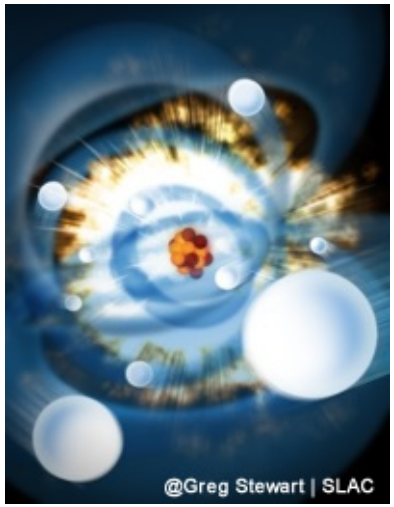
LCLS experiments and x-ray atomic physics

Linda Young



CAMOS Meeting
Keck Center, Washington D.C.
6 April 2011

Outline

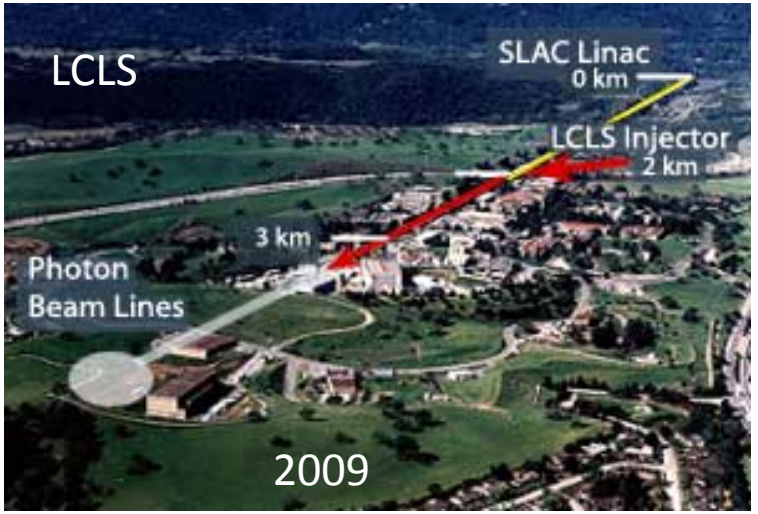
- Context & motivation
- Survey of AMO-related LCLS experiments
- Non-resonant high intensity x-ray phenomena
  LCLS Experiment 1: Oct 1 - 6, 2009
- Resonant high intensity x-ray processes
  LCLS Experiment 5: Oct 29 - Nov 3, 2009
- Outlook/wider impacts/final thoughts

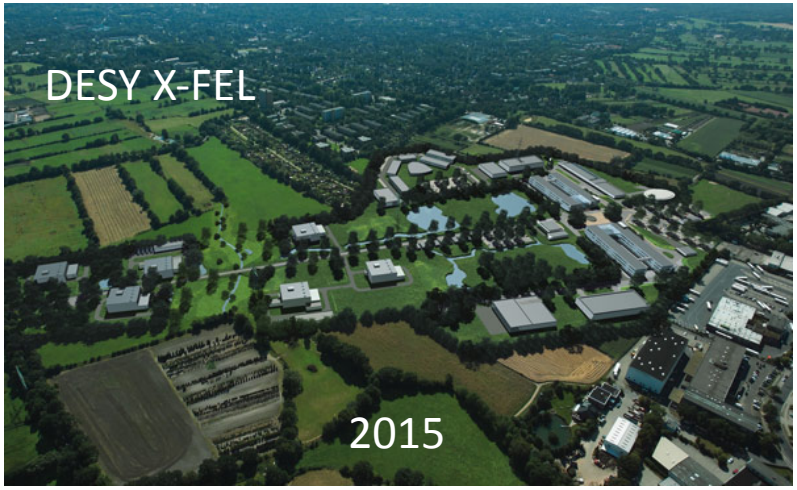
XFELs are here (LCLS) and more are coming soon!

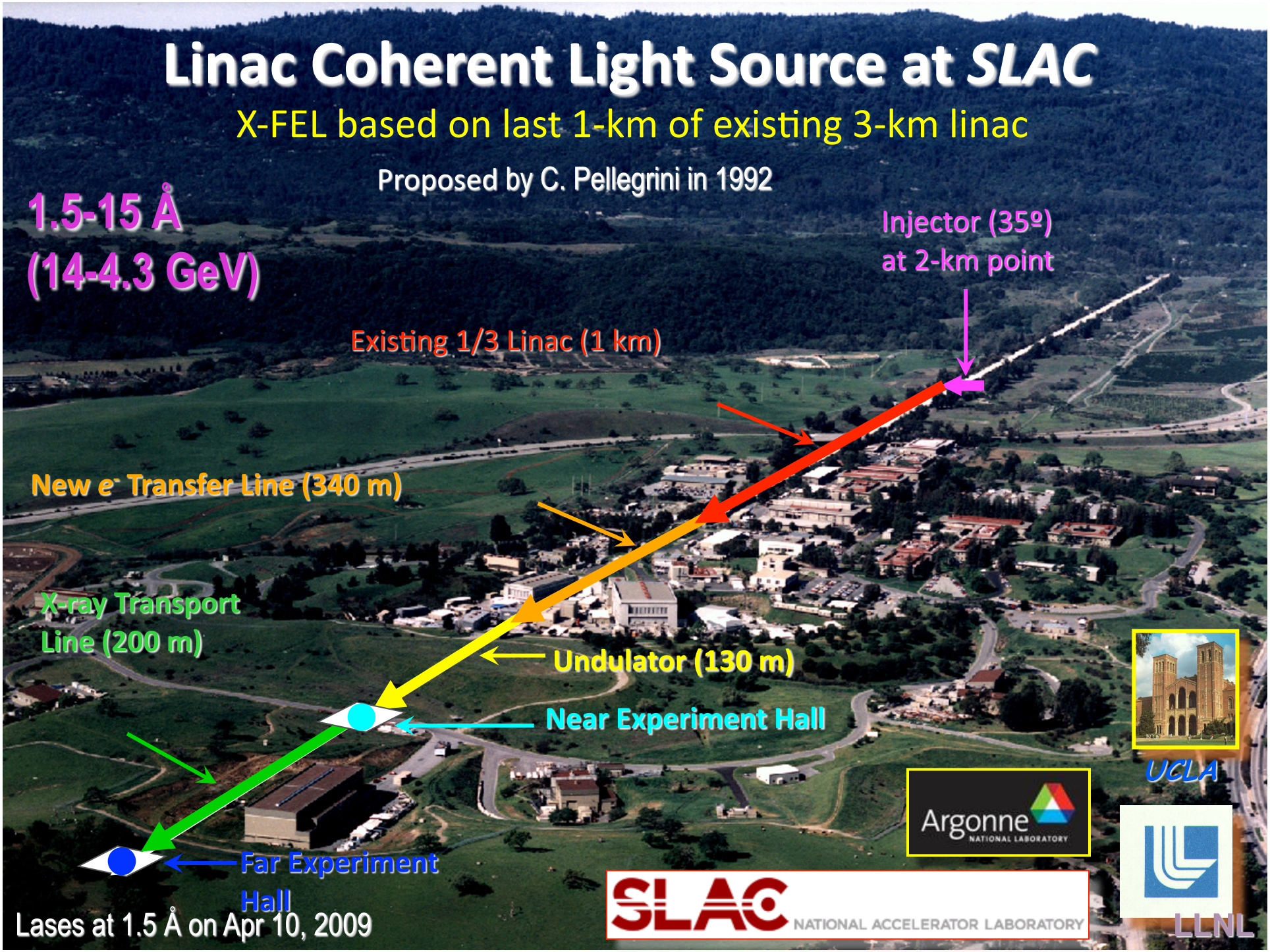
Physics Today, May 2005

X-Ray Free-Electron Laser Projected Parameters			
	LCLS (US)	DESY XFEL (Europe)	SCSS (Japan)
Pulse duration	<230 fs	100 fs	80 fs
Wavelength	1–64 Å	1–15 Å	1–50 Å
Repetition rate	120 Hz	10 Hz	60 Hz
Electron bunches per pulse	1	≤3000	1
Electron beam energy	4–14 GeV	≤20 GeV	≤8 GeV
Photons per pulse ($\times 10^{12}$)	1.2 (at 1.5 Å)	1.2 (at 1 Å)	0.76 (at 1 Å)
Linac length	1 km	2 km	350 m
Estimated cost*	$379 million	$1 billion	$330 million
Estimated start date	2009	2012	2010

*Estimates include varying amounts of instrumentation and different methods of accounting.



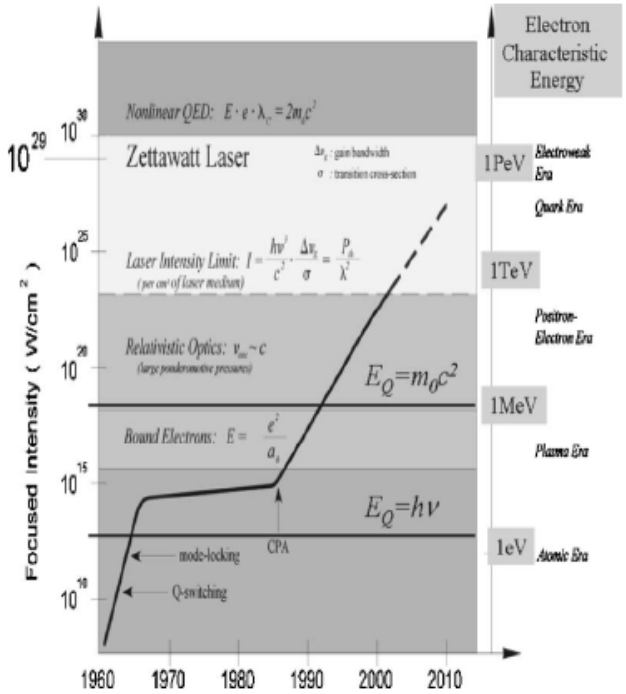



Linac Coherent Light Source at SLAC
X-FEL based on last 1-km of existing 3-km linac
Proposed by C. Pellegrini in 1992
1.5-15 Å
(14-4.3 GeV)
Injector (35º)
at 2-km point
Existing 1/3 Linac (1 km)
New e- Transfer Line (340 m)
X-ray Transport
Line (200 m)
Undulator (130 m)
Near Experiment Hall
Far Experiment
Hall
Lases at 1.5 Å on Apr 10, 2009
UCLA
Argonne
NATIONAL LABORATORY
SLAC
NATIONAL ACCELERATOR LABORATORY
LLNL

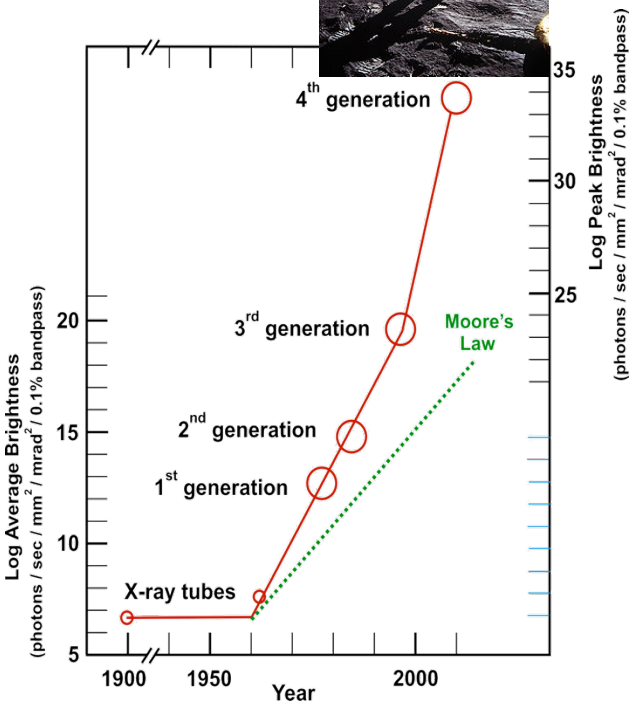
Compare the evolution of high intensity optical and x-ray sources

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

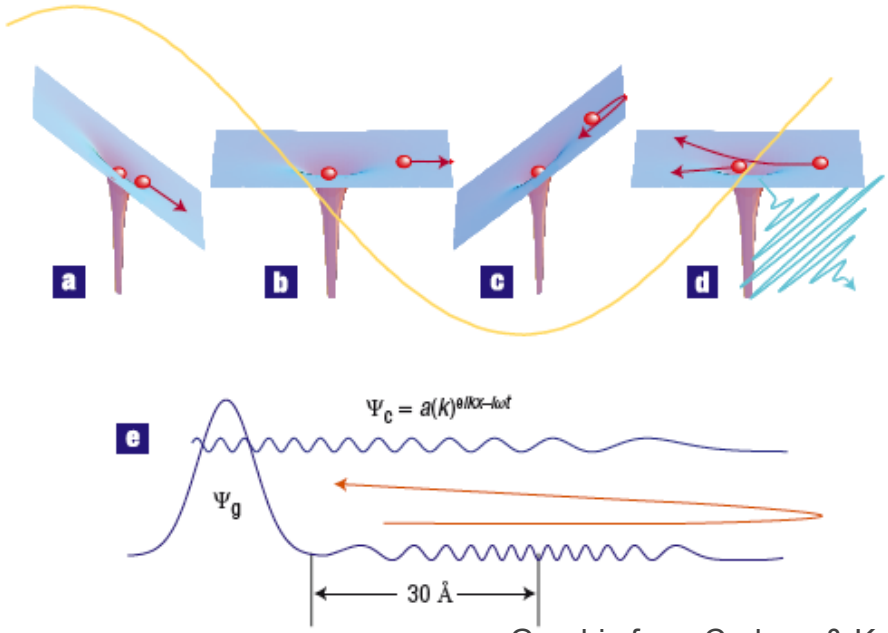
At long wavelengths - laser-driven electron dynamics is dominant
… not so at short wavelengths

electron ponderomotive energy (au)

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

displacement

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

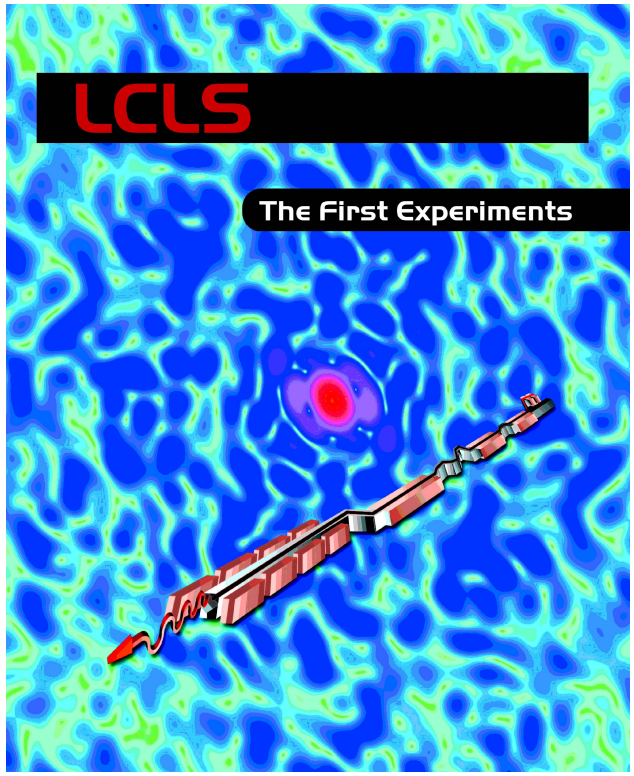


Graphic from Corkum & Krausz
Nature Physics (2007)

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

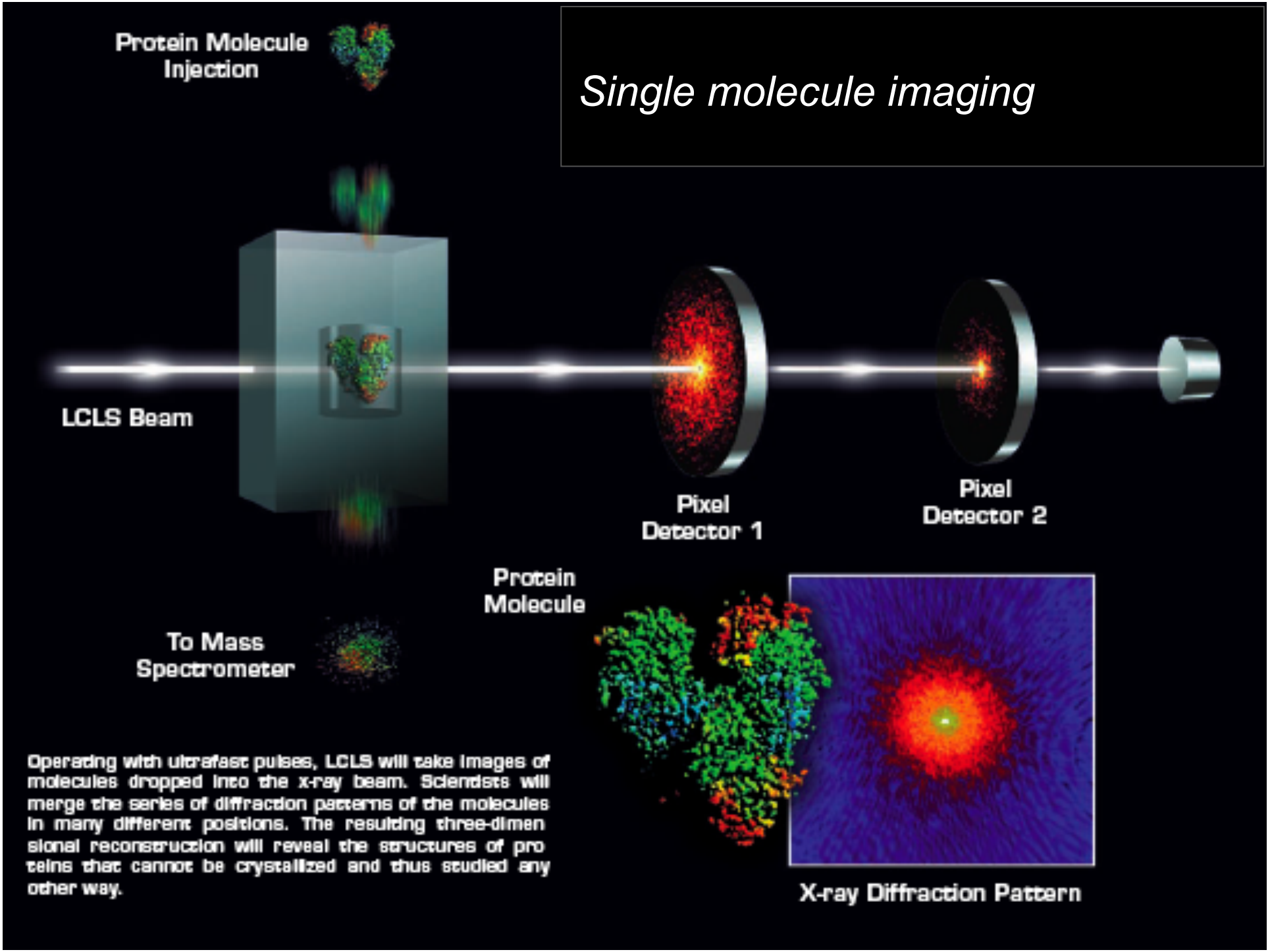
XCS: X-ray Correlation Spectroscopy

CXI: Coherent X-ray Imaging

MEC: Materials in Extreme Conditions

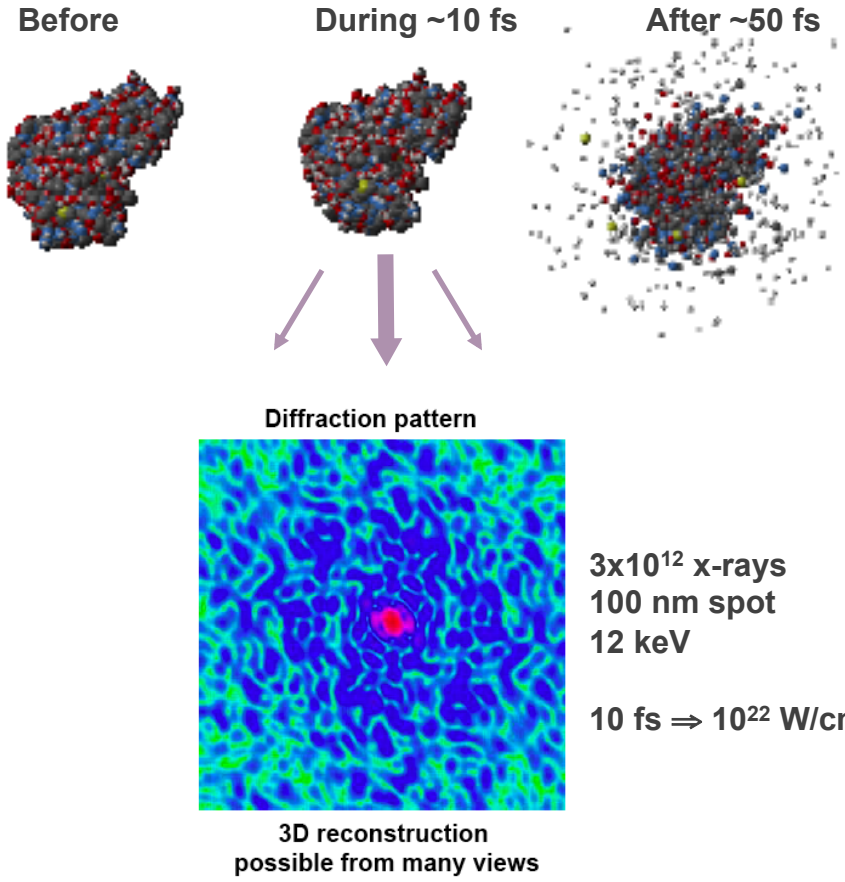
AMO

- 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


Protein Molecule Injection
Single molecule imaging
LCLS Beam
Pixel Detector 1
Pixel Detector 2
Protein Molecule
To Mass Spectrometer
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.
X-ray Diffraction Pattern

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



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

AMO-related LCLS publications to date

X-ray Atom Interactions

- *Femtosecond electronic response of atoms to ultraintense x-rays*
  L. Young et al., Nature **446**, 56 (2010).
- *Nonlinear atomic response to intense ultrashort x-rays*
  G. Doumy et al., Phys. Rev. Lett. **106**, 083002 (2011).

X-ray Molecule Interactions

- *Ultraintense X-Ray Induced Ionization, Dissociation, and Frustrated Absorption in Molecular Nitrogen*
  M.Hoener et al., Phys. Rev. Lett. **104**, 253002 (2010).
- *Auger Electron Angular Distribution of Double Core-Hole States in the Molecular Reference Frame*
  J. Cryan et al., Phys. Rev. Lett. **105**, 083004 (2010).
- *Double Core-Hole Production in N2: Beating the Auger Clock*
  L. Fang et al., Phys. Rev. Lett. **105**, 083005 (2010).

X-ray / optical laser timing and synchronization

- *Time-resolved pump-probe experiments at the LCLS*
  J. M. Glownia et al. Opt. Express **18**, 17621 (2010).

AMO-related LCLS pubs in the works …

X-ray pulse characterization using two color interactions

- *Femtosecond X-ray Pulse Length Characterization at the LCLS Free Electron Laser*
  S. Düsterer M. Meyer et al.
- *Investigation of the time structure of LCLS's few fs x-ray pulses*
  R. Kienberger et al.

X-ray Cluster Interactions

- *Explosions of clusters in intense X-ray pulses*
  T. Ditmire et al.
- *Ultrafast imaging of X-ray excited clusters*
  C. Bostedt, T. Möller et al.

Resonant x-ray interactions: Control of inner-shell processes

- *Modifying Auger Decay with Femtosecond X-ray Pulses*
  E. P. Kanter et al.

Electron-Beam Ion Trap: highly-charged ion spectroscopy

Bernitt, Brown, J. Crespo, P. Beiersdorfer, J. Ullrich

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 x 10^{12} x-rays/(100 nm)^2 = 3 x 10^6 x-rays/Å^2

10 fs

~10^{22} W/cm^2

Our approach to understanding ultraintense x-ray interactions

- Start with a well-characterized target

Binding energies in neutral neon

2p : ~21 eV

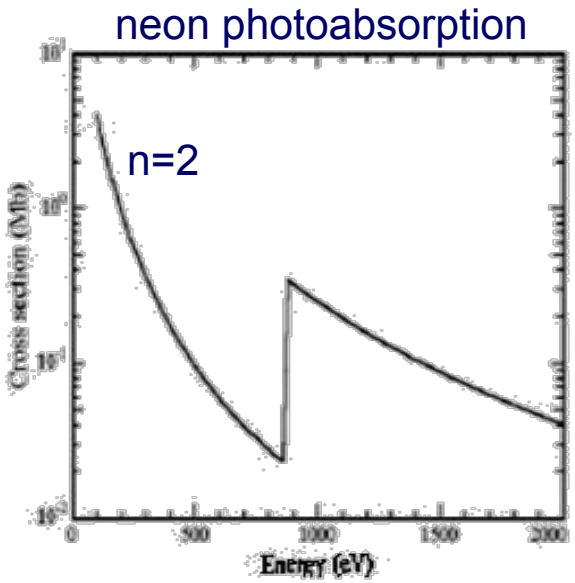
2s : ~48 eV

1s : ~870 eV

Inner-shell excitation

Auger yield 98%

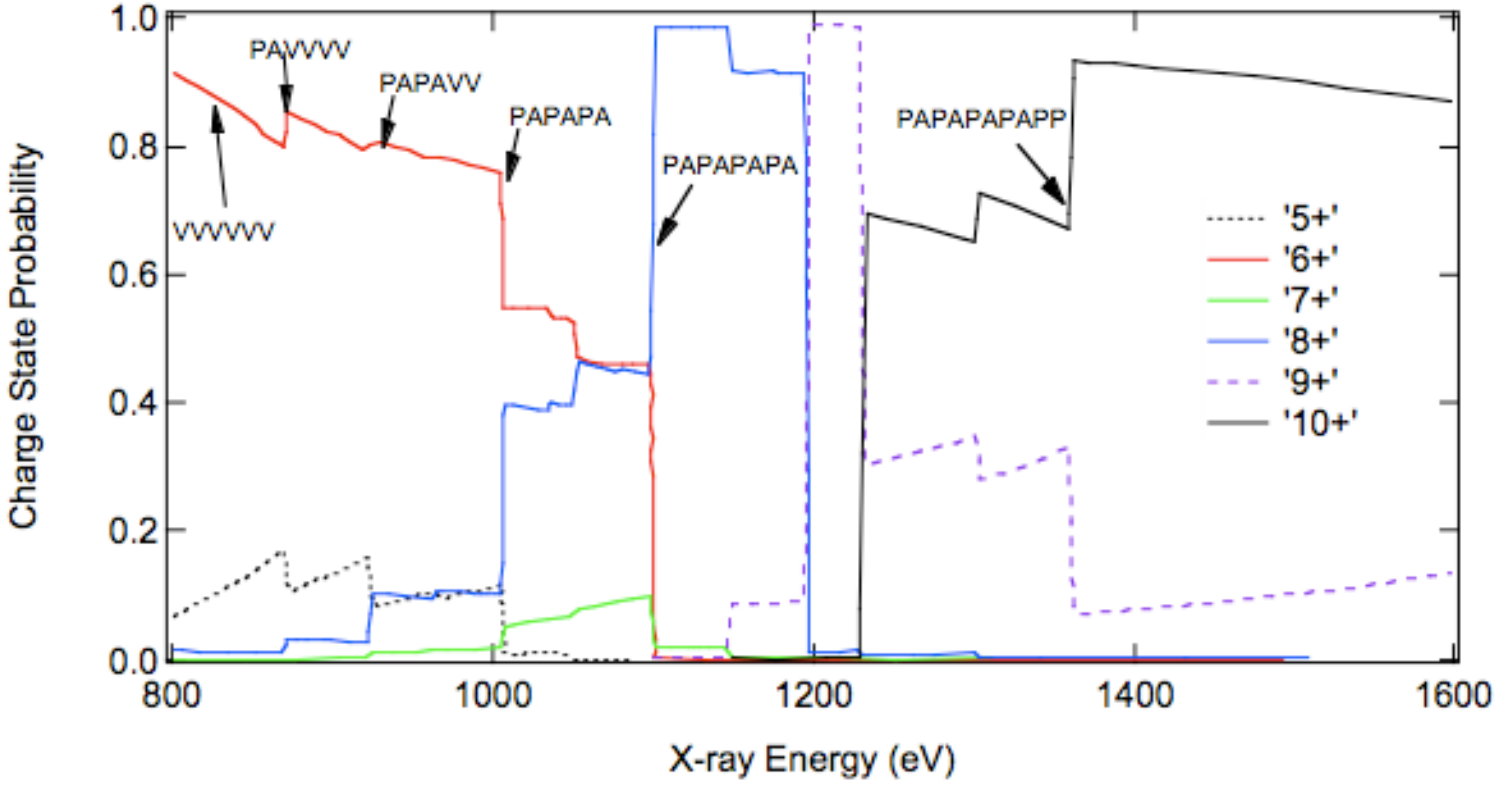
Auger clock - τ_{1s}: 2.4 fs



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

Guided by theory

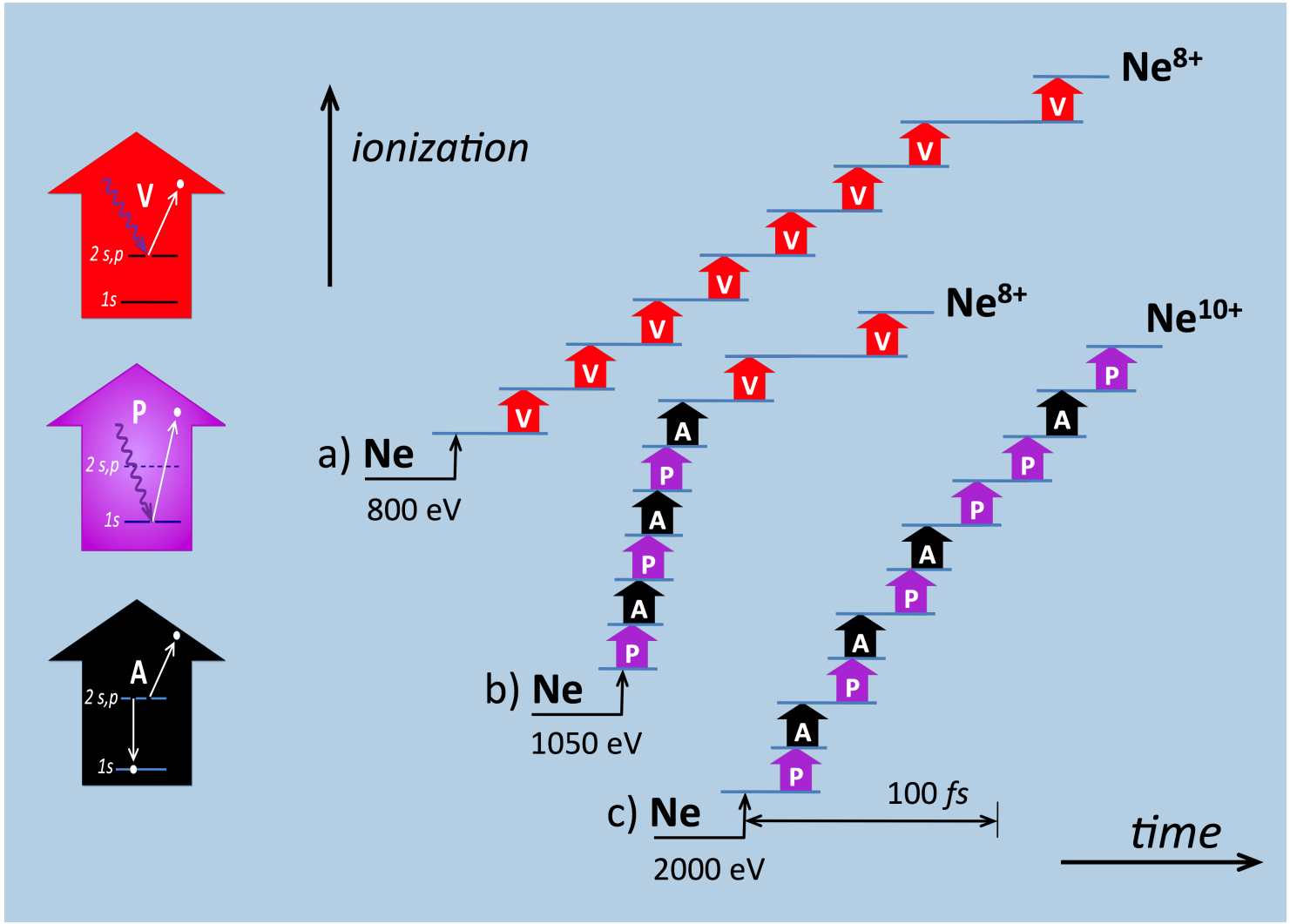
Theory: Rohringer & Santra, PRA 76, 033416 (2007)



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

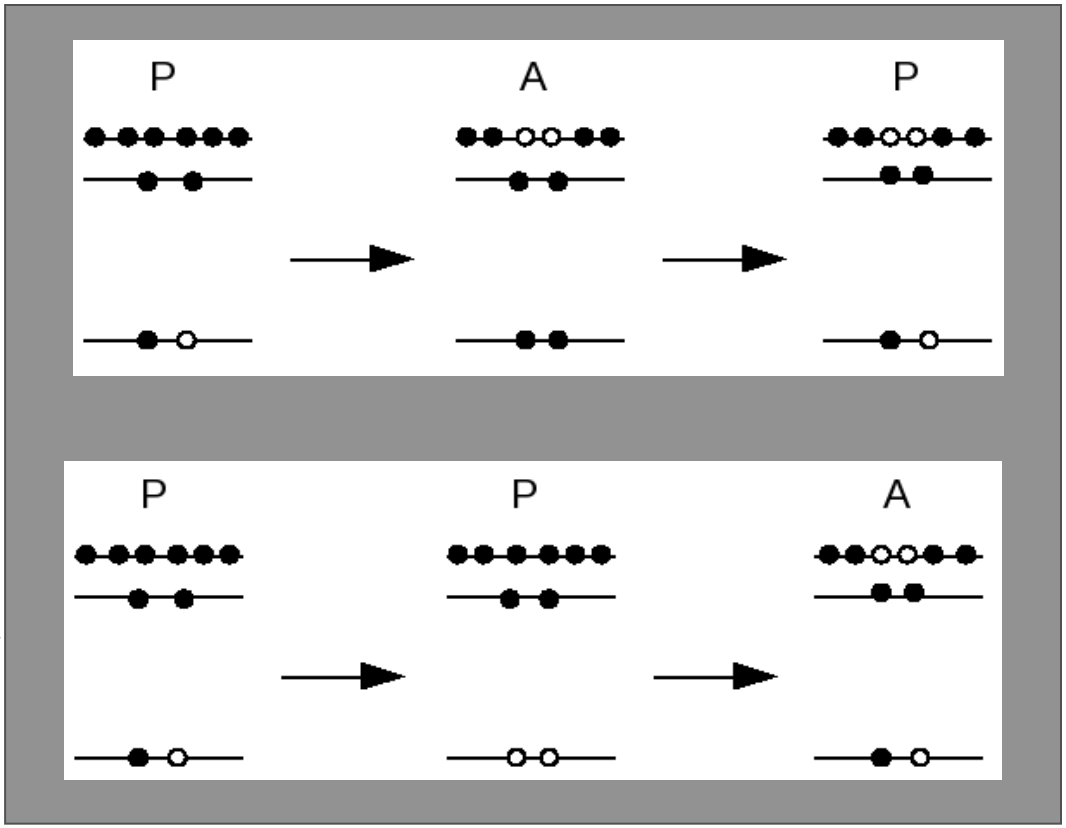


Sequential single photon processes dominate the interaction

How does one arrive at a particular charge state?

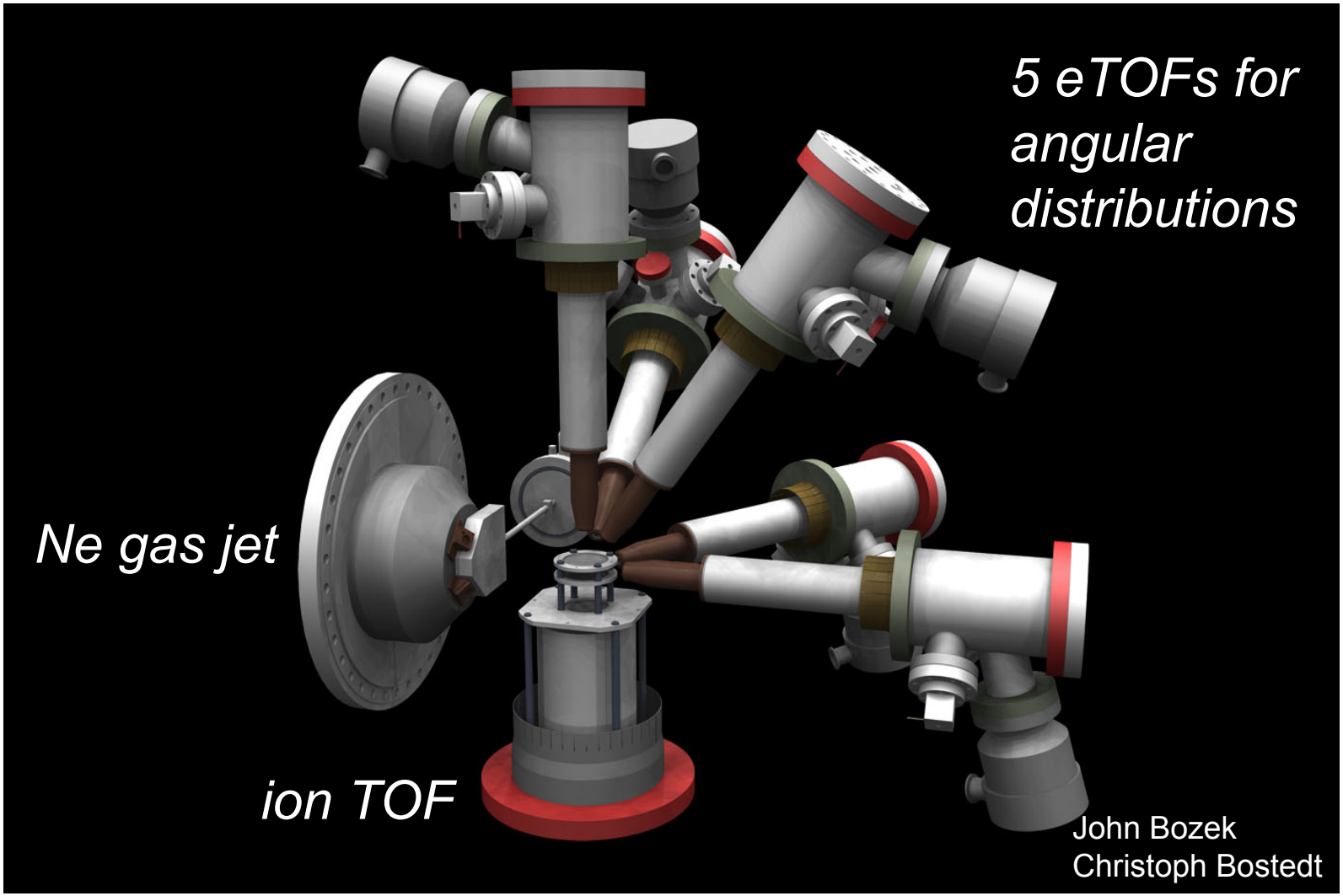
Low Intensity
PAP

High Intensity
PPA



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

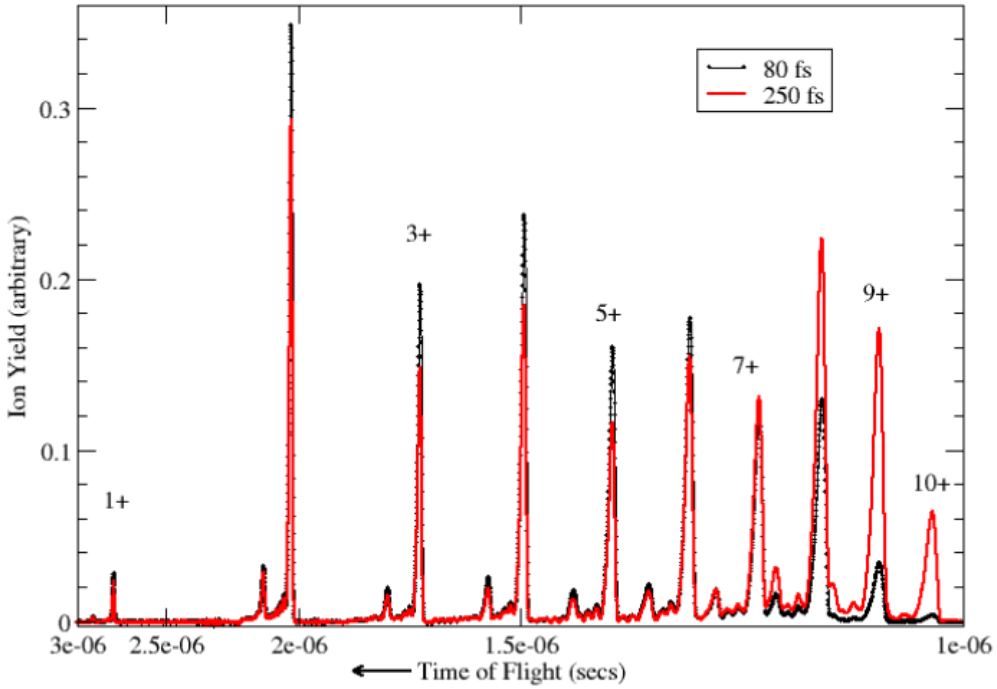
High field physics chamber



Day 1 – two interesting observations

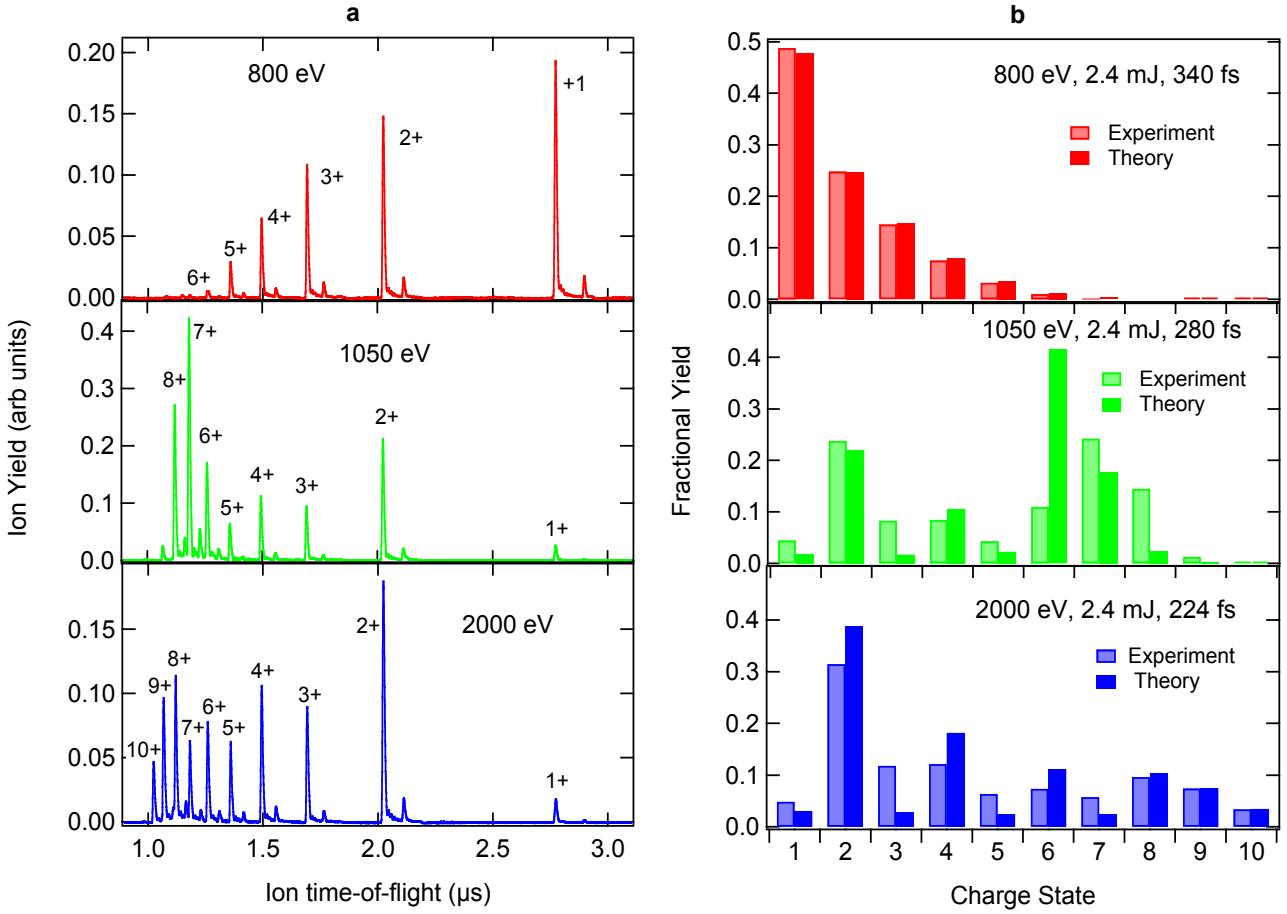
- Single ~100 fs pulse at 2000 eV fully strips neon

  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

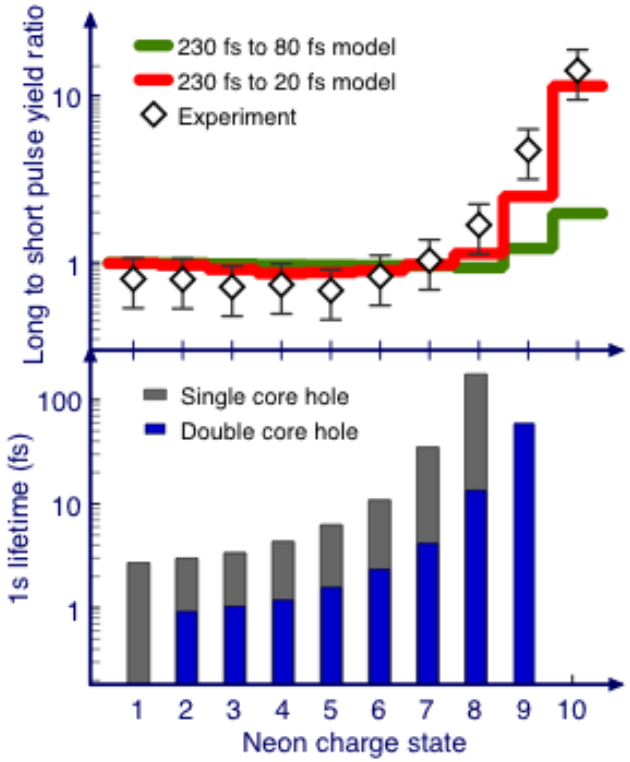


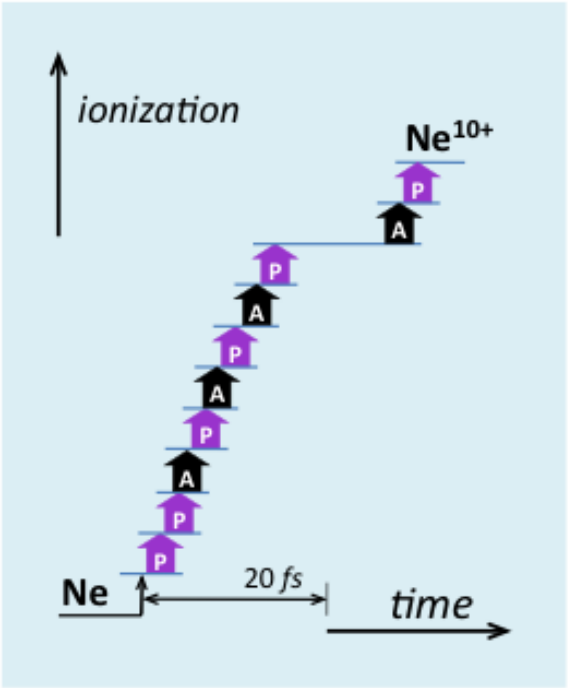
Theory
- Intensity averaged
- Fluence determined by experiment

Consistent with "measured" pulse energy and focus.

Sang-Kil Son, Robin Santra – refined calcs include shakeoff – G. Doumy et al, PRL 2011)

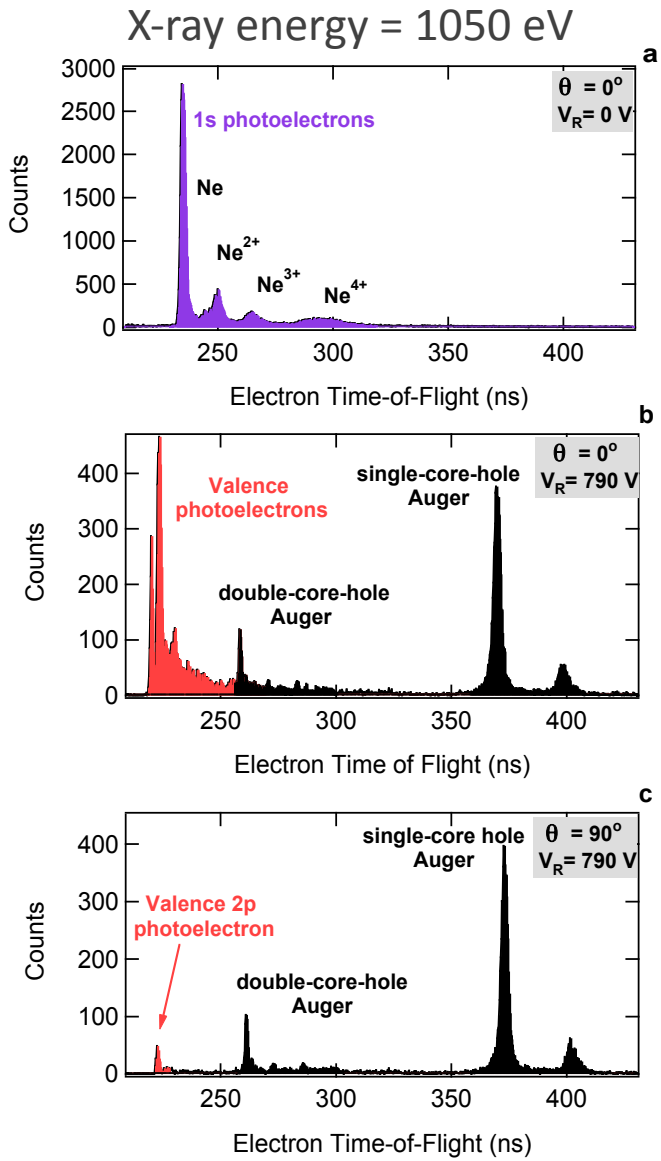
Atoms become transparent at high x-ray intensity !





- 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



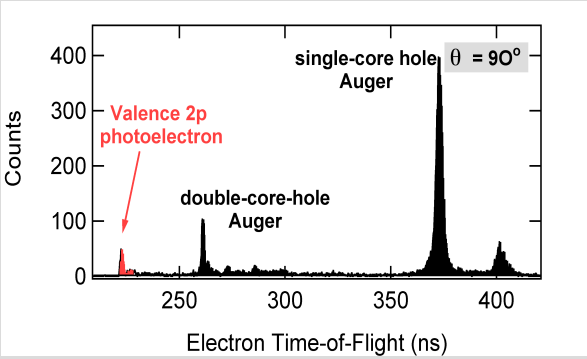
"Slow" 1s photoelectrons along x-ray polarization axis

"Fast" valence photoelectrons and Augers along polarization axis

Clean hollow atom signature double-core-hole Auger

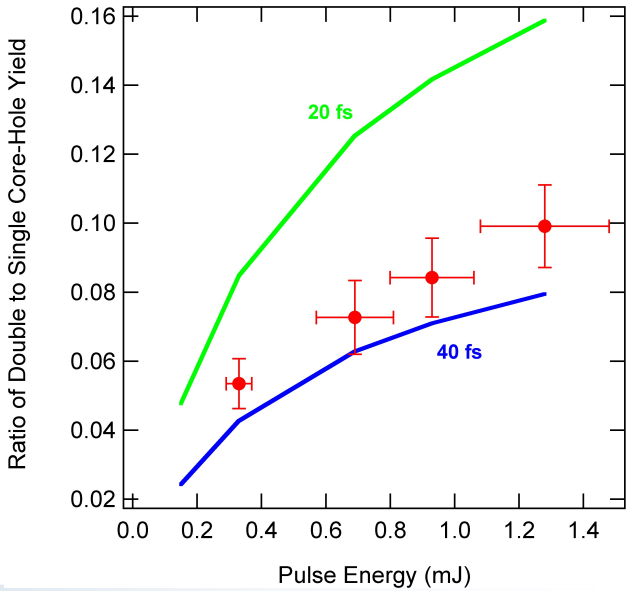
$\theta = 90^{o}$

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

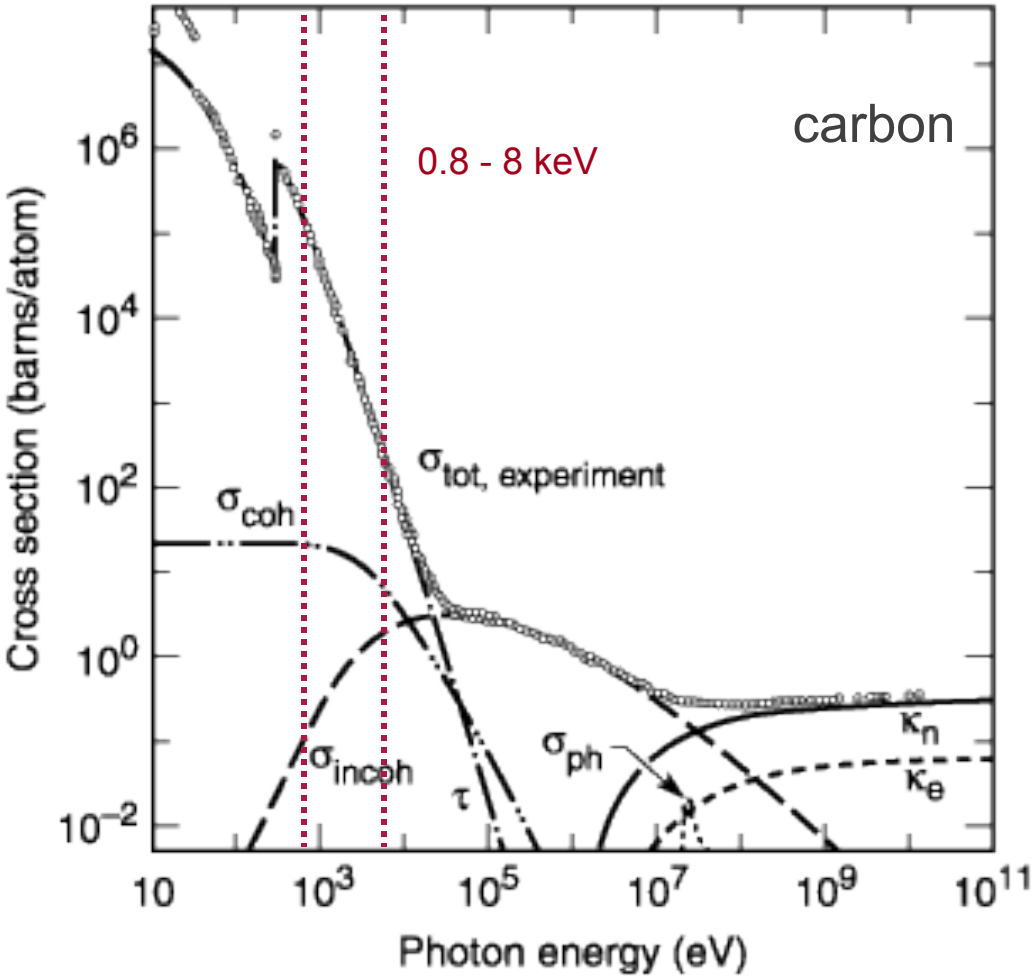


Hollow atom yield
@ LCLS ~10%
@ synchrotron ~0.3%
due to electron correlation

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	

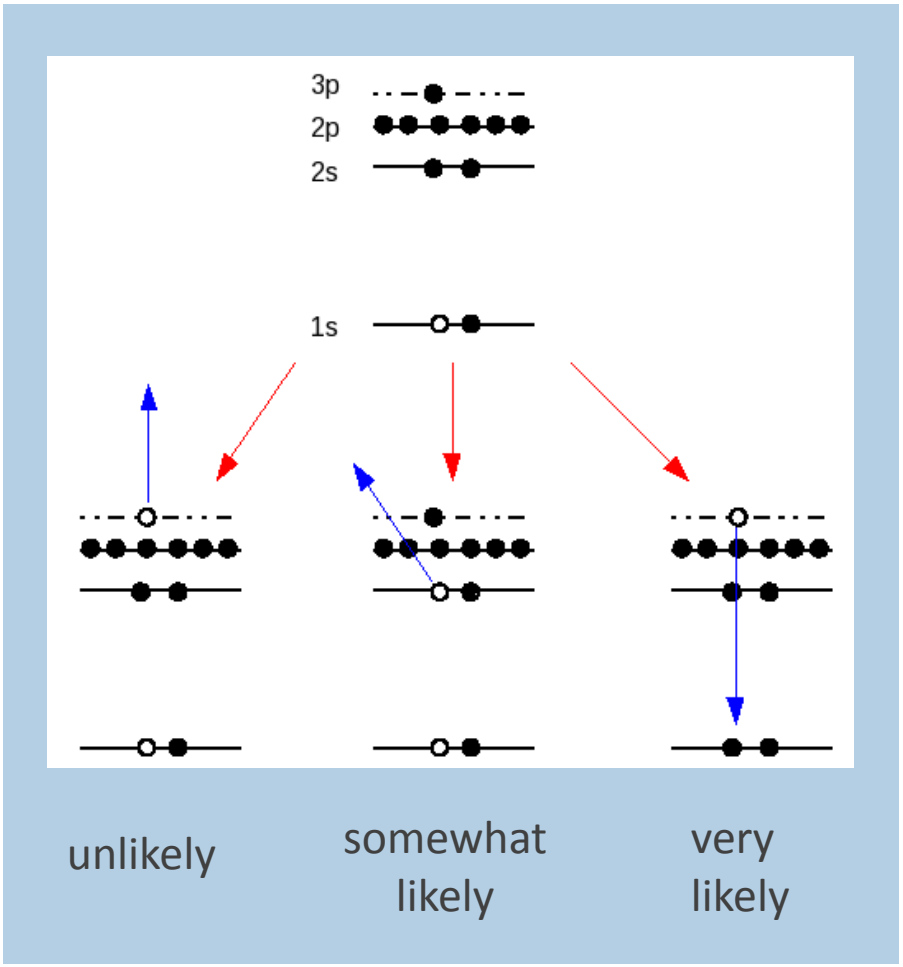
Impact of hollow atom formation on coherent x-ray scattering
Sang-Kil Son, L Young, R Santra
Phys. Rev A. **83**, 033402 (2011)

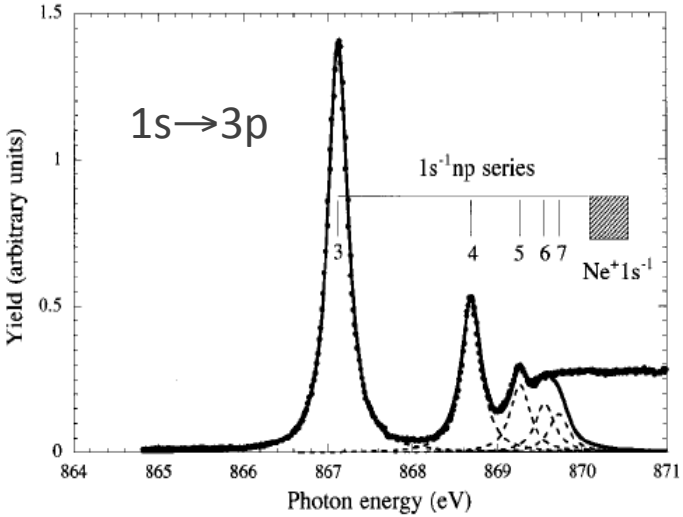
LCLS Experiment 5

Resonant x-ray processes at high intensity

Can we control inner-shell electron dynamics?

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





- Strong 1s→3p resonance

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

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

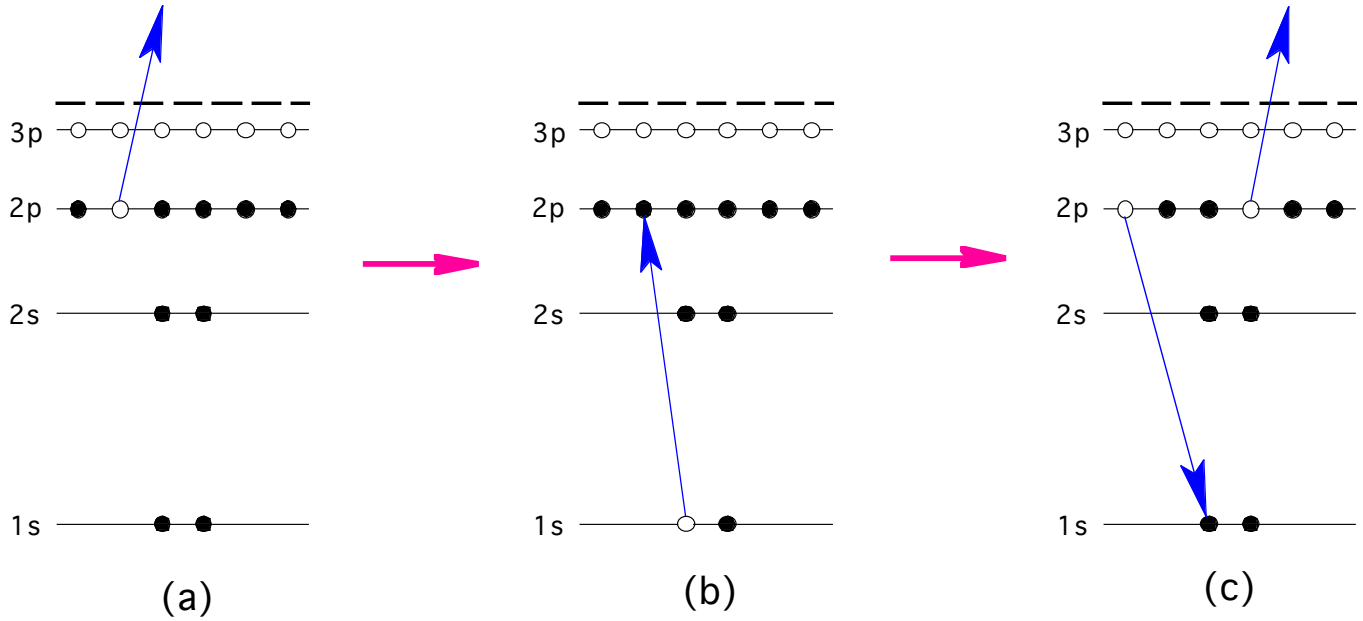
- Rabi flopping possible

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

  $I_{Ne} \sim 1.4 \times 10^{18}\ W/cm^2$

But LCLS linewidth ~ 8 eV!

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

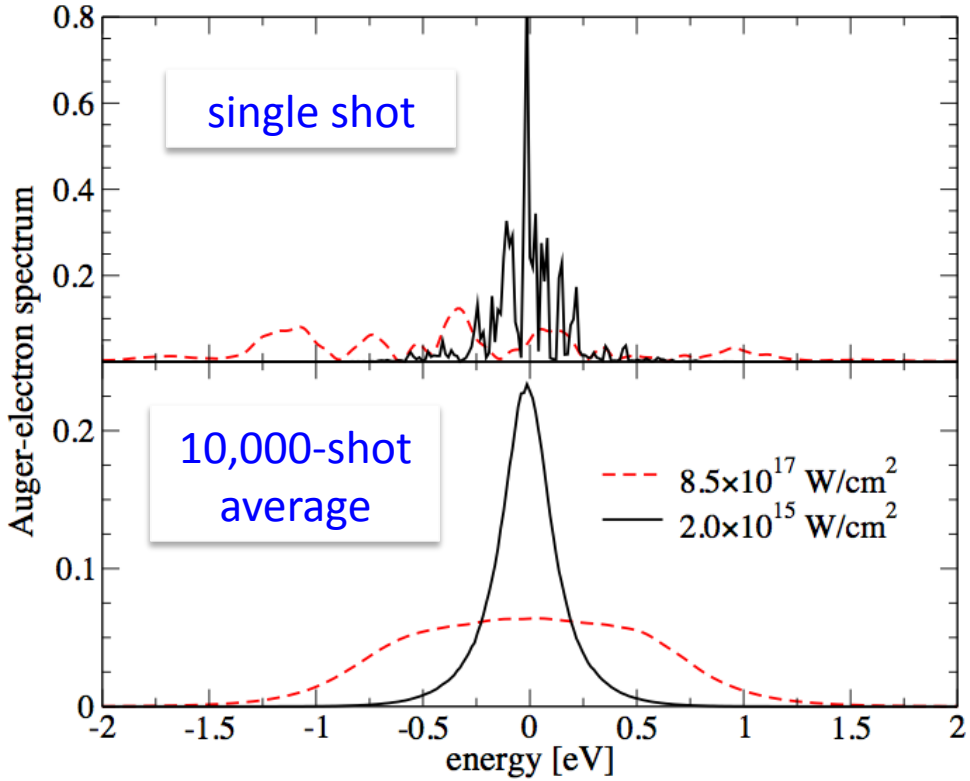


$E_{x\text{-}ray} = 848.6\ eV$

$\sigma_{1s\text{-}2p} = 500\sigma_{2p\text{-}\infty} = 30\ \sigma_{1s\text{-}3p}$

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”

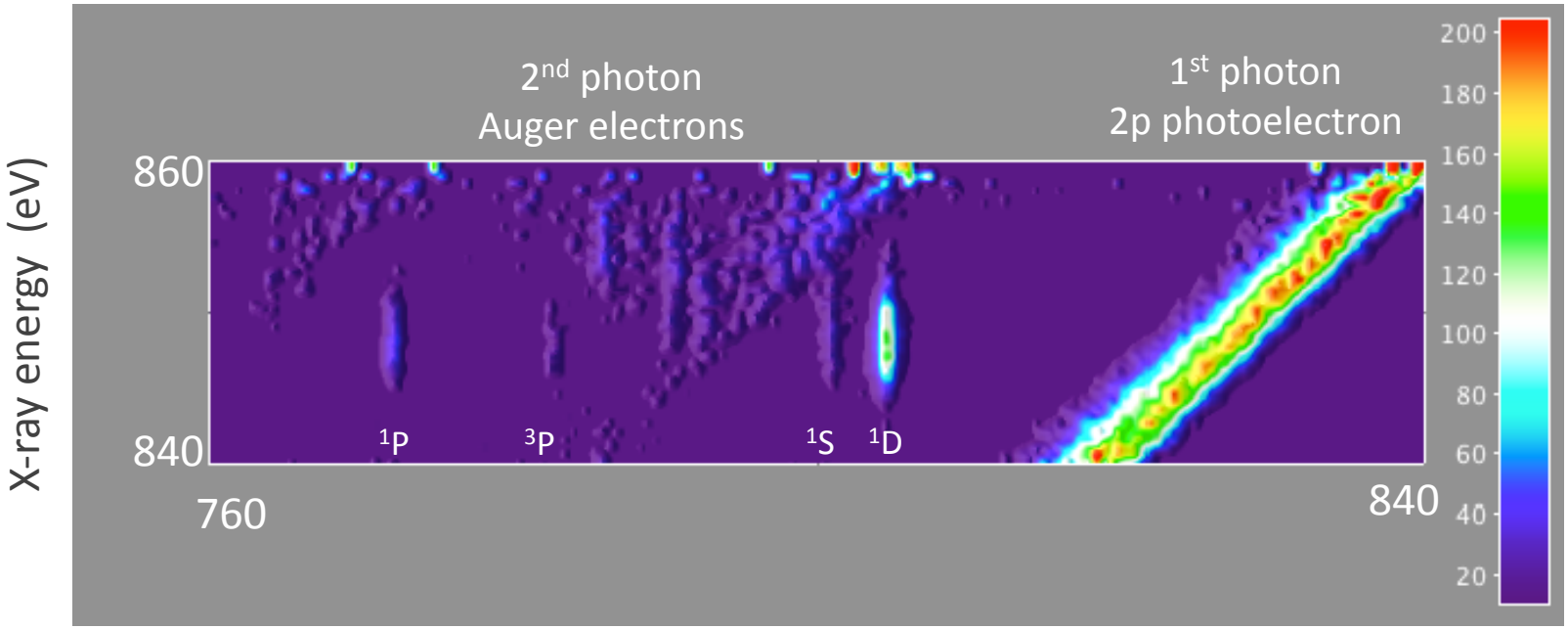


-> Look for Auger line broadening on resonance

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

Electron spectra vs photon energy

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



Electron Kinetic Energy (eV)

"Hidden" resonance revealed in single SASE pulse
Preliminary from Bertold Krässig

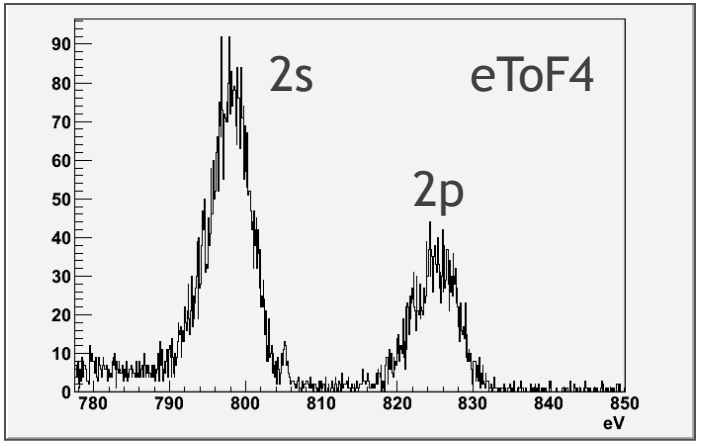
Characterization of X-ray SASE pulse

- Shot-to-shot photon energy jitter

  -Photoelectron energy spectra give x-ray energy centroid

  -Jitter derived from shot-to-shot GeV electron beam energy measurements

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

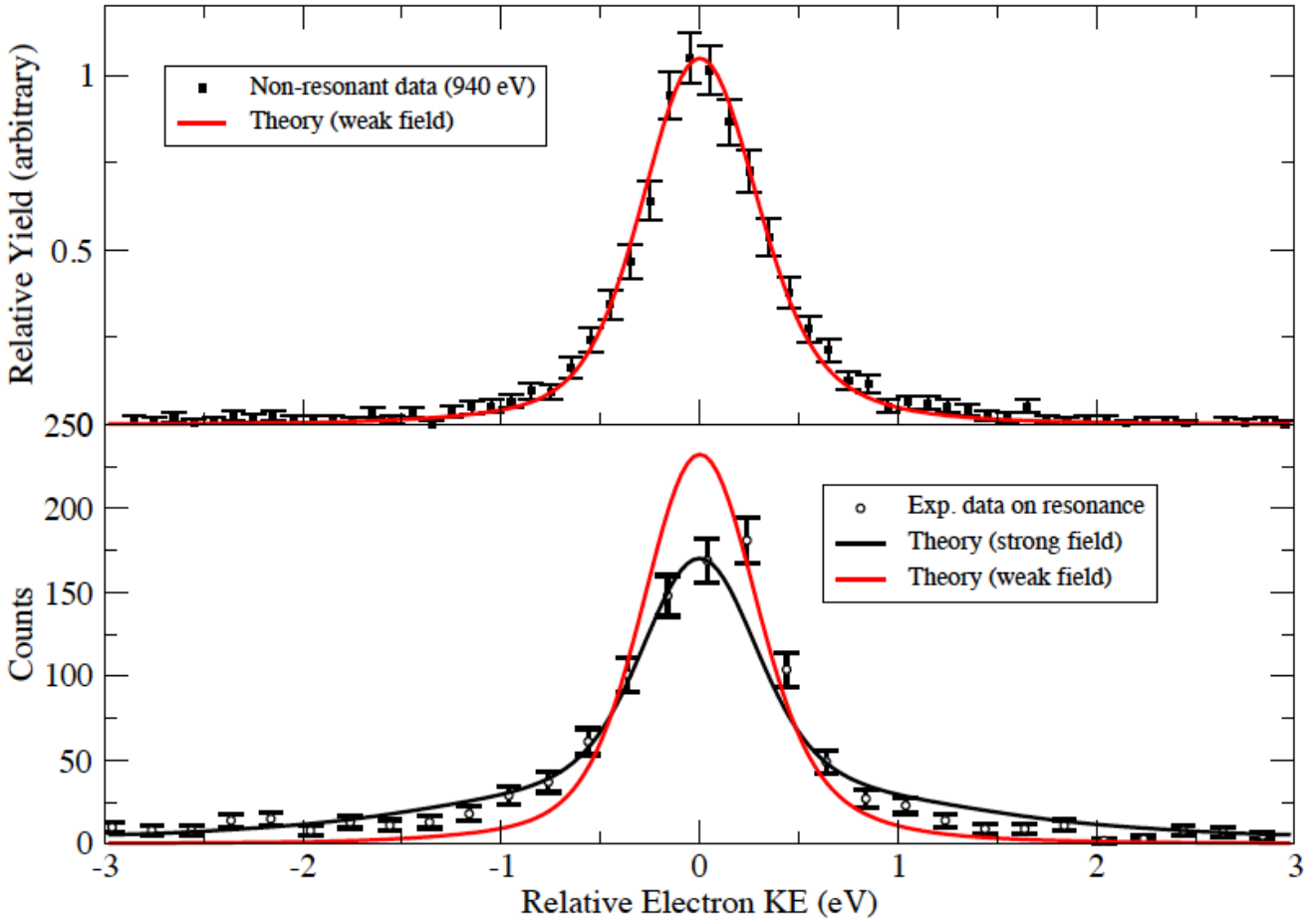


Electron kinetic energy (eV)

- Intrinsic x-ray bandwidth

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%

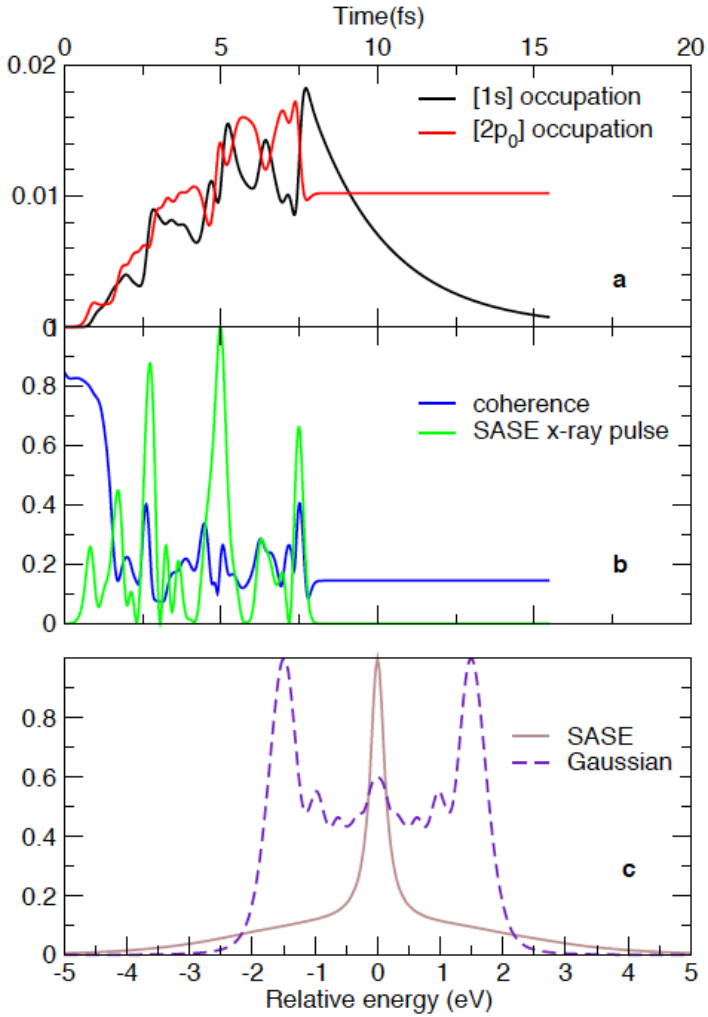
Is the ^1D Auger line broadened on 1s-2p resonance?



X-ray parameters
0.3 mJ, 8.5 fs, 2 μm^2
(20% transmission)

Theory from N. Rohringer and R. Santra
Preliminary Analysis – E. Kanter

Improved control of inner-shell dynamics with Gaussian pulse



~2 x 10^{17} W/cm^2

0.3 mJ, 8.5 fs, 2 μm^2

To be submitted
Kanter et al.
Calcs from N. Rohringer

Summary

- Insight into ultraintense x-ray interations
  - six-photon, ten-electron stripping of neon (~$10^{12}/\mu m^2$)
  - 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

- Hidden resonances are potential complications in using intense SASE pulses

- Intense x-rays can “control” inner-shell electron dynamics

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Argonne AMO group Oct 2009

Heroes at AMO Control

Heroes at MCC

Outlook/Wider Impact/Final Thoughts

The chaotic truth about current XFELs

European XFEL

Self-seeding techniques and their importance for XFELs

SASE pulses, baseline mode of operation: poor longitudinal coherence

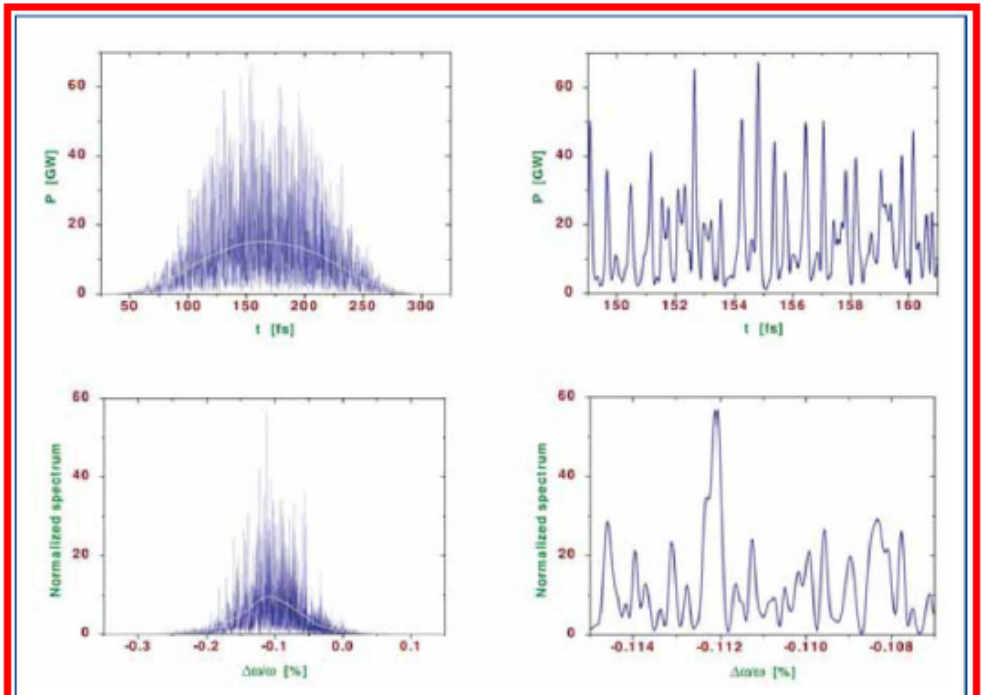


Figure 5.2.4 *Temporal (top) and spectral (bottom) structure for 12.4 keV XFEL radiation from SASE 1. Smooth lines indicate averaged profiles. Right side plots show enlarged view of the left plots. The magnetic undulator length is 130 m.*

Source: The European XFEL TDR – DESY 2006-097 (2006)

$$\frac{\Delta\omega}{\omega} \sim 2\rho \sim 10^{-3}$$

$$\left(\frac{\Delta\omega}{\omega}\right)_{spike} \sim \frac{1}{\sigma_T \omega} \sim 10^{-5}$$

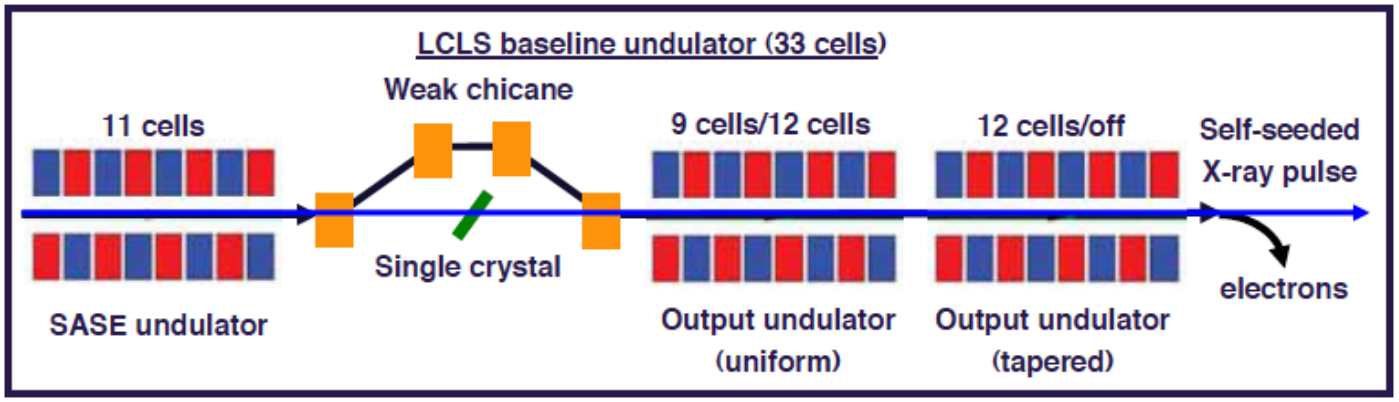
- Hundreds of longitudinal modes
- A lot of room for improvement
- Self-seeding schemes answer the call for increasing longitudinal coherence

How to obtain longitudinally coherent x-ray laser pulses?

European XFEL

Feasibility study for LCLS (I)

10



100 GW-level
Fully-coherent
Self-seeded
X-ray pulse

$4 \cdot 10^{11}$ photons
5 fs (FWHM)
at 0.15 nm

low charge
(0.02 nC) mode of
operation

Single mode hard X-ray FEL

Parameters for the low-charge mode of operation at LCLS

	Units	
Undulator period	mm	30
K parameter (rms)	-	2.466
Wavelength	nm	0.15
Energy	GeV	13.6
Charge	nC	0.02
Bunch length (rms)	μm	1
Normalized emittance	mm mrad	0.4
Energy spread	MeV	1.5

- 4m-long magnetic chicane
- R_{56}=12μm

Implementation in progress – SLAC/Argonne

LETTER

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Femtosecond X-ray protein nanocrystallography

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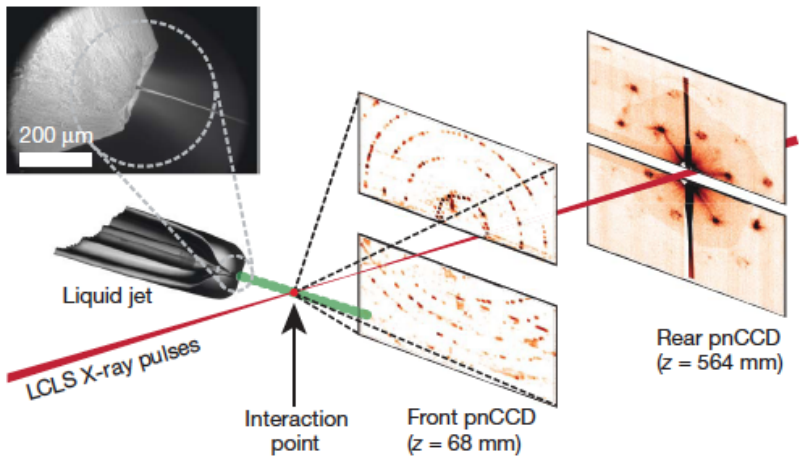

Photosystem I

membrane protein, 1 MDa

3,000,000 diffraction patterns

Crystals 200 nm - 2µm

Resolution 8.5Å

Match synchrotron structure

Dose: 700MGy

Typical synch dose: 30 MGy

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Single mimivirus particles intercepted and imaged with an X-ray laser

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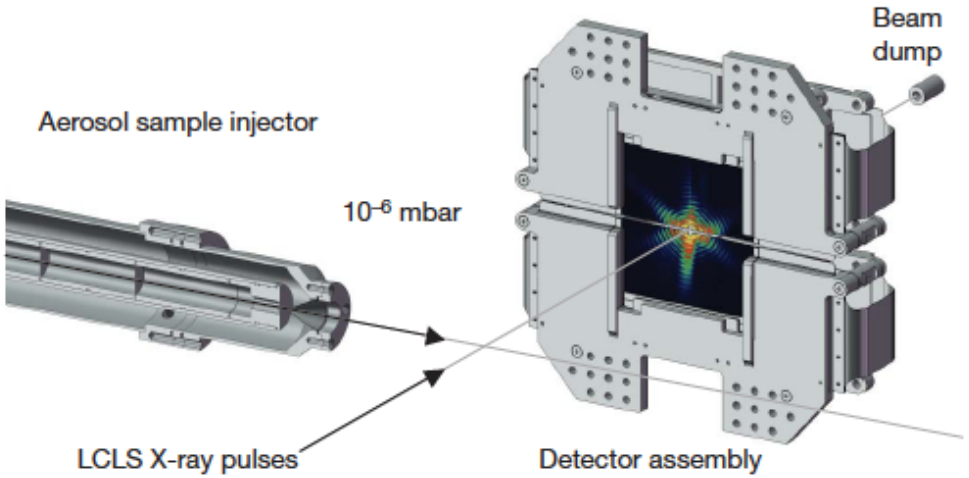

Mimivirus

-Largest known virus – 0.75 µm

-Does not crystallize

-Too large for 3D cryoelectron microscopy

Single Shot Scattering Pattern

32 nm resolution

Final thoughts

- XFELs open an entirely new intensity, wavelength regime for exploration
  - multiphoton effects are prominent
  - a complete contrast to any other x-ray source on earth
  - can create and probe exotic forms of matter

- XFEL research is in its infancy
  - Just learning how to use and control this source of radiation
  - LCLS is a superb opportunity to understand basics and explore novel ideas
  - Nanocrystallography appears to be an early success

- XFEL access is very limited
  - One beamline at LCLS currently
  - Beamlines at XFEL by 2015