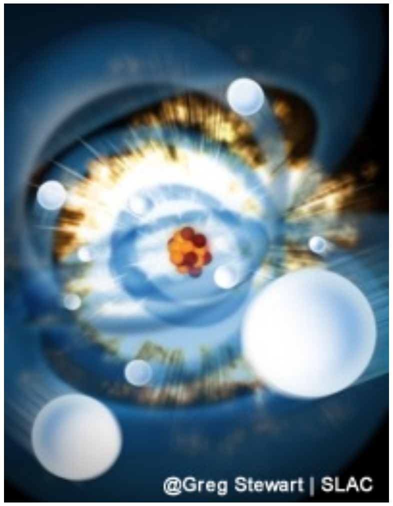
The electronic response of atoms to ultraintense x-rays

Linda Young



X-ray Science in the 21st Century
Kavli Institute for Theoretical Physics
Santa Barbara, California
2-6 August 2010

Outline

- Context & motivation

- Non-resonant high intensity x-ray phenomena
  LCLS Experiment 1: Oct 1 - 6, 2009

- Resonant high intensity x-ray processes
  LCLS Experiment 5: Oct 29 - Nov 3, 2009

- X-ray two-photon photoelectron spectroscopy
  LCLS Experiment X: Aug 5-10, 2010

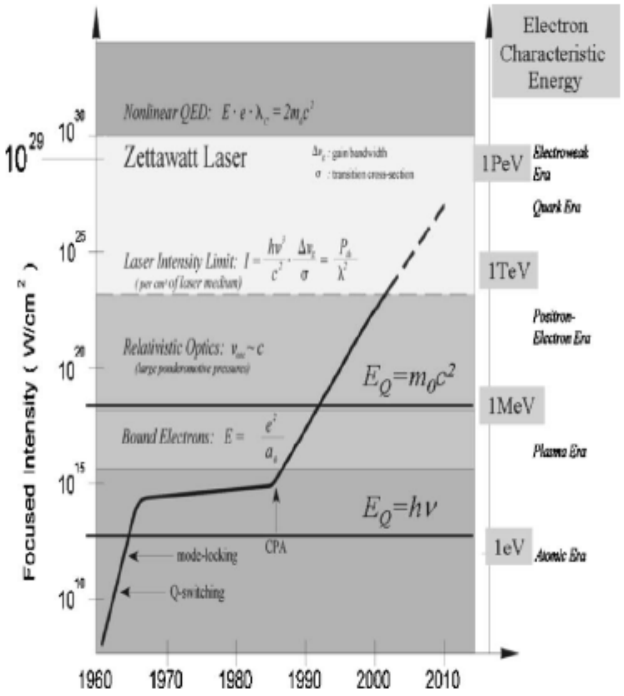
- Summary

Compare the evolution of high intensity optical and x-ray sources

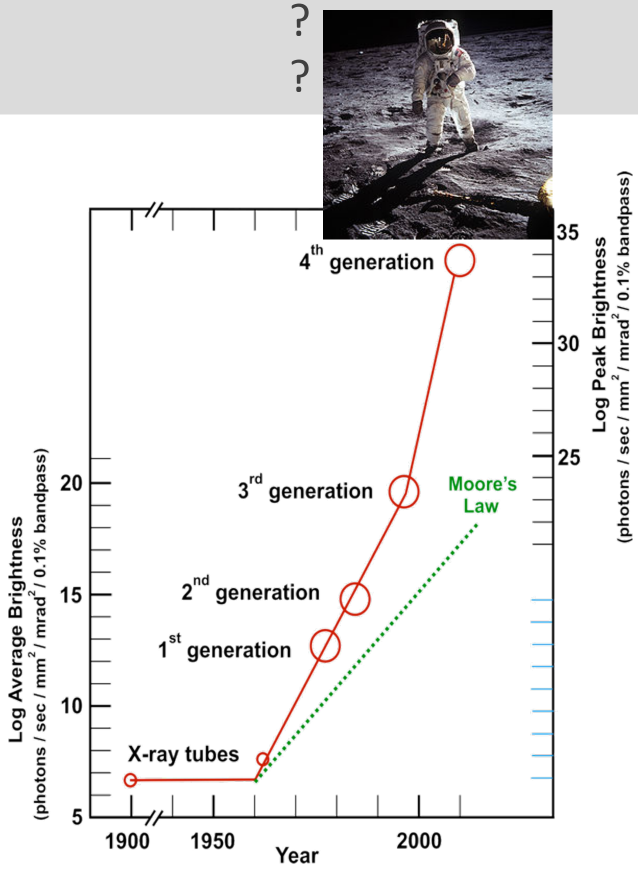
Hign-intensity at optical wavelengths
- high harmonic generation
- tabletop coherent x-ray radiation
- attosecond pulses

High-intensity at x-ray wavelengths

?
?
?



G. Mourou RMP 2006



D. Moncton, George Brown

Contrast optical and x-ray interactions at high intensity

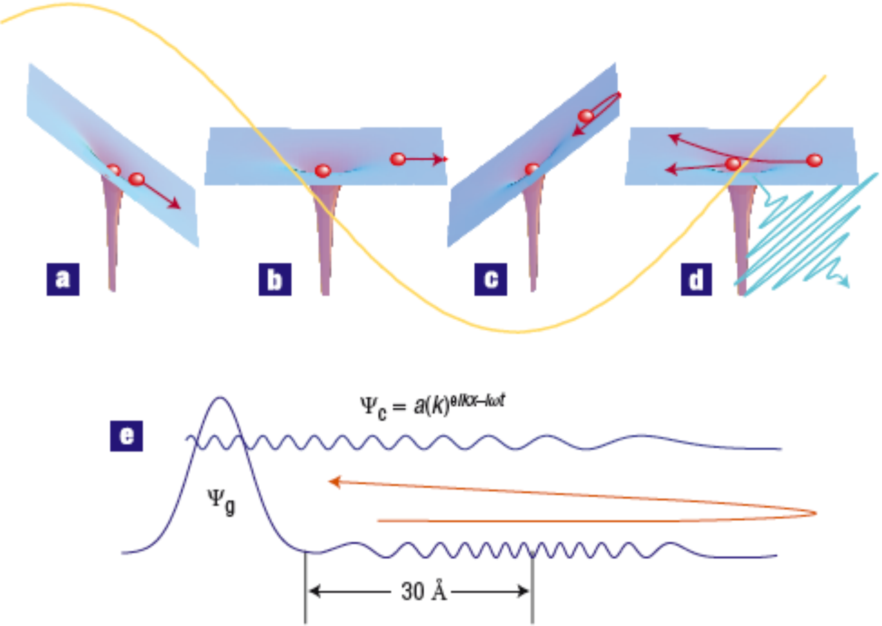
At long wavelengths - laser-driven electron dynamics is dominant

electron ponderomotive energy (au)

$$U_p = I/4\omega^2$$

displacement

$$\alpha = E/\omega^2$$



Graphic from Corkum & Krausz
Nature Physics (2007)

Ti:sapphire laser (1.55 eV) PW/cm^2
U_p ~ 60 eV & α ~ 50 au

LCLS (800 eV) 100 PW/cm^2
U_p ~ 25 meV & α ~ 0.003 au

Science Drivers for LCLS

AMO: Atomic Molecular and Optical

SXR: Soft X-ray Materials Science

XPP: X-ray Pump-Probe

XCS: X-ray Correlation Spectroscopy

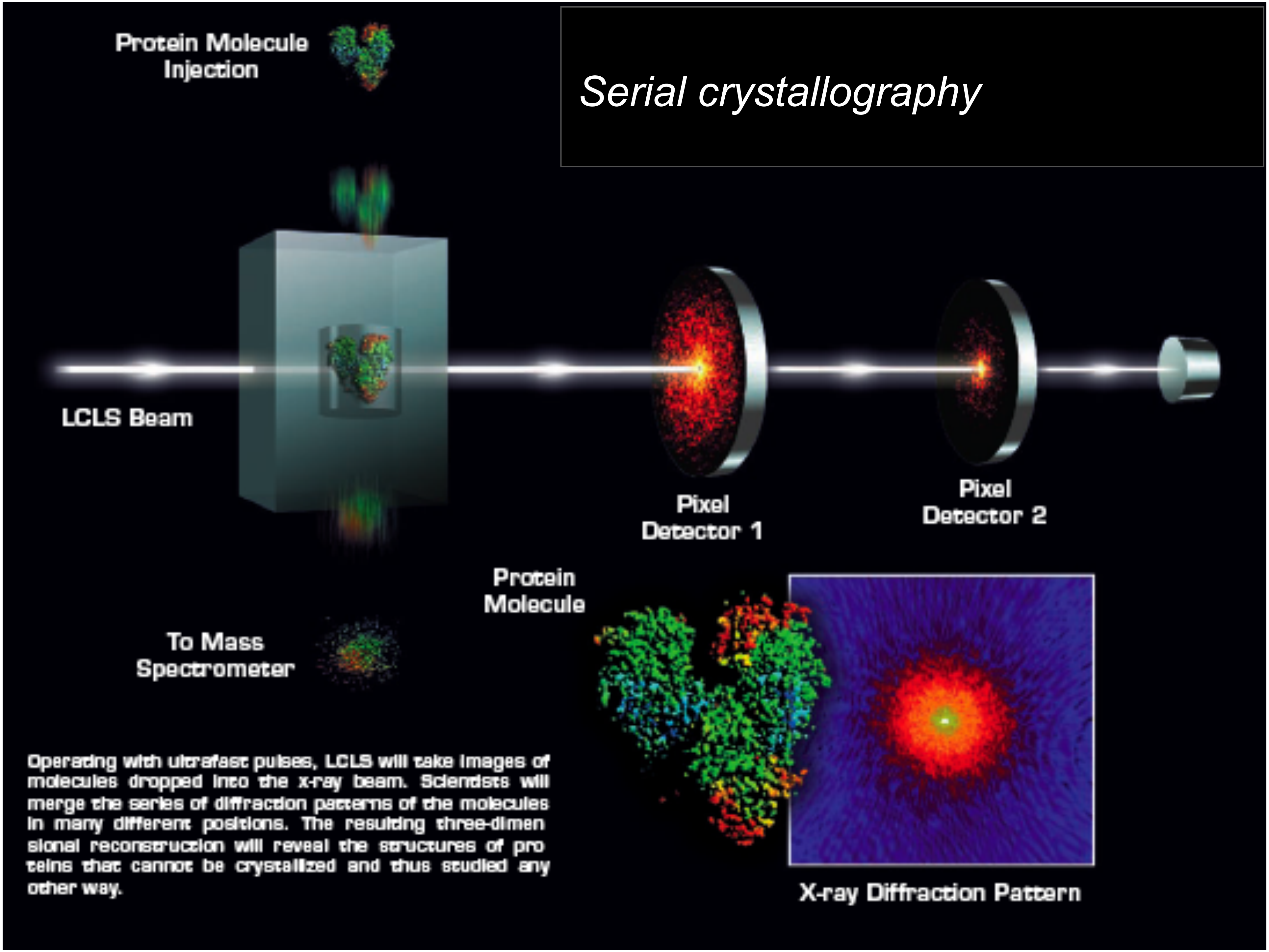
CXI: Coherent X-ray Imaging

MEC: Materials in Extreme Conditions

AMO

- Understand and control x-ray atom/molecule interactions at ultrahigh x-ray intensity as a foundation for other applications.
- Provide diagnostics of the LCLS radiation

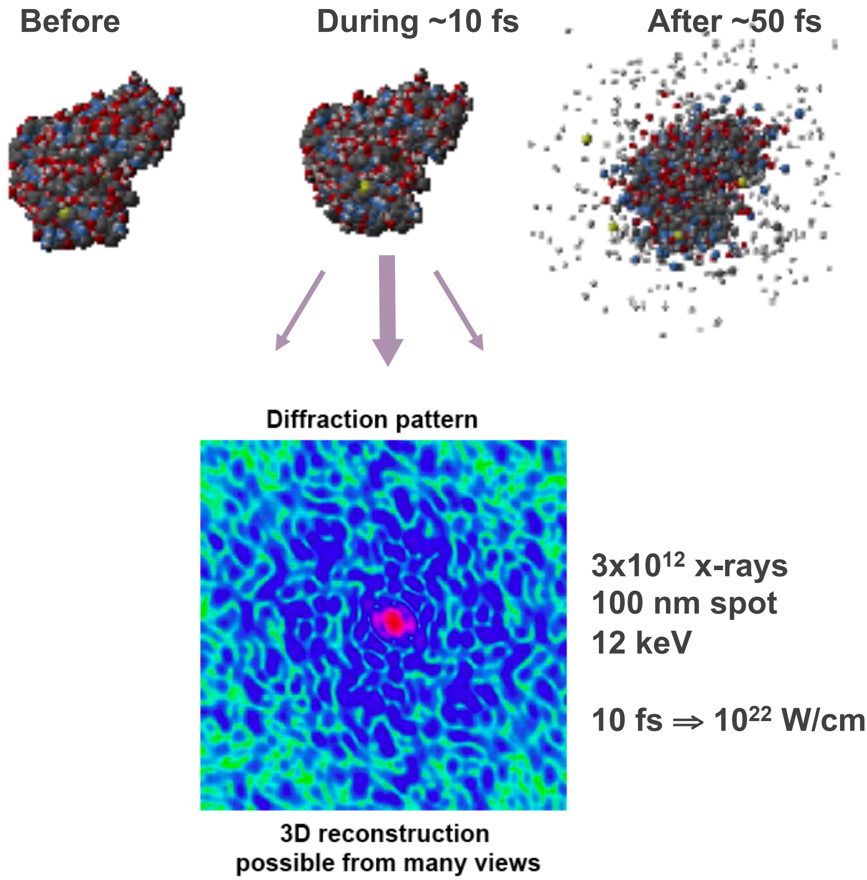
Serial crystallography



Operating with ultrafast pulses, LCLS will take images of molecules dropped into the x-ray beam. Scientists will merge the series of diffraction patterns of the molecules in many different positions. The resulting three-dimensional reconstruction will reveal the structures of proteins that cannot be crystallized and thus studied any other way.

AMO questions at the ultraintense x-ray frontier

- fundamental nature of x-ray damage at high intensity
  - -Coulomb explosion
  - -electronic damage
  - -behavior at 10^{22} W/cm^2 - 1Å
- nonlinear x-ray processes
  - role of coherence
- quantum control of inner-shell processes



3×10^{12} x-rays
100 nm spot
12 keV

10 fs ⇒ 10^{22} W/cm^2

Neutze, Wouts, van der Spoel, Weckert, Hajdu Nature 406, 752 (2000)

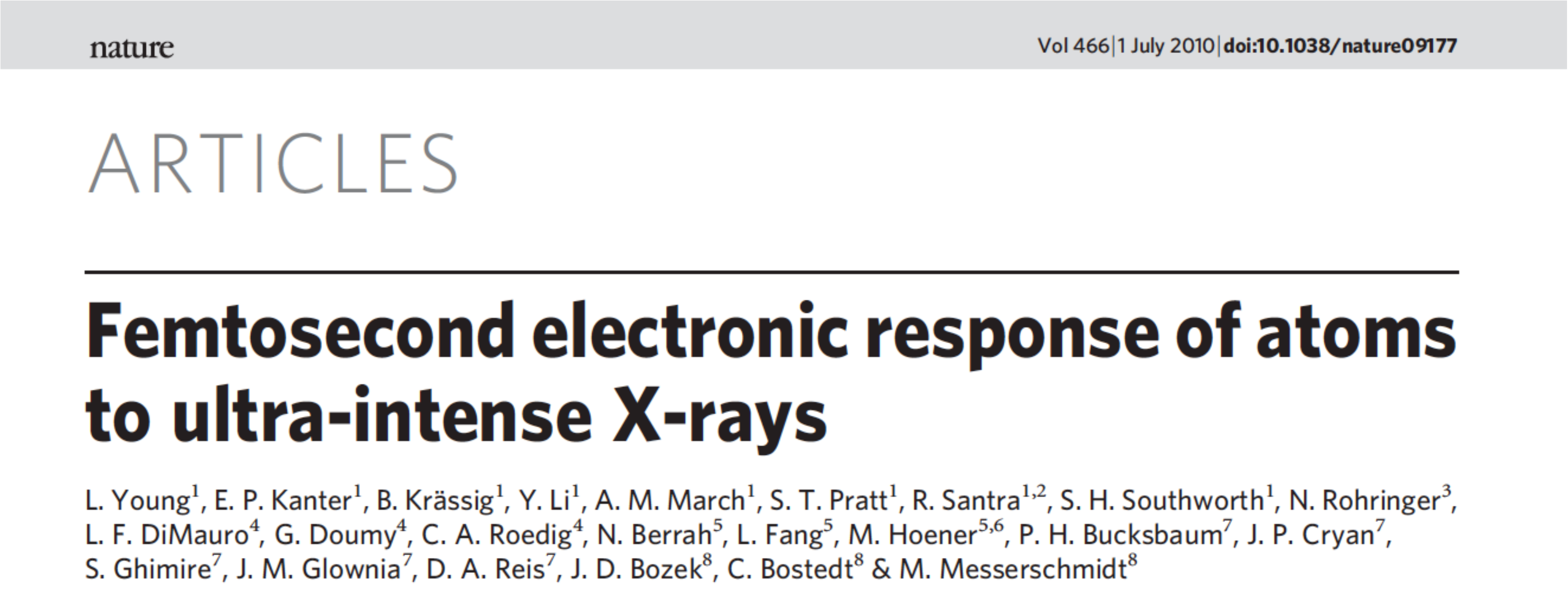
LCLS Experiment 1 - Oct 1, 2009

Nature of the electronic response to

10^5 x-rays/Å^2
80 - 340 fs
800 - 2000 eV

~10^{18} W/cm^2

Original single molecule imaging parameters, Neutze et al. Nature (2000)
3×10^{12} x-rays/(100 nm)^2 = 3×10^6 x-rays/Å^2
10 fs
~10^{22} W/cm^2

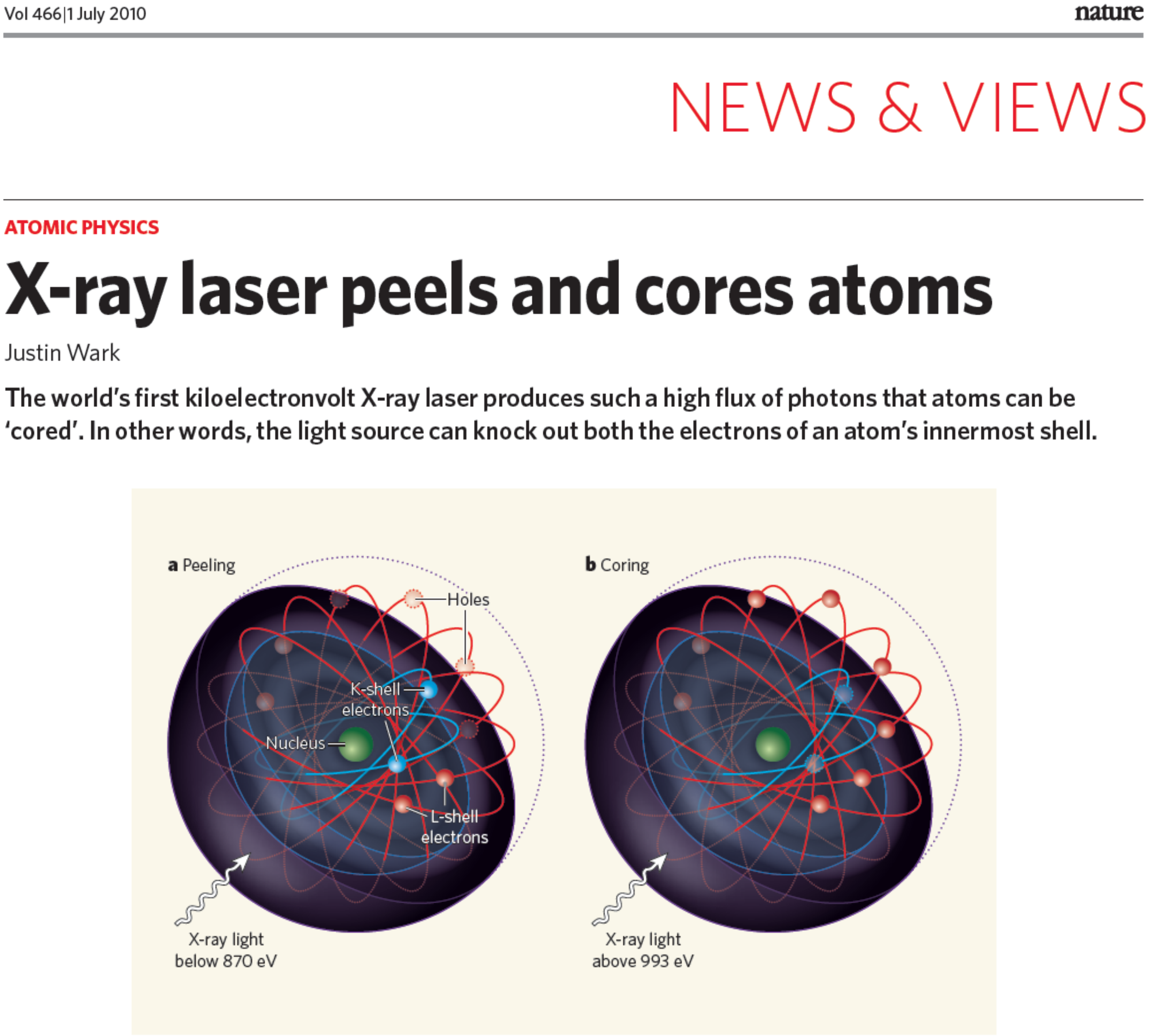


nature Vol 466|1 July 2010|**doi:10.1038/nature09177**

ARTICLES

Femtosecond electronic response of atoms to ultra-intense X-rays

L. Young[1], E. P. Kanter[1], B. Krässig[1], Y. Li[1], A. M. March[1], S. T. Pratt[1], R. Santra[1,2], S. H. Southworth[1], N. Rohringer[3], L. F. DiMauro[4], G. Doumy[4], C. A. Roedig[4], N. Berrah[5], L. Fang[5], M. Hoener[5,6], P. H. Bucksbaum[7], J. P. Cryan[7], S. Ghimire[7], J. M. Glownia[7], D. A. Reis[7], J. D. Bozek[8], C. Bostedt[8] & M. Messerschmidt[8]



Vol 466|1 July 2010 **nature**

NEWS & VIEWS

ATOMIC PHYSICS

X-ray laser peels and cores atoms

Justin Wark

The world's first kiloelectronvolt X-ray laser produces such a high flux of photons that atoms can be 'cored'. In other words, the light source can knock out both the electrons of an atom's innermost shell.

Our approach to understanding ultraintense x-ray interactions

- Start with a well-characterized target

Binding energies in neutral neon

2p : ~21 eV

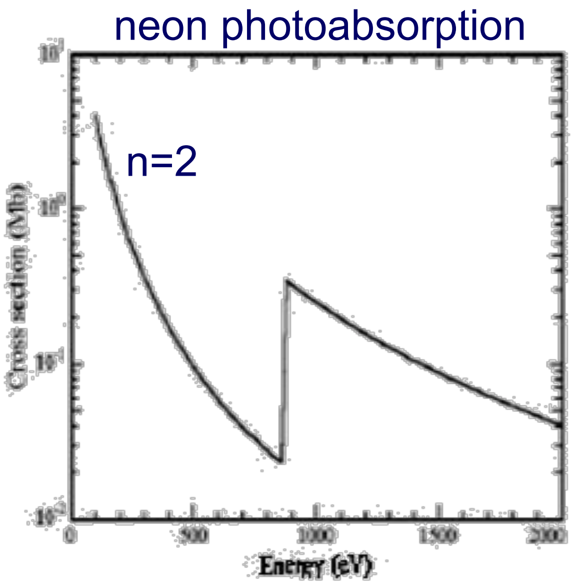
2s : ~48 eV

1s : ~870 eV

Inner-shell excitation

Auger yield 98%

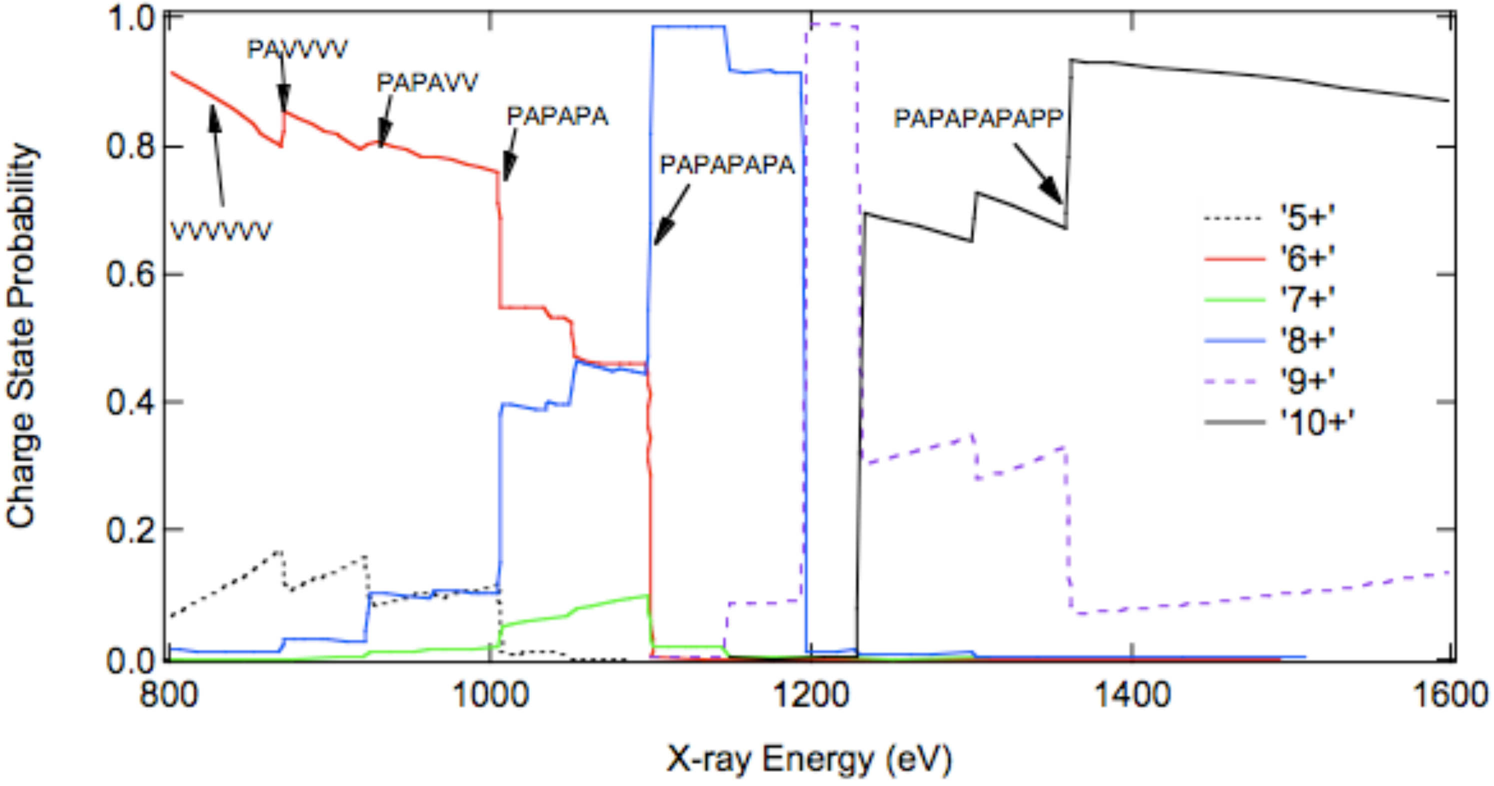
Auger clock - τ_{1s}: 2.4 fs



- Probe changes in interaction from outer- to inner-shell between 800-2000 eV

Guided by theory

Theory: Rohringer & Santra, PRA 76, 033416 (2007)



LCLS specs

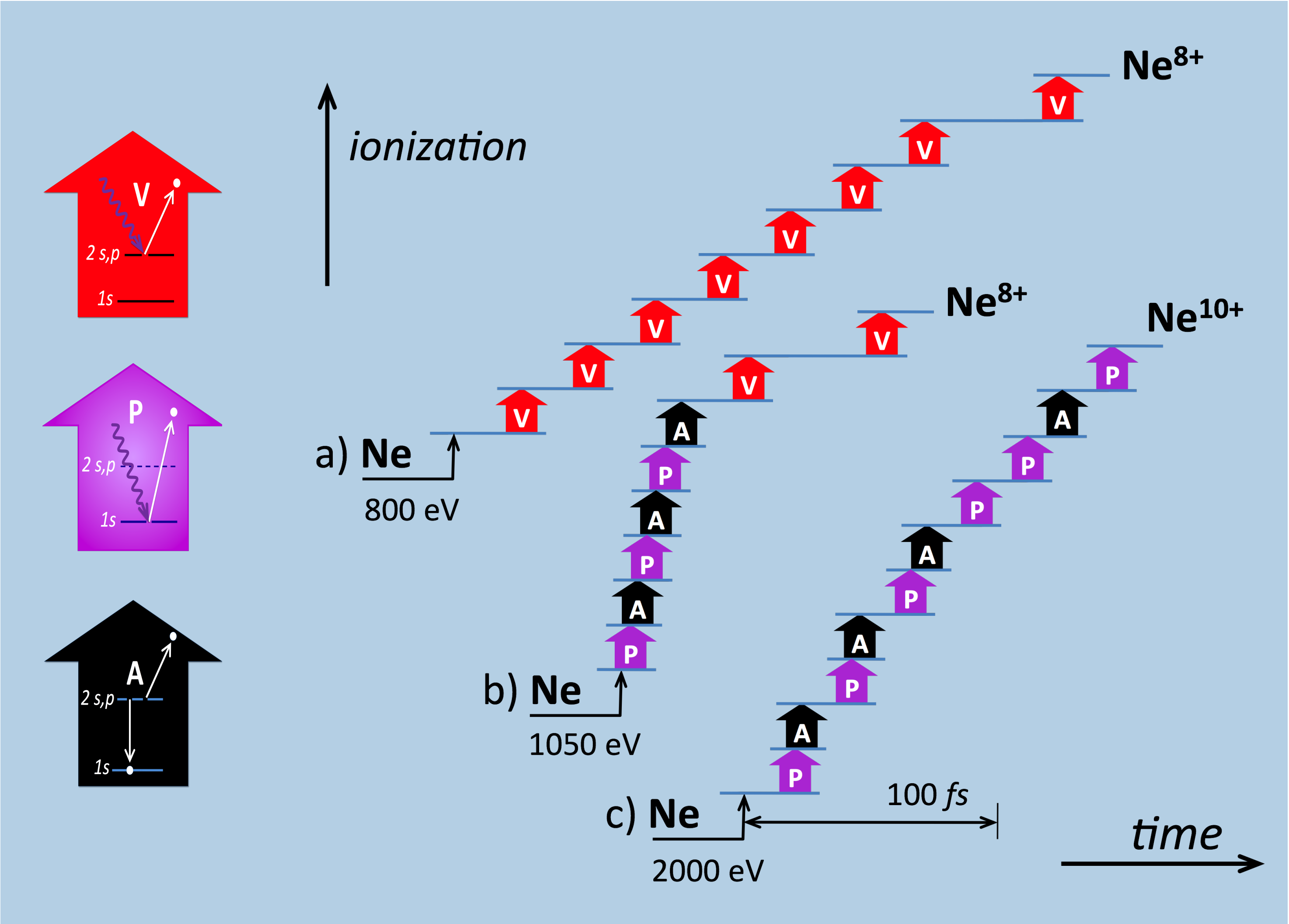
10^{13} x-rays

230 fs

1 µm spot

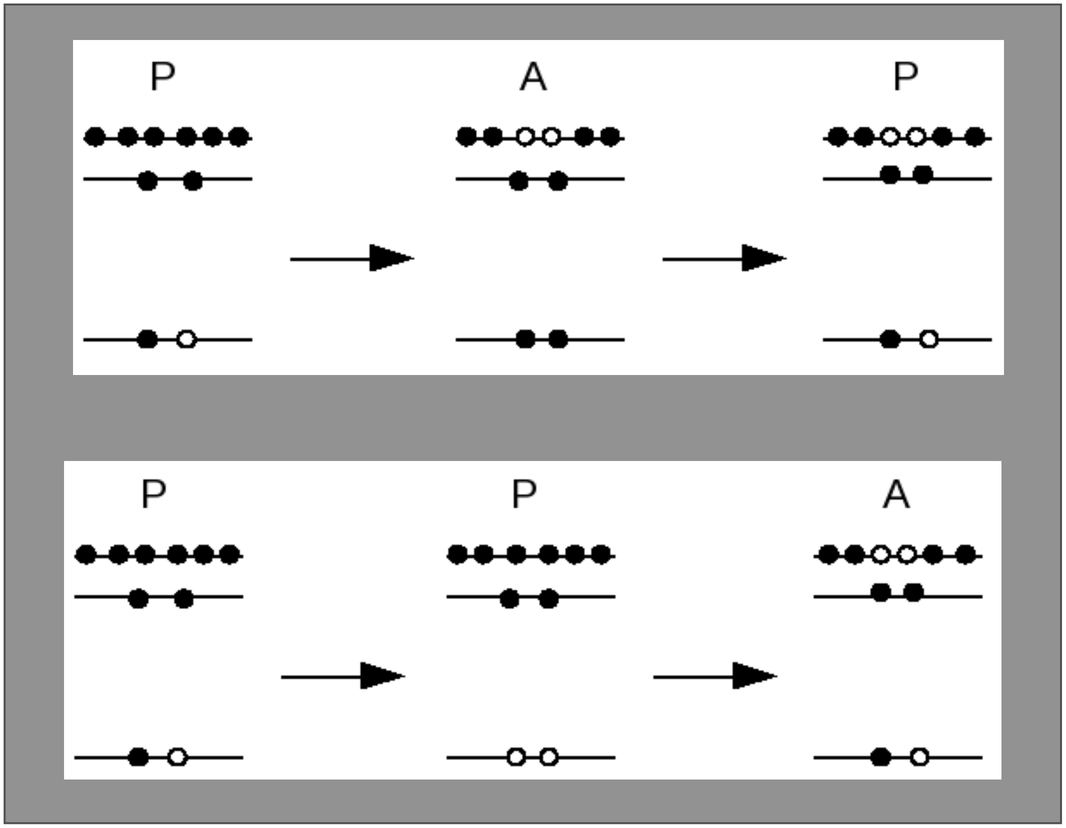
Three target energies: 800 eV, 1050 eV, 2000 eV

Valence ionization, core ionization and Auger decay



Sequential single photon processes dominate the interaction

How does one arrive at a particular charge state?

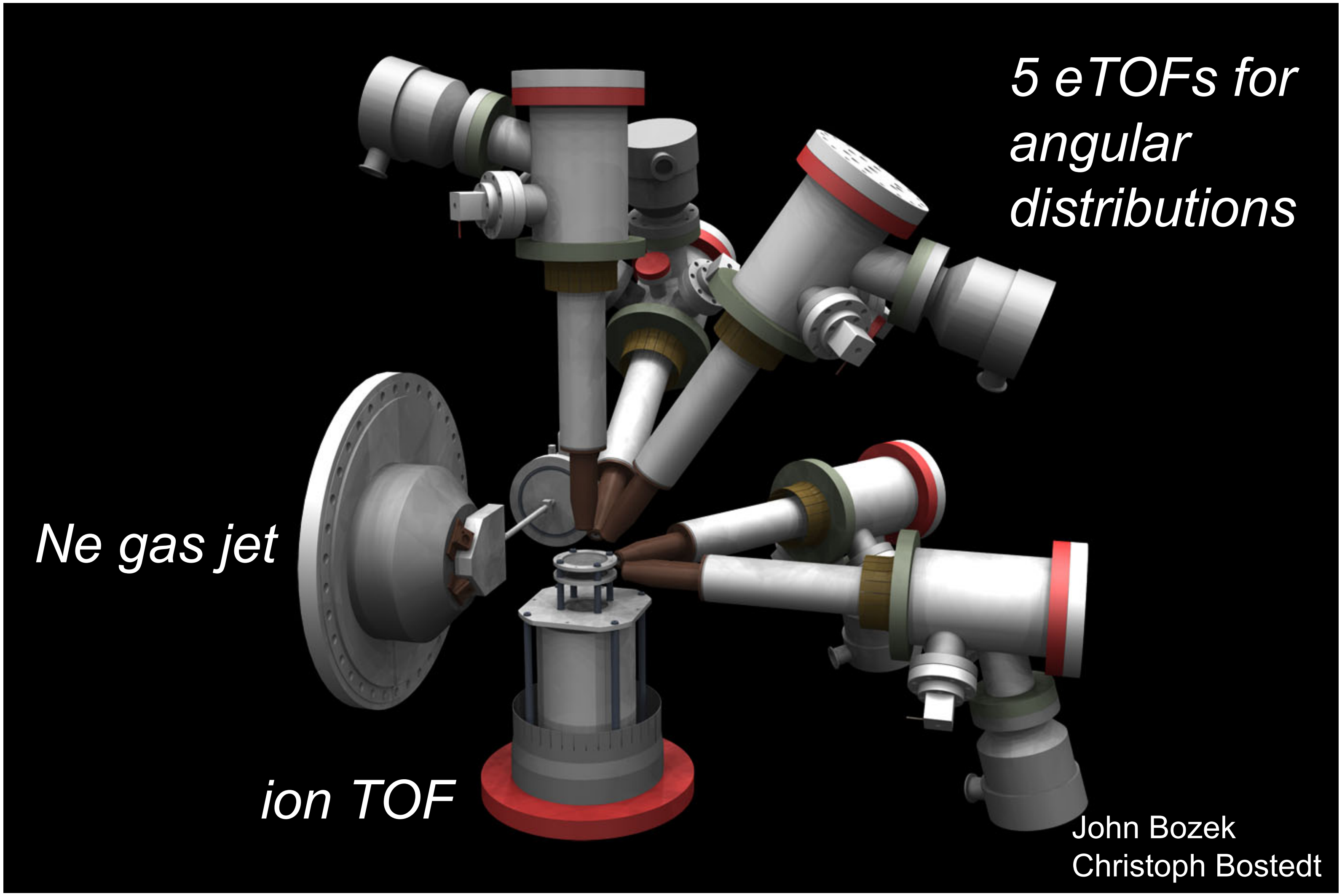


Low Intensity *PAP*

High Intensity *PPA*

- Hollow atoms produced at high x-ray intensity
- Electron spectroscopy can define the mechanism

High field physics chamber

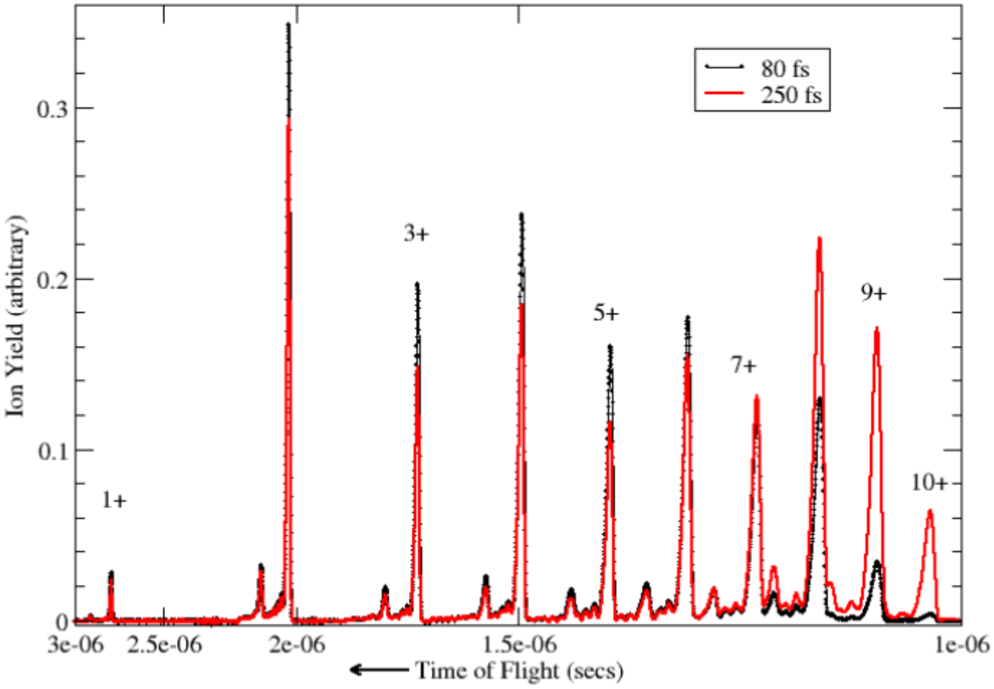


John Bozek
Christoph Bostedt

Day 1 – two interesting observations

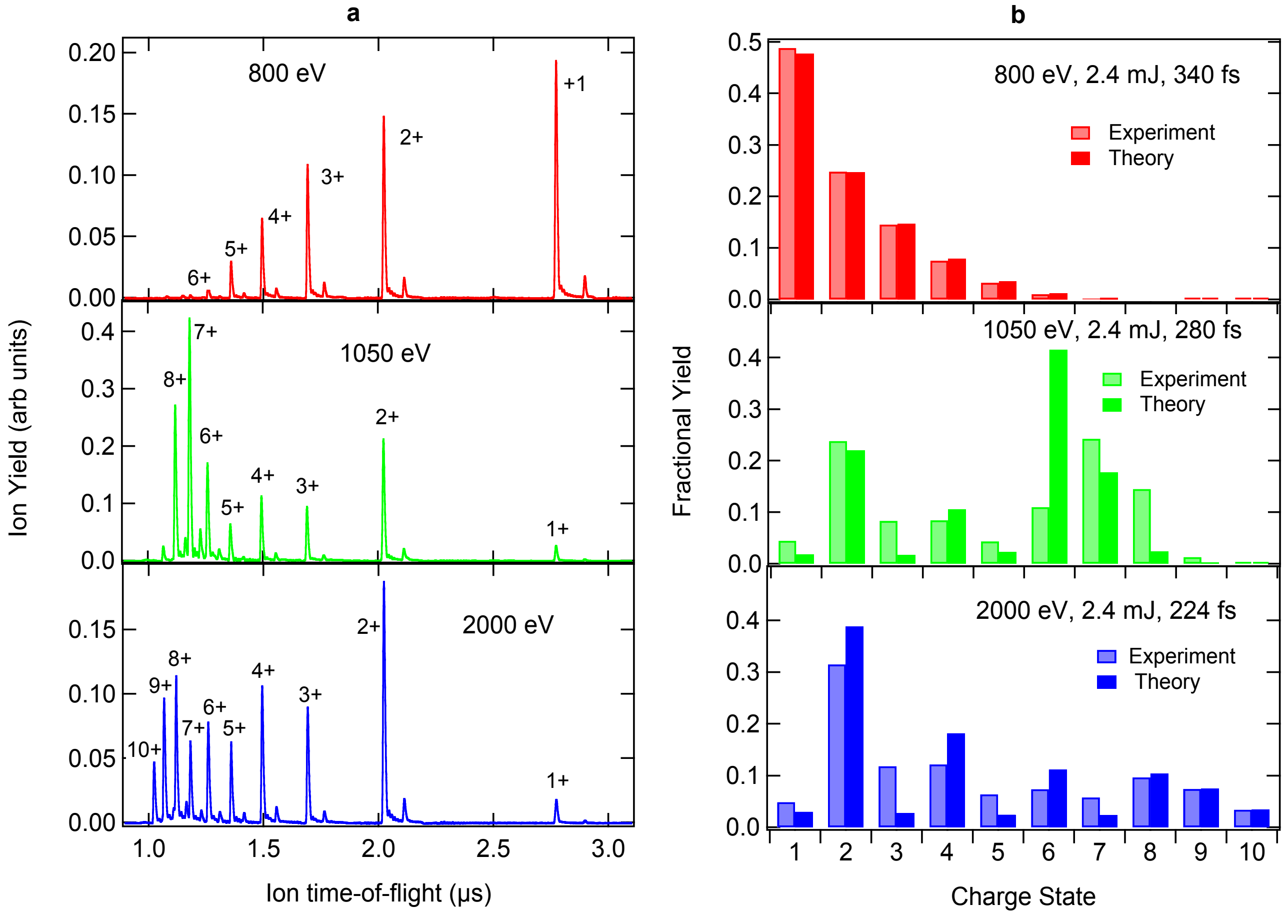
- Single ~100 fs pulse at 2000 eV fully strips neon

  6-photon, 10-electron process



- Shorter pulses with equal pulse energy & fluence suppress absorption & damage.

Theory can model ultraintense x-ray-induced electronic damage in neon

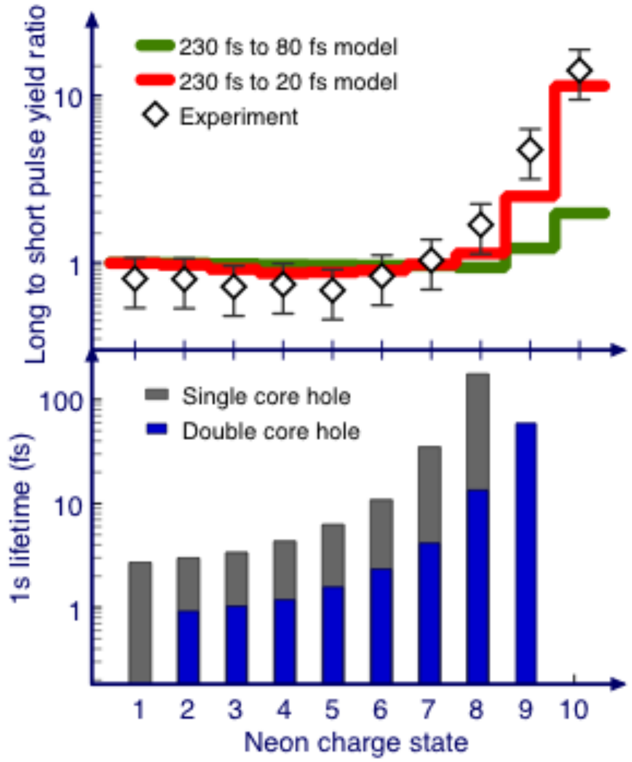


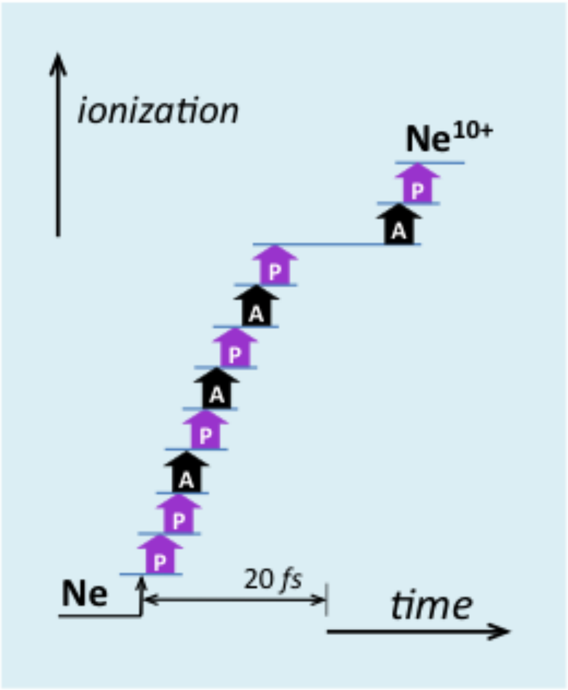
Theory

- Intensity averaged
- Fluence determined by experiment

Consistent with "measured" pulse energy and focus.

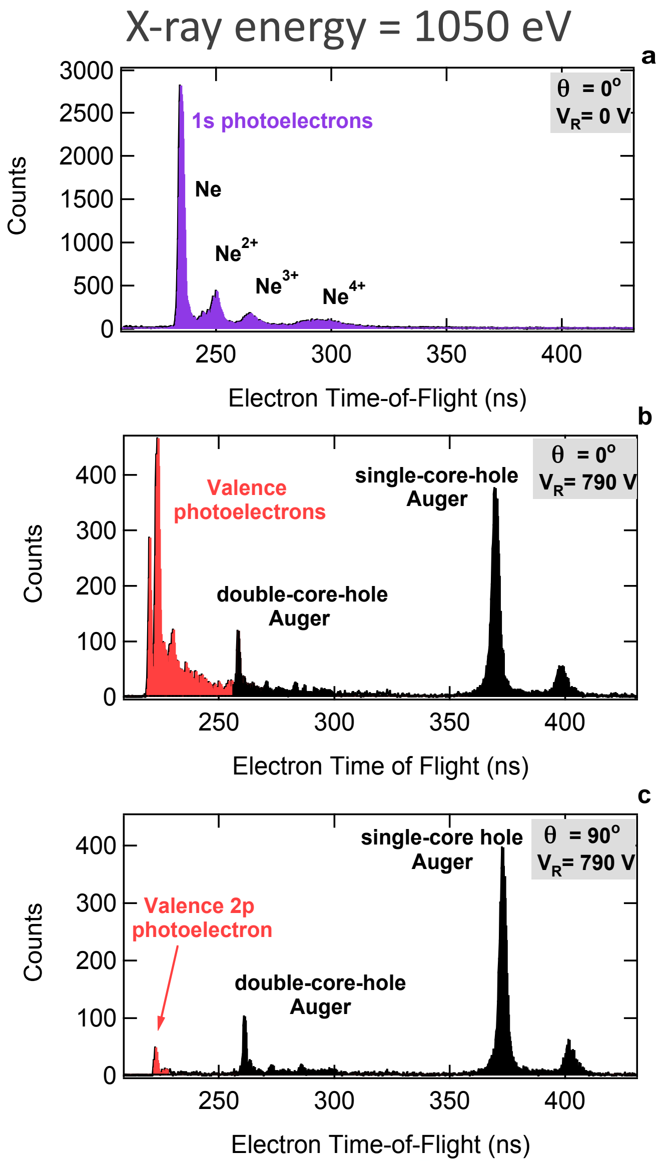
Atoms become transparent at high x-ray intensity !





- x-ray absorption is due to the presence of 1s electrons
- high x-ray intensities eject 1s electrons rendering the atom transiently transparent
- slowing atomic clocks create transparency at surprisingly long timescales

Electron spectrometers track ionization mechanism



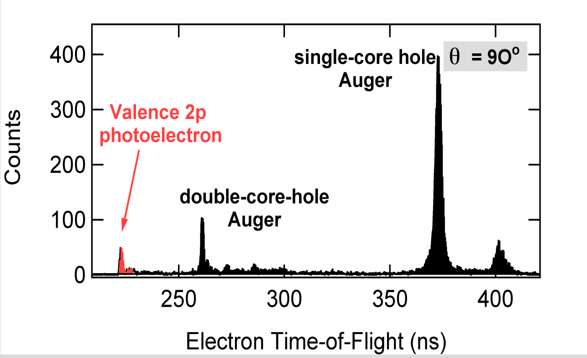
"Slow" 1s photoelectrons along x-ray polarization axis

"Fast" valence photoelectrons and Augers along polarization axis

Clean hollow atom signature double-core-hole Auger

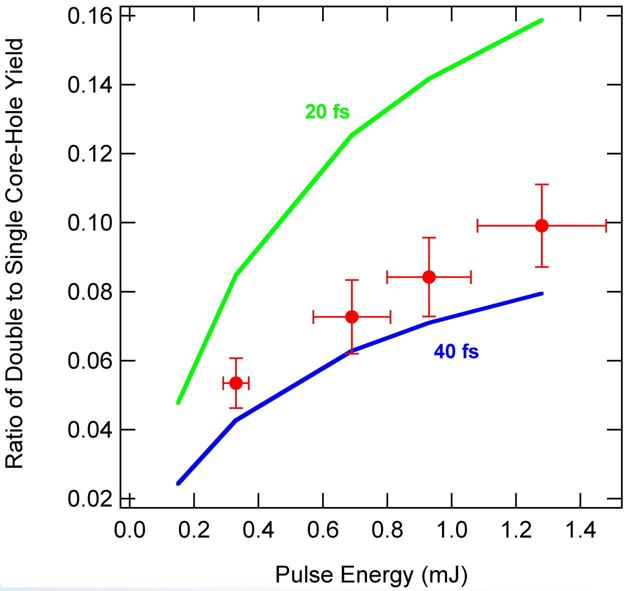
θ = 90°

Hollow atom production: deliberate, huge and an a an indicator of x-ray pulse duration

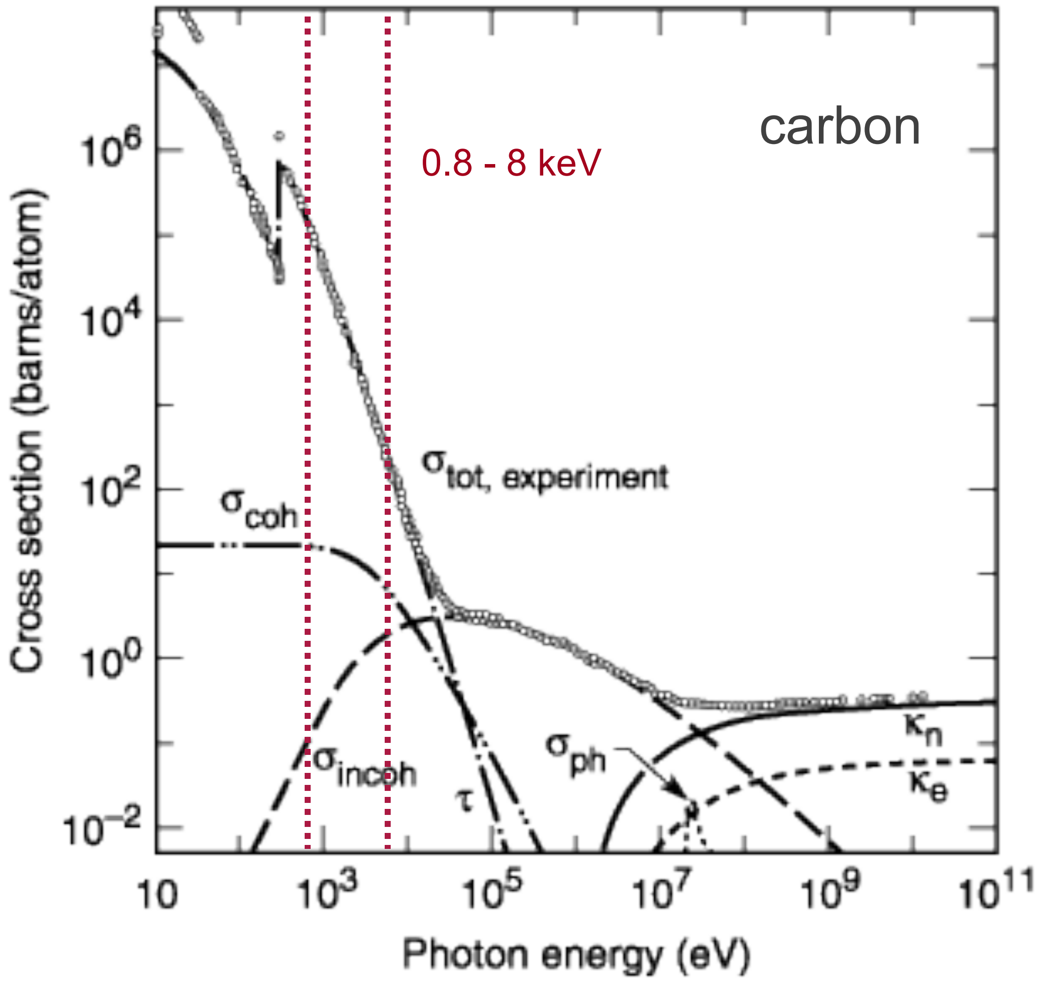


Hollow atom yield
@ LCLS ~10%
@ synchrotron ~0.3%
due to electron correlation

1050 eV,
nominal electron bunch
duration ~80 fs



Absorption vs scattering: normal and hollow atoms



	$\frac{\sigma_{photo}}{\sigma_{elastic}}$	$\frac{\sigma_{Compton}}{\sigma_{elastic}}$
2 keV	360	0.05
8 keV	20	0.60
8 keV hollow	2	

Summary of ultra-intense x-ray interaction phenomena

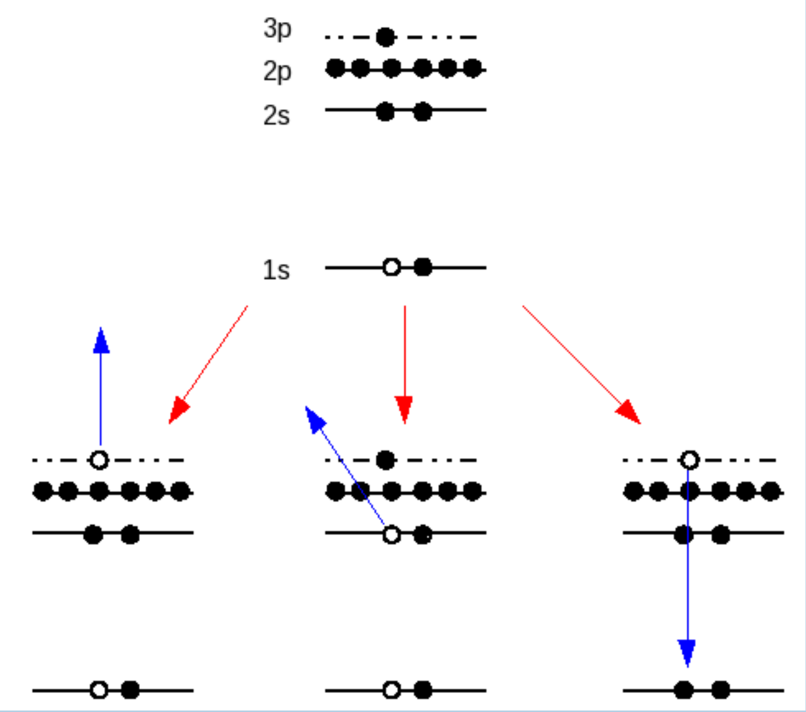
- Target changes during a 100 fs x-ray pulse at fluences similar to that for single molecule imaging
  - six-photon, ten-electron stripping of neon ($\sim 10^{12}/\mu m^2$)
  - multiphoton absorption probability high when fluence > $1/\sigma$
- Intensity-induced x-ray transparency – a general phenomena
  - transient x-ray transparency caused by formation of hollow atoms
  - hollow atoms $\sigma_{scatt}/\sigma_{abs}$ is increased – advantageous for imaging
- Straightforward rate equation calculations capture essential physics
- Femtosecond time-scale atomic processes provide FEL diagnostics

LCLS Experiment 5

Resonant x-ray processes at high intensity

Can we control inner-shell electron dynamics?

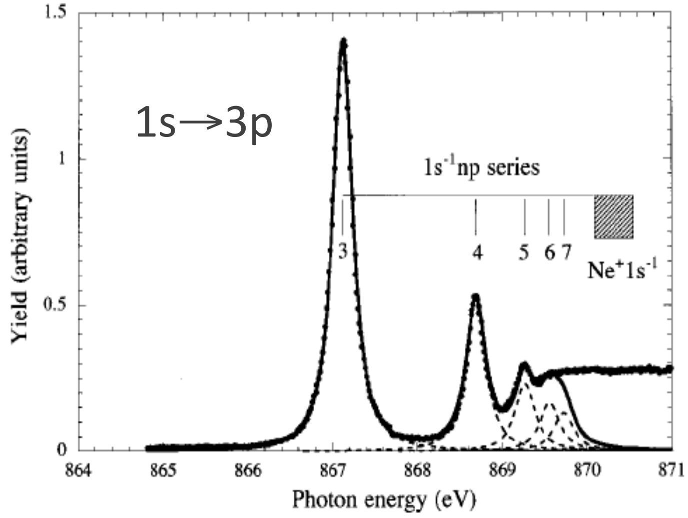
“Rabi flopping” may inhibit Auger decay & x-ray damage.



unlikely

somewhat likely

very likely



- Strong 1s→3p resonance

  $\mu_{Ne\ 1s\text{-}3p} = 0.01\ ea_0$

  $\tau_{Ne\ 1s^{-1}} = 2.4\ fs = 100\ a.u.$

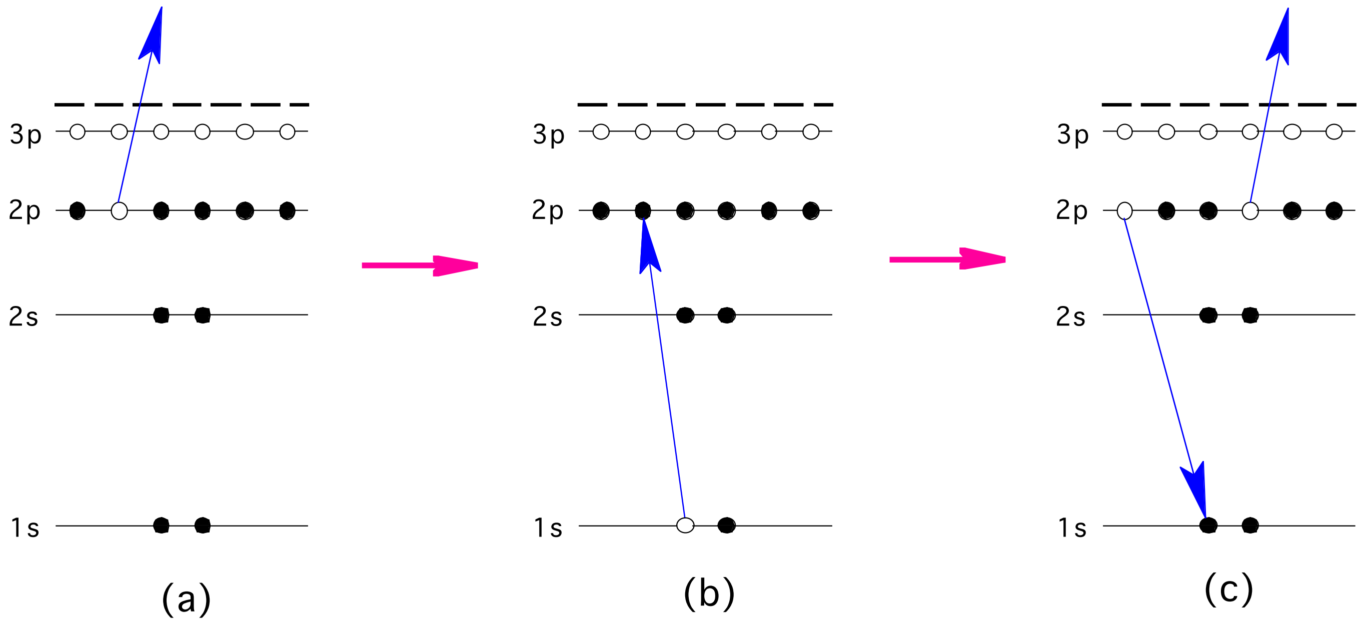
- Rabi flopping possible

  $E_{Ne} \sim 6.3\ a.u.$

  $I_{Ne} \sim 1.4\ x10^{18}\ W/cm^2$

But LCLS linewidth ~ 8 eV!

Rabi-flopping on 1s - 2p resonance more probable



$E_{x\text{-}ray} = 848.6\ eV$

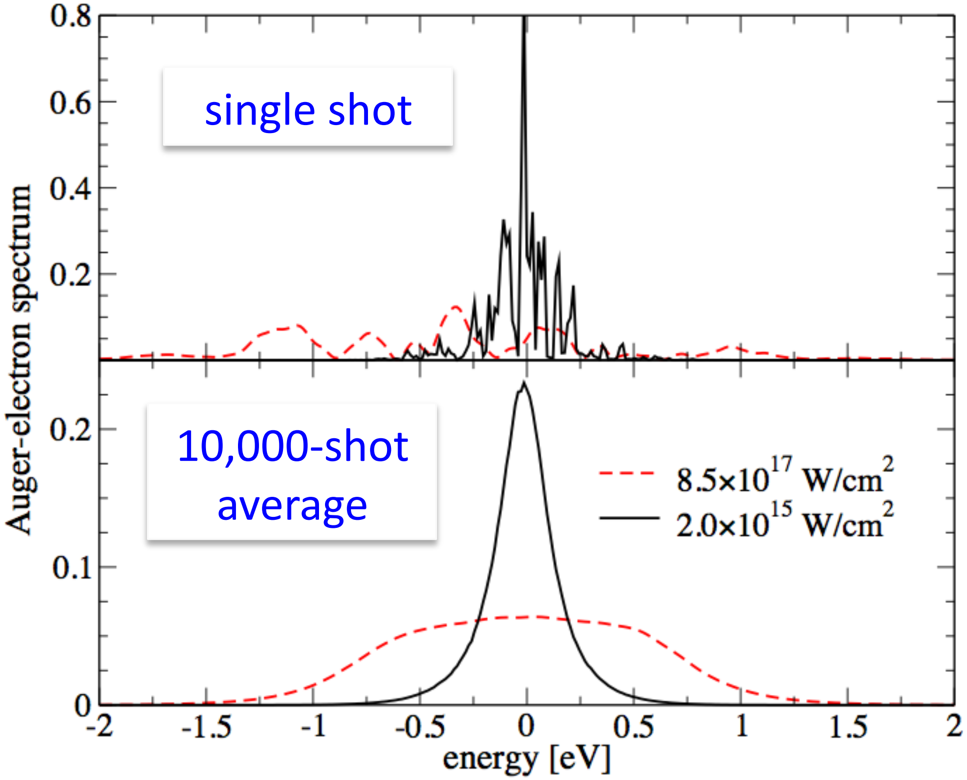
$\sigma_{1s\text{-}2p} = 500\sigma_{2p\text{-}\infty} = 5\ \sigma_{1s\text{-}3p}$

Observe Auger yield when x-rays scanned over 1s - 2p resonance.

Observe broadening at resonance to indicate Rabi flopping

Theory: Rohringer & Santra PRA (2008).

Calculated “Resonant Auger effect at high x-ray intensity”

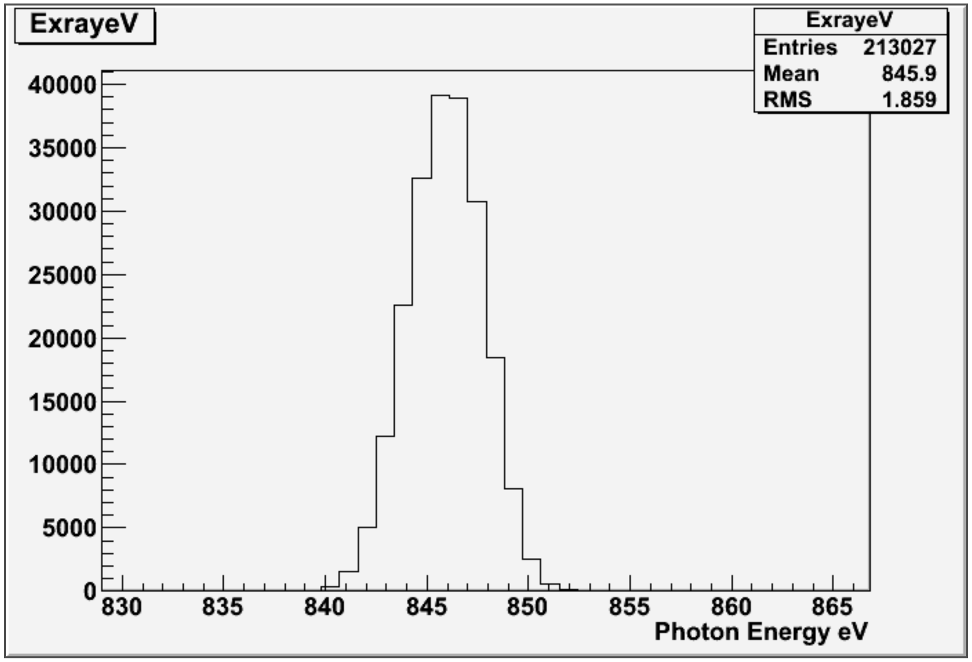


-> Look for Auger line broadening on resonance

N. Rohringer & R. Santra, PRA 77, 053404 (2008)

Shot-to-shot photon energy jitter

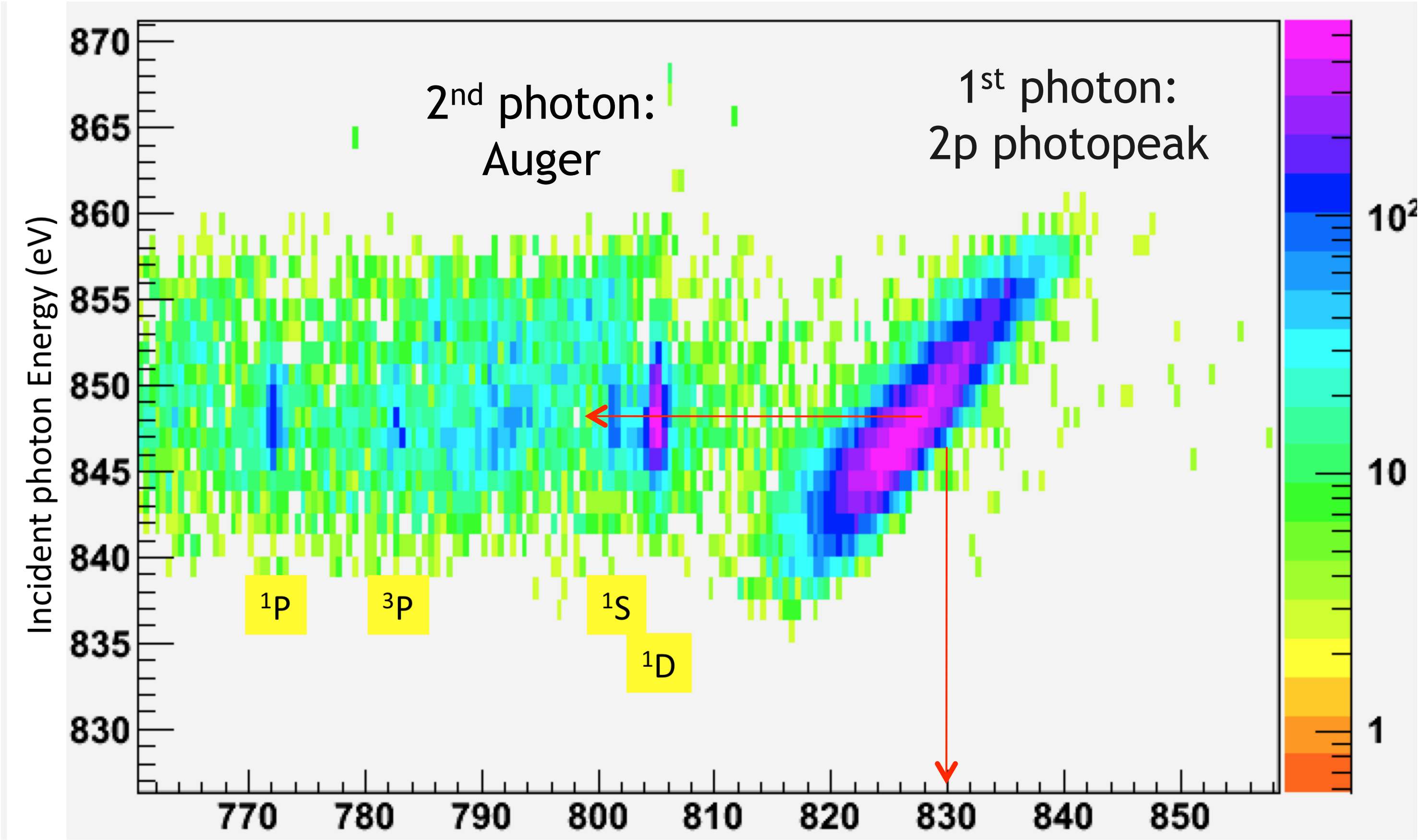
derived from GeV electron bunch energy measurement



Conditions	FWHM photon energy jitter (eV)
40 pC (<10 fs) 850 eV 0.3 mJ 4500 A	4.25
250 pC (100 fs) 787 eV 1.5 mJ 2500 A	4.79
250 pC (100 fs) 769 eV 1.5 mJ 2500 A	5.24

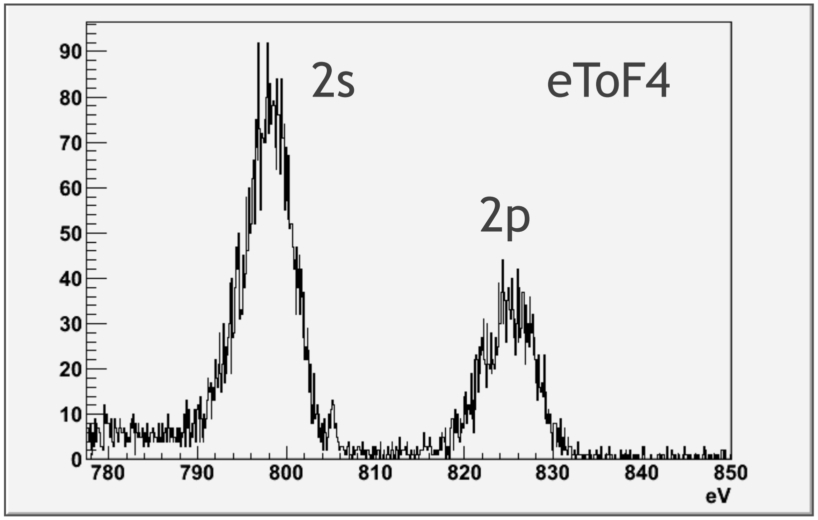
Auger-electrons observed at the 1s -> 2p resonance of Ne$^+$

using eTOF1 perpendicular to photon polarization to suppress 2s photoelectrons (40 pC/bunch)

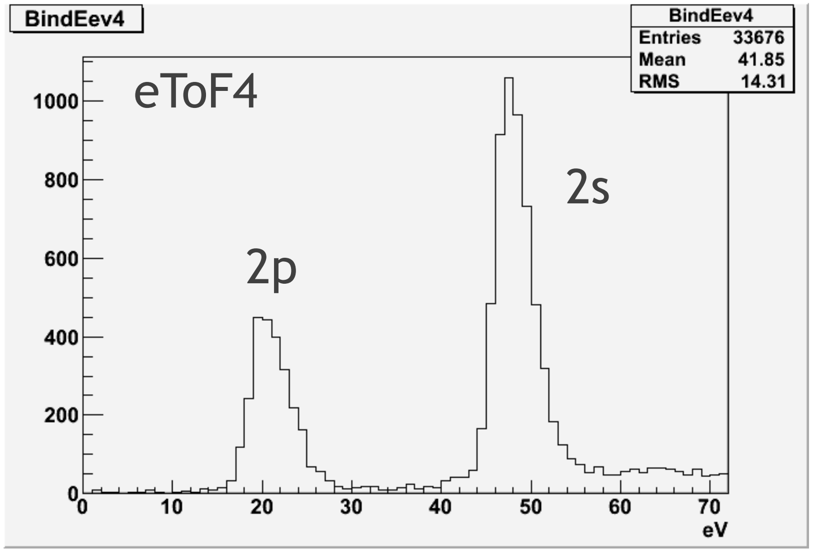


Electron kinetic energy (eV)

Determination of intrinsic x-ray bandwidth from electron spectra



Electron kinetic energy (eV)

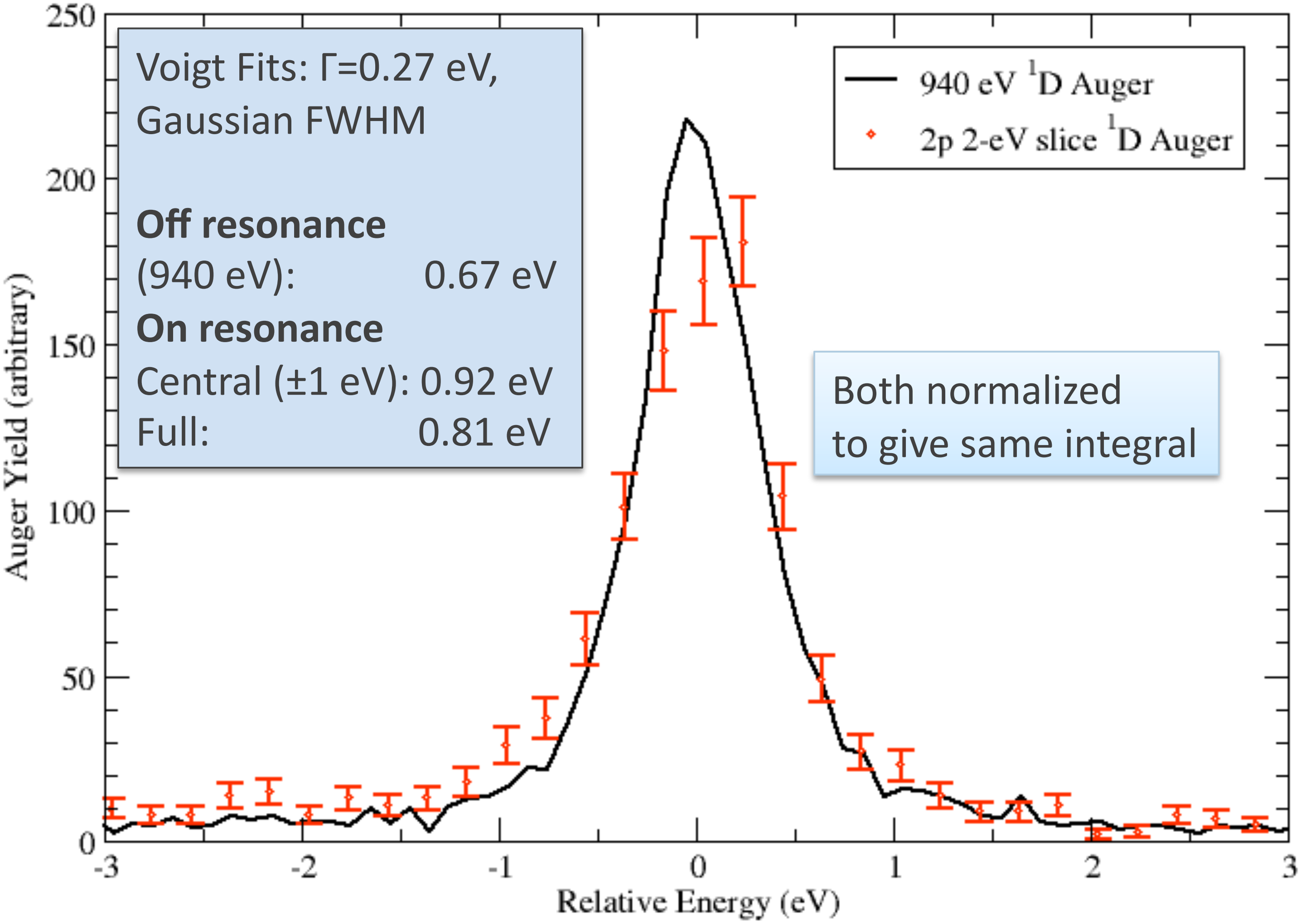


Electron binding energy (eV)

Conditions	Intrinsic x-ray pulse bandwidth (from 2s photopeak) (eV) (FWHM)	Intrinsic x-ray pulse bandwidth (from 2p photopeak) (eV) (FWHM)	Average bandwidth (eV) (FWHM)	%
40 pC (<10 fs) 850 eV 0.3 mJ 4500 A	4.3	4.5	4.4	0.5 %
250 pC (100 fs) 787 eV 1.5 mJ 2500 A	7.1	7.8	7.45	0.9%
250 pC (100 fs) 769 eV 1.5 mJ 2500 A	7.7	7.8	7.77	1%

Is the ^{1}D Auger line broadened on resonance?

Preliminary Result: Yes!



Voigt Fits: Γ=0.27 eV, Gaussian FWHM

Off resonance
(940 eV): 0.67 eV
On resonance
Central (±1 eV): 0.92 eV
Full: 0.81 eV

Both normalized to give same integral

Are multiple core holes within a single molecule useful?

LCLS experiment X
5-10 Aug 2010

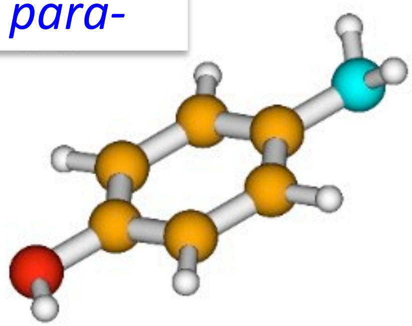
S. Southworth et al.

X-ray two photon photoelectron spectroscopy

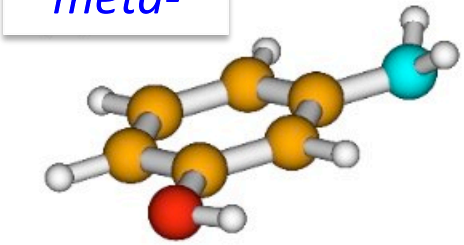
a probe of molecular electronic structure

early work by Cederbaum and coworkers: C_2H_n and Benzene

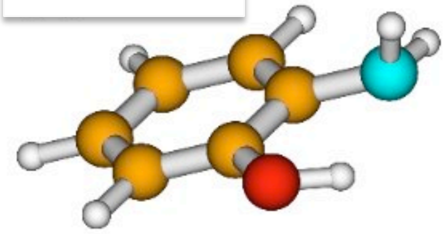
para-



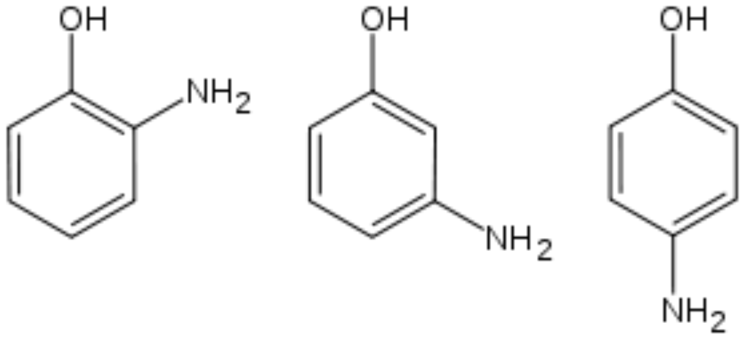
meta-



ortho-



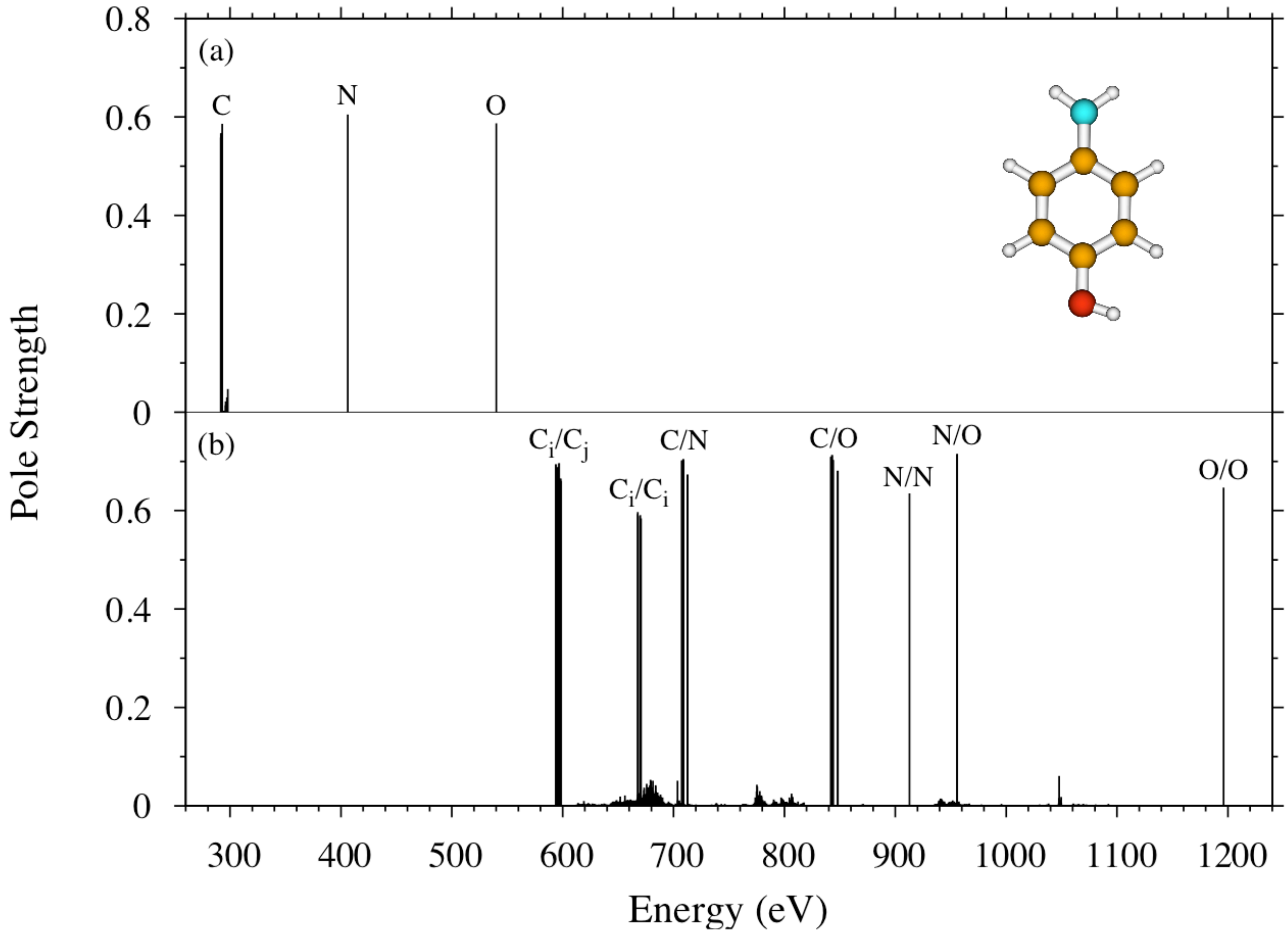
structural isomers of aminophenol



5 – 7 fs core-hole lifetimes on C, N, O

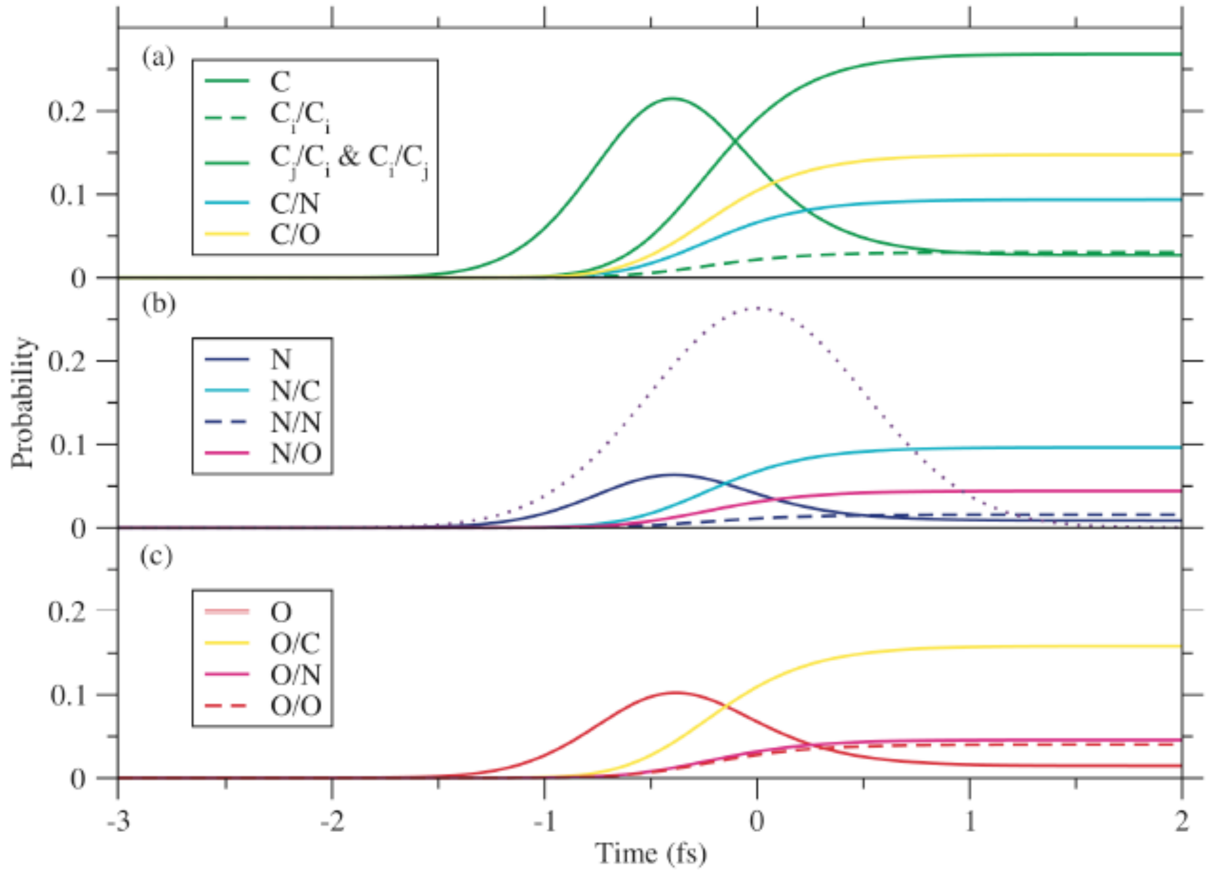
R. Santra, N. V. Kryzhevoi, and L. S. Cederbaum, PRL 103, 013002 (2009)

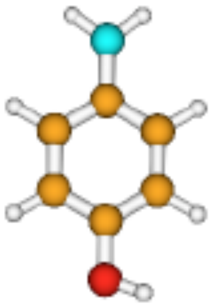
Inner-shell single- and double-ionization potentials of p-aminophenol



R. Santra, N. V. Kryzhevoi, and L. S. Cederbaum, PRL 103, 013002 (2009)

Time evolution of single and double hole states in p-aminophenol

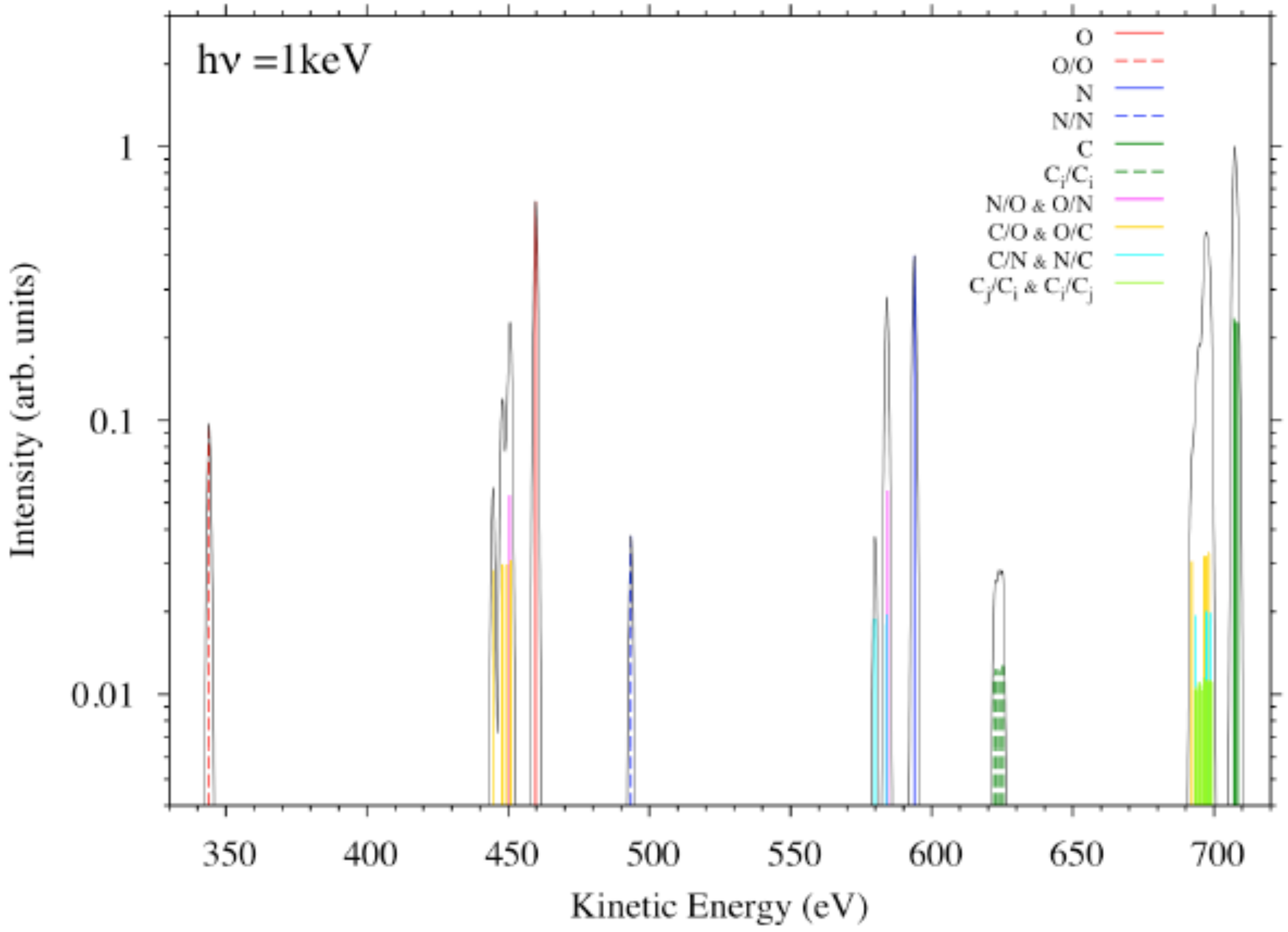


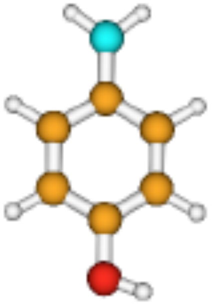


X-ray parameters
- 1 keV, 1.2 fs
- 10^{11} photons
- 1μm spot size

R. Santra, N. V. Kryzhevoi, and L. S. Cederbaum, PRL 103, 013002 (2009)

Calc’d x-ray photoelectron spectrum p-aminophenol





X-ray parameters
1 keV, 1.2 fs
10^{11} photons
1μm spot size

Distinct PE Groups: O: 340-460 eV, N: 480-600 eV, C: 620-710 eV

Summary

- Insight into ultraintense x-ray interations
  - six-photon, ten-electron stripping of neon ($\sim 10^{12}/\mu m^2$)
  - multiple photon absorption probability high when fluence > $1/\sigma$
- Intensity-induced x-ray transparency – a general phenomena
  - transient x-ray transparency caused by formation of hollow atoms
  - hollow atoms $\sigma_{scatt}/\sigma_{abs}$ is increased
- Femtosecond time-scale atomic processes provide FEL diagnostics
- Straightforward rate equation calculations capture essential physics
- Looking forward to extension toward more complex, yet tractable, systems …

Argonne AMO group Oct 2009

Heroes at AMO Control

Heroes at MCC