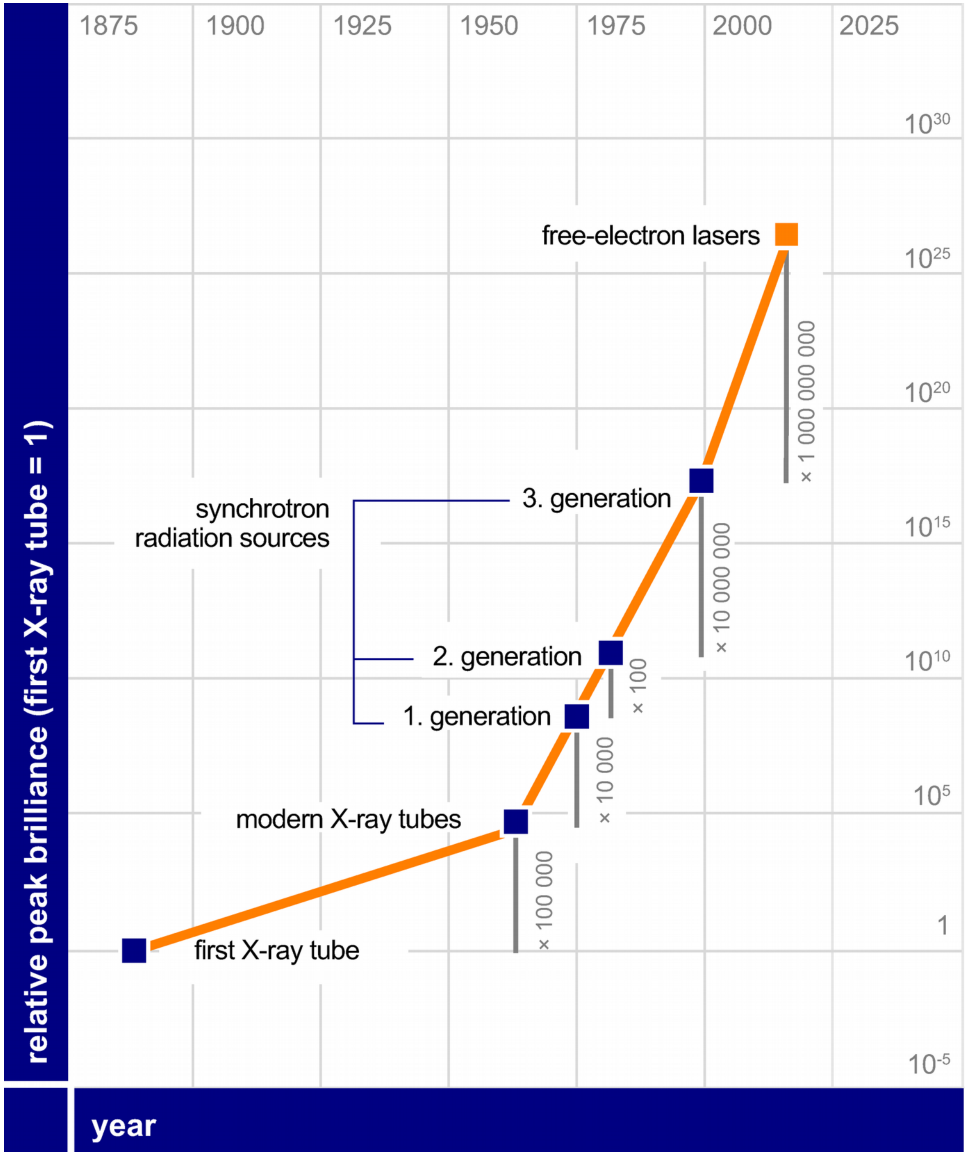
“absorption first”

“emission first”

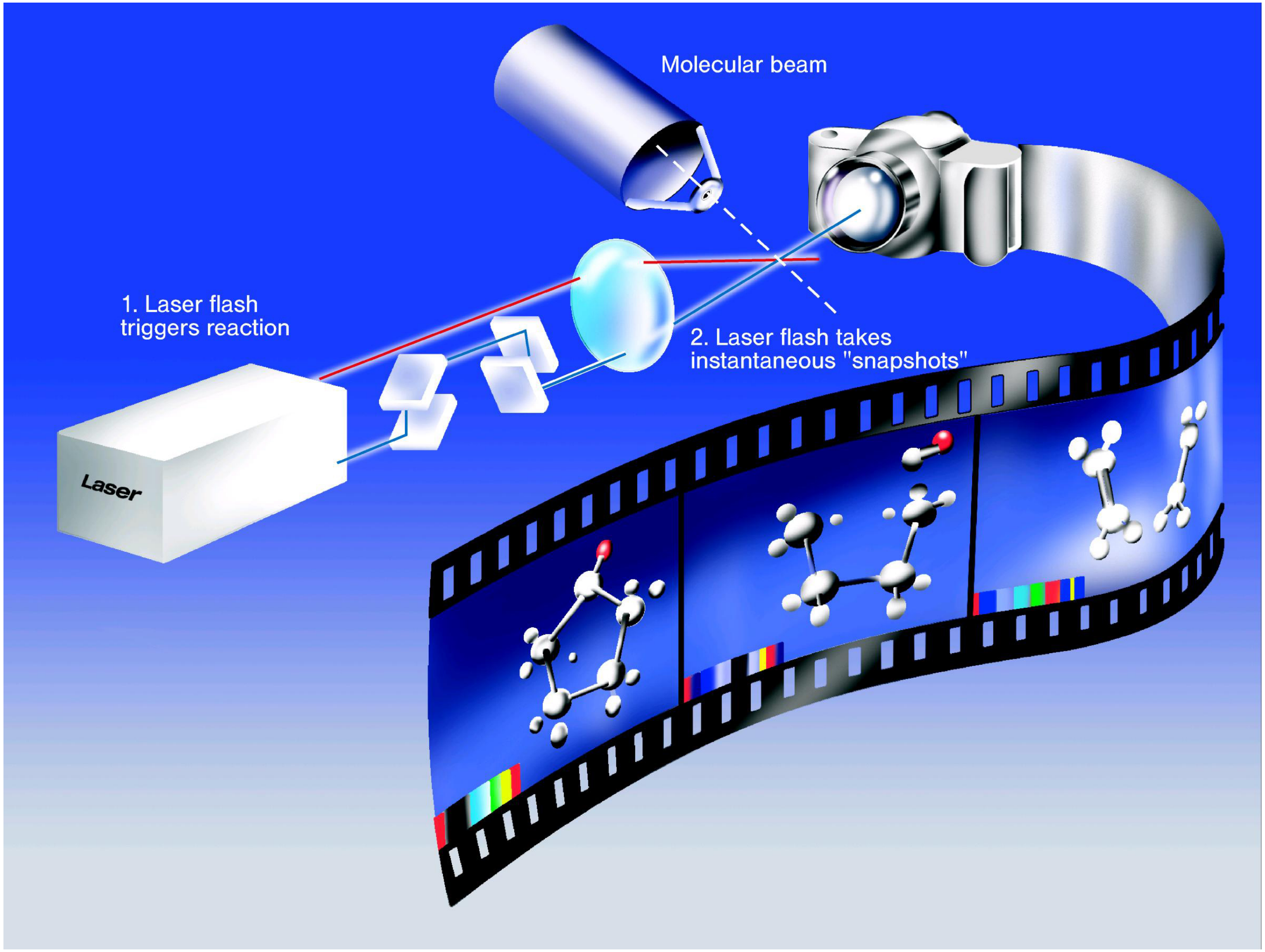


Relativistic origin of A^2

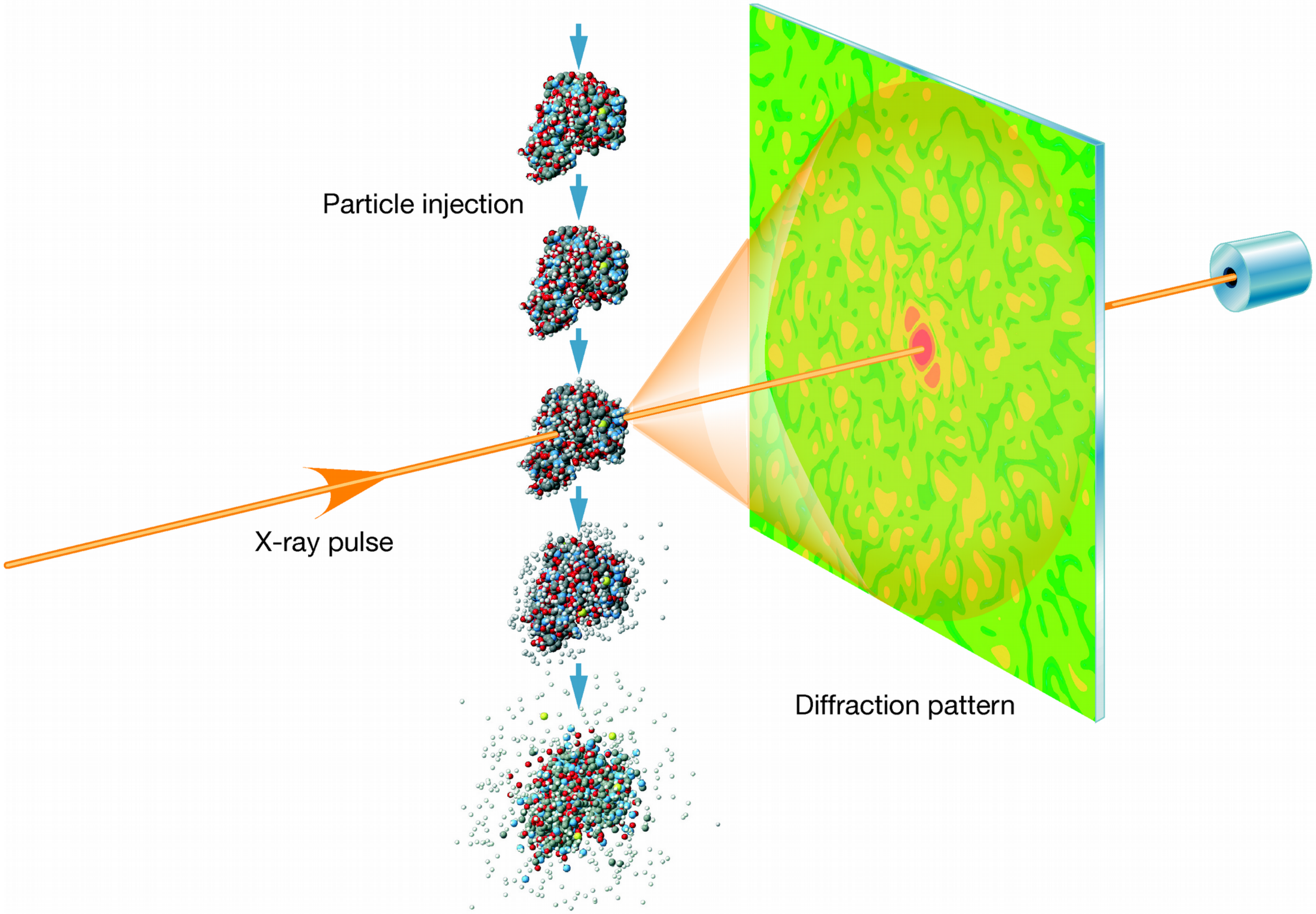
A brief history of x-ray intensity



Making molecular movies: a new tool for femtochemistry

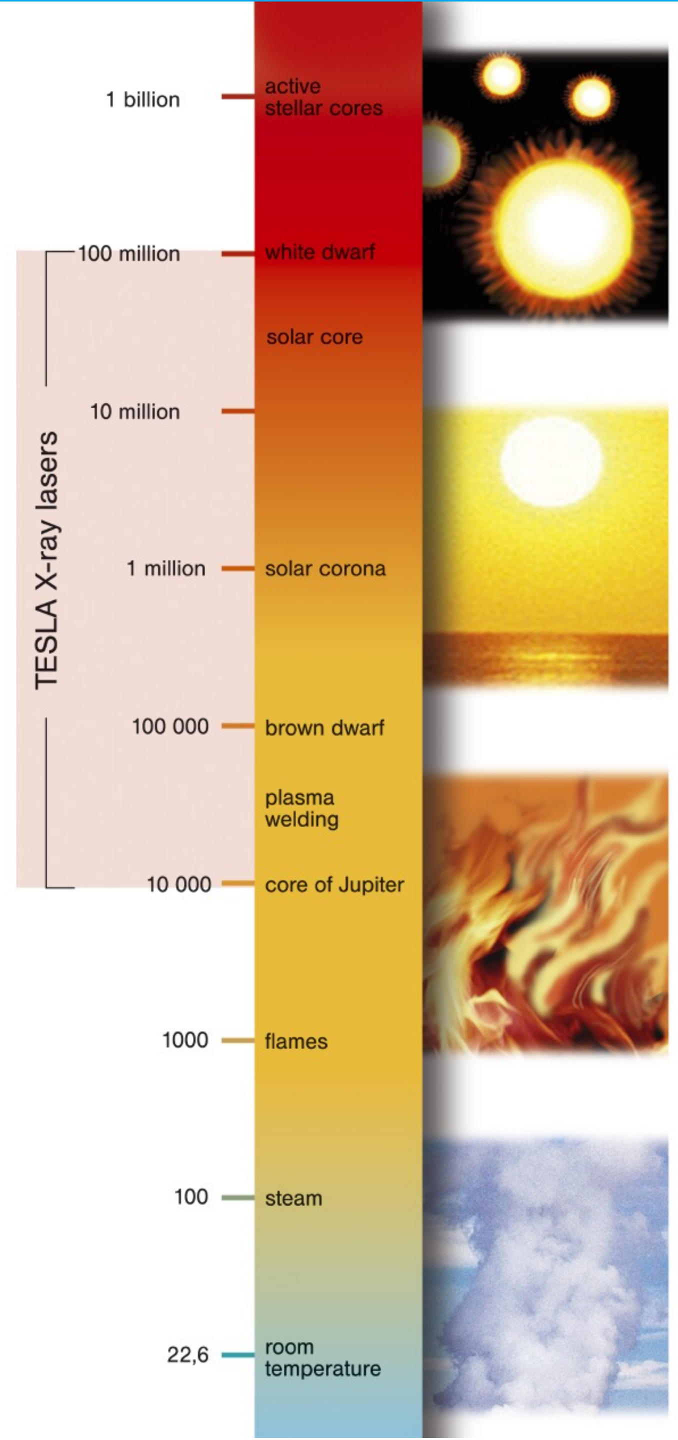


Single-shot structure determination of biomolecules



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

Generating and probing extreme states of matter



The first atomic-physics experiments at LCLS (fall 2009)

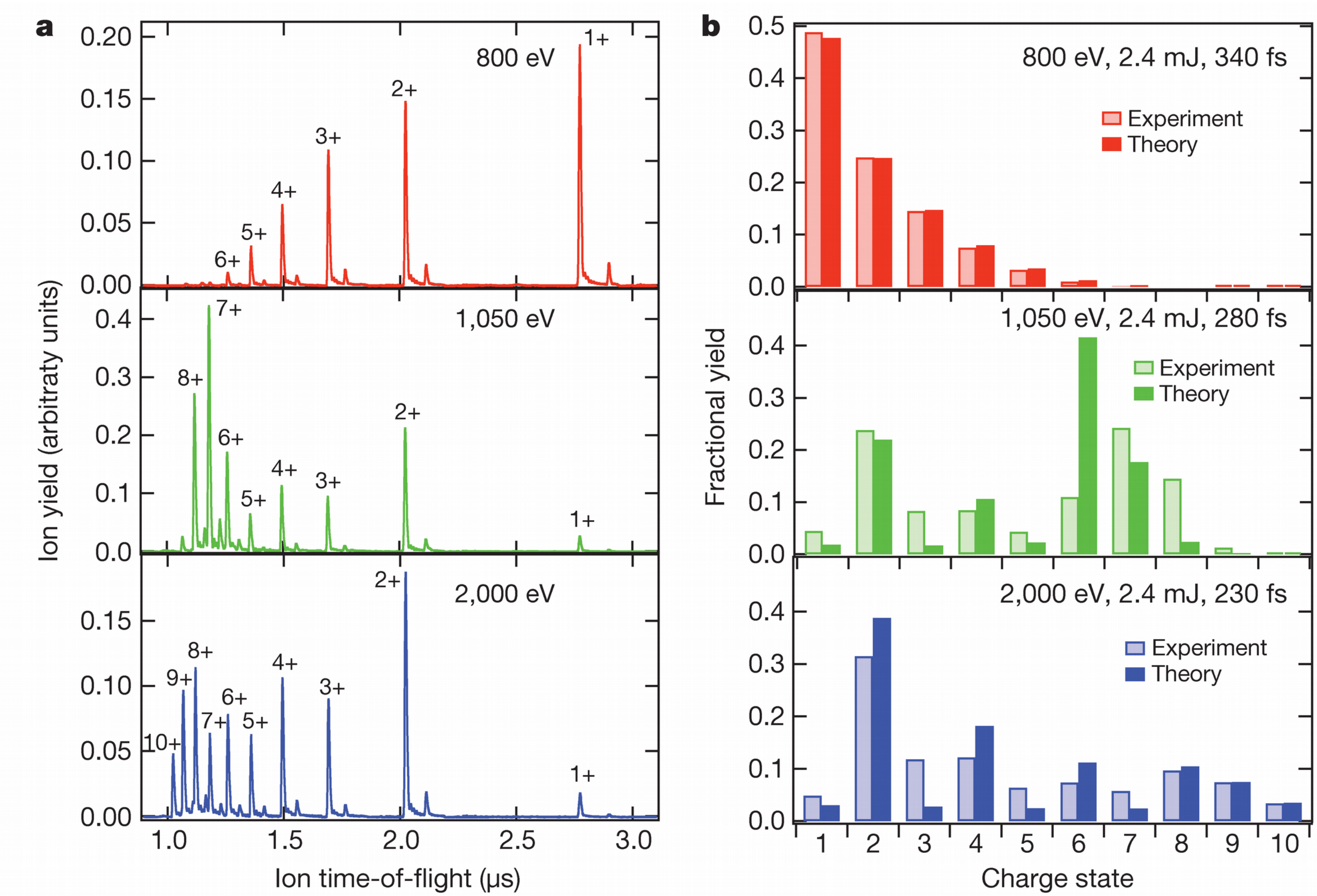
- Complete stripping of neon in a single x-ray pulse (removal of all 10 electrons)
  [L. Young *et al.*, Nature **466**, 56 (2010)]

- Double-core-hole formation in neon by beating the Auger decay of 1s-ionized Ne^{1+} (decay lifetime of 2.4 fs)
  [L. Young *et al.*, Nature **466**, 56 (2010)]

- Nonsequential two-photon ionization of Ne^{8+}
  [G. Doumy *et al.*, Phys. Rev. Lett. **106**, 083002 (2011)]

- Modification of Auger line profile in neon via x-ray-driven Rabi oscillations
  [E. P. Kanter *et al.*, Phys. Rev. Lett. **107**, 233001 (2011)]

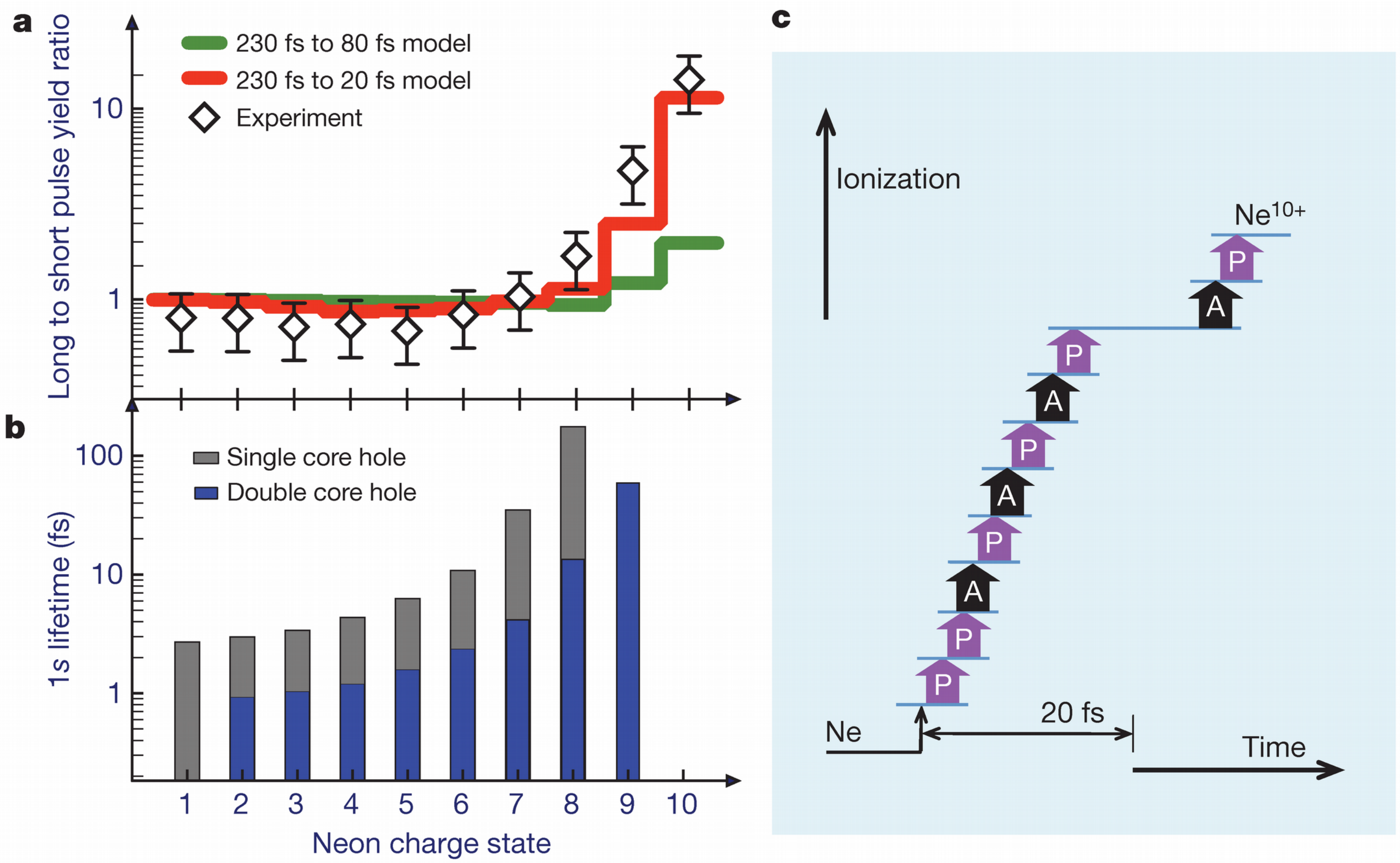
Neon charge states as a function of the photon energy



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

Counterintuitive impact of pulse duration

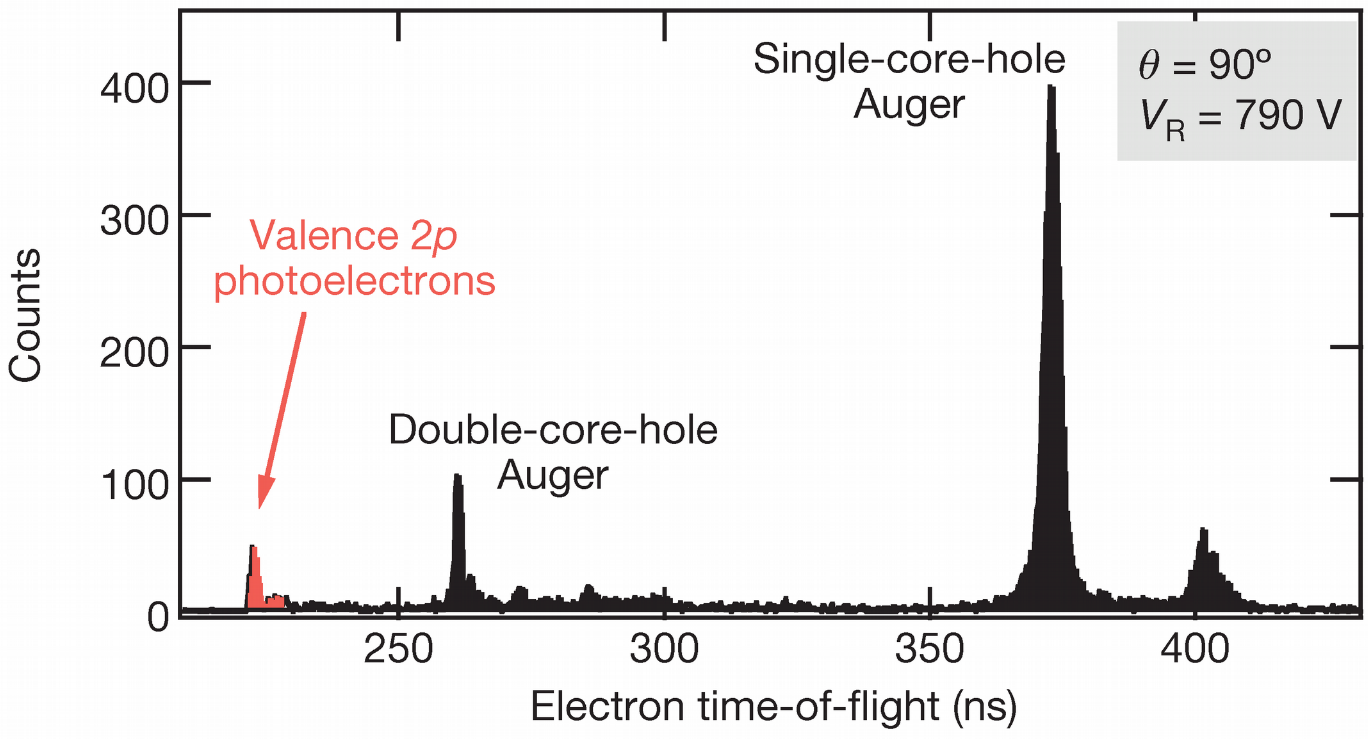
photon energy 2 keV, pulse energy 2 mJ



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

Observation of double-core-hole formation

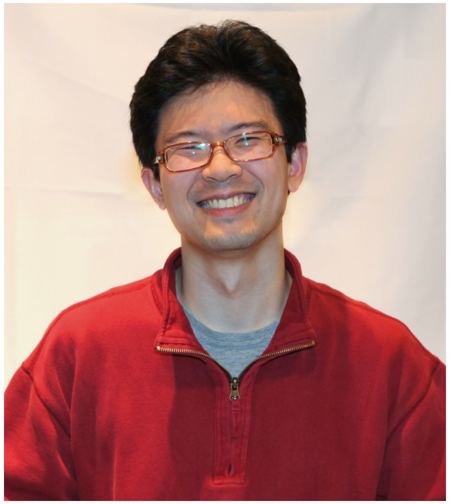
photon energy 1050 eV, pulse energy 2 mJ, nominal pulse duration 80 fs, electrons emitted perpendicular to x-ray polarization axis



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

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)

Number of active configurations = number of coupled rate equations

- C: $1s^2\ 2s^2\ 2p^2$

  → ***27*** *configurations*

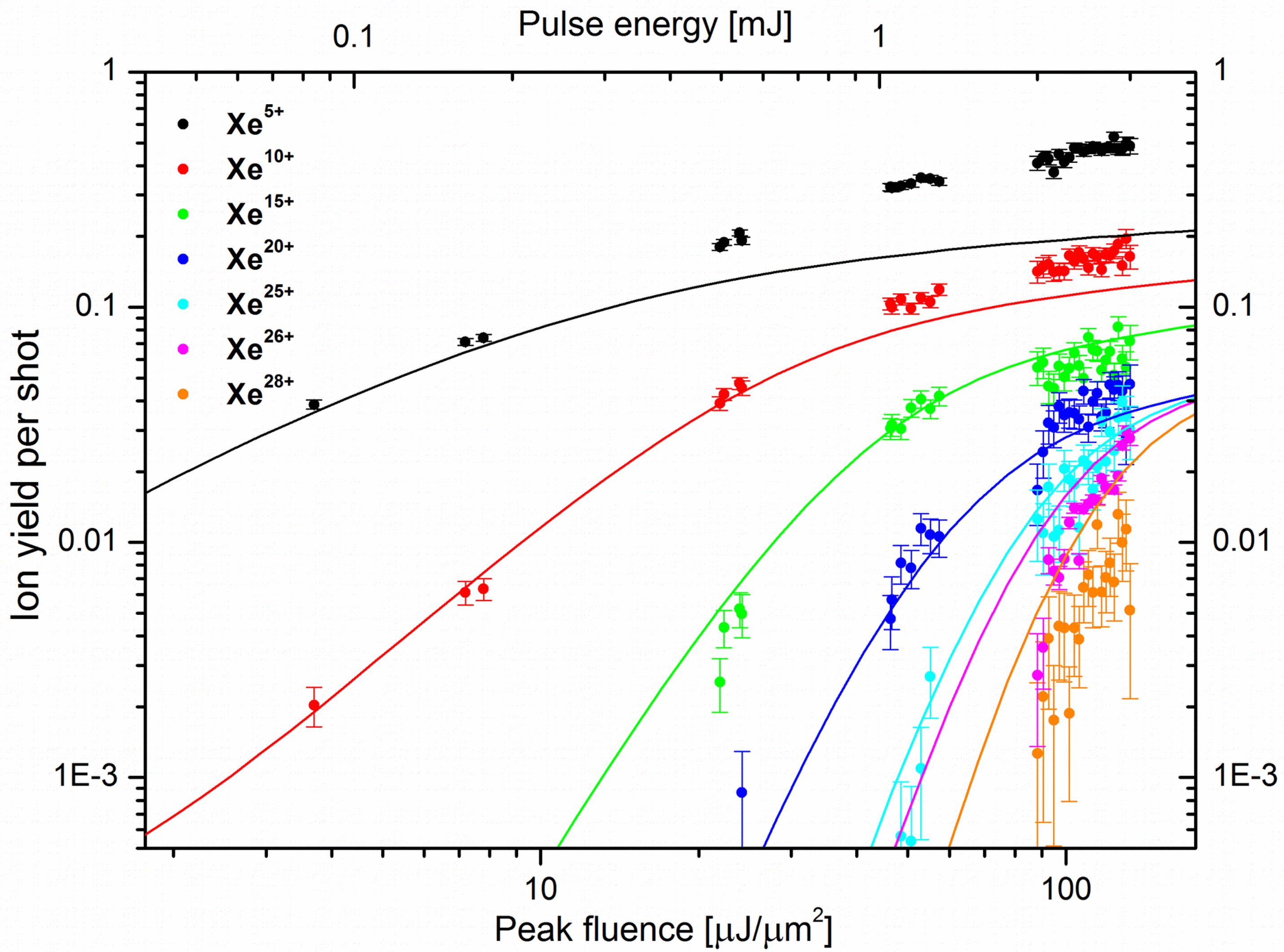
- Ne: $1s^2\ 2s^2\ 2p^6$

  → ***63*** *configurations*

- Xe: [$1\mathrm{s}^2\ 2\mathrm{s}^2\ 2\mathrm{p}^6$] $3s^2\ 3p^6\ 3d^{10}\ 4s^2\ 4p^6\ 4d^{10}\ 5s^2\ 5p^6$

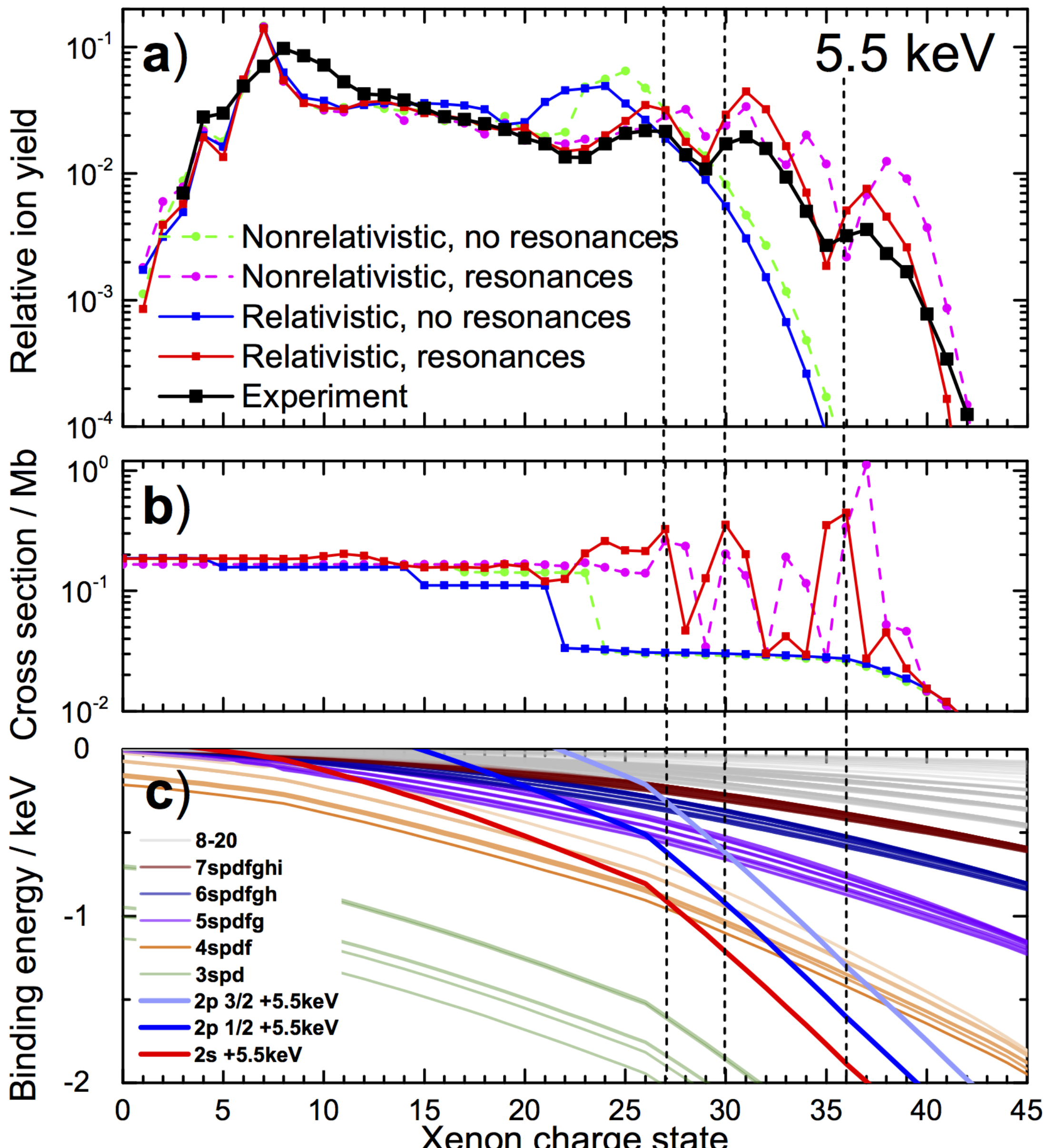
  → ***1,120,581*** *configurations*
  (excluding ionization from the K and L shells)

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*

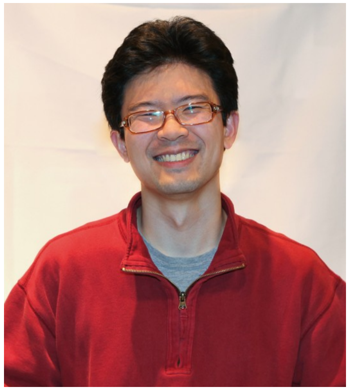
- Relativistic, including resonances (n_{max} = 30, l_{max} = 7)

  → $\boldsymbol{2.6 \times 10^{68}}$ *configurations*

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

XMDYN

Zoltan Jurek



Sang-Kil Son

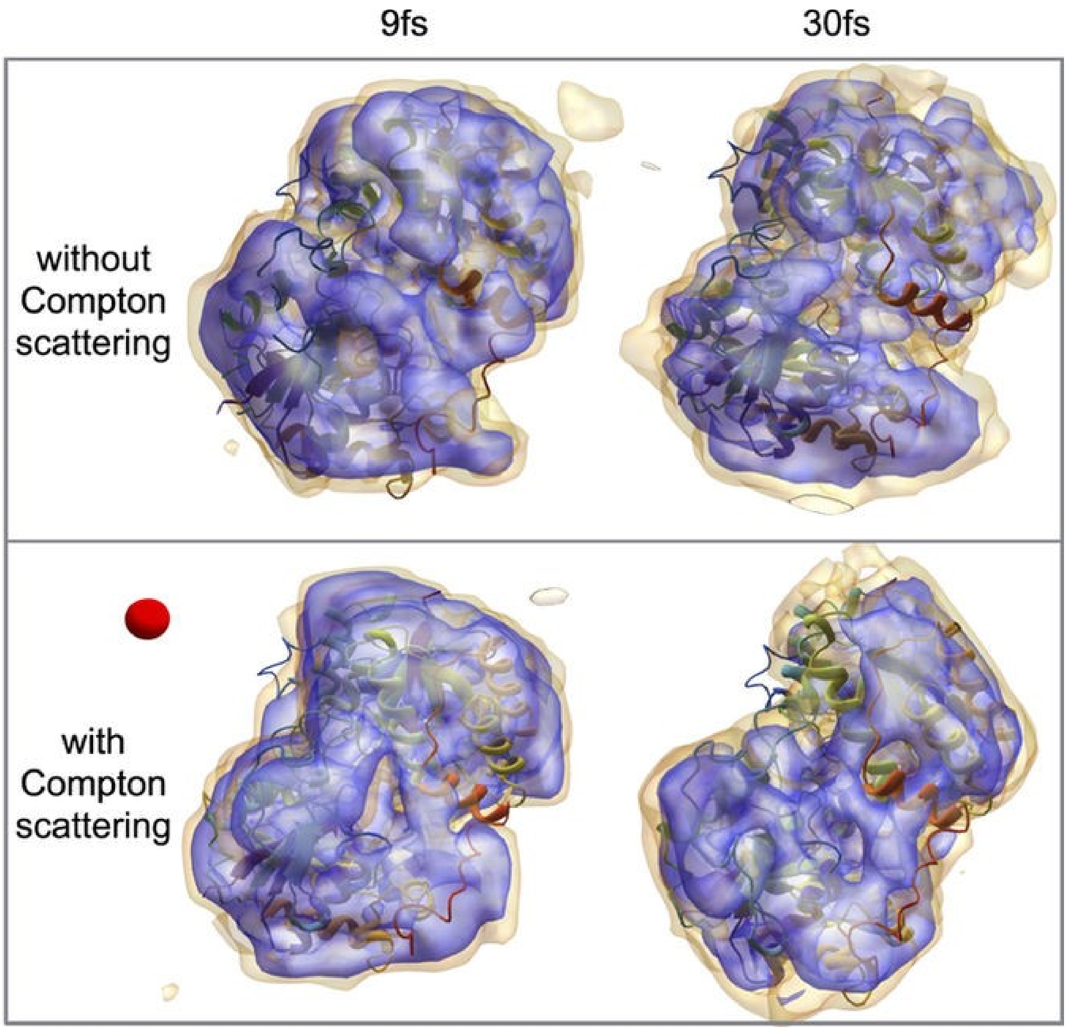
→ ab-initio calculation of atomic parameters (subshell photoionization cross sections, electronic decay rates, x-ray scattering cross sections) for arbitrary electronic configurations → uses XATOM

→ 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

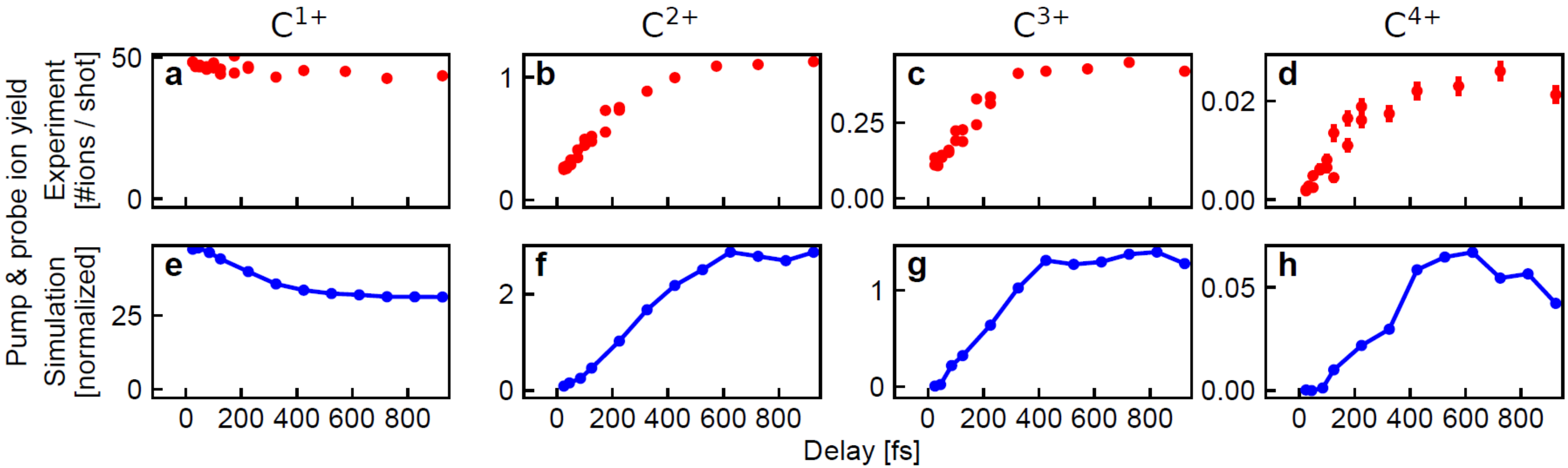


nitrogenase iron protein

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

X-ray pump / x-ray probe study of C_{60}

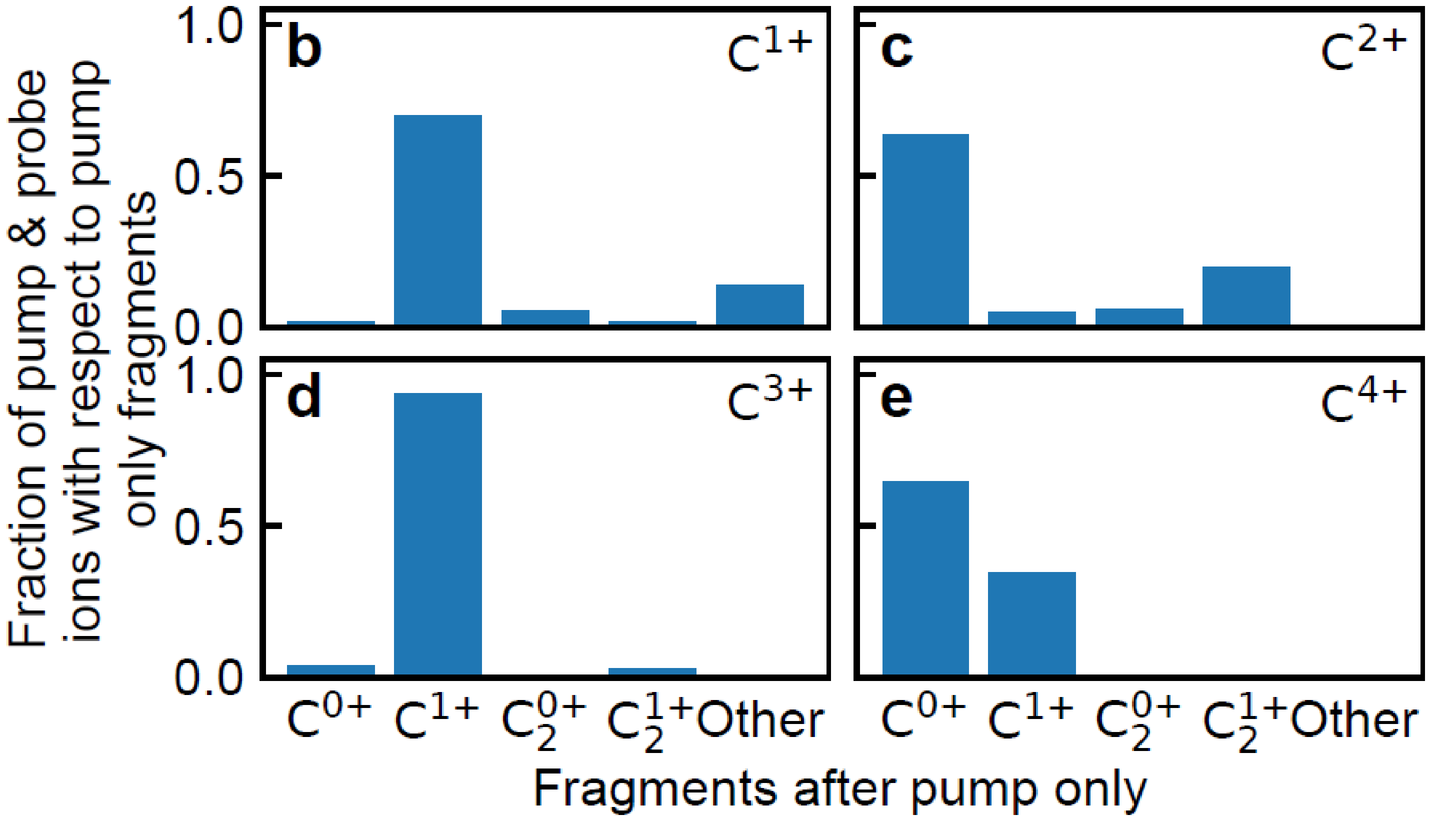
Collaboration with **Nora Berrah, Jon Marangos**, et al.
Experiment carried out at LCLS



Photon energy: 640 eV
Focal area: 400 μm^2
Pump (probe) pulse duration: 20 fs (10 fs)
Combined pulse energy: 0.77 mJ, shared 45%/55% (pump/probe)
Pump peak intensity: 4 x 10^{15} W/cm^2

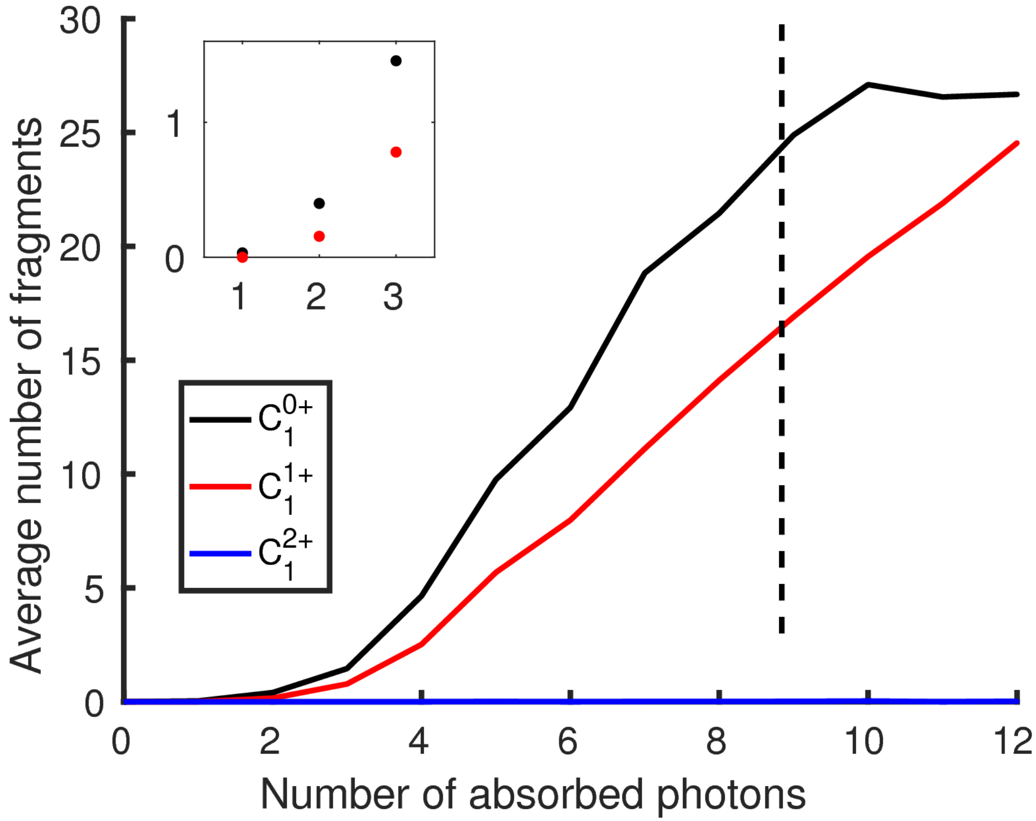
N. Berrah *et al.*, Nature Phys. **15**, 1279–1283 (2019).

Relative weight of the parent of the final C^{1+}, C^{2+}, C^{3+}, C^{4+}



N. Berrah *et al.*, Nature Phys. **15**, 1279–1283 (2019).

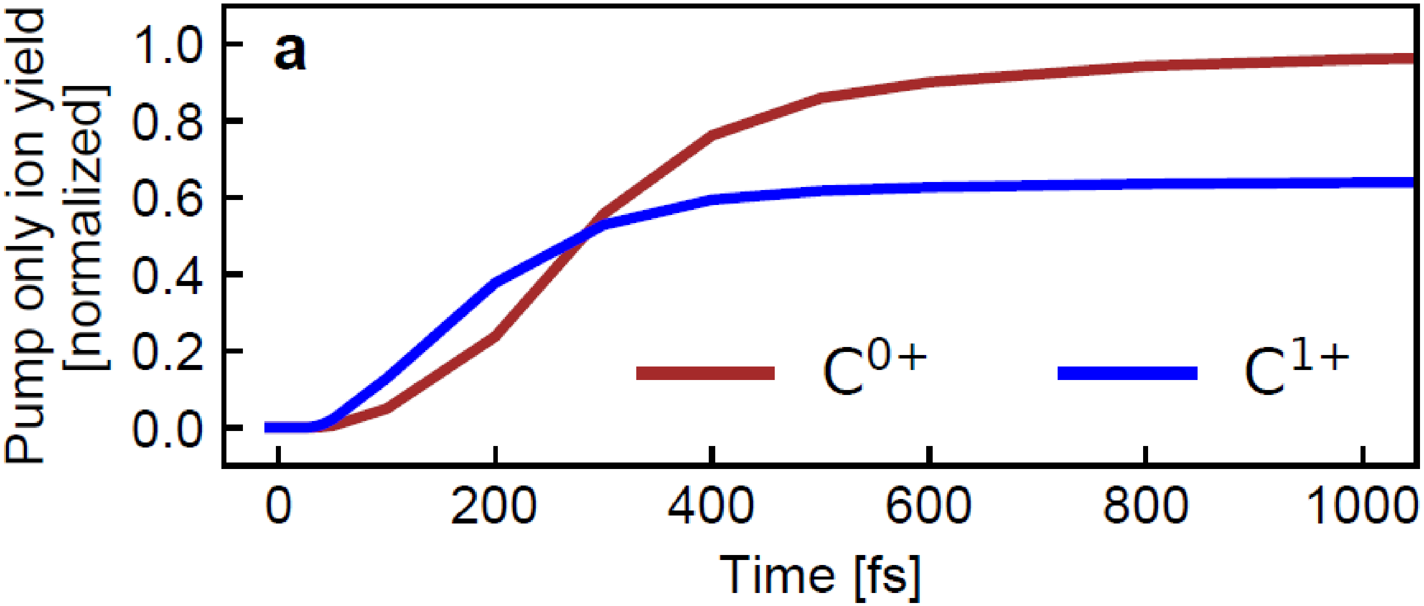
Appearance of the neutral and singly charged atomic fragments, as a function of the number of absorbed x-ray photons



The dashed line corresponds to the average response of the fullerene to the pump pulse when the molecule is in the center of the focus.

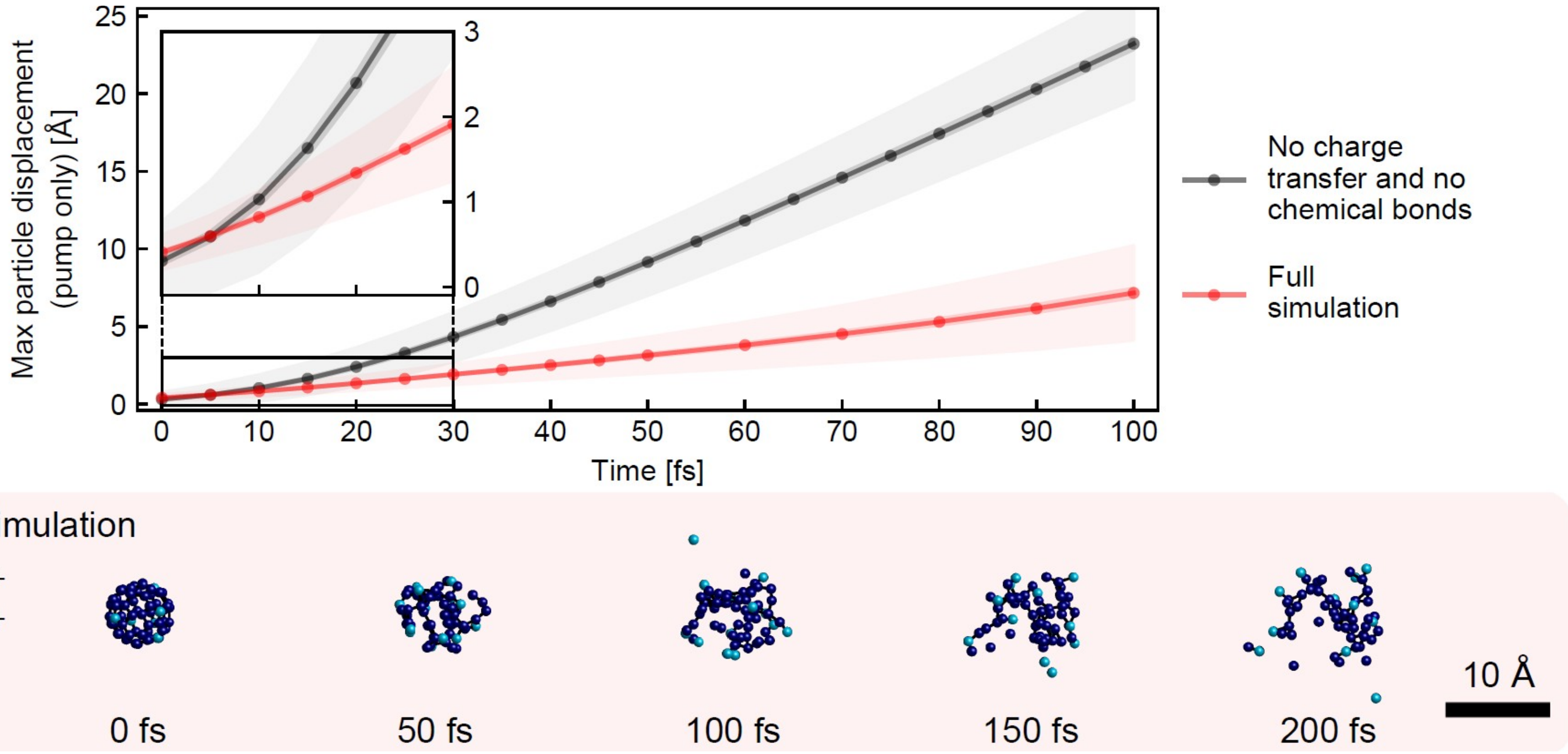
N. Berrah *et al.*, Nature Phys. **15**, 1279–1283 (2019).

Simulated real-time evolution of the volume-integrated yield using the pump pulse only



N. Berrah *et al.*, Nature Phys. **15**, 1279–1283 (2019).

Evolution of the molecular structure (pump only)



N. Berrah *et al.*, Nature Phys. **15**, 1279–1283 (2019).

Conclusions

- Radiation damage at high x-ray intensity of relevance to applications of XFELs.
- 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.
- At x-ray intensities used for SFX (serial femtosecond x-ray crystallography), there is hardly any atomic displacement during the x-ray pulse.