Atoms in intense light fields

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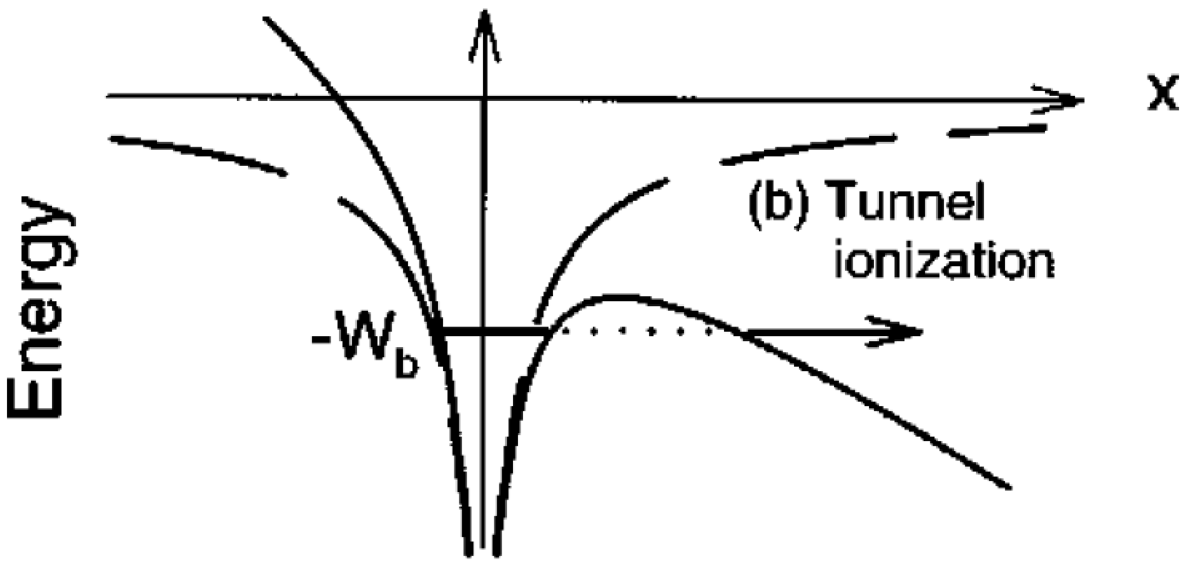
Hamburg, Germany

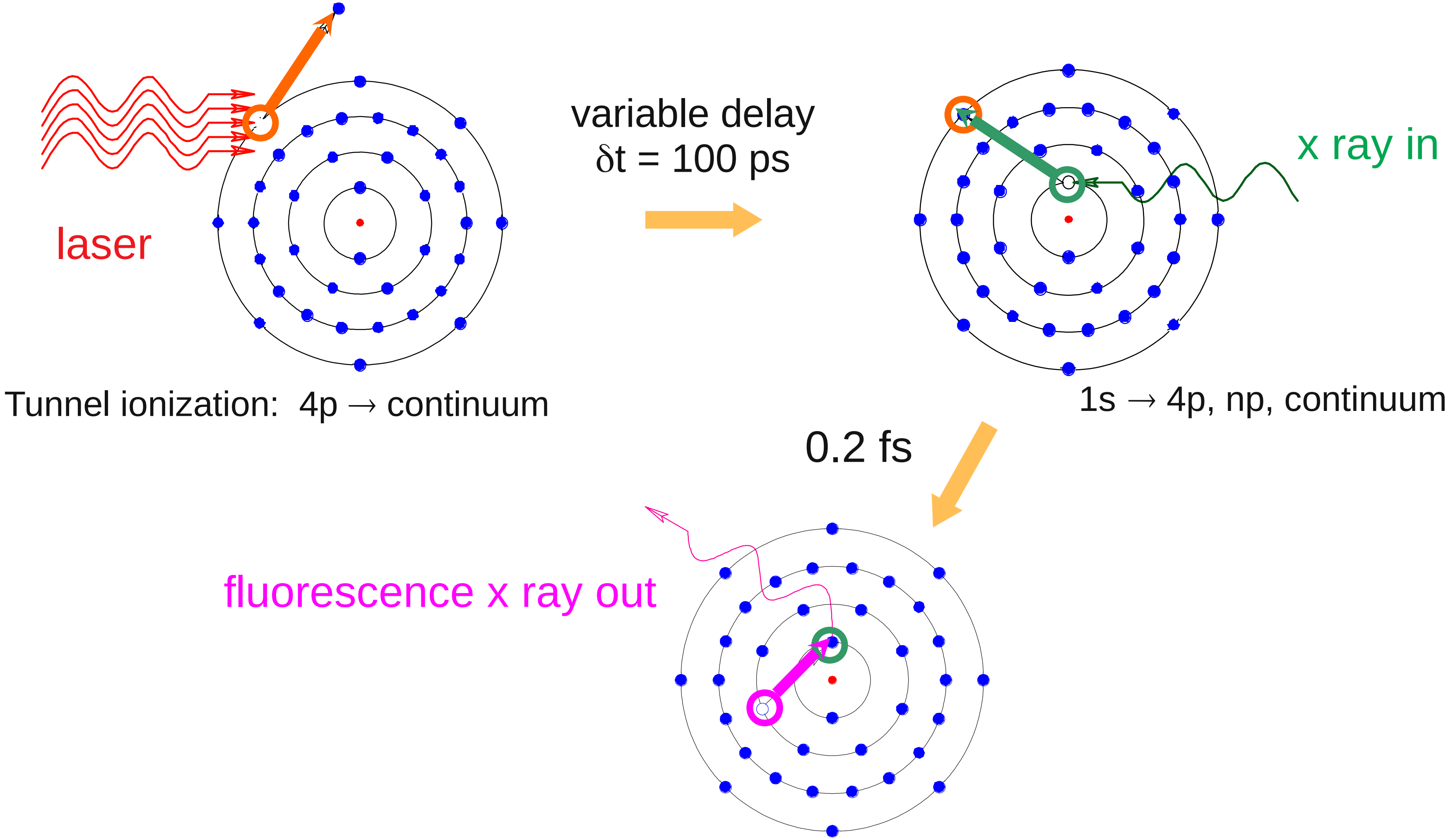
Ionization in nonperturbatively strong optical fields

Brabec and Krausz, Rev. Mod. Phys. **72**, 545 (2000).

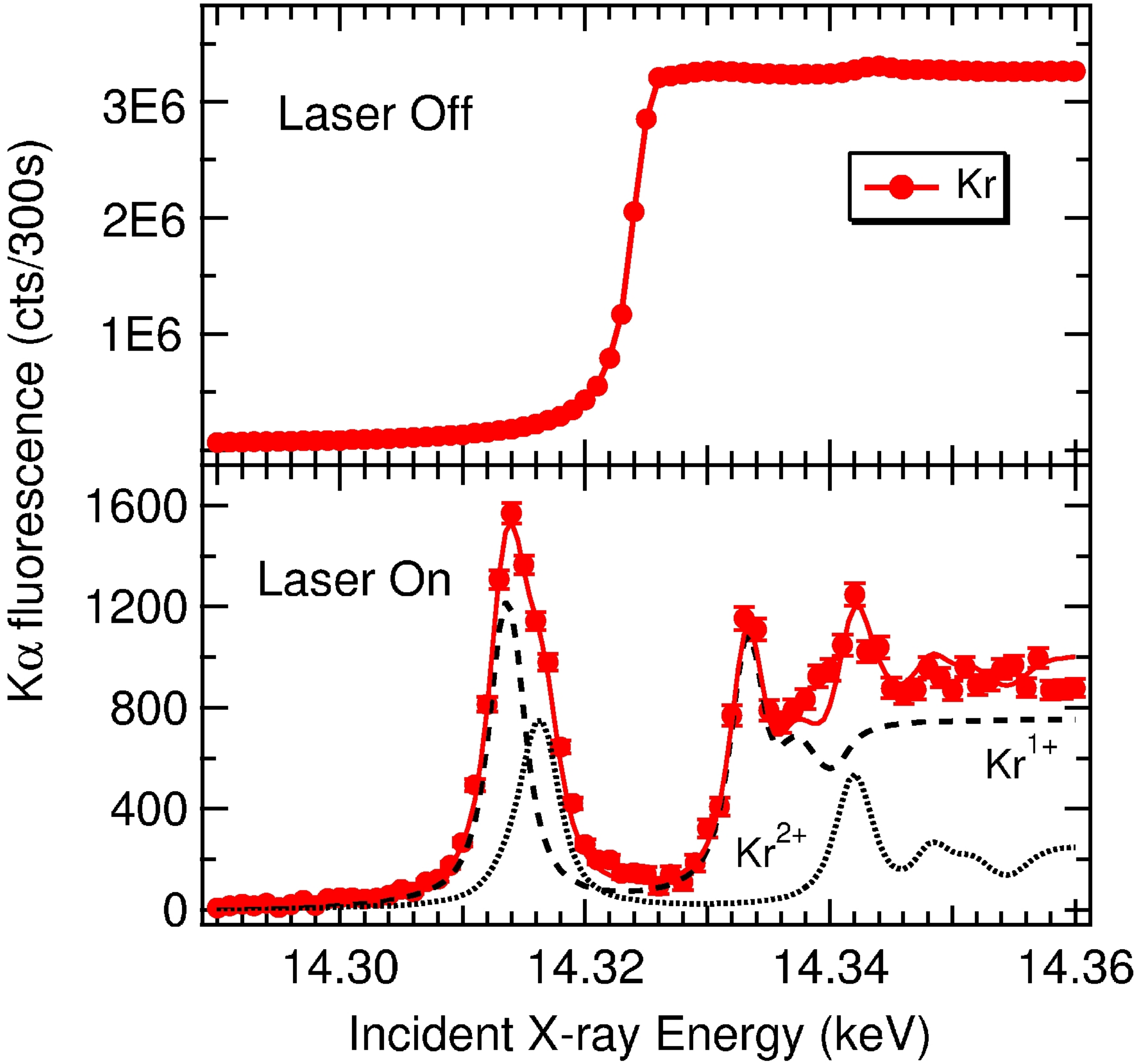


Assumptions:
- the photon energy is much smaller than the electron binding energy
- electric dipole approximation is valid
- multipolar Hamiltonian is used (“length form” or “length gauge”)
- field is so strong that the electronic response at a given time t during the optical cycle is fast in comparison to optical period (2.7 fs at 800 nm).

Resonant core excitation of laser-ionized krypton atoms

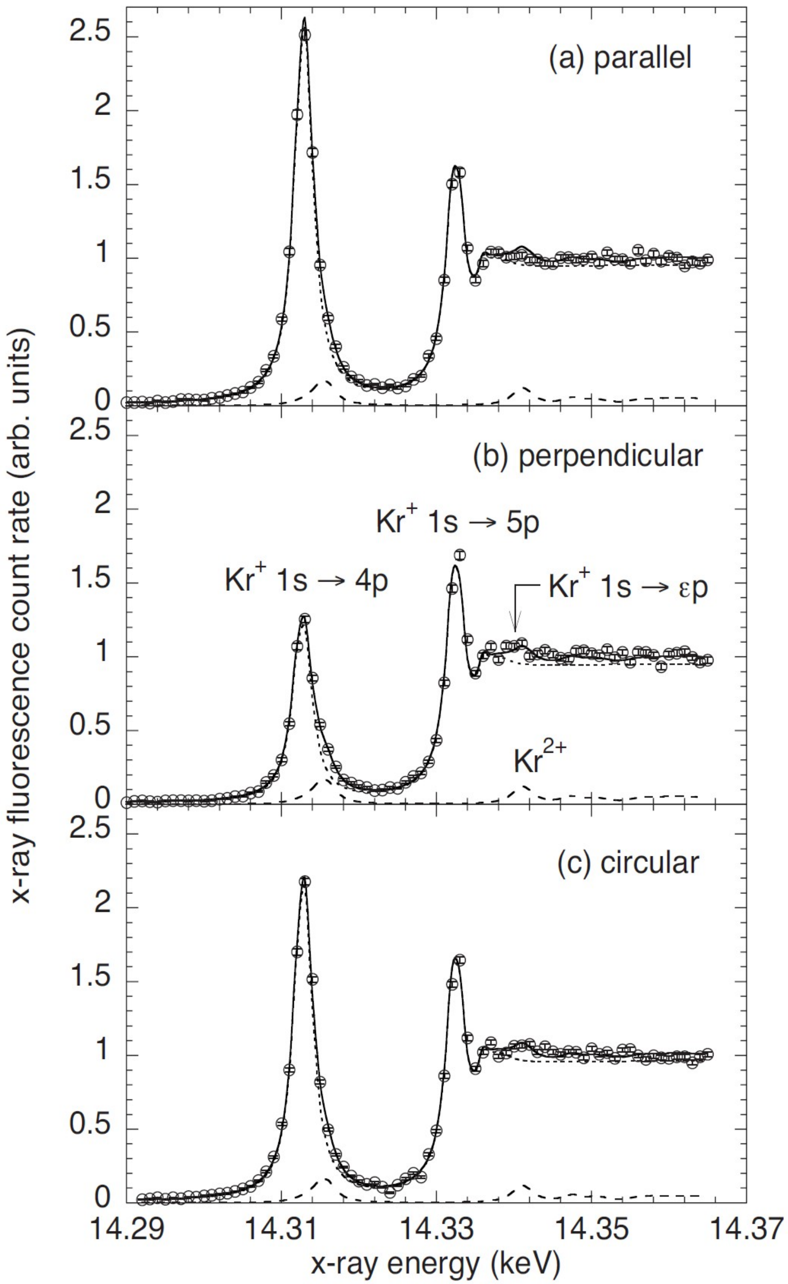


Kr near-edge absorption spectrum



Young *et al.*,
Phys. Rev. Lett. **97**, 083601 (2006).

Circularly polarized laser



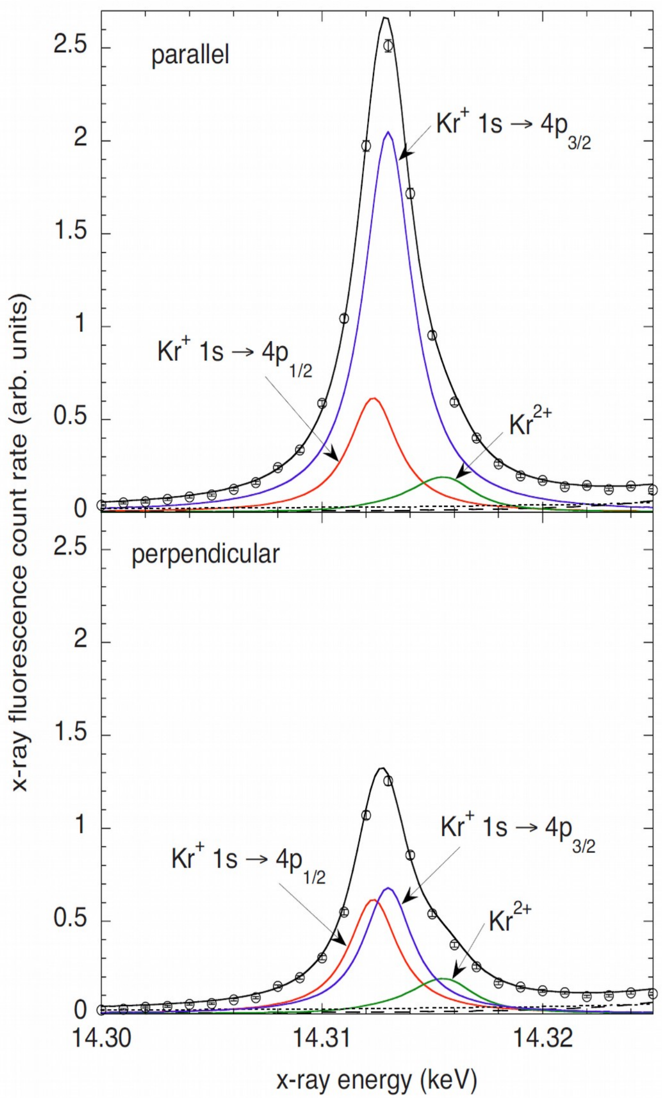
Polarized x-ray absorption probes aligned ion states

observe R = 2 : 1 ratio for || vs. ⊥ (at the 1s → 4p resonance)

Young *et al.*, Phys. Rev. Lett. **97**, 083601 (2006).

Southworth *et al.*, Phys. Rev. A **76**, 043421 (2007).

Quantum-state populations of strong-field-generated Kr^+



$$\sigma(\omega_x, 0^\circ) = 2\rho_{3/2,1/2}\sigma_{3/2}(\omega_x) + \rho_{1/2,1/2}\sigma_{1/2}(\omega_x)$$

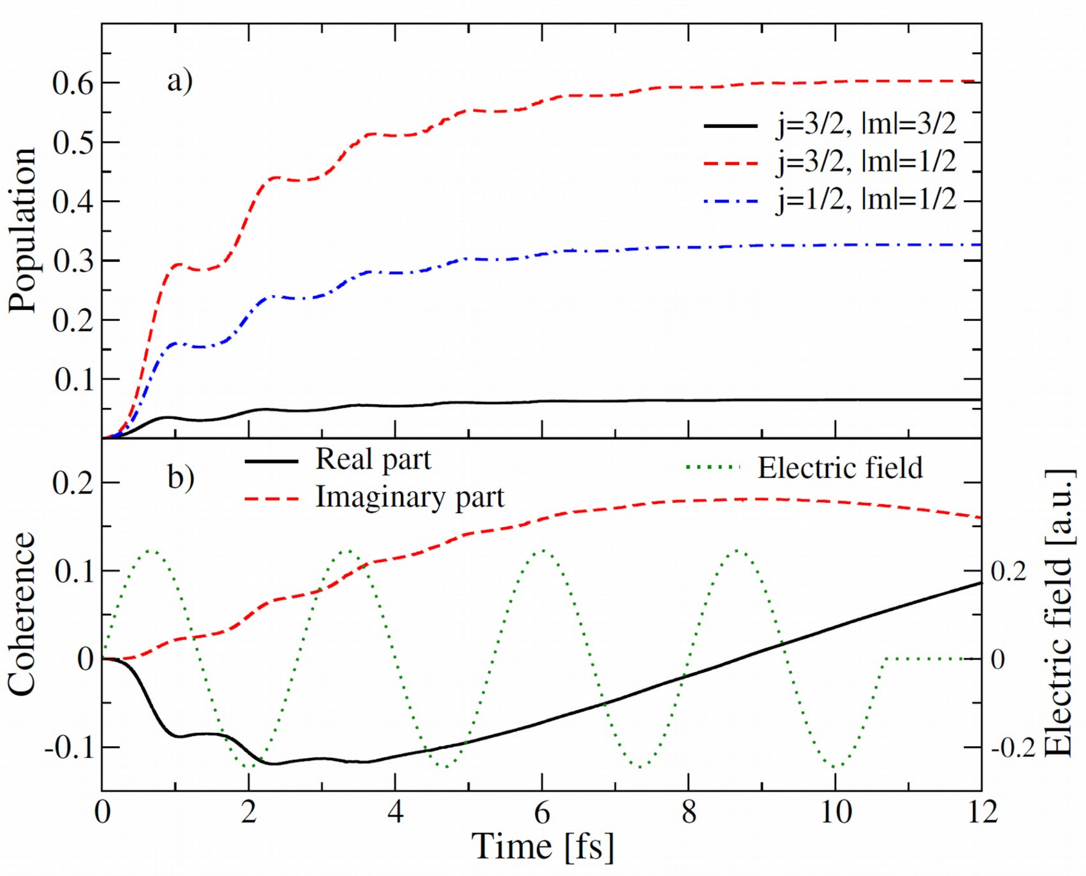
$$\sigma(\omega_x, 90^\circ) = \frac{1}{2}\{\rho_{3/2,1/2} + 3\rho_{3/2,3/2}\}\sigma_{3/2}(\omega_x) + \rho_{1/2,1/2}\sigma_{1/2}(\omega_x)$$

	$\rho_{j,\|m\|}(\%)$	
$\|j,m\rangle$	Experimental	Theoretical
$\|\frac{3}{2},\pm\frac{1}{2}\rangle$	59±6	71
$\|\frac{1}{2},\pm\frac{1}{2}\rangle$	35±4	25
$\|\frac{3}{2},\pm\frac{3}{2}\rangle$	6±6	4

Southworth *et al.*, Phys. Rev. A **76**, 043421 (2007).

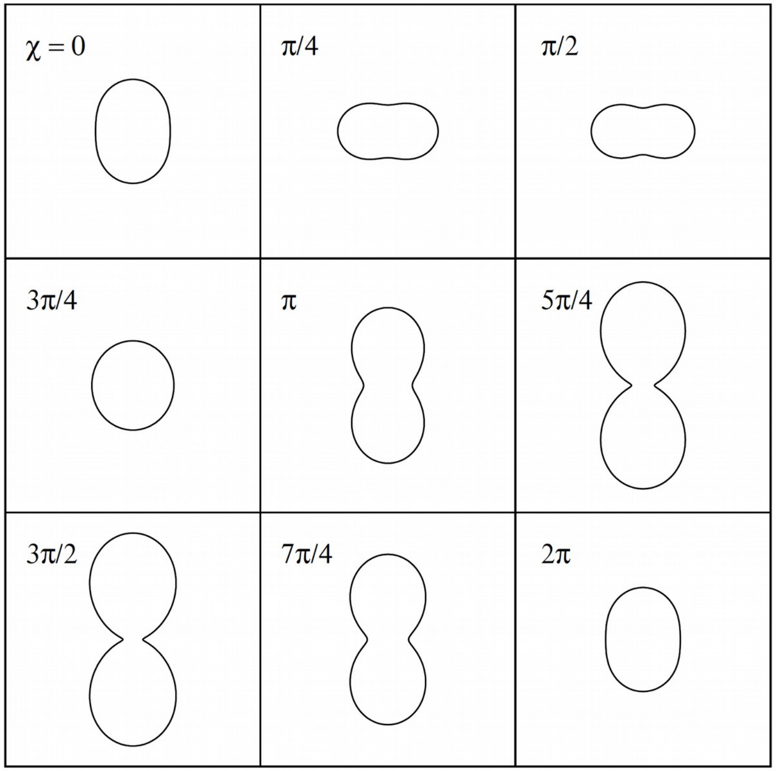
Time evolution of the ion density matrix of neon

800 nm; four cycles; field amplitude 0.245 a.u.



N. Rohringer and R. Santra, Phys. Rev. A **79**, 053402 (2009).

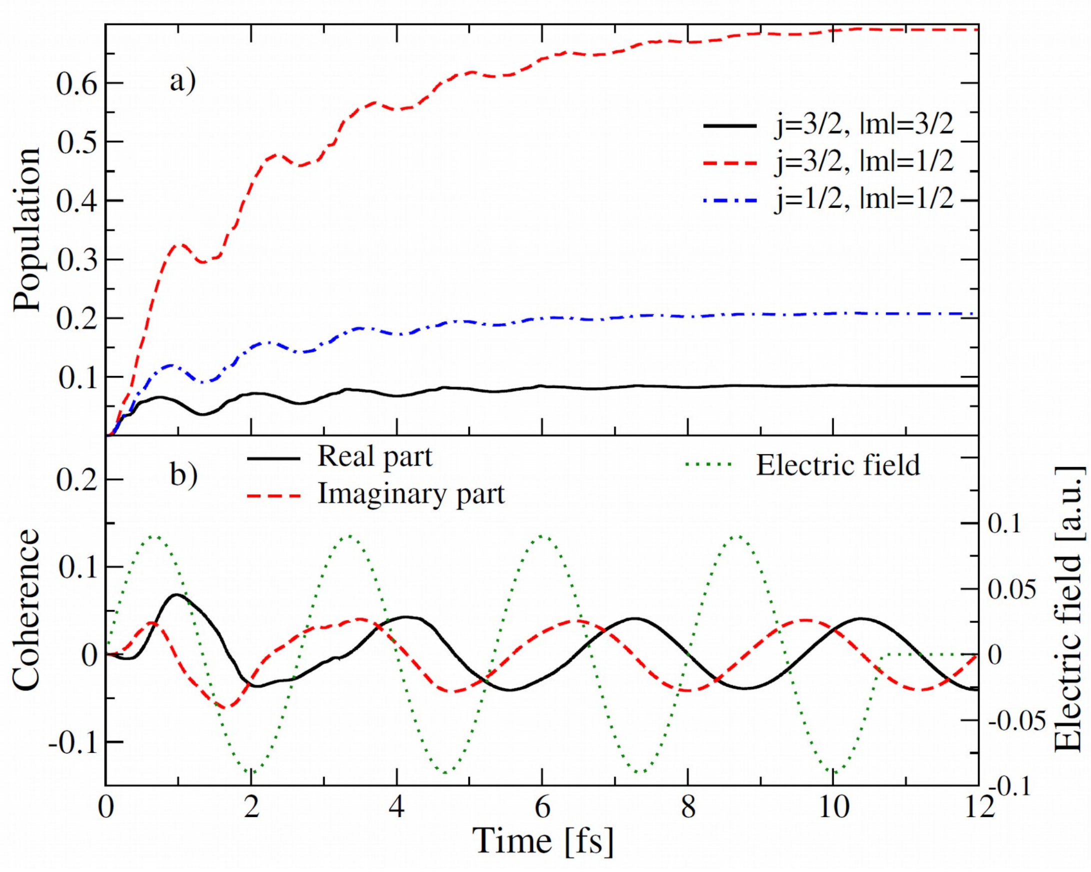
Time evolution of the hole density of laser-generated Ne^+



N. Rohringer and R. Santra, Phys. Rev. A **79**, 053402 (2009).

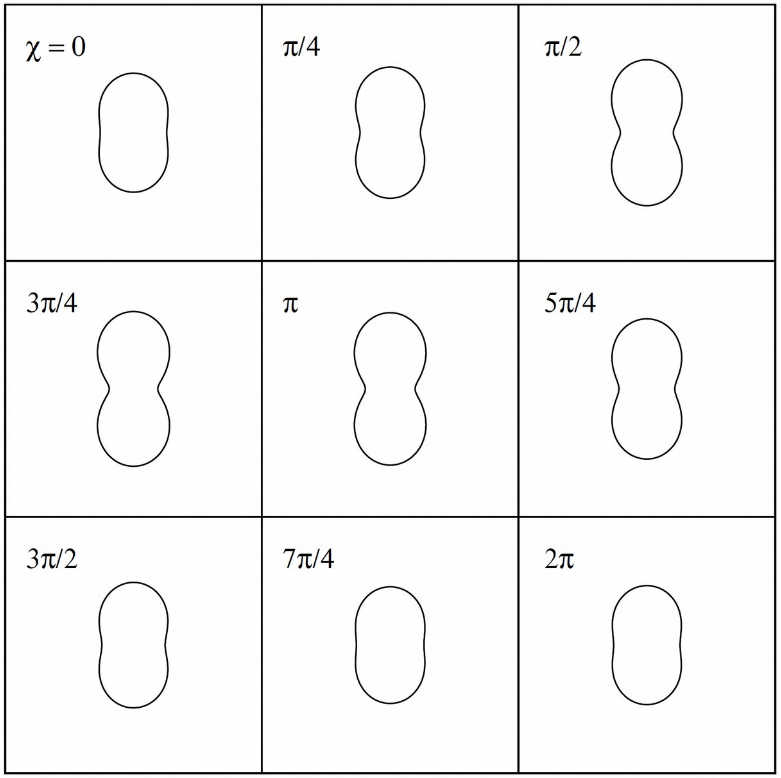
Time evolution of the ion density matrix of xenon

800 nm; four cycles; field amplitude 0.09 a.u.



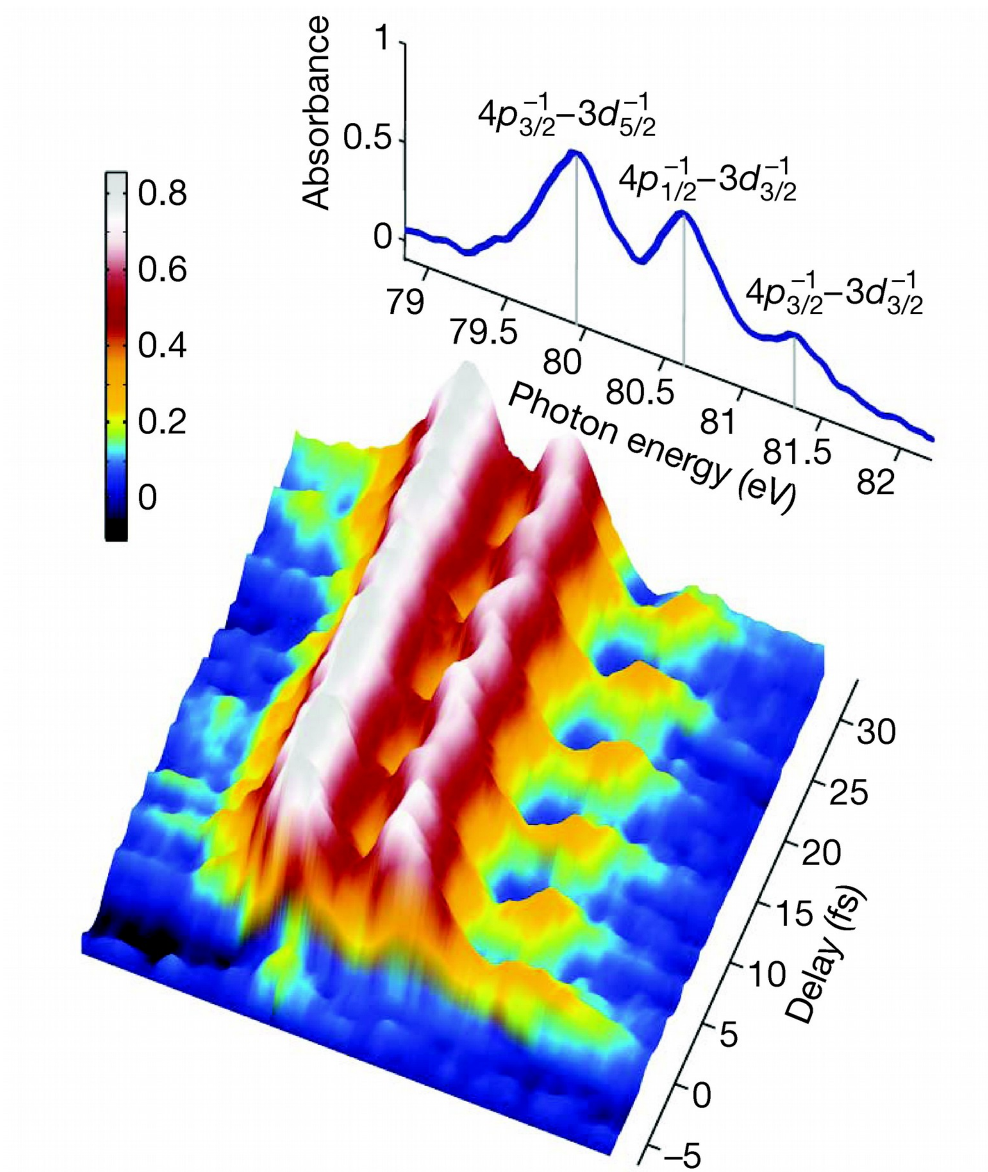
N. Rohringer and R. Santra, Phys. Rev. A **79**, 053402 (2009).

Time evolution of the hole density of laser-generated Xe^+



N. Rohringer and R. Santra, Phys. Rev. A **79**, 053402 (2009).

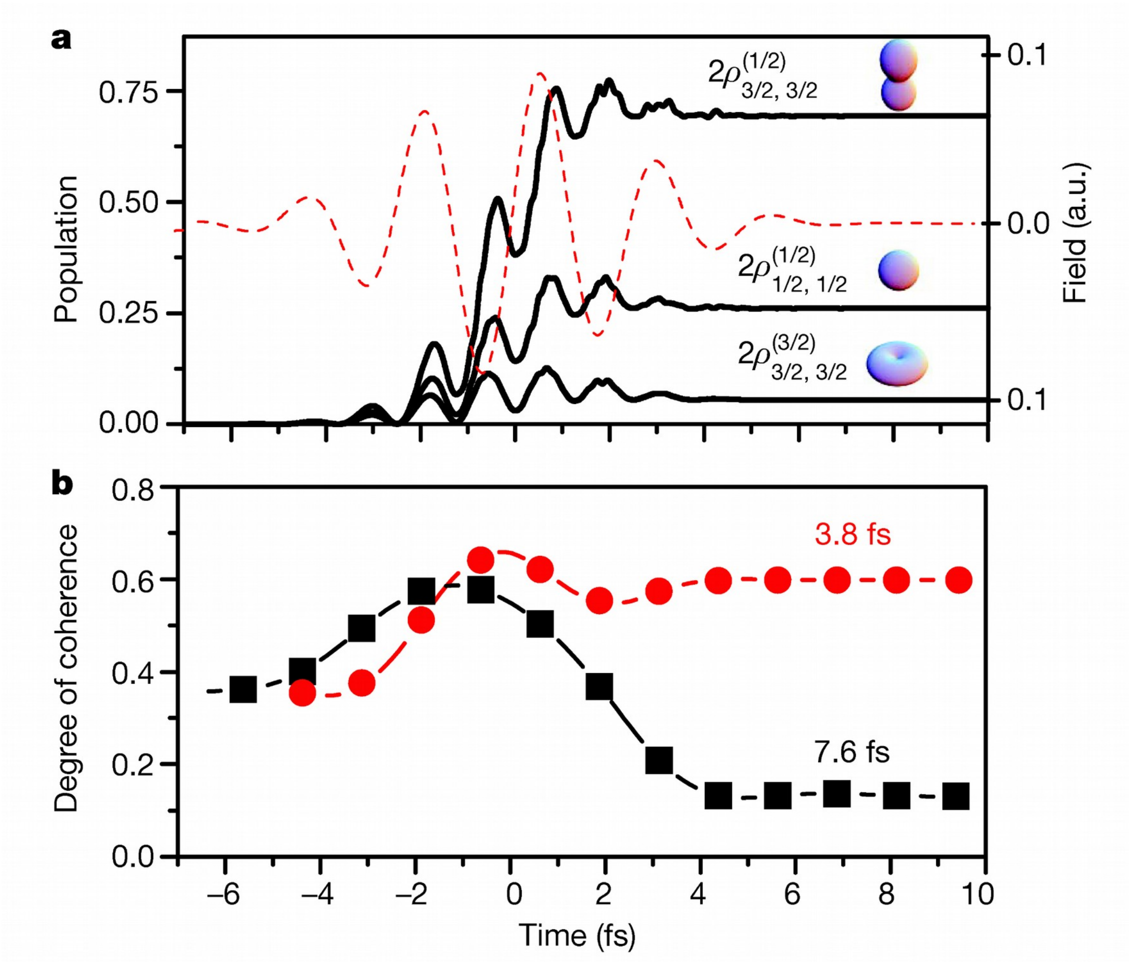
Attosecond transient absorption spectroscopy



Experiment on krypton at a near-IR peak intensity near 10^{14} W/cm^2

E. Goulielmakis *et al.*, Nature **466**, 739 (2010).

Calculated hole populations and degree of coherence

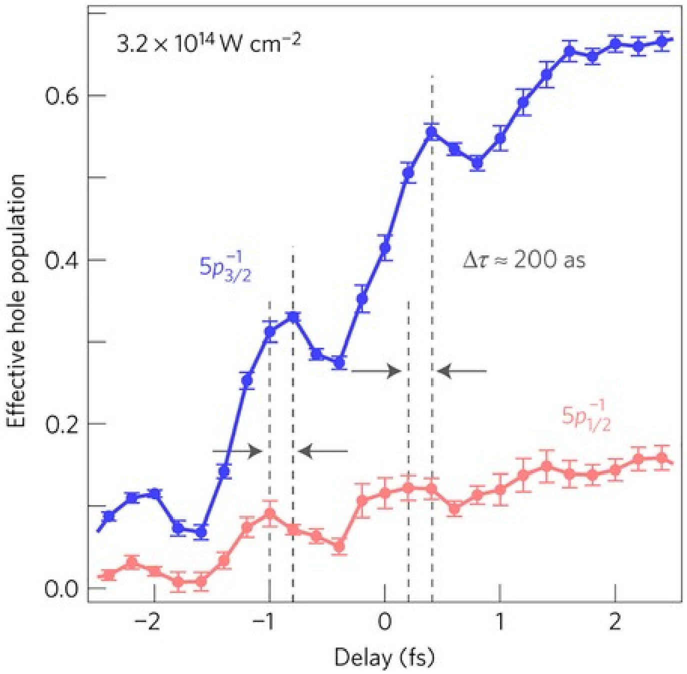


E. Goulielmakis *et al.*, Nature **466**, 739 (2010).

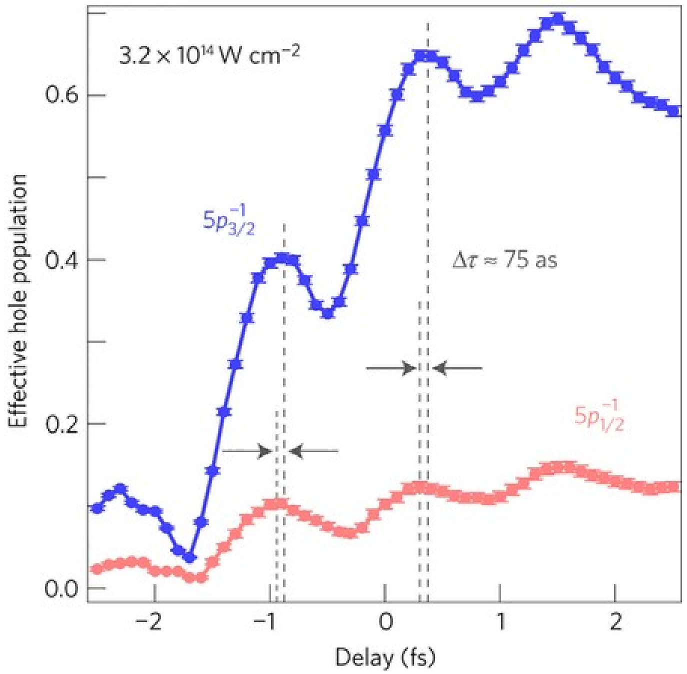
Calculated degree of electronic coherence (~ 0.6) is consistent with the transient-absorption data

Evolution of the hole population (in xenon)

Experiment

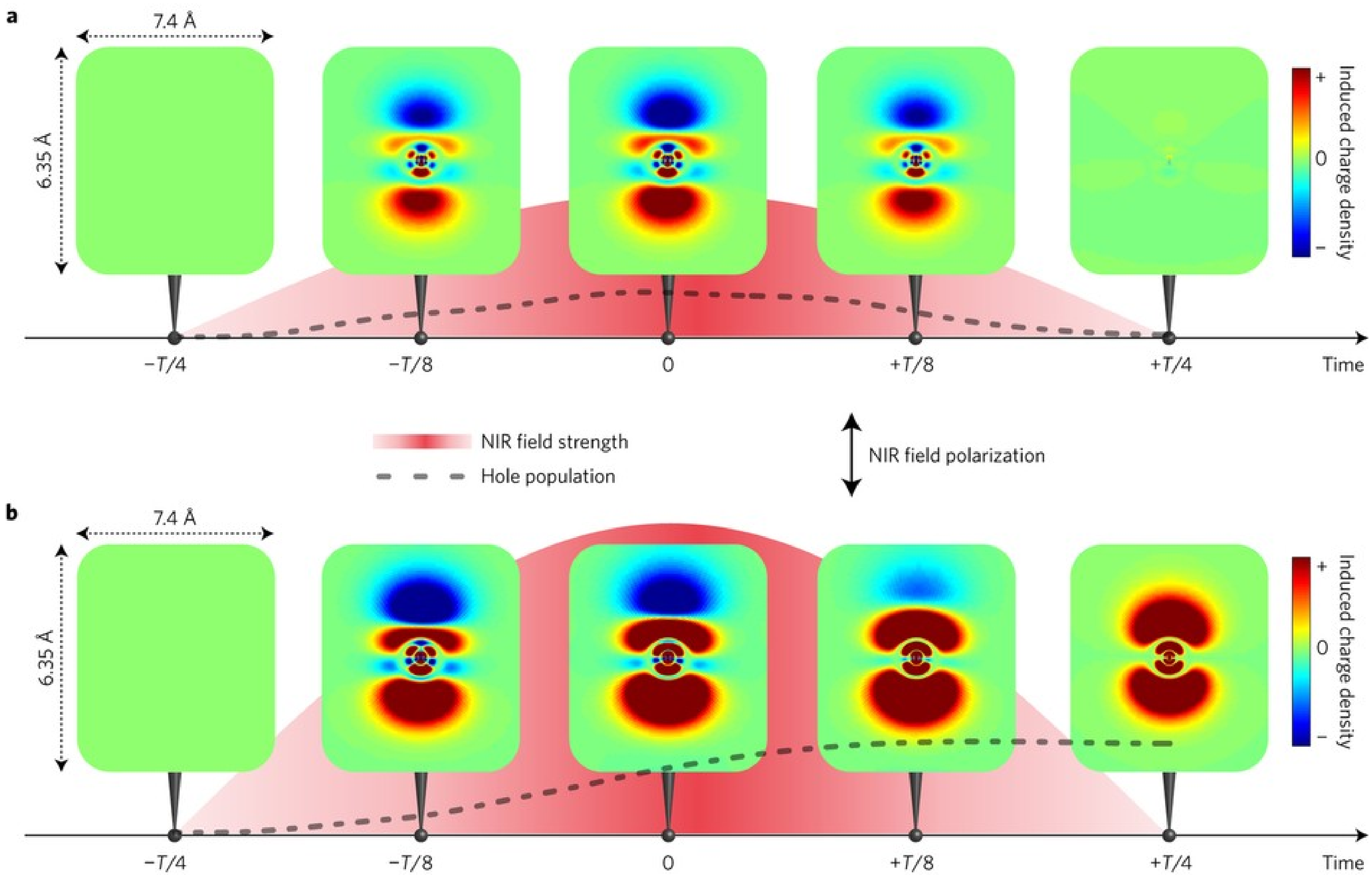


Theory



M. Sabbar *et al.*, Nature Phys. **13**, 472 (2017).

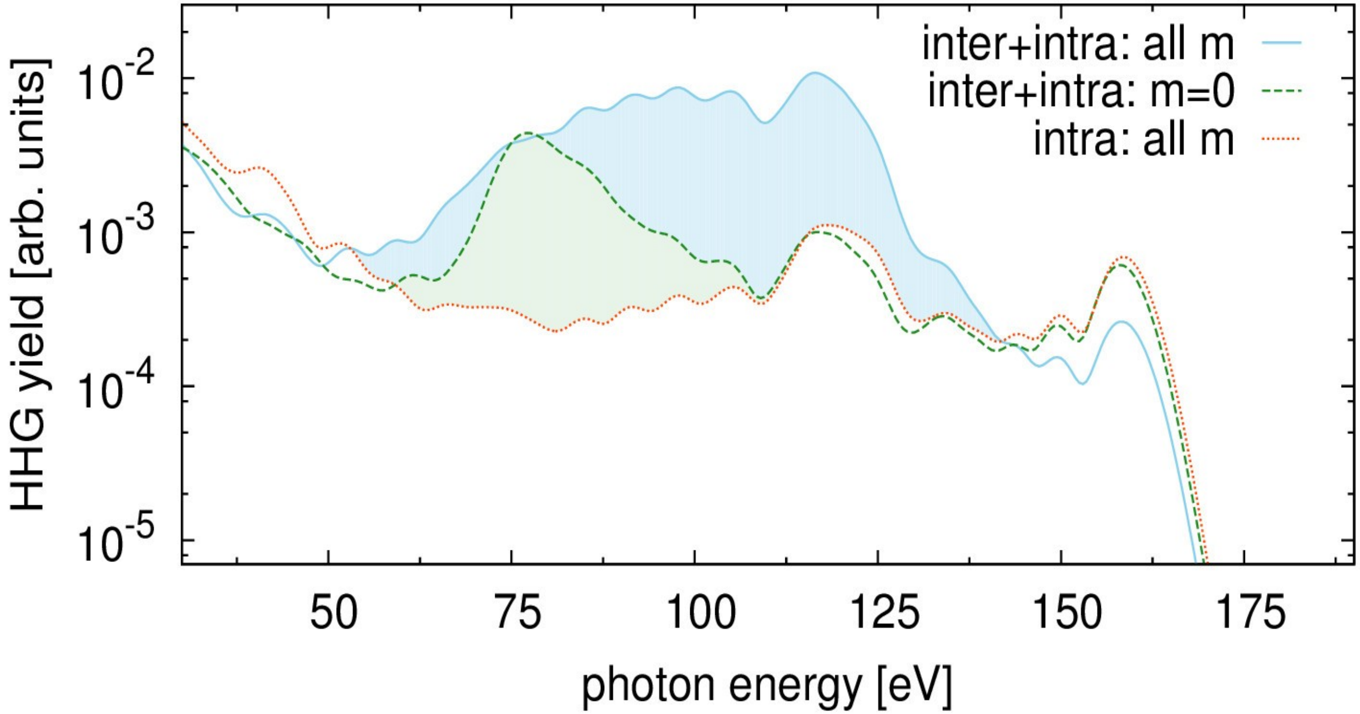
Reversible and irreversible processes



M. Sabbar *et al.*, Nature Phys. **13**, 472 (2017).

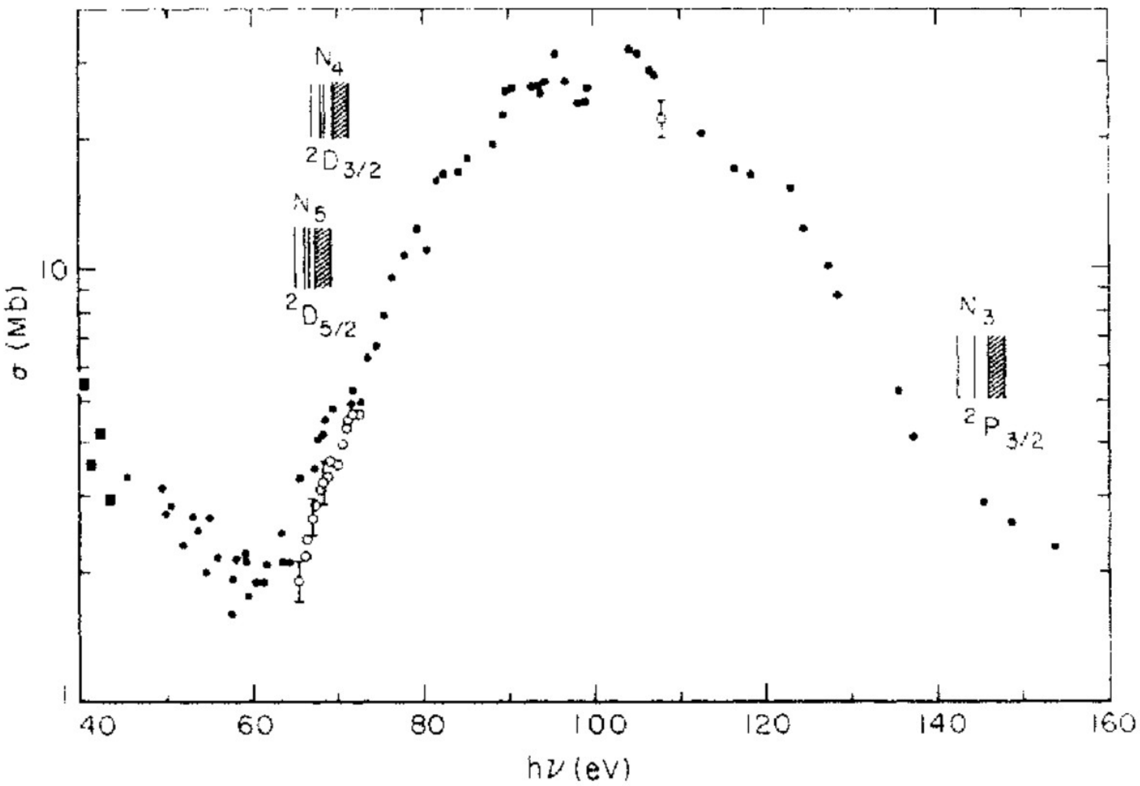
Some other applications of our methodology

- S. Pabst *et al.*, Phys. Rev. Lett. **106**, 053003 (2011).
- A. Wirth *et al.*, Science **334**, 195 (2011).
- S. Pabst and R. Santra, Phys. Rev. Lett. **111**, 233005 (2013).
- T. Mazza *et al.*, Nature Commun. **6**, 6799 (2015).



The Xe giant dipole resonance (GDR)

2014 marked the 50th anniversary of the discovery of the giant dipole resonance in the XUV photoabsorption spectrum of atomic xenon.

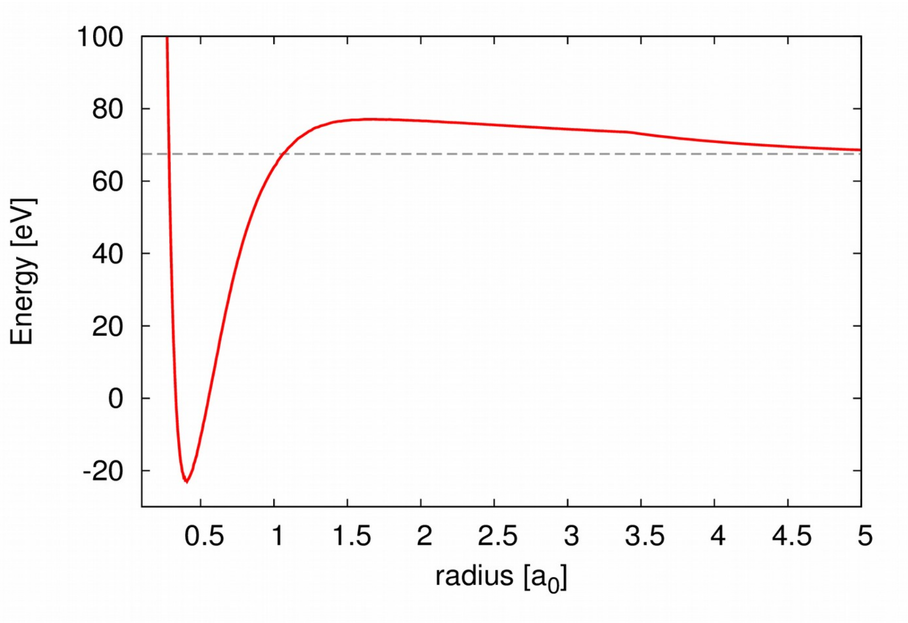


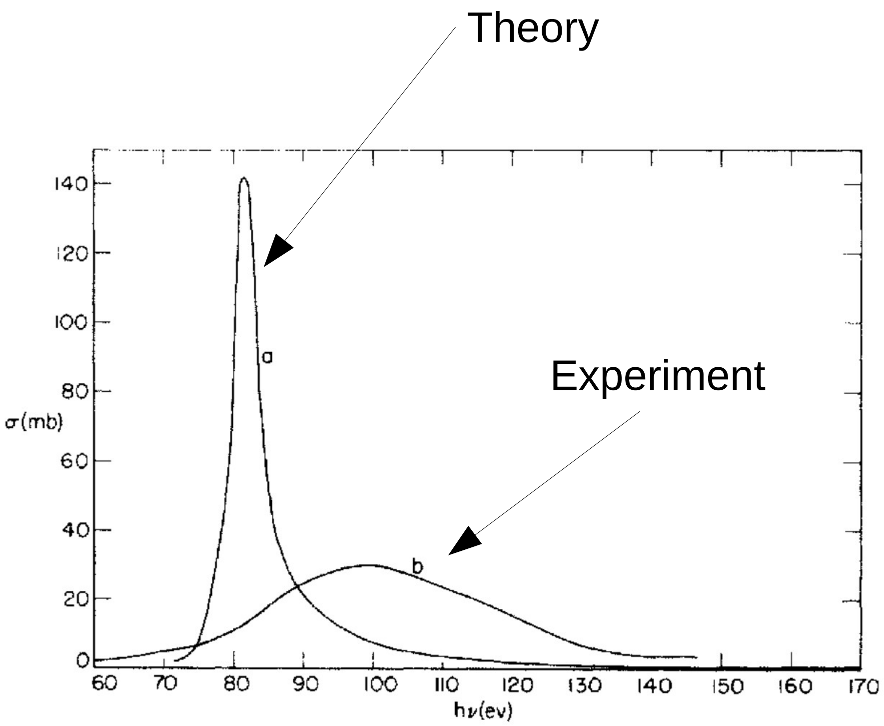
D. L. Ederer, Phys. Rev. Lett. **13**, 760 (1964).

A. P. Lukirskii, I. A. Brytov, and T. M. Zimkina, Opt. Spectrosc. **17**, 234 (1964).

The effective radial potential giving rise to the Xe GDR

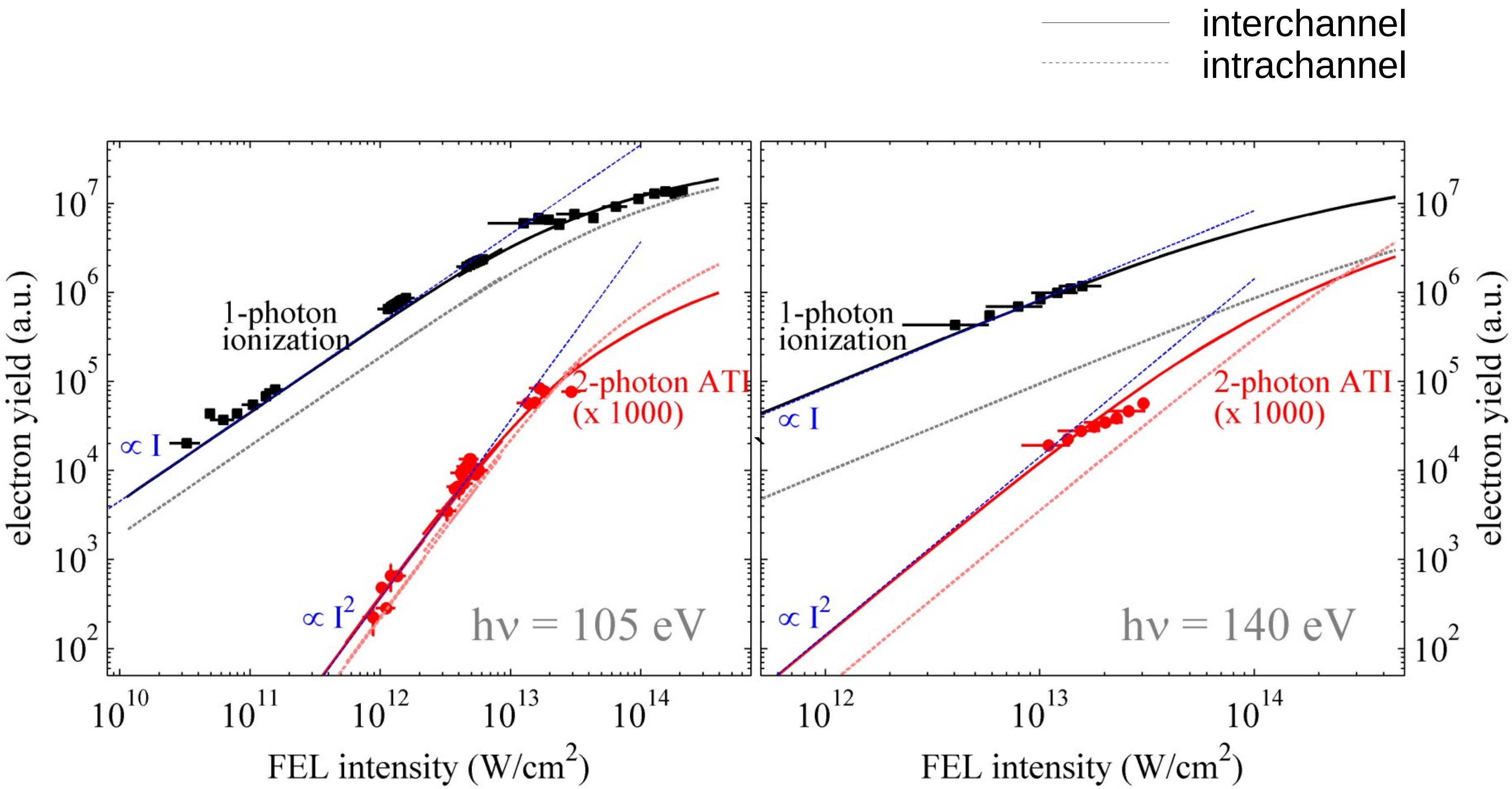
$$V(r) = V_{\mathrm{HS}}(r) + \frac{l(l+1)}{2r^2}, \quad l = 3$$





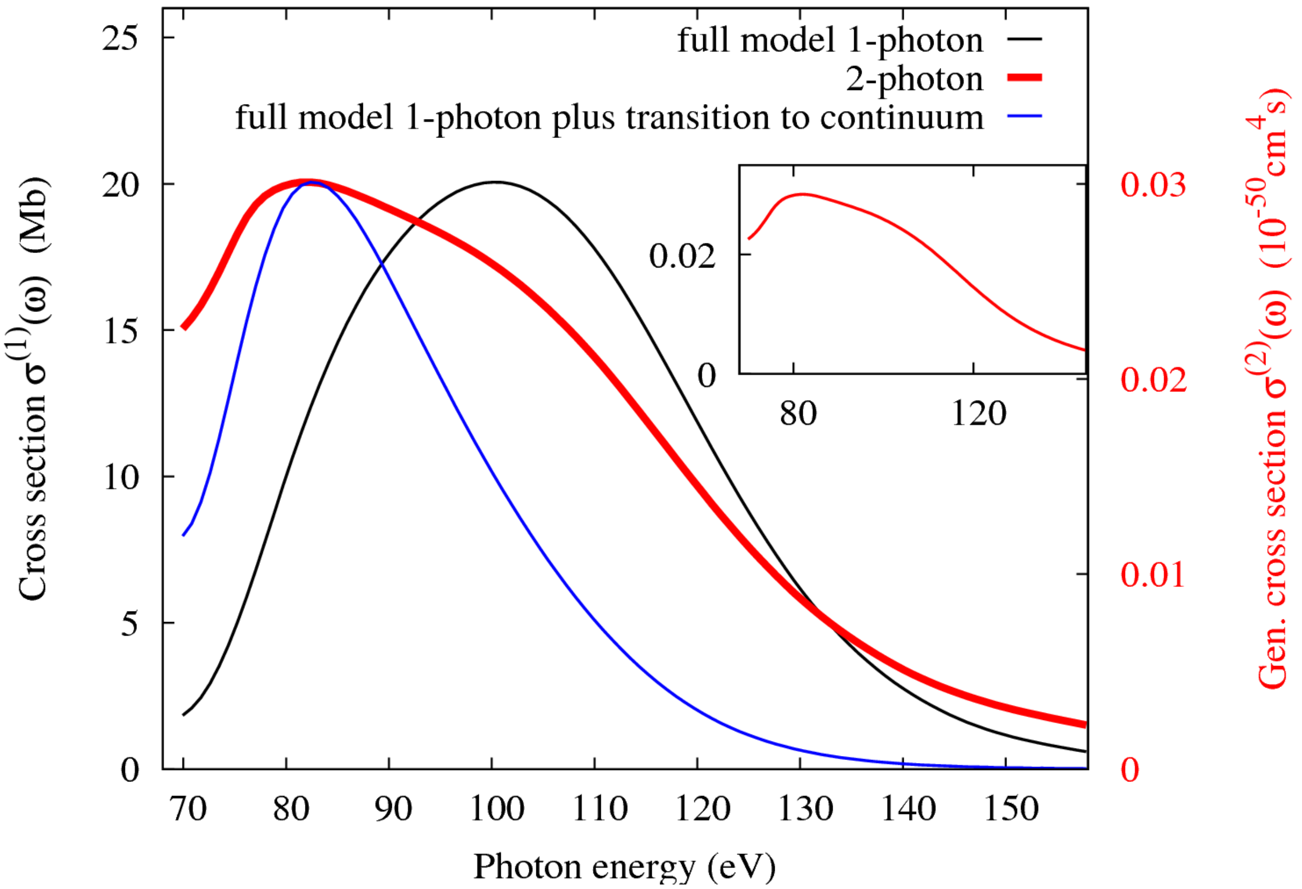
J. W. Cooper, Phys. Rev. Lett. **13**, 762 (1964).

One- and two-photon absorption at the Xe GDR



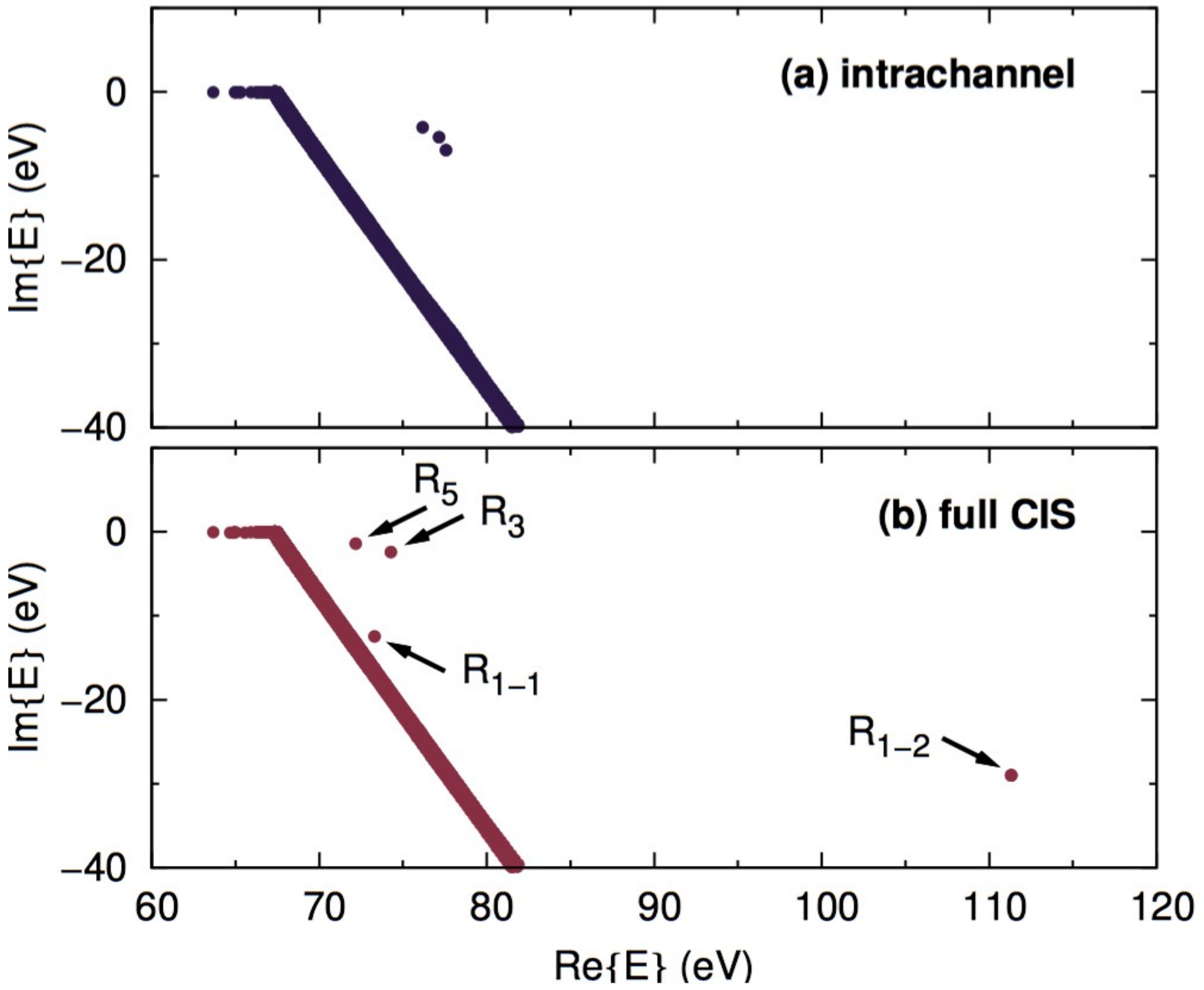
T. Mazza *et al.*, Nature Commun. **6**, 6799 (2015).

Nonlinear process (ATI) uncovers resonance substructure



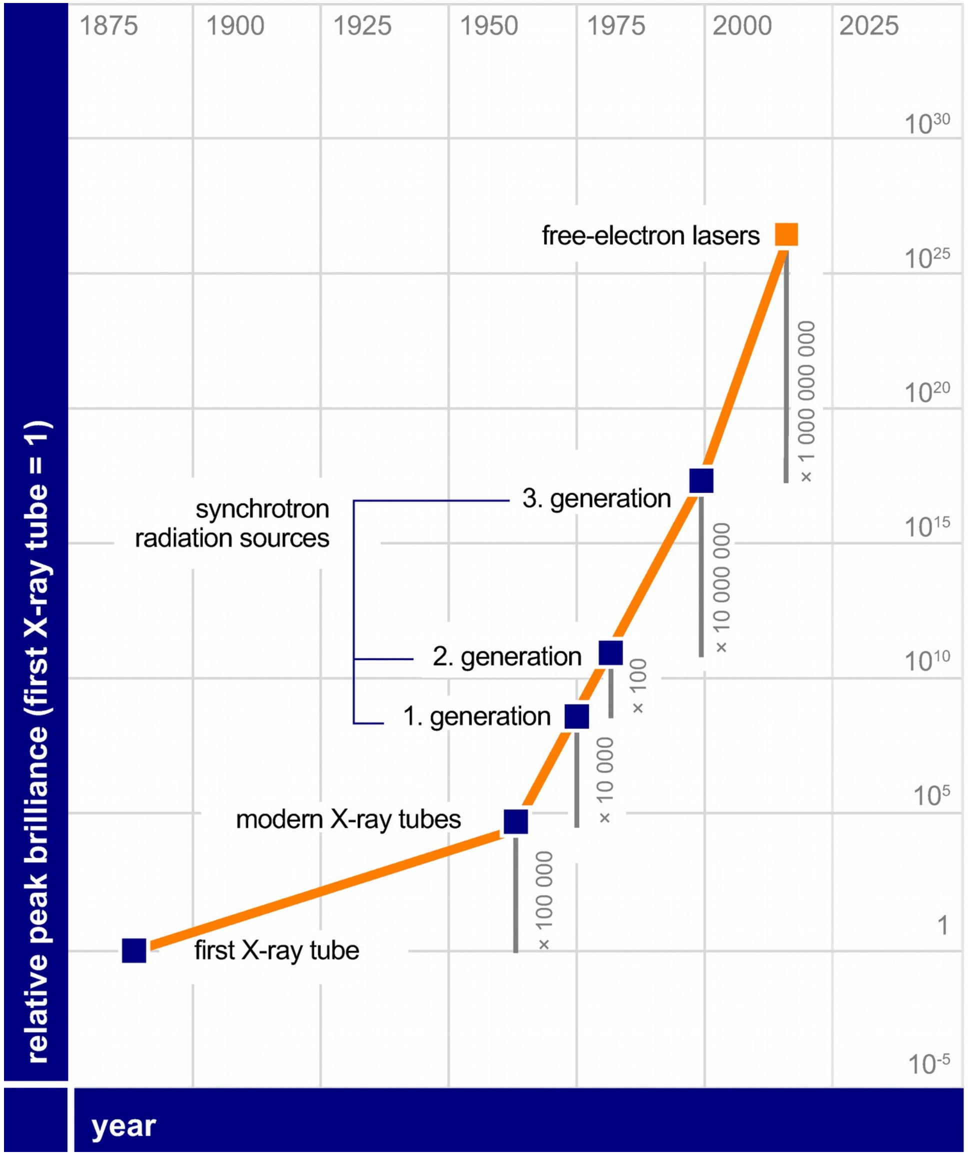
T. Mazza *et al.*, Nature Commun. **6**, 6799 (2015).

Discovery of new resonances in the 4d regime

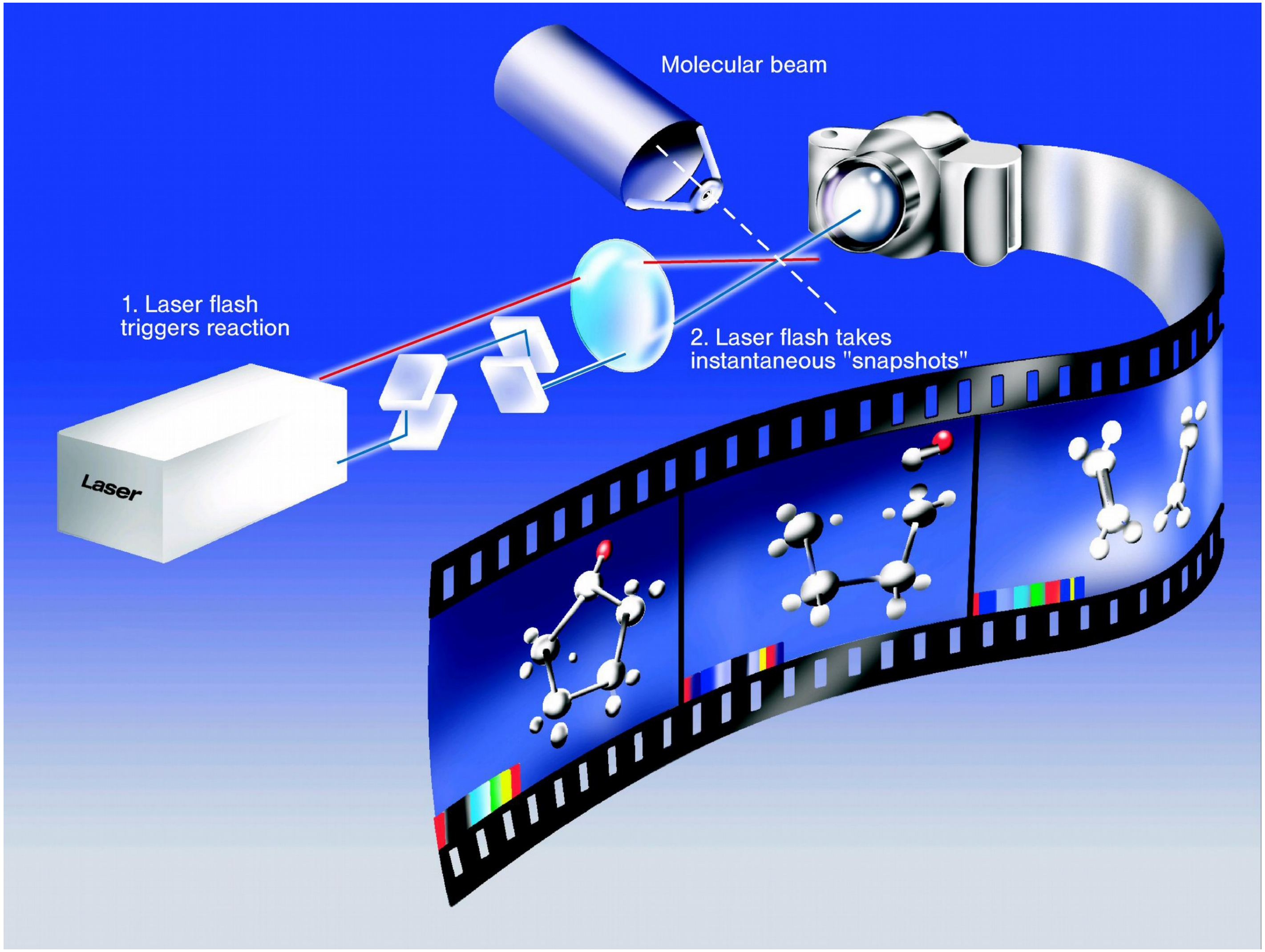


Y.-J. Chen *et al.*, J. Phys. Commun. **2**, 045024 (2018).

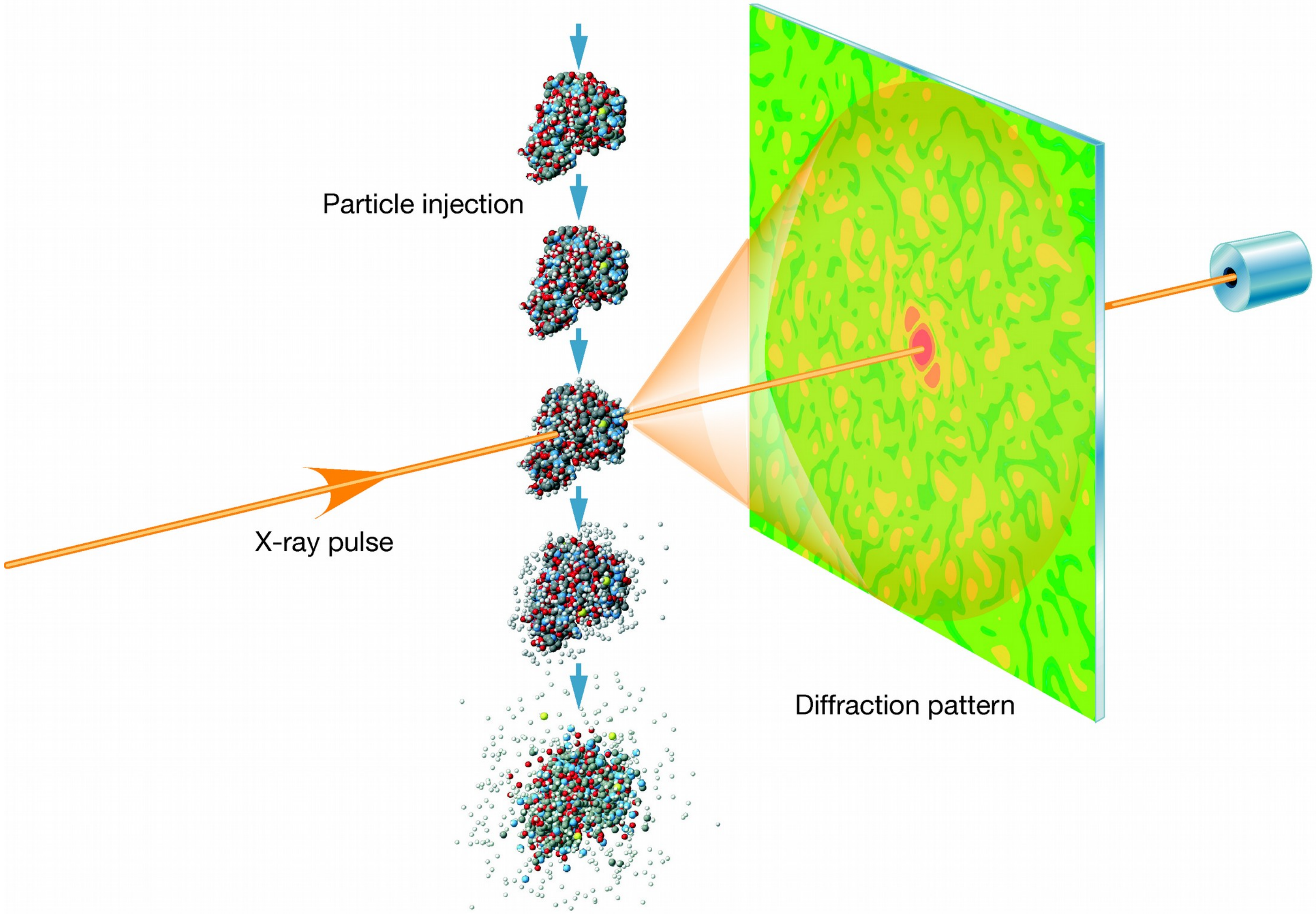
A brief history of x-ray intensity



Making molecular movies: a new tool for femtochemistry

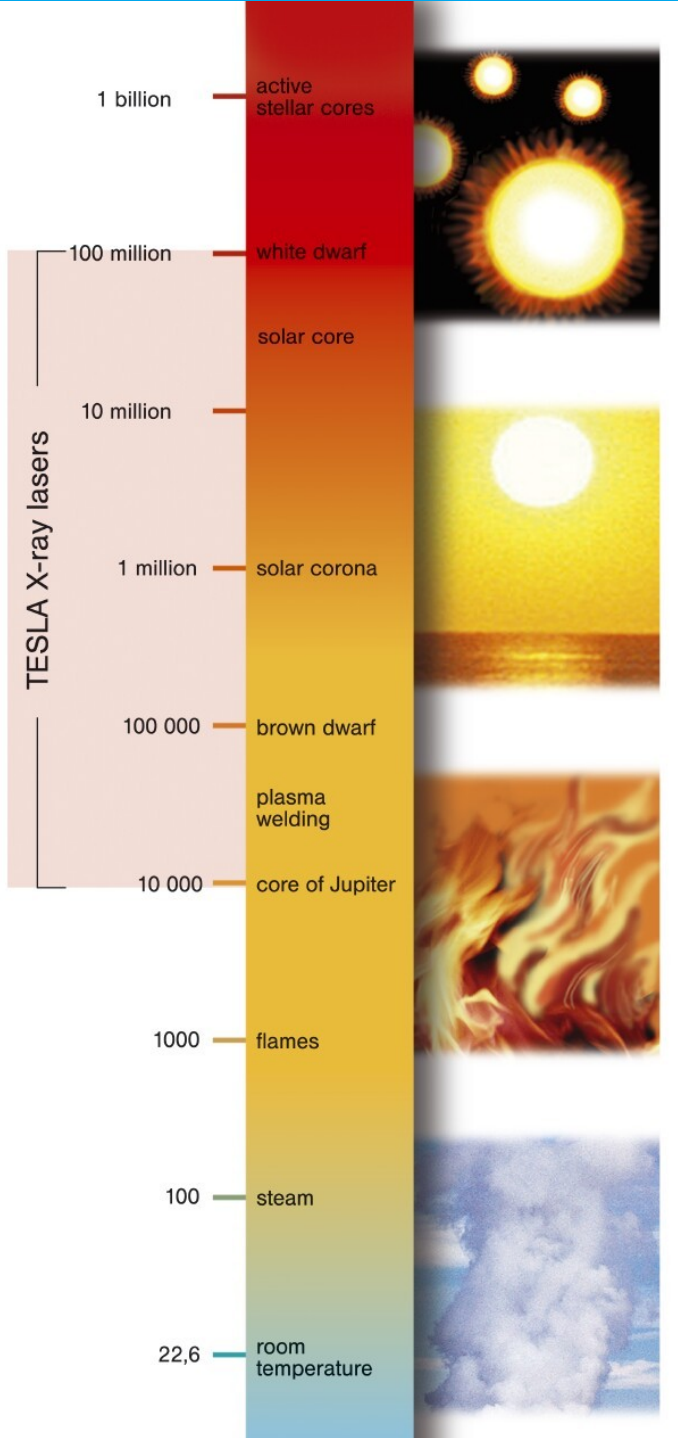


Single-shot structure determination of biomolecules



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

Generating and probing extreme states of matter



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

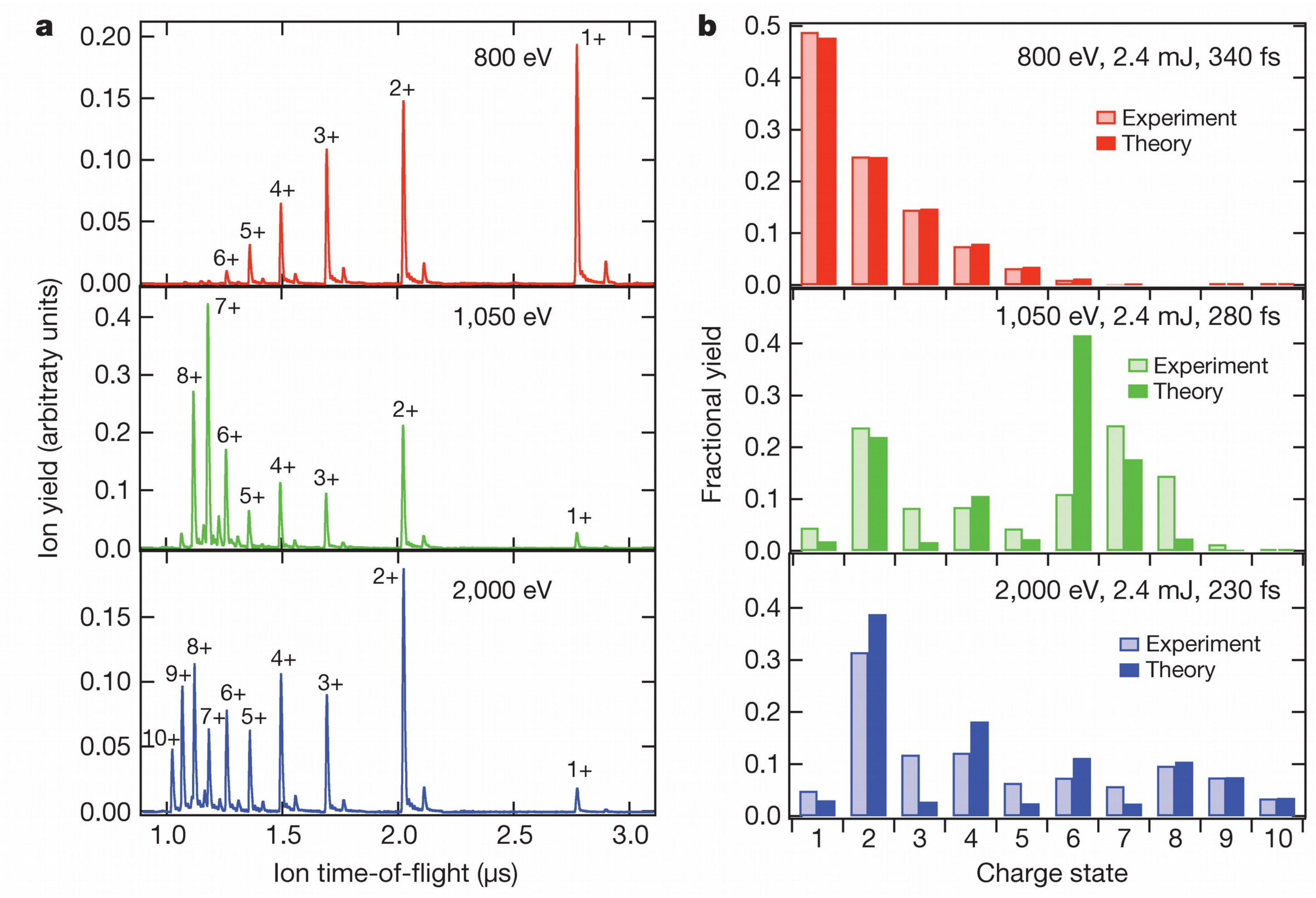
- Complete stripping of neon in a single x-ray pulse (removal of all 10 electrons)
  [L. Young *et al.*, Nature **466**, 56 (2010)]

- 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)]

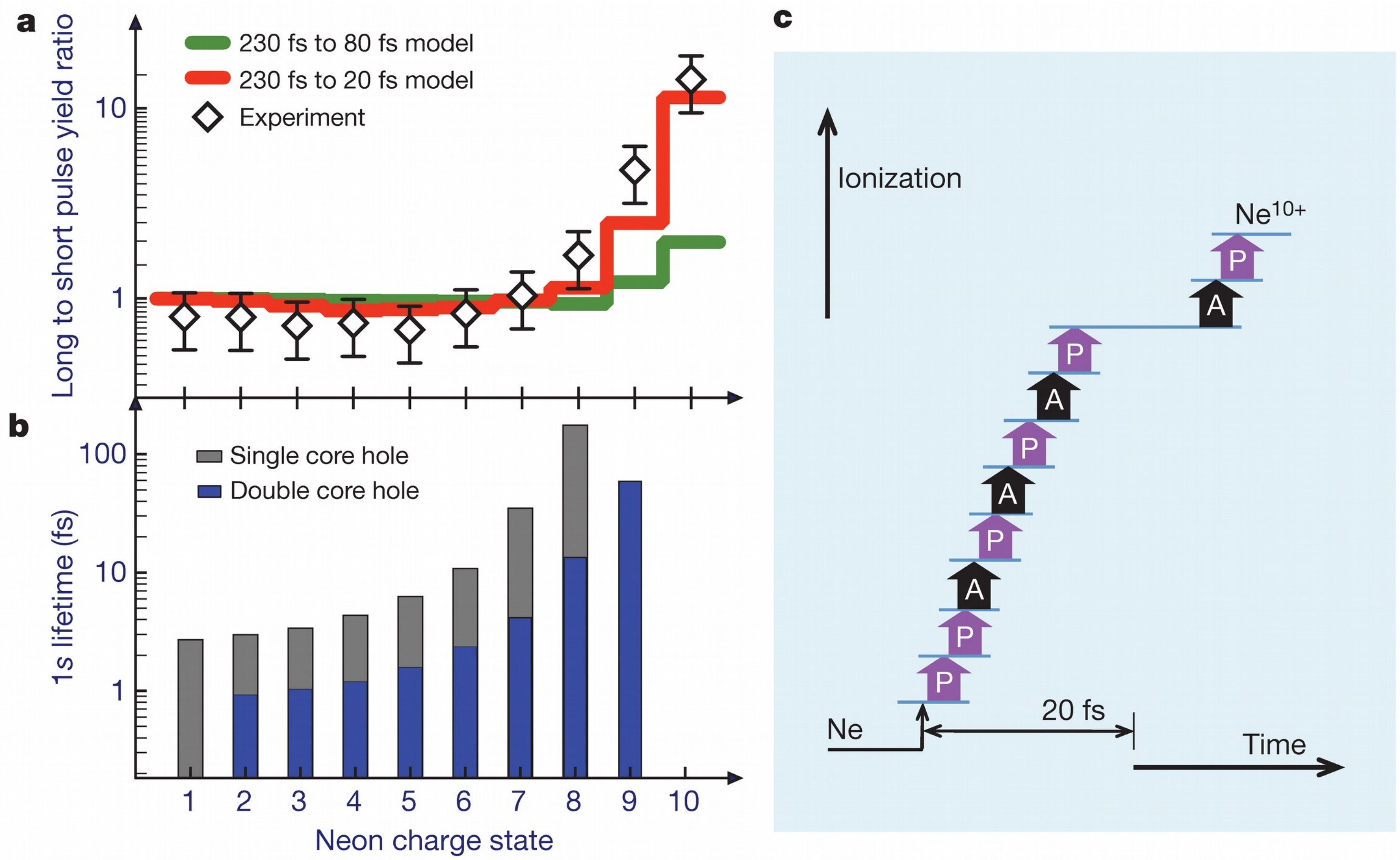
Neon charge states as a function of the photon energy



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

Counterintuitive impact of pulse duration

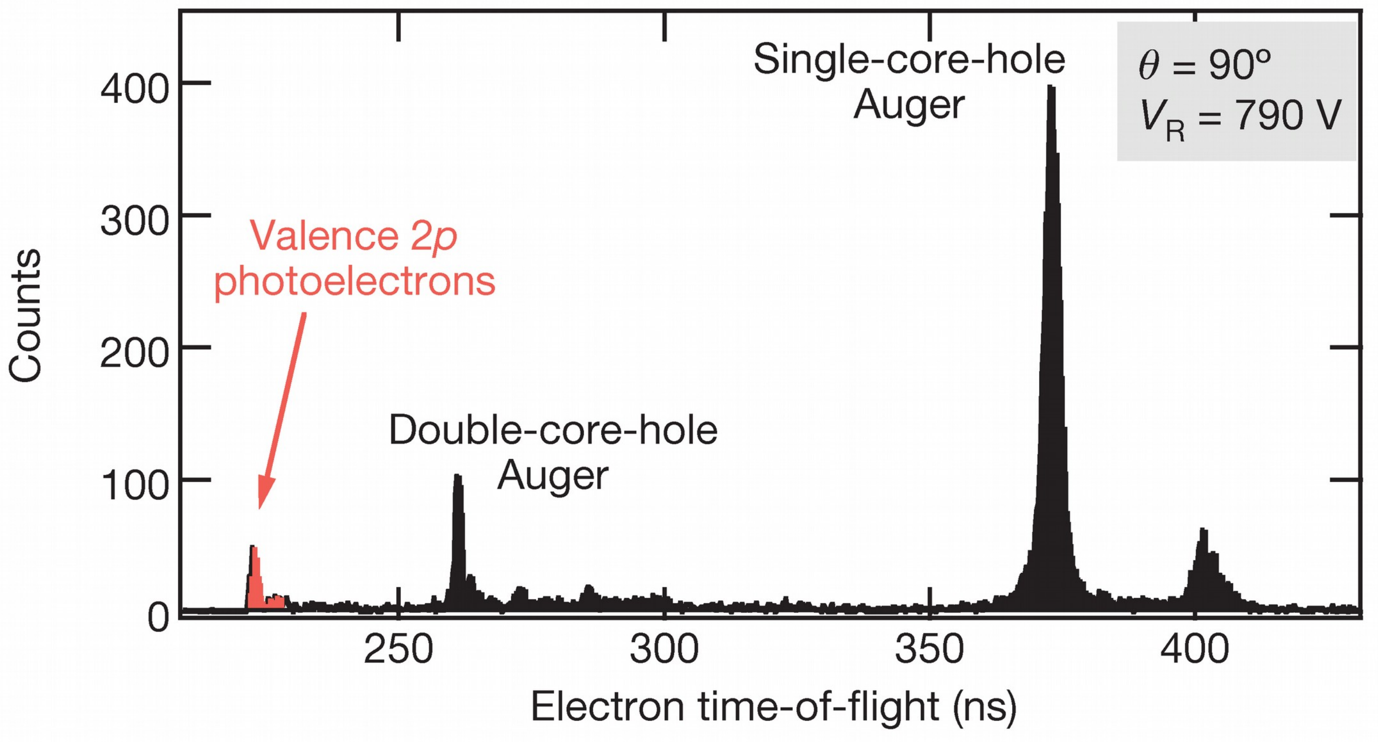
photon energy 2 keV, pulse energy 2 mJ



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

Observation of double-core-hole formation

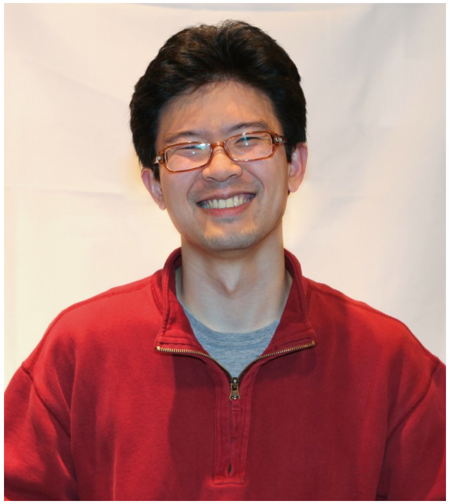
photon energy 1050 eV, pulse energy 2 mJ, nominal pulse duration 80 fs, electrons emitted perpendicular to x-ray polarization axis



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*

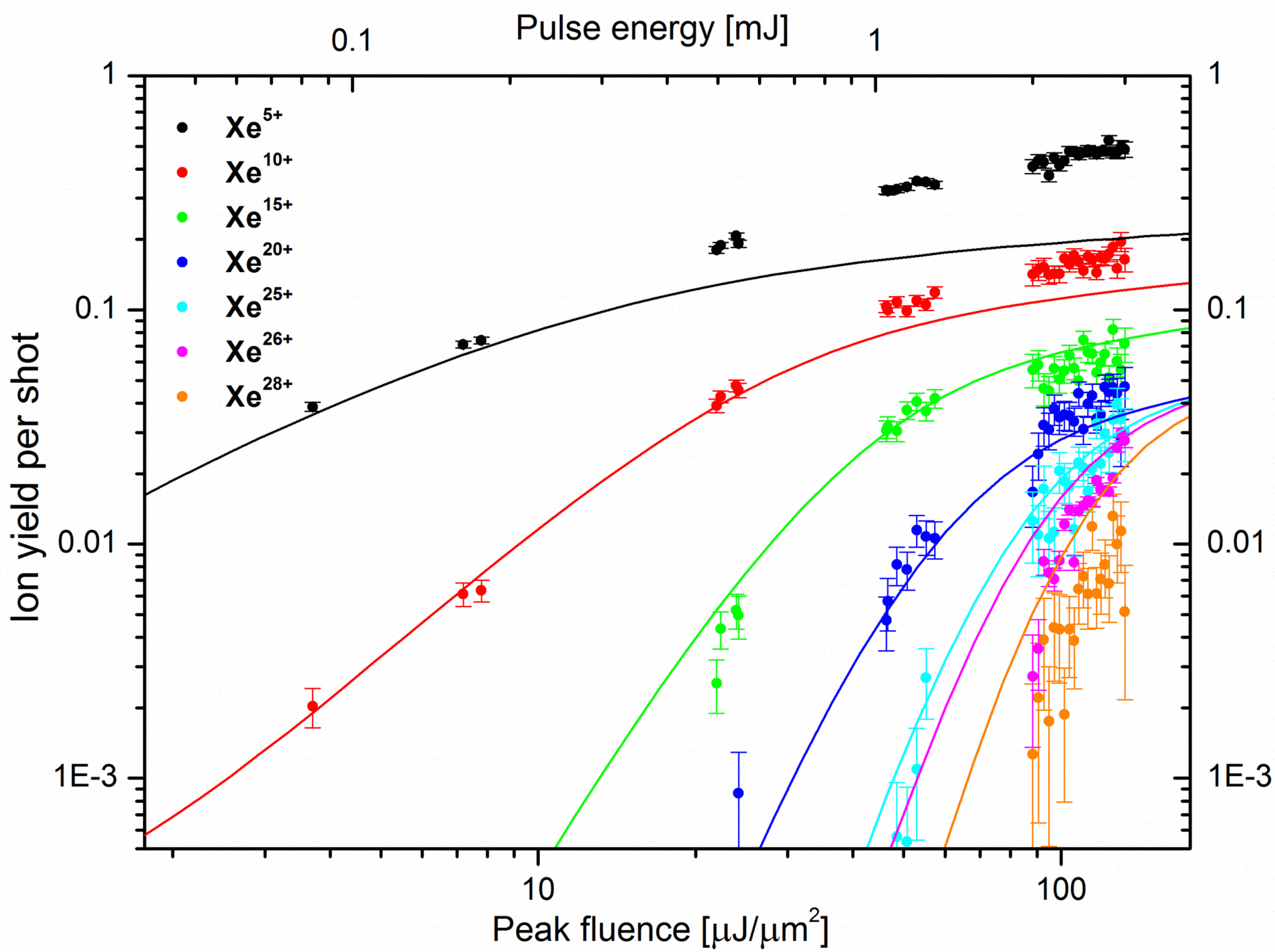
- Ne: $1s^2\ 2s^2\ 2p^6$

  → ***63*** *configurations*

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

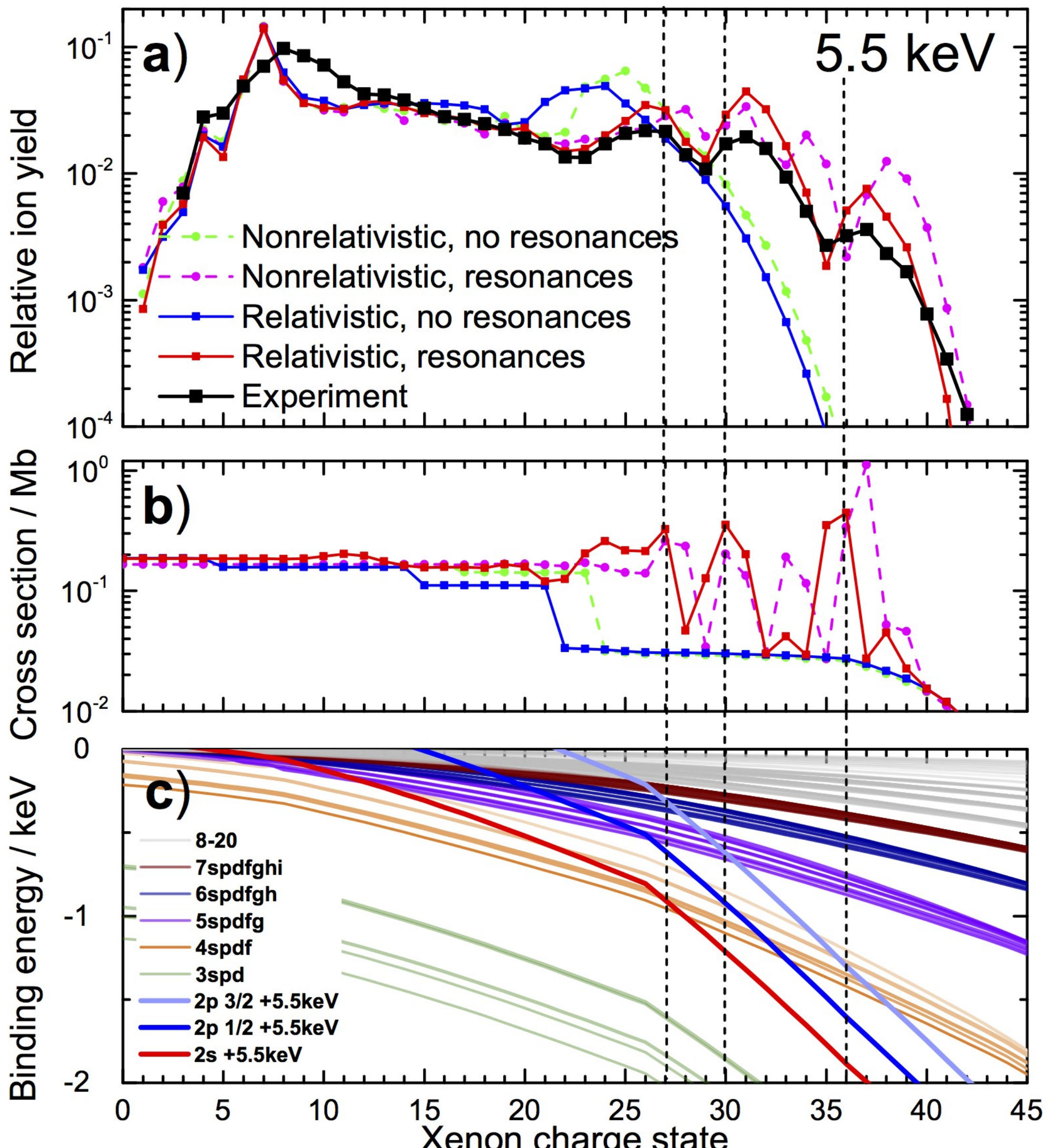
  → ***1,120,581*** *configurations*
  (excluding ionization from the K and L shells)

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)

  → ***2.6 × 10⁶⁸*** *configurations*

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

Conclusions

- Hole alignment and dynamics following optical tunnel ionization.
- Competition of reversible and irreversible processes in this regime.
- Discovery of the substructure of the Xe giant dipole resonance through XUV nonlinear spectroscopy.
- Radiation damage at high x-ray intensity of relevance for 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.