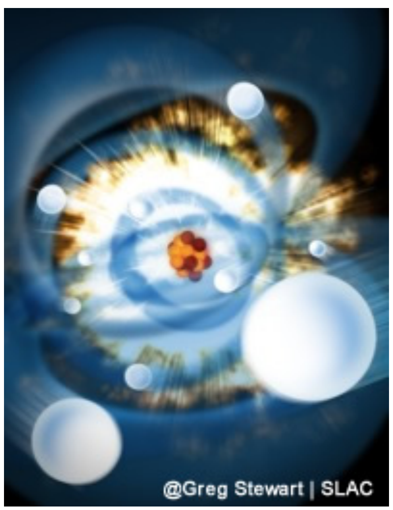
Harnessing ultraintense x-rays: from atomic response to applications



Linda Young
Nobel Symposium 158
Sigtunahöjden, Sigtuna, Sweden
14 Jun 2015

Compare ultraintense optical and x-ray sources

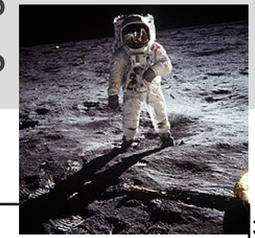
Hign-intensity at optical wavelengths
- high harmonic generation
- tabletop coherent x-ray radiation
- attosecond pulses

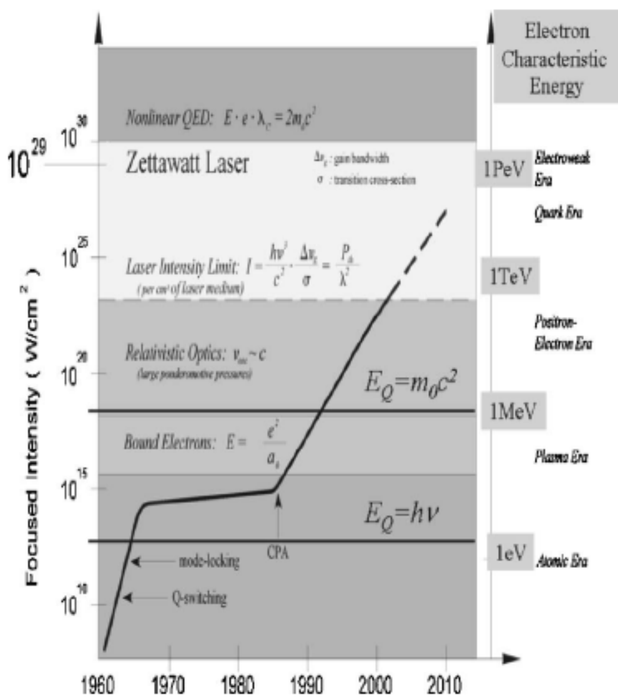
High-intensity at x-ray wavelengths

?

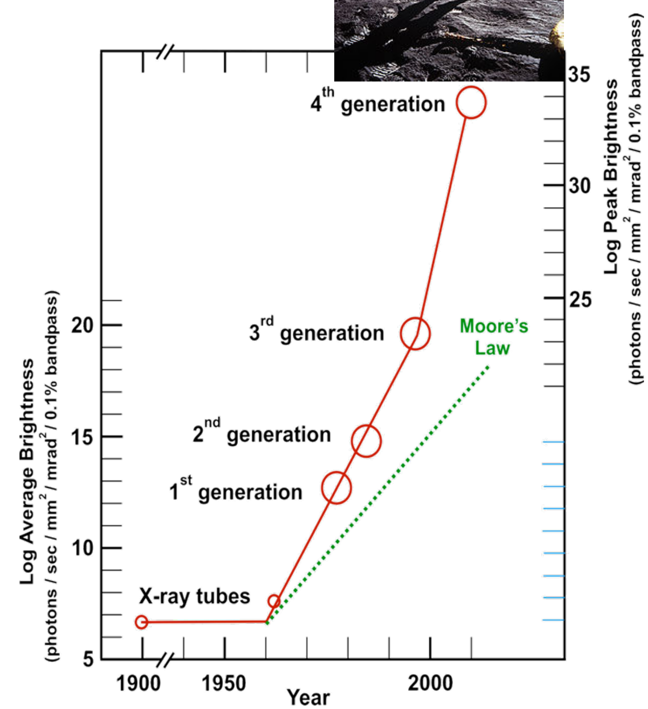
?

?



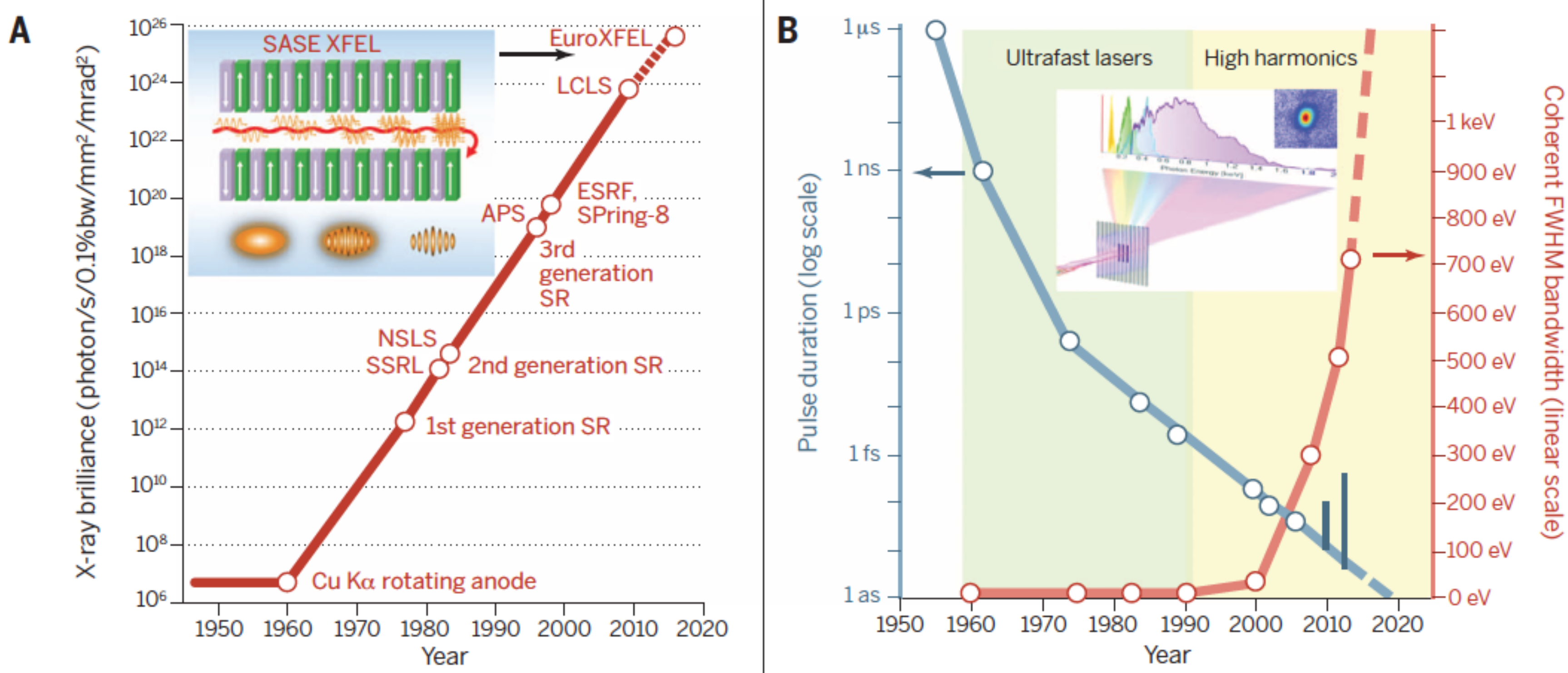


G. Mourou RMP 2006



D. Moncton, George Brown

X-ray sources: accelerator-based vs laser-based HHG



10^{13} x-rays/pulse/1% BW @ 1 keV
~100 fs

10^{5} x-rays/pulse/1% BW @ 1 keV
~1 fs

XFELs 10^{8} "brighter" than HHG sources

From: Miao, Ishikawa, Robinson, Murnane, Science **348**, 530 (2015)

Outline

- Birth of world's first hard x-ray FEL - LCLS

- Non-resonant high intensity x-ray phenomena

  LCLS Experiment 1: Oct 1 - 6, 2009

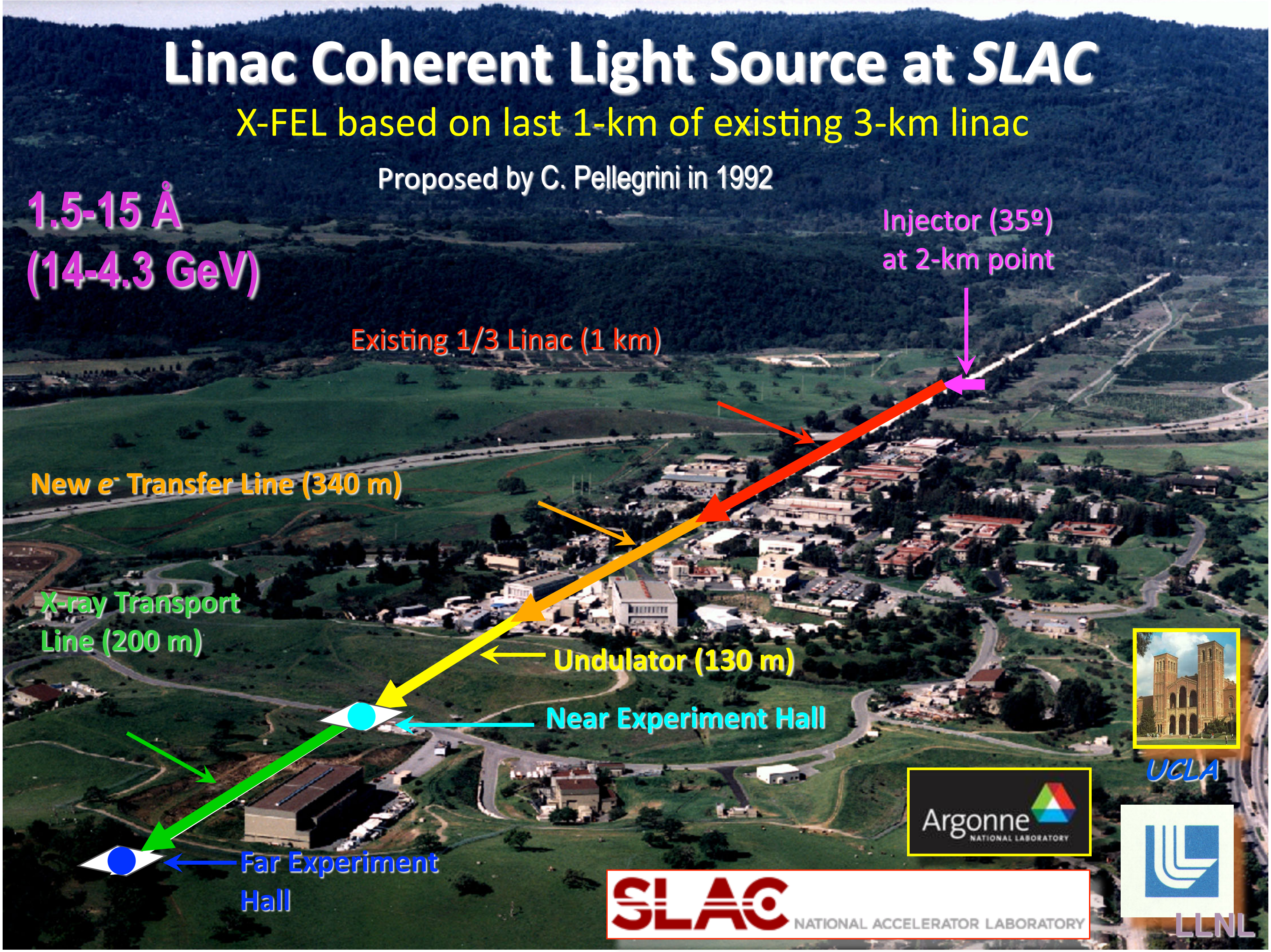
  L. Young *et al.*, Nature 466, 56 (2010)

- Resonant high intensity x-ray processes

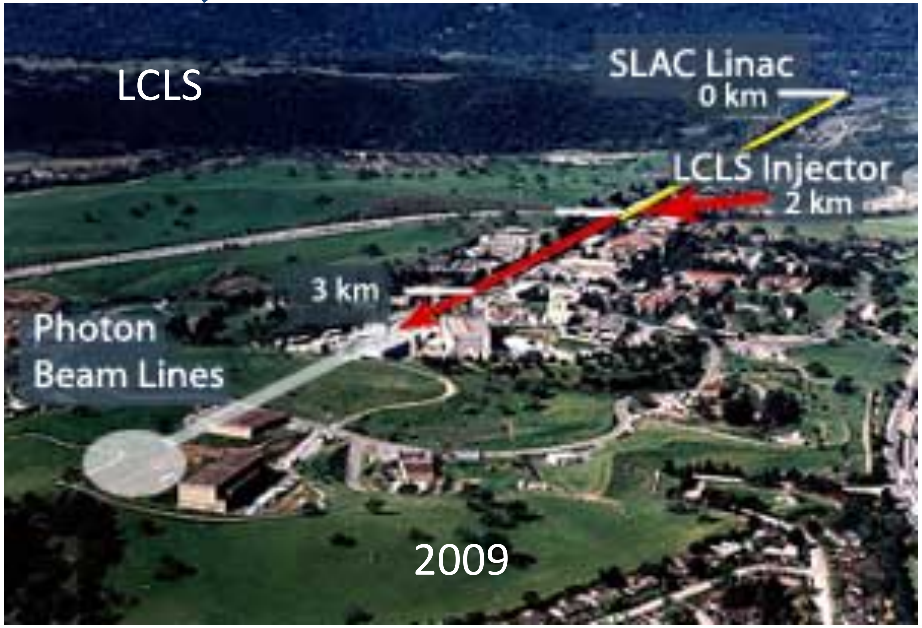
  LCLS Experiment 5: Oct 1 - 6, 2009

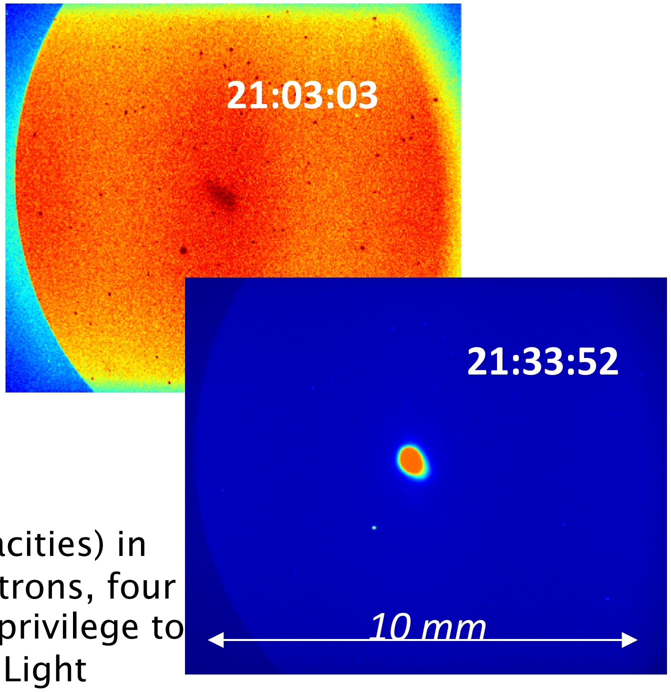
  E.P. Kanter *et al.*, PRL 107, 233001 (2011)

- Towards single particle imaging


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
UCLA
Argonne
NATIONAL LABORATORY
SLAC
NATIONAL ACCELERATOR LABORATORY
LLNL

April 10, 2009: LCLS lases at 1.5 Angstroms





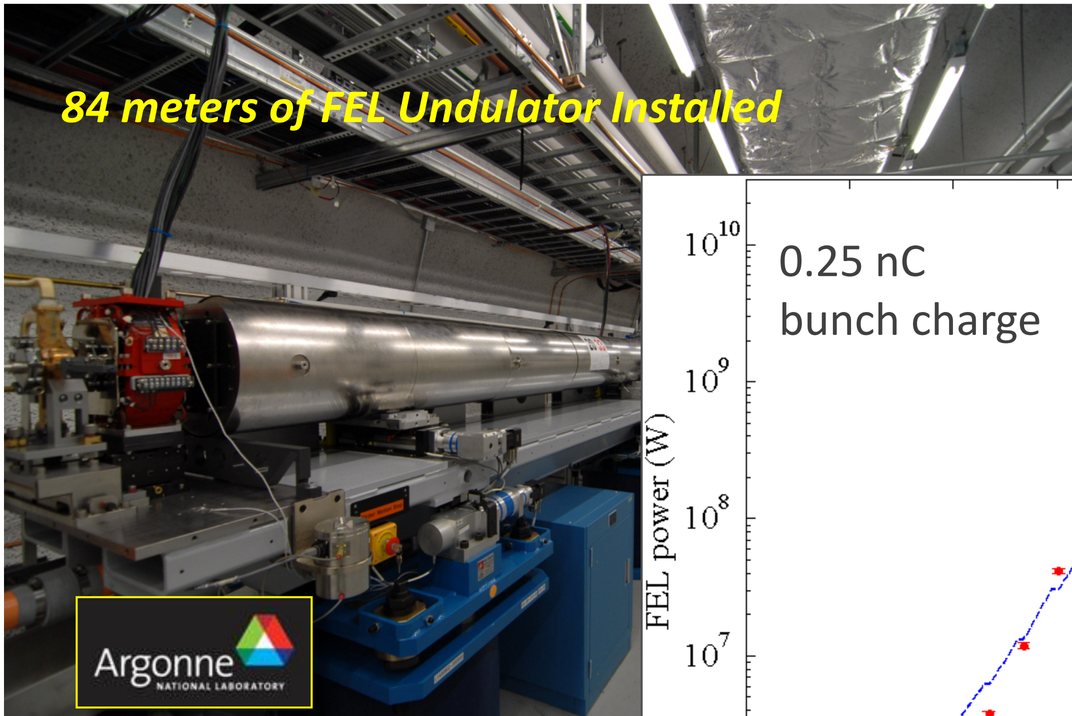
In my life before SLAC, I had the privilege to participate (in various capacities) in the design, construction and commissioning of two linacs, two synchrotrons, four storage rings and three FELs (free-electron lasers). Now I have had the privilege to be in SLAC's Main Control Center on April 10, when the Linac Coherent Light Source became a 1.5 Ångstrom laser. I don't expect I will ever, as long as I live, see such a beautiful, smooth turn-on of any light source. With each undulator placed on the beam path, the FEL power increased by a factor of about 2.3; two hours into the first attempt at lasing, the pinpoint of FEL light from twelve undulators was nearly 2,000-fold more intense than plain old undulator radiation. The team called it quits at 11:30 p.m. that night. When they returned at 8:00 a.m. the next morning, the FEL light came back as soon as the shutter was opened.

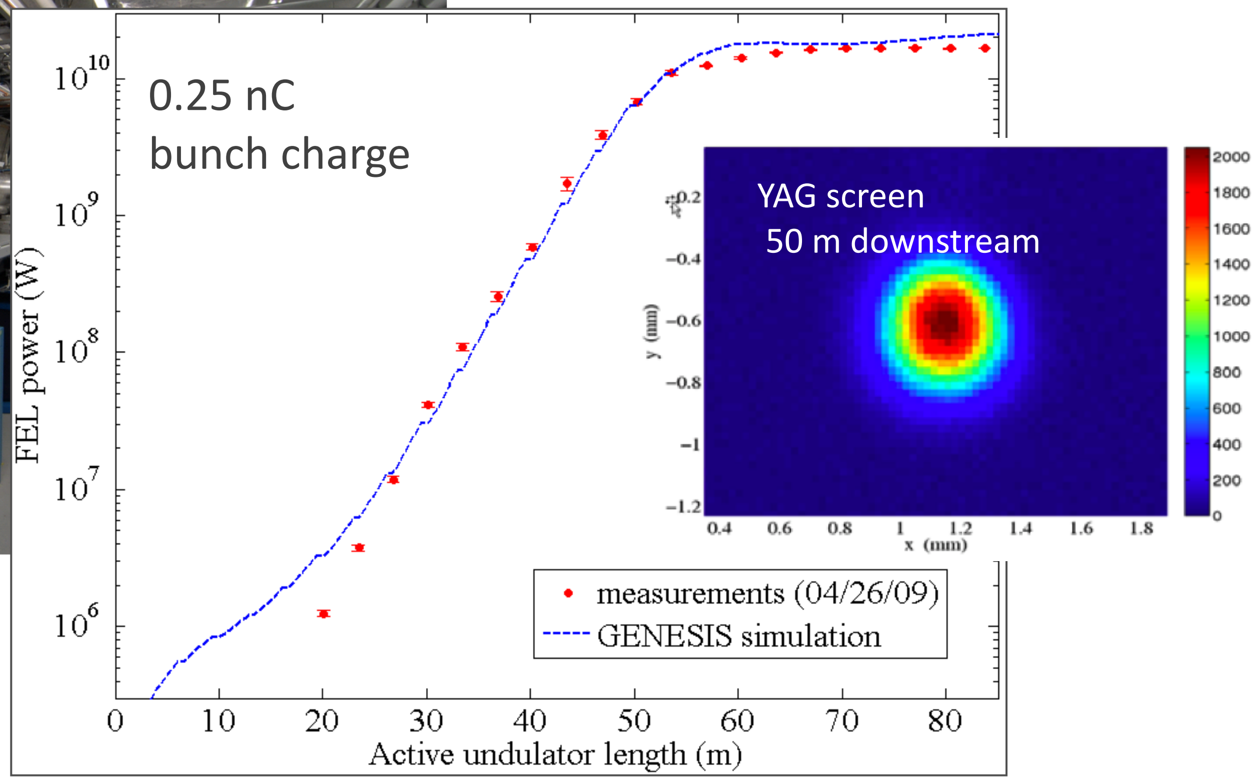
– John Galayda

From John Galayda

Yes I Do Smile on Occasion

LCLS saturation at 1.5 Å





- Saturation after ~65 meters of undulator!

Paul Emma PAC 2009 proceedings

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

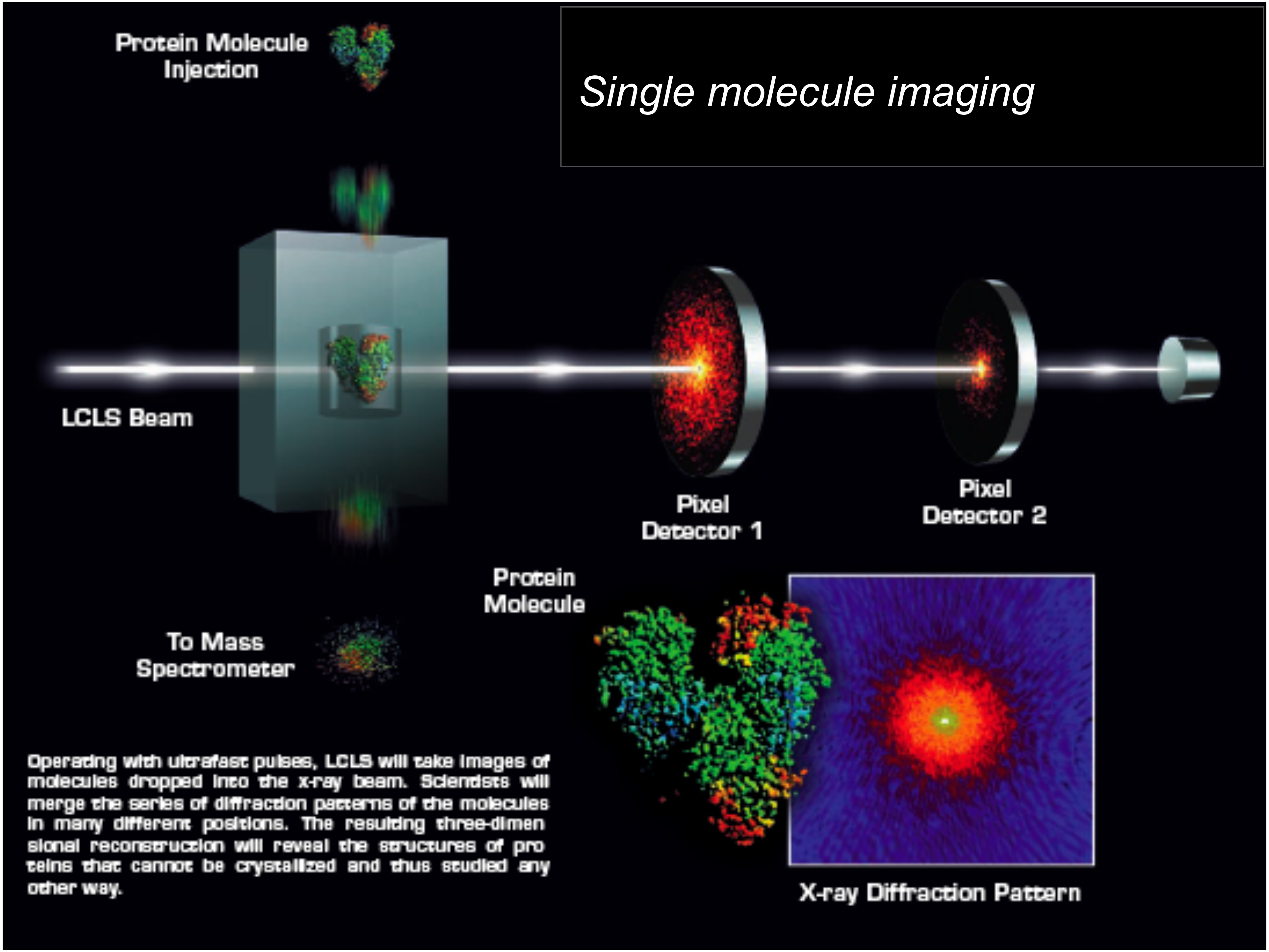
CXI: Coherent X-ray Imaging

MEC: Materials in Extreme Conditions

AMO

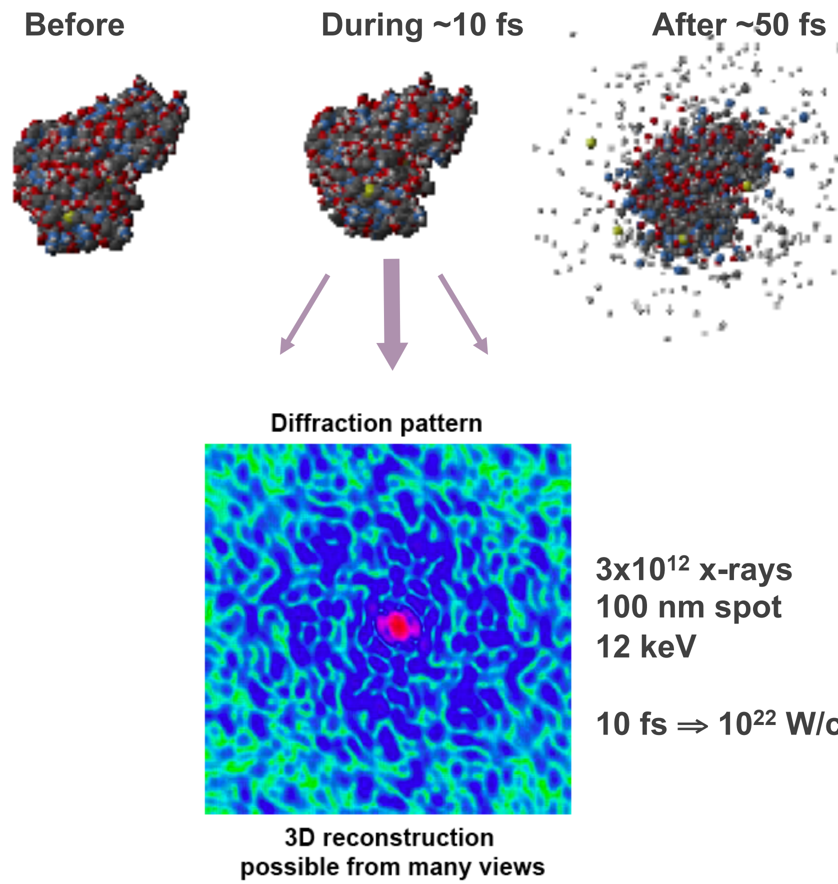
- Understand and control x-ray atom/molecule interactions at ultrahigh x-ray intensity as a foundation for other applications.
- Provide diagnostics of the LCLS radiation

Single molecule imaging



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



3x10^{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

Our approach to understanding ultraintense x-ray interactions

- Start with a well-characterized target

Binding energies in neutral neon

2p : ~21 eV

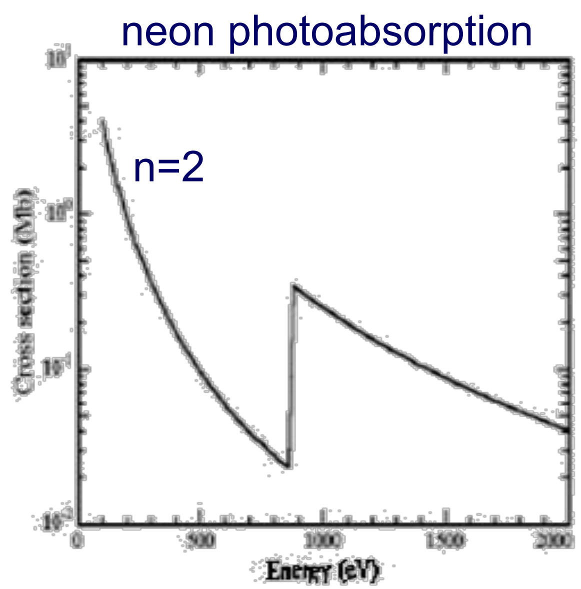
2s : ~48 eV

1s : ~870 eV

Inner-shell excitation

Auger yield 98%

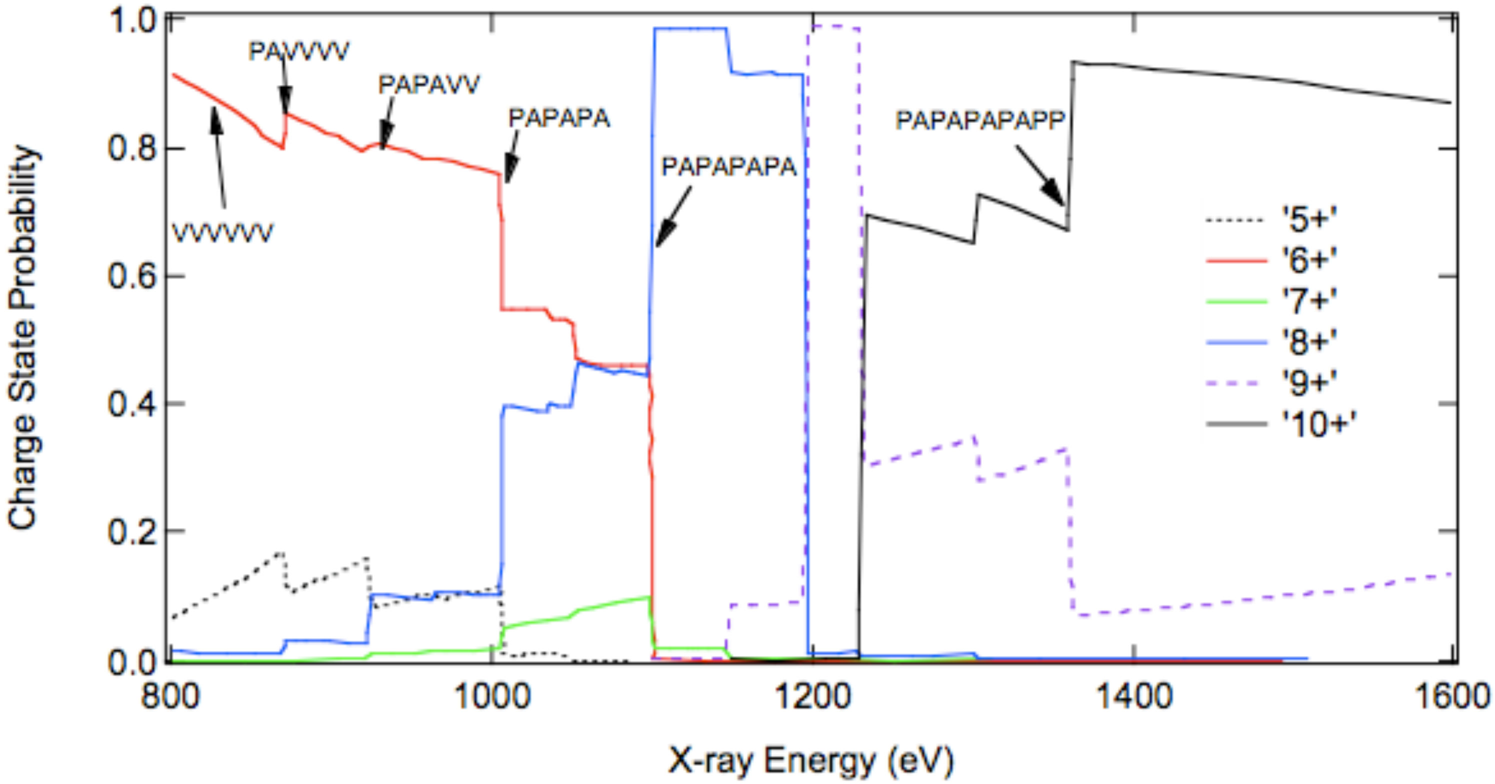
Auger clock - τ_{1s}: 2.4 fs



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

Guided by theory

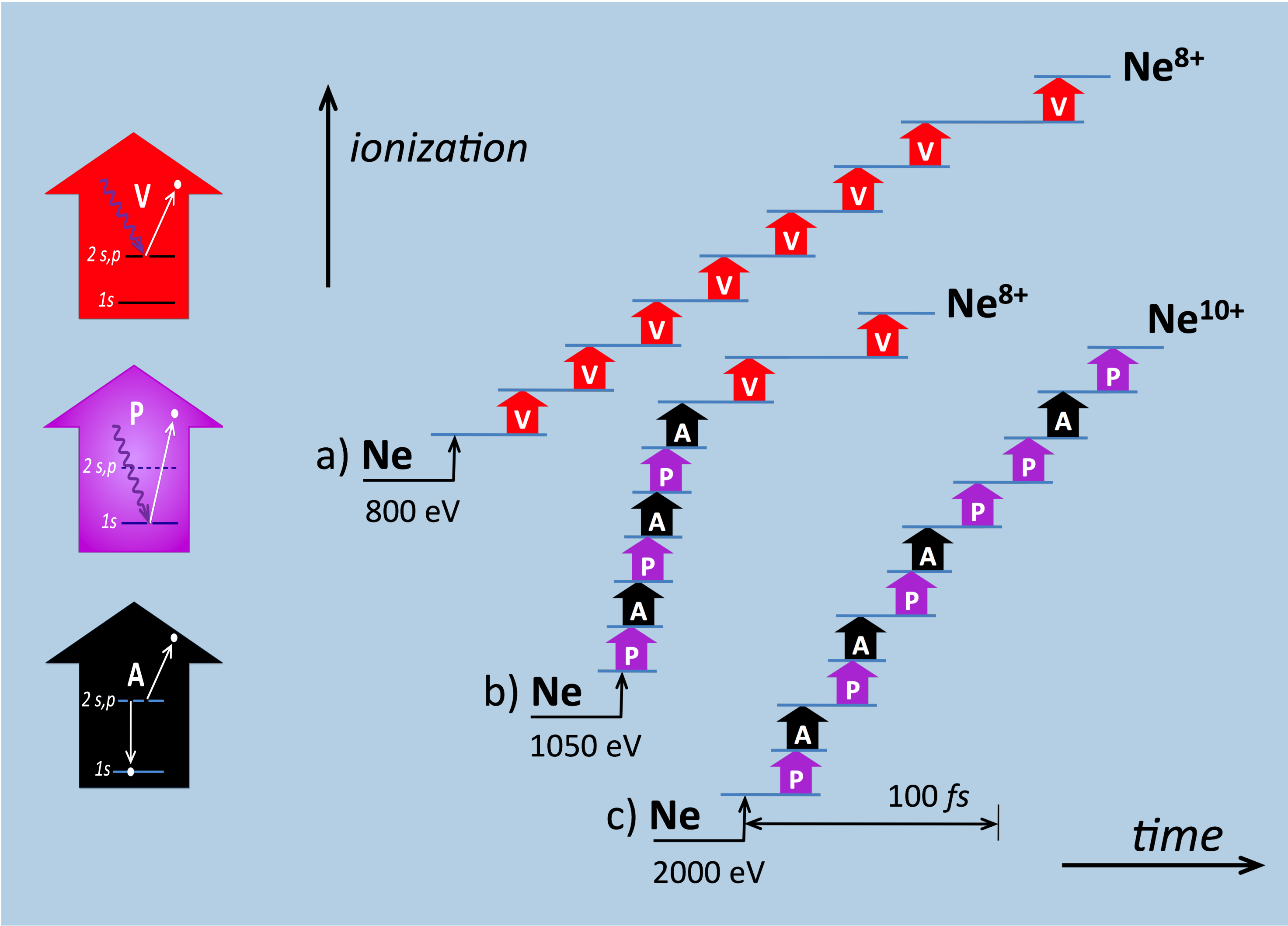
Theory: Rohringer & Santra, PRA 76, 033416 (2007)



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

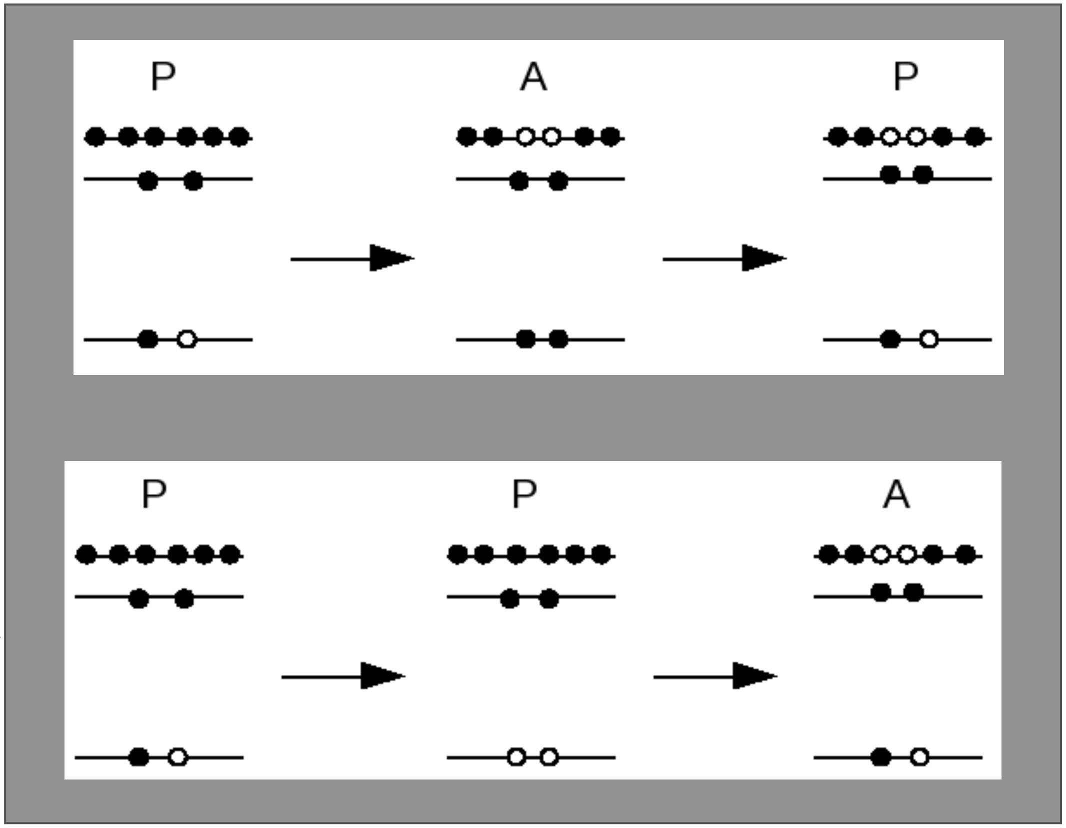


Sequential single photon processes dominate the interaction

How does one arrive at a particular charge state?

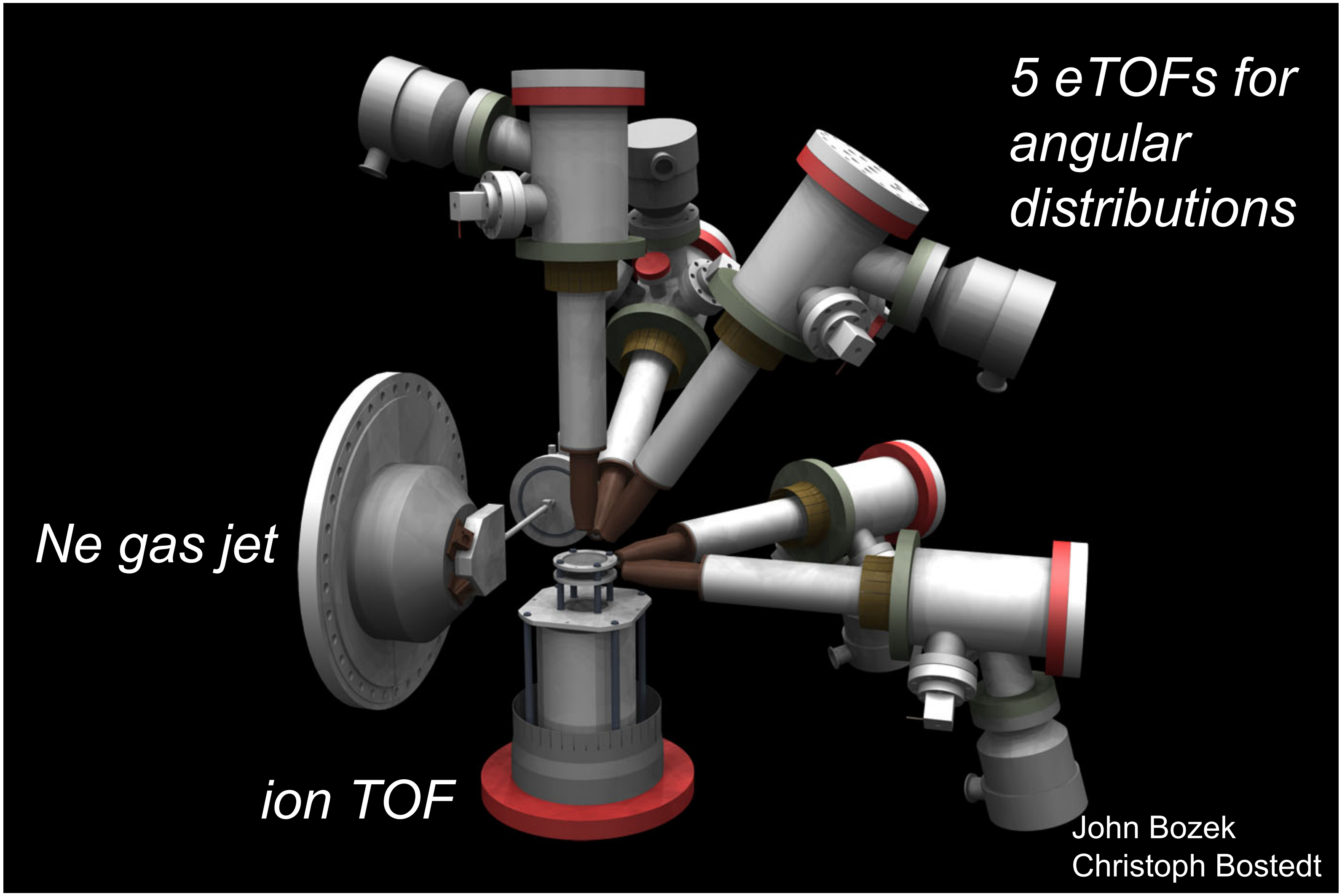
Low Intensity *PAP*

High Intensity *PPA*



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

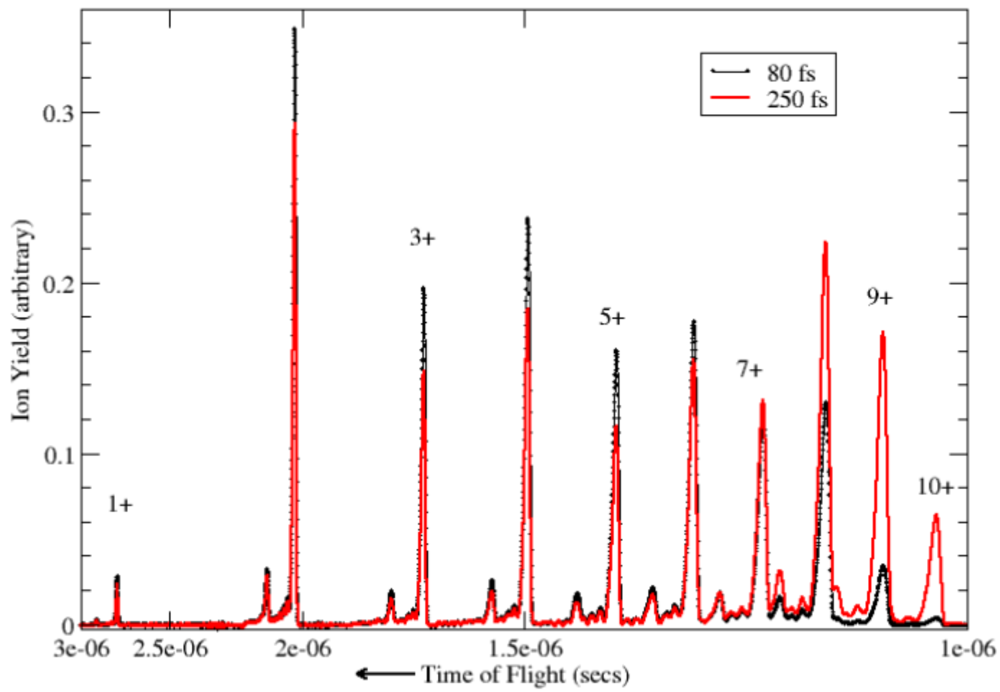
High field physics chamber



Day 1 – two interesting observations

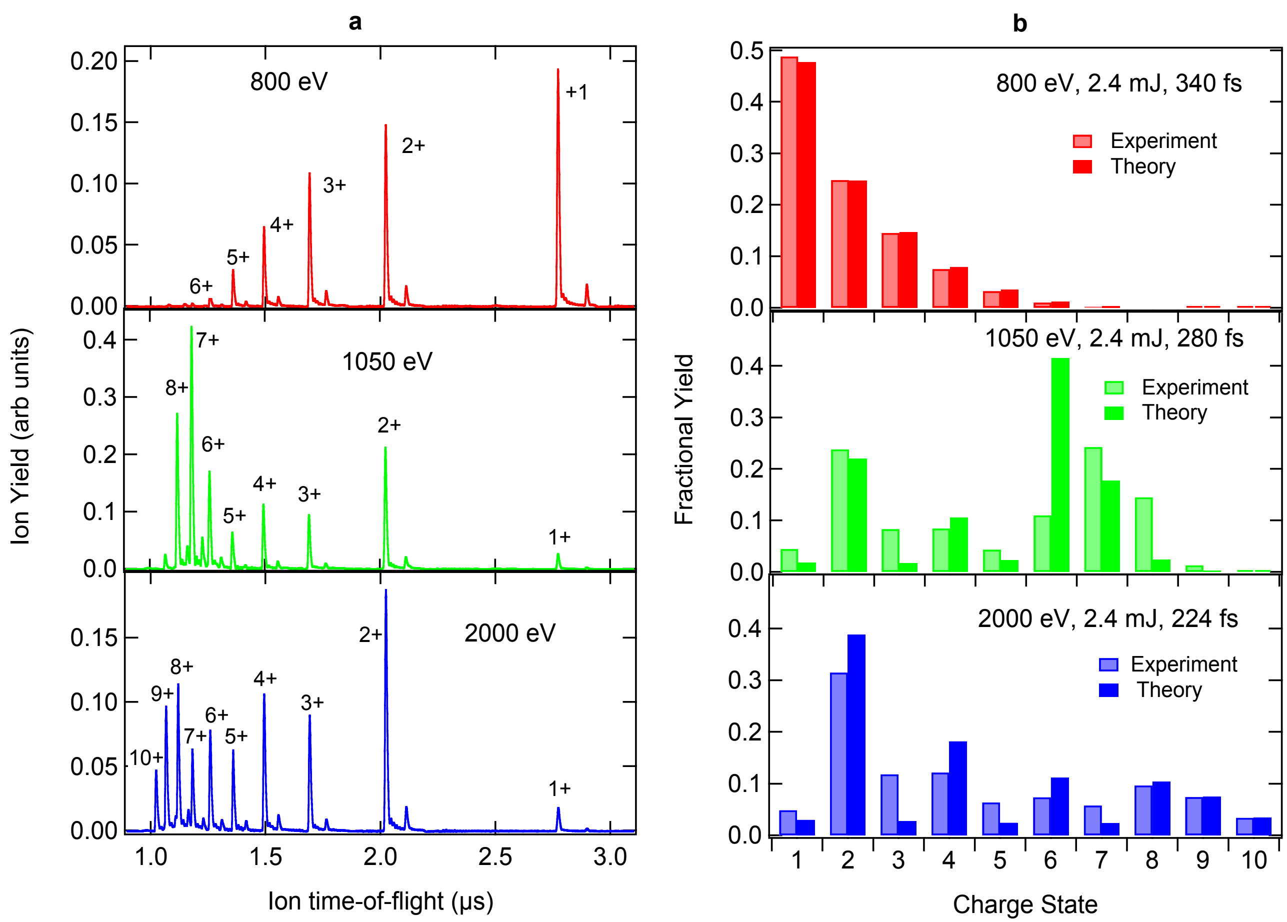
- Single ~100 fs pulse at 2000 eV fully strips neon

  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

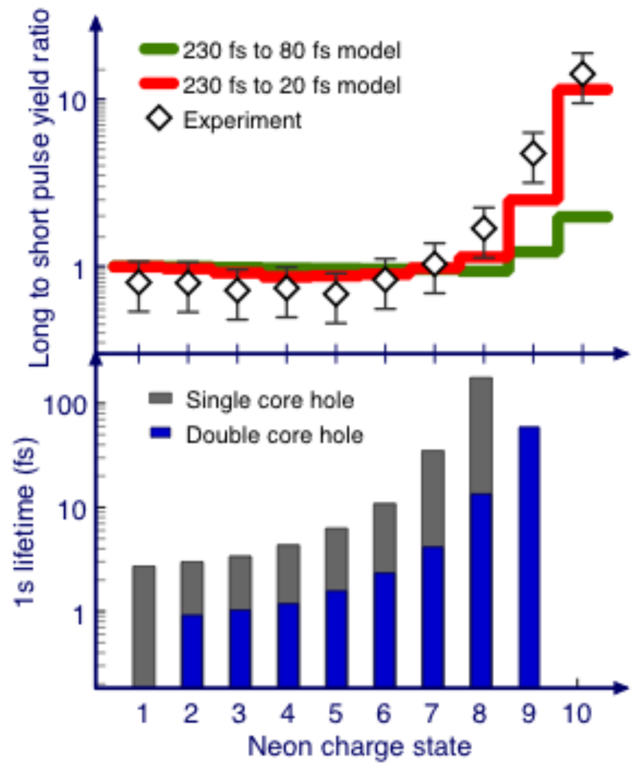


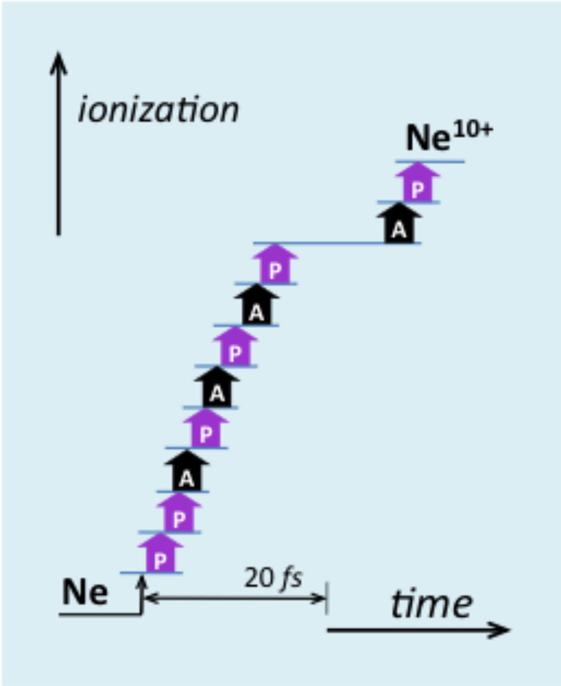
Theory
- Intensity averaged
- Fluence determined by experiment

Consistent with "measured" pulse energy and focus.

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

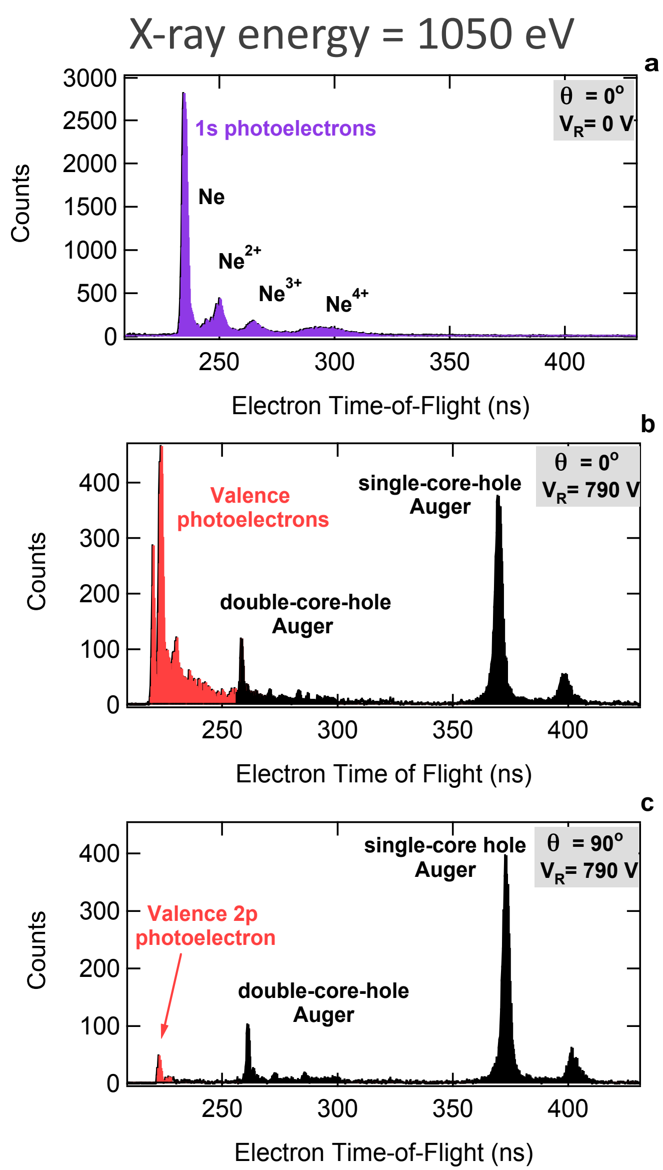
Atoms become transparent at high x-ray intensity !





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

Electron spectrometers track ionization mechanism



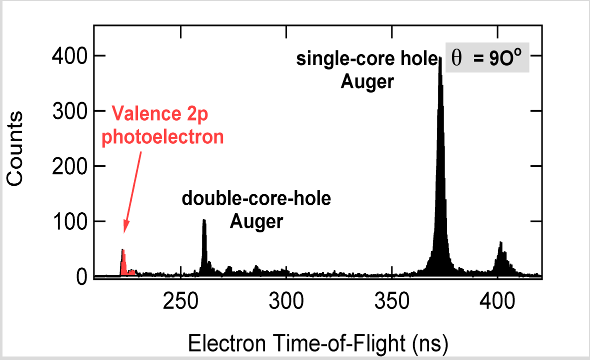
"Slow" 1s photoelectrons along x-ray polarization axis

"Fast" valence photoelectrons and Augers along polarization axis

Clean hollow atom signature double-core-hole Auger

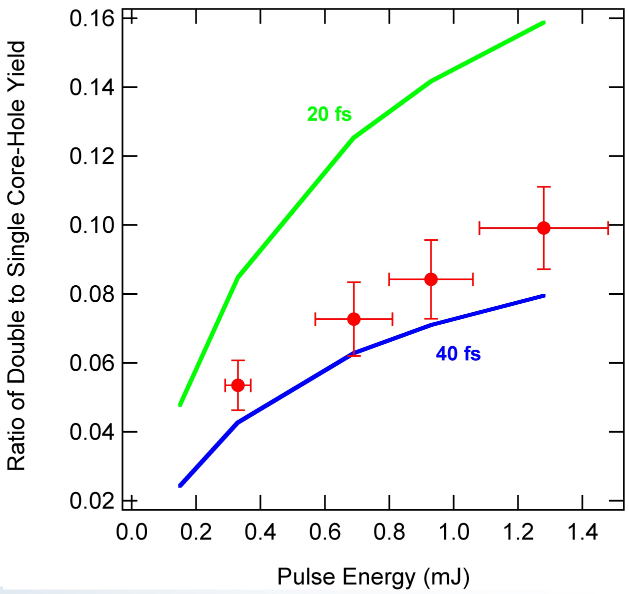
θ = 90°

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

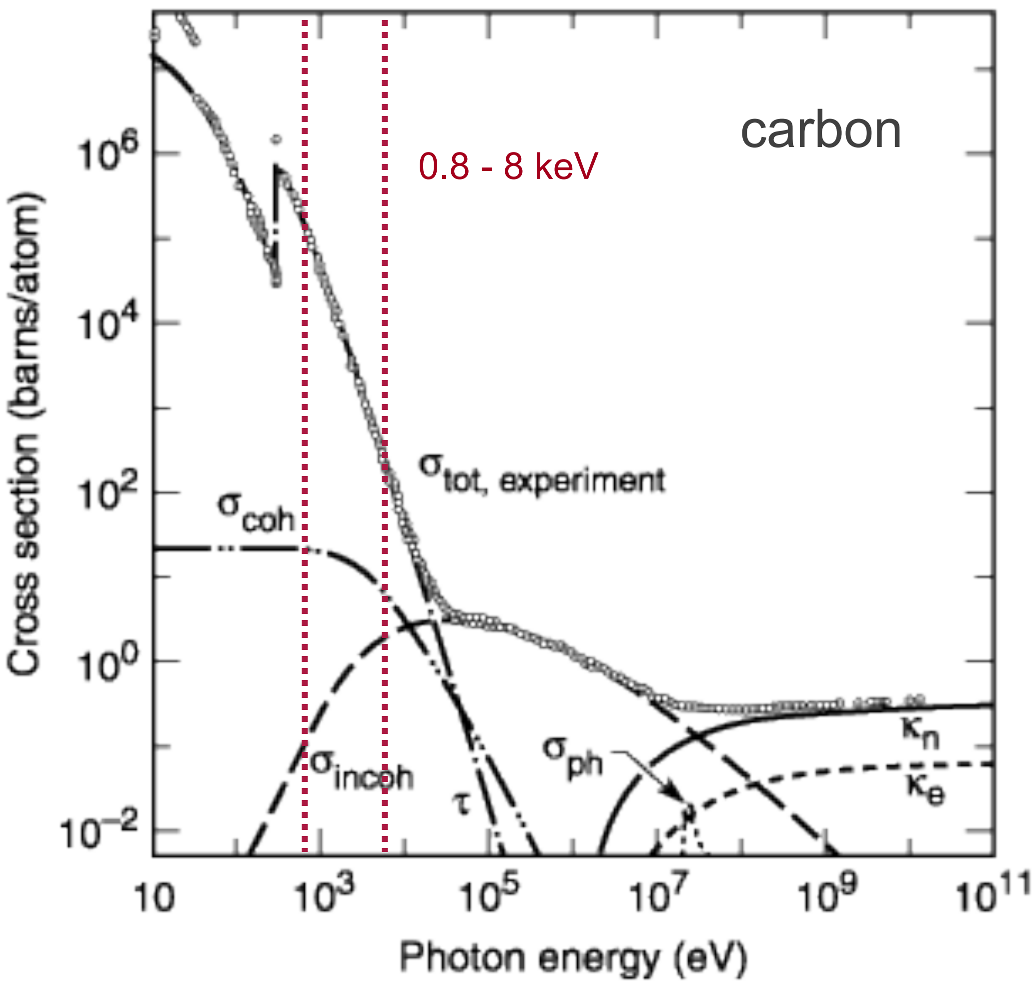


Hollow atom yield
@ LCLS ~10%
@ synchrotron ~0.3%
due to electron correlation

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	

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

Prescient prediction

"Furthermore, during short intense pulses numerous K-holes may be present at any one time, reducing the photoelectric cross-sections of atoms in which they were produced and thus lowering the total number of primary ionization events in the sample (see trend in Fig. 1). This effect makes the system radiation hardened to photo-ionization during very short exposures, and is more pronounced at higher radiation intensities."

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

Vol 466|1 July 2010 nature

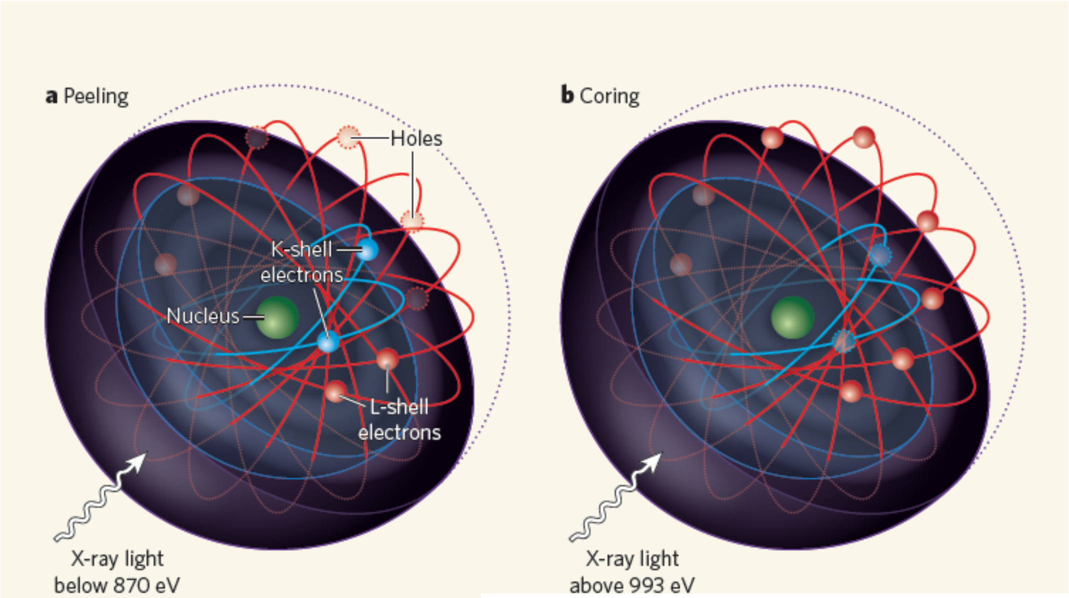
NEWS & VIEWS

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.



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

Nature **466**, 56 (2010).

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]

Summary: non-resonant ultraintense x-ray interactions

- Ultraintense x-ray interactions – multiphoton processes rule!
  - establish sequential single photon absorption as dominant ionization mechanism
    - fully stripped neon: six-photon, ten-electron (~$10^{12}/\mu m^2$)
  - multiple photon absorption probability high when fluence > $1/\sigma$
  - controlled electron stripping (outer v inner shells)

- X-ray induced transparency – a general phenomena
  - transient x-ray transparency caused by ejection of inner-shell electrons
  - induced transparency = frustrated absorption = core-level bleaching
    - molecules: Hoener *et al.*, PRL **104**, 253002 (2010)
    - clusters: Schorb *et al.*, PRL **108**, 233401 (2012)
    - solids: Yoneda *et al.*, Nat. Comm. (2014), Rackstraw *et al.*, PRL (2015)
  - implications for imaging: $\sigma_{scatt}/\sigma_{abs}$ is increased

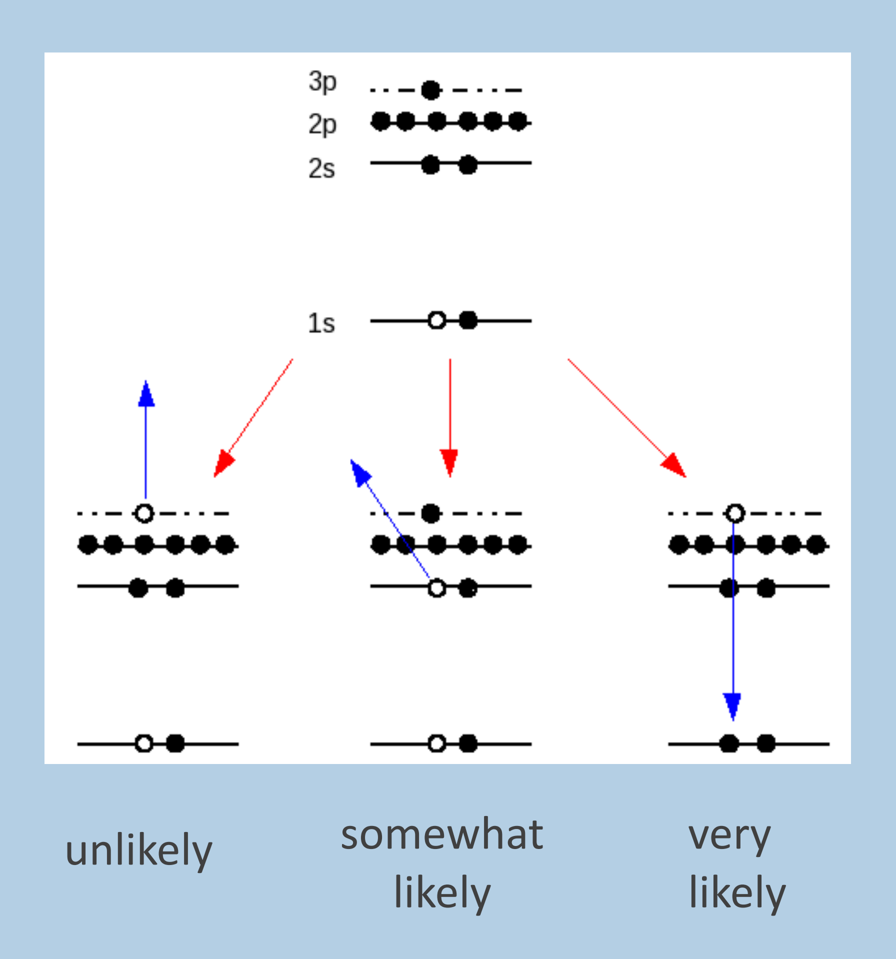
- Femtosecond time-scale atomic processes provide FEL diagnostics

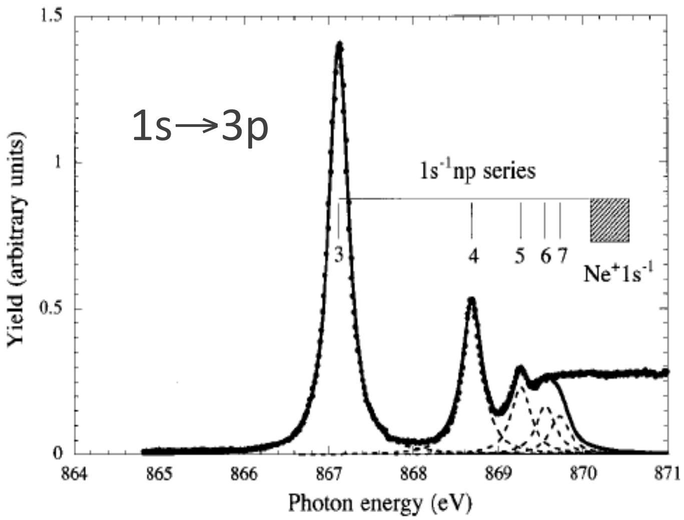
LCLS Experiment 5

Resonant x-ray processes at high intensity

Can we control inner-shell electron dynamics?

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





- Strong 1s→3p resonance

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

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

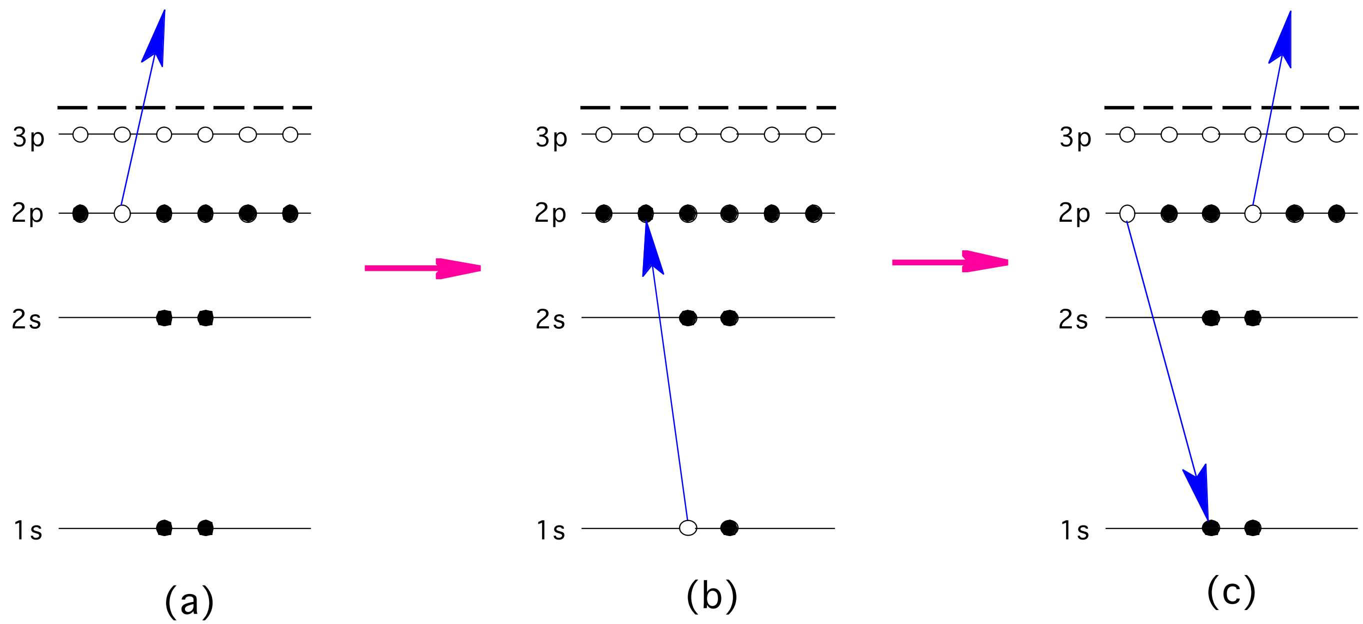
- Rabi flopping possible

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

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

But LCLS linewidth ~ 8 eV!

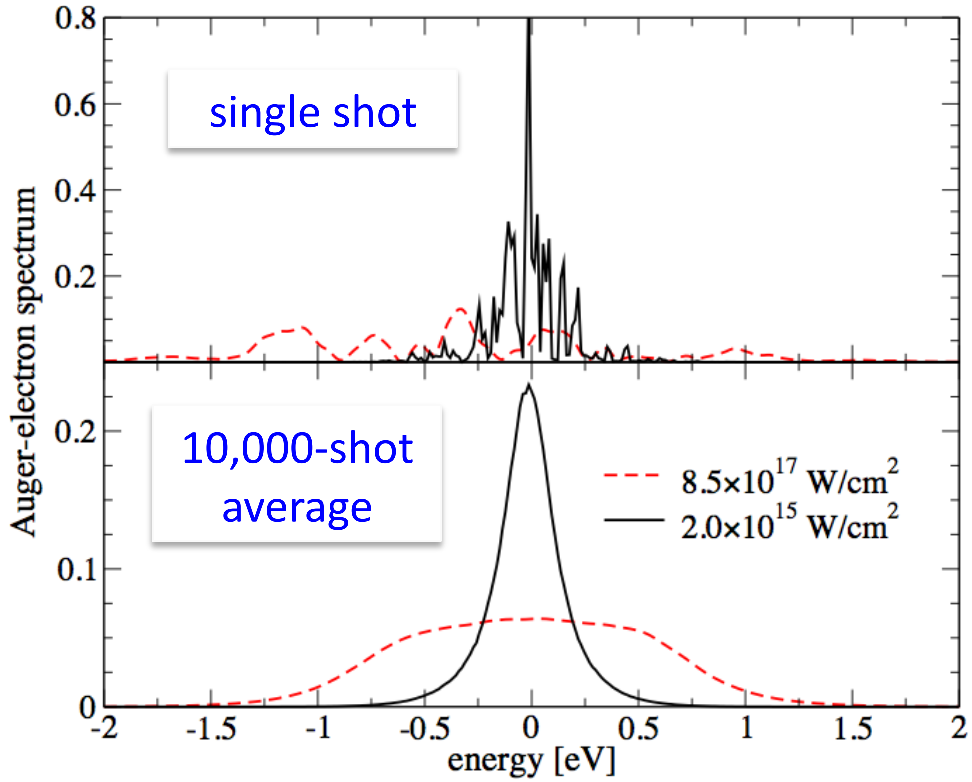
Rabi-flopping on 1s - 2p resonance more feasible



$E_{x\text{-}ray} = 848.6\ eV$
$\sigma_{1s\text{-}2p} = 500\sigma_{2p\text{-}\infty} = 30\ \sigma_{1s\text{-}3p}$

Observe Auger yield when x-rays scanned over 1s - 2p resonance.
Observe broadening at resonance to indicate Rabi flopping
Theory: Rohringer & Santra PRA (2008).

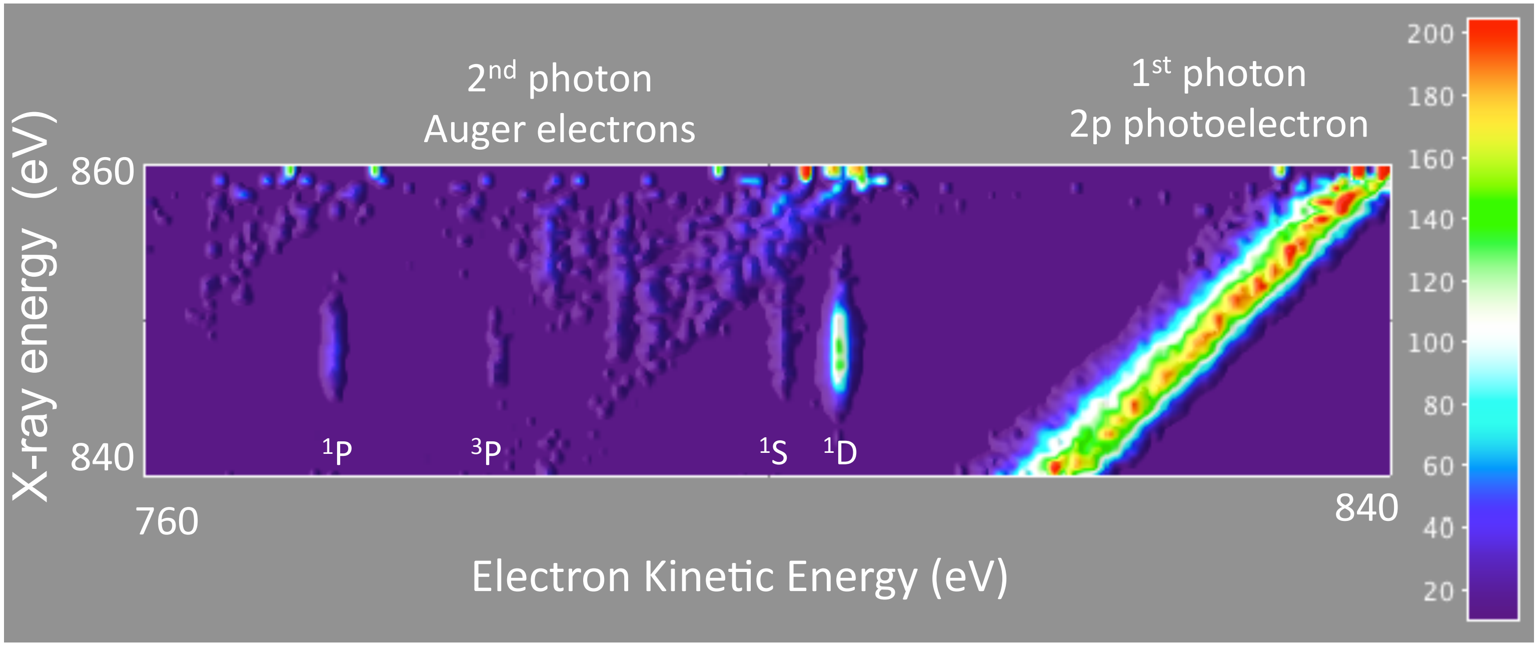
Calculated "Resonant Auger effect at high x-ray intensity"



-> Look for Auger line broadening on resonance

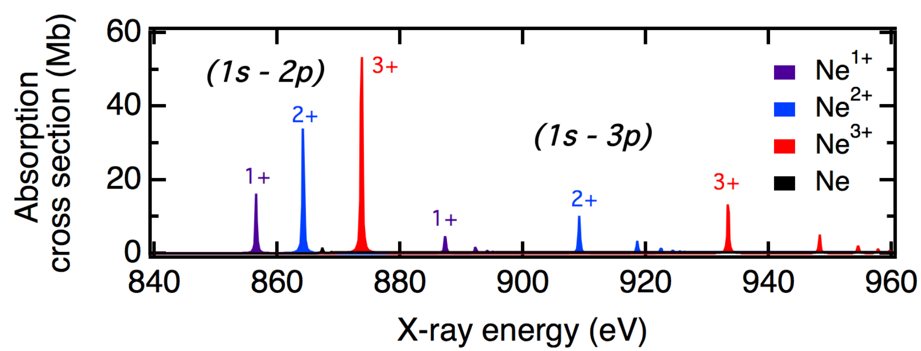
N. Rohringer & R. Santra, PRA 77, 053404 (2008)

Looking for Rabi flopping: unveiling and driving hidden resonances with LCLS pulses



- High fluence pulse alters target to reveal enormous “hidden” resonances ~1000x larger than background

- X-ray absorption spectrum changes rduring the fs duration pulse



E. P. Kanter et al., PRL (2011)

Characterization of SASE x-ray pulse required

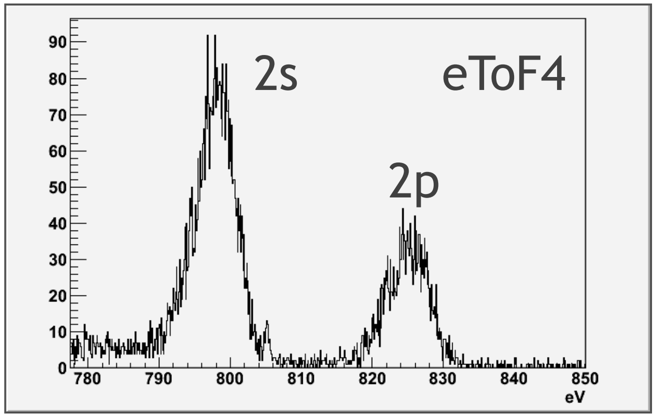
- Shot-to-shot photon energy jitter

  -Photoelectron energy spectra give x-ray energy centroid

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

  E_x (eV) = 44.25 $E_{e(}$(GeV)^2

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

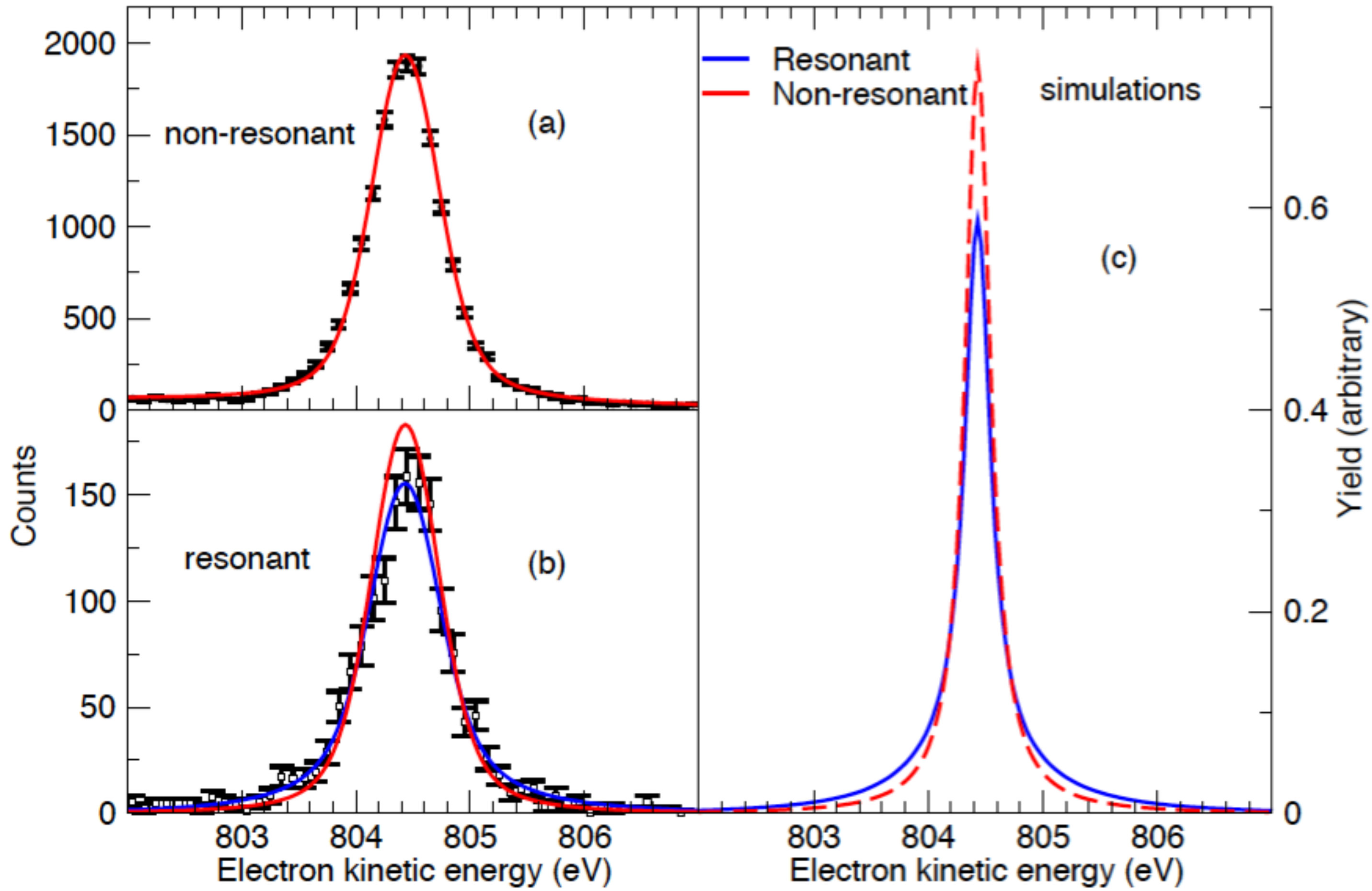


Electron kinetic energy (eV)

- Intrinsic x-ray bandwidth

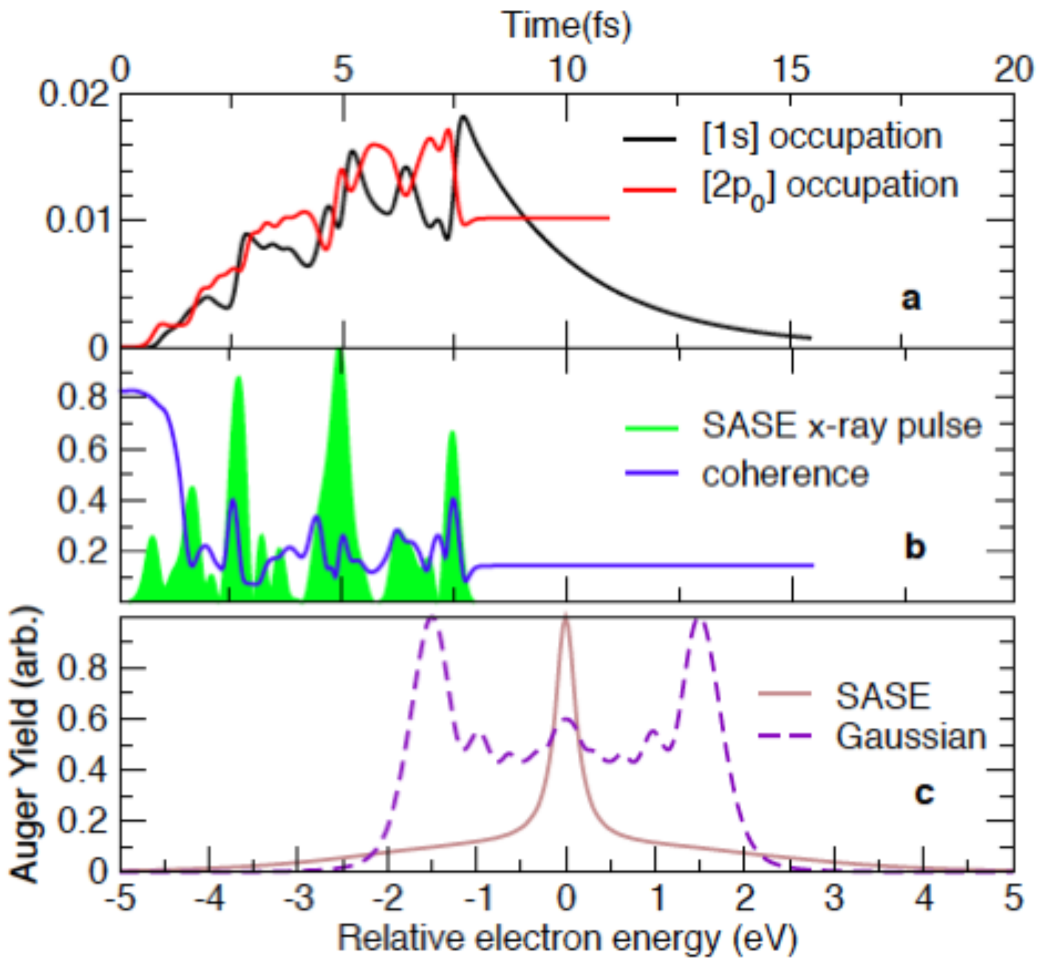
Conditions	Intrinsic x-ray pulse bandwidth (from 2s photopeak) (eV) (FWHM)	Intrinsic x-ray pulse bandwidth (from 2p photopeak) (eV) (FWHM)	Average bandwidth (eV) (FWHM)	%
40 pC (<10 fs) 850 eV 0.3 mJ 4500 A	4.3	4.5	4.4	0.5 %
250 pC (100 fs) 787 eV 1.5 mJ 2500 A	7.1	7.8	7.45	0.9%
250 pC (100 fs) 769 eV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

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



E.P. Kanter *et al.*, PRL (2011)   Theory from N. Rohringer and R. Santra

SASE vs Gaussian pulse for Rabi flopping



Summary: resonant x-ray processes at high intensity

- First hint of Rabi cycling for inner-shell electrons: Ne 1s – 2p resonance

- Need XFEL with improved longitudinal coherence – SEEDING
  - Quantum control – multidimensional spectroscopies
  - Single particle imaging (reduced radiation damage & increased x-ray intensity)

- "Hidden" resonances critical in atomic response to ultraintense x-rays
  - Enhanced two-photon absorption probability
    - Doumy *et al.*, PRL **106**, 083002 (2011)
  - Ionization beyond sequential single photon model
    - Schorb *et al.*, PRL **108**, 233401 (2012) - Ar
    - Rudek *et al.*, Nat. Phot. **6**, 858 (2012) – Xe
    - Rudek *et al.*, Phys. Rev. A **87**, 023413 (2013) – Kr

Improving the X-ray Laser

Self-seeding techniques and their importance for XFELs

SASE pulses, baseline mode of operation: poor longitudinal coherence

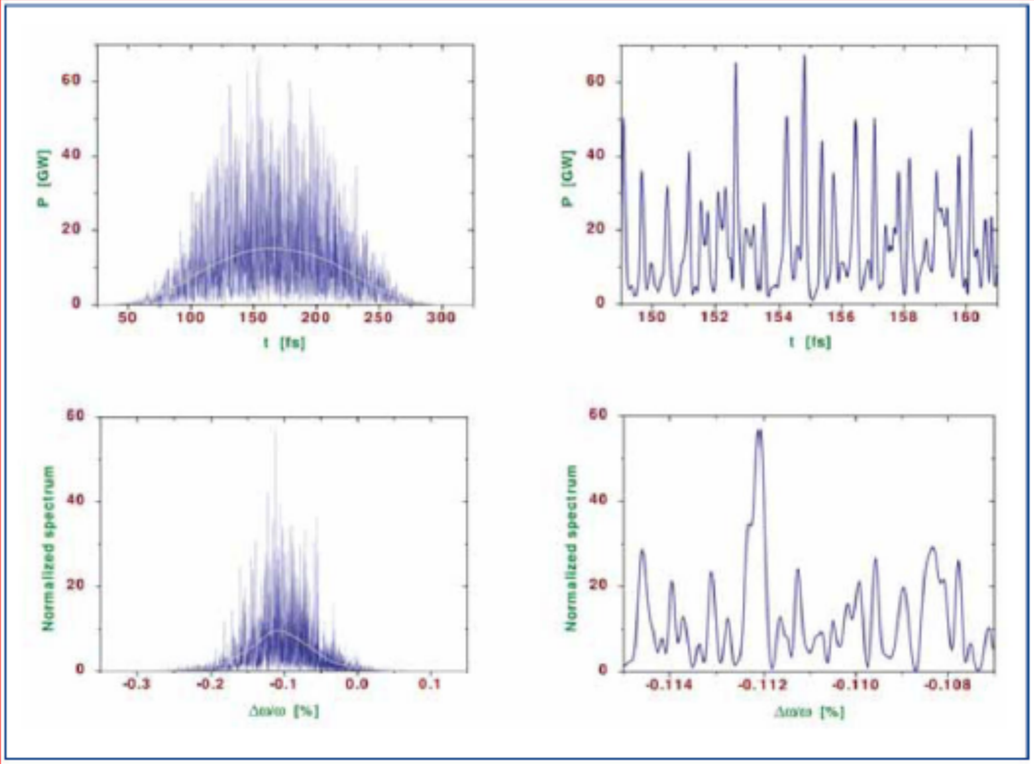


Figure 5.2.4 *Temporal (top) and spectral (bottom) structure for 12.4 keV XFEL radiation from SASE 1. Smooth lines indicate averaged profiles. Right side plots show enlarged view of the left plots. The magnetic undulator length is 130 m.*

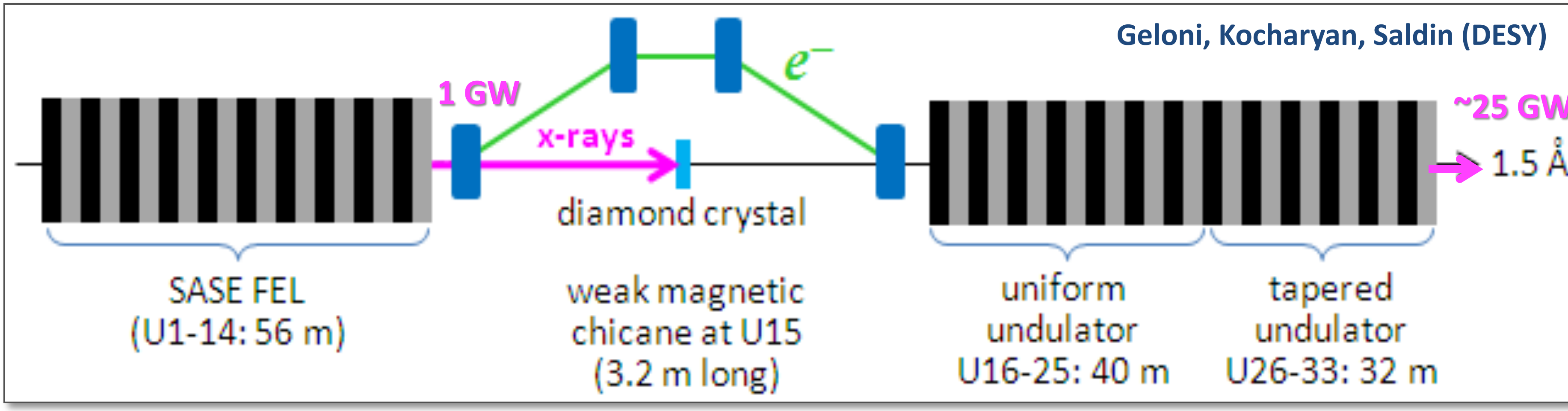
Source: The European XFEL TDR – DESY 2006-097 (2006)

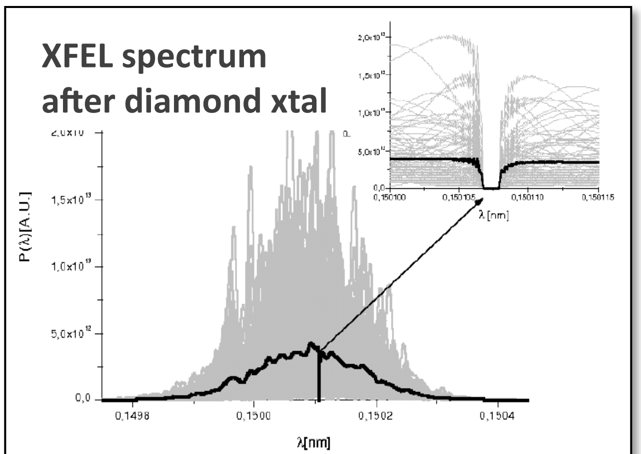
$$\frac{\Delta\omega}{\omega} \sim 2\rho \sim 10^{-3}$$

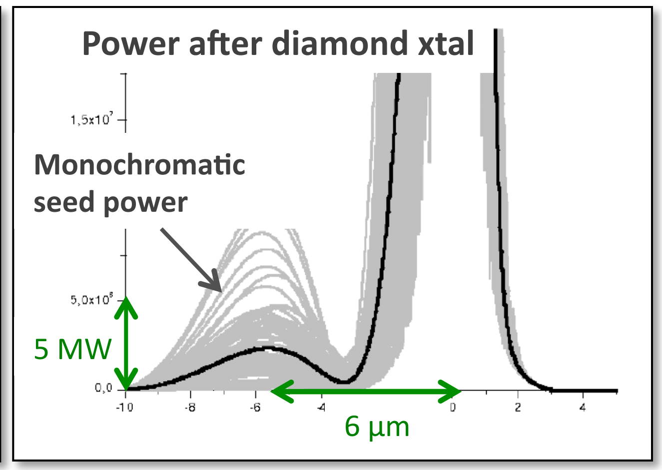
$$\left(\frac{\Delta\omega}{\omega}\right)_{spike} \sim \frac{1}{\sigma_T \omega} \sim 10^{-5}$$

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

Hard x-ray self-seeding proposed 2010





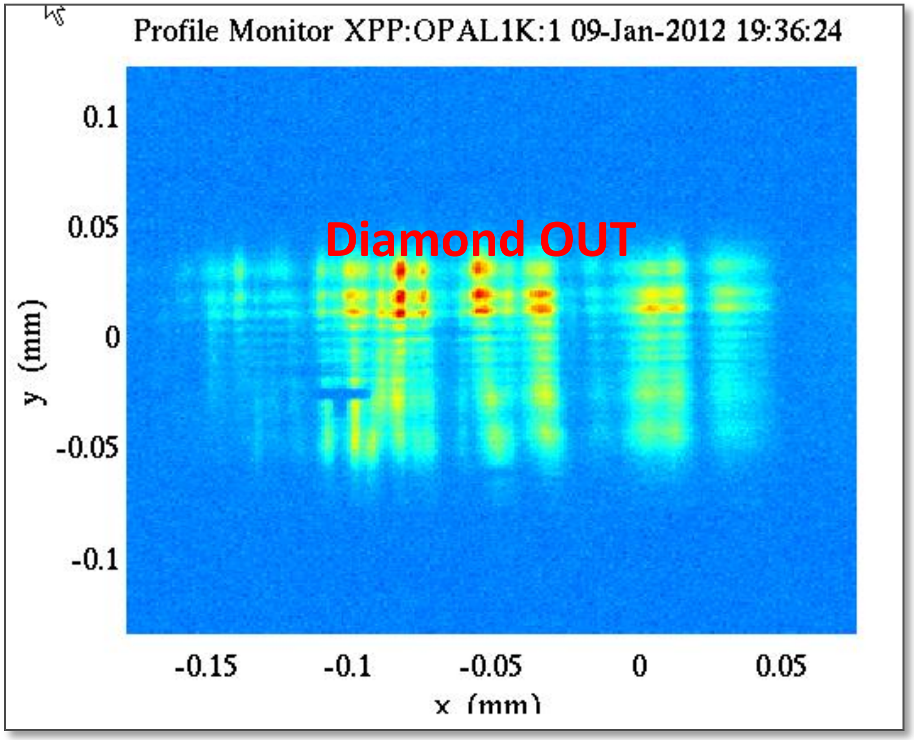


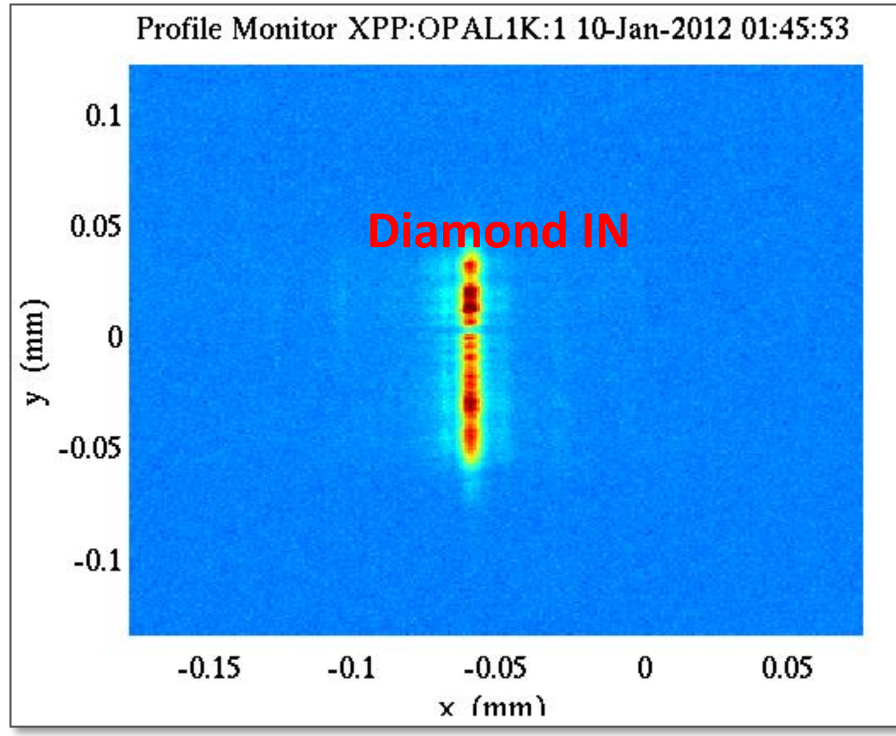
Diamond C(004): 100μm

λ = 0.15 nm, θ_B = 57°

arXiv:1008.3036v1 Geloni

Hard x-ray self seeding realized Jan 2012 – P. Emma et al.





Bandwidth <10^{-4} at 8-9 keV and tunable

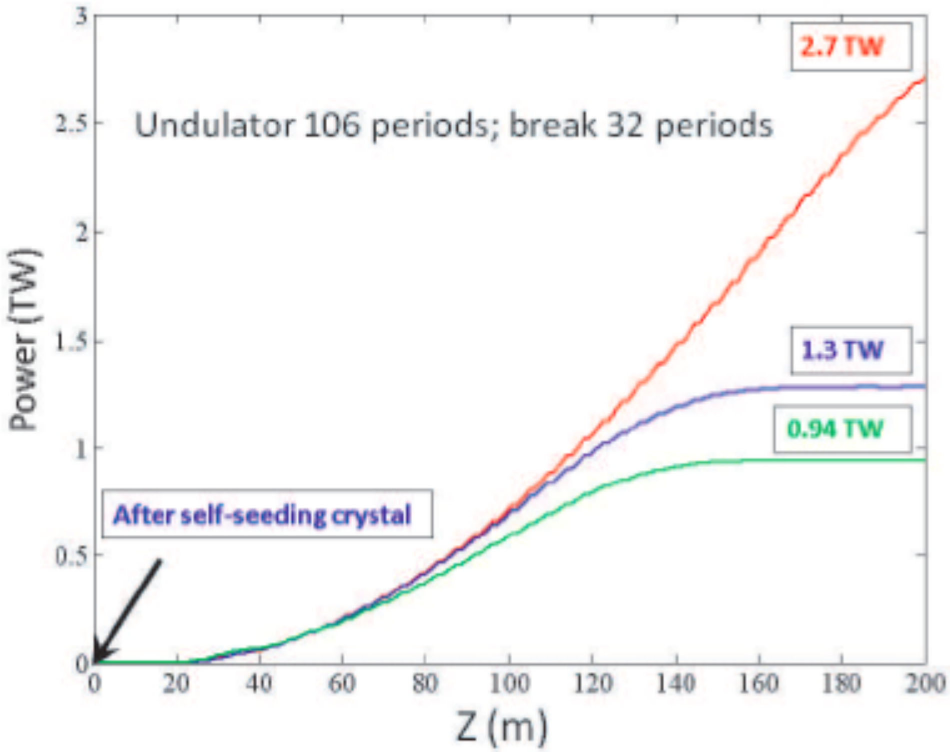
But … did not achieve saturation and power jitter still present

Three lab collaboration: SLAC Argonne NATIONAL LABORATORY ТИСНУМ

J. Amann *et al.*, Nature Photonics 6, 693 (2012)

On the road to a TW FEL : LCLS-TN-11-3

GENESIS simulation



200 m undulator tapered

10^{13} x-rays @ 8-10 keV
10 fs

W.M. Fawley[1], J. Frisch[1], Z. Huang[1], Y. Jiao[1], H.-D. Nuhn[1], C. Pellegrini[1,2], S. Reiche[3], J. Wu[1†]
[1]SLAC National Accelerator Laboratory, Menlo Park, CA 94025, USA
[2]Department of Physics and Astronomy, UCLA, Los Angeles, CA 90095-1547, USA
[3]Paul Scherrer Institute, Villigen PSI, 5232, Switzerland

Also arXiv Jun 2013: S. Serkez *et al.* 10 TW FEL, 10^{14} x-rays, 10 fs @ 3.5 keV

Towards single particle imaging

LETTER

doi:10.1038/nature09748

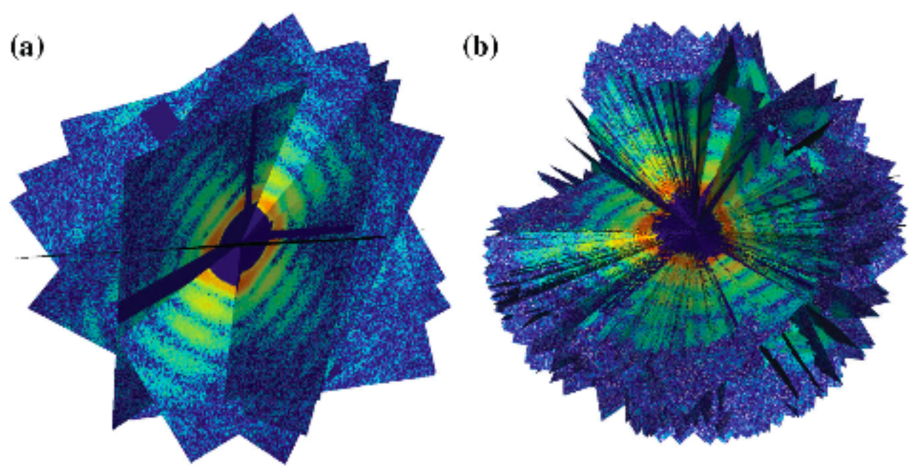
Single mimivirus particles intercepted and imaged with an X-ray laser

Seibert et al., Nature 470, 78 (2011)

PRL **114,** 098102 (2015) Selected for a Viewpoint in *Physics* PHYSICAL REVIEW LETTERS week ending 6 MARCH 2015

Three-Dimensional Reconstruction of the Giant Mimivirus Particle with an X-Ray Free-Electron Laser

Ekeberg et al., PRL (2015)



Mimivirus

-Largest known virus – 0.75 µm

-Does not crystallize

-Large for 3D cryoelectron microscopy

Single Shot Scattering Pattern

2D: 32 nm resolution

Set of 198 scattering patterns

3D: reconstruction to 120 nm

Physics

on Ekeberg et al., PRL (2015)

Viewpoint

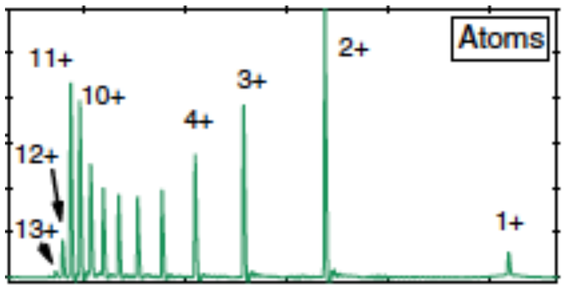
X-Ray Imaging of a Single Virus in 3D

"And there are still open questions on the impact of electronic damage on x-ray scattering on femtosecond time scales: The above-mentioned work by Neutze et al. tracked the movements of the atomic nuclei of the biomolecule, showing they don't move on the few-femtosecond timescale of an x-ray pulse. Electrons, however, move faster than nuclei. Since electrons are what scatters x rays, it is yet to be confirmed that few-femtosecond pulses can probe an unperturbed electronic structure." - Keith Nugent

March 2, 2015

Beyond the sequential single photon ionization model

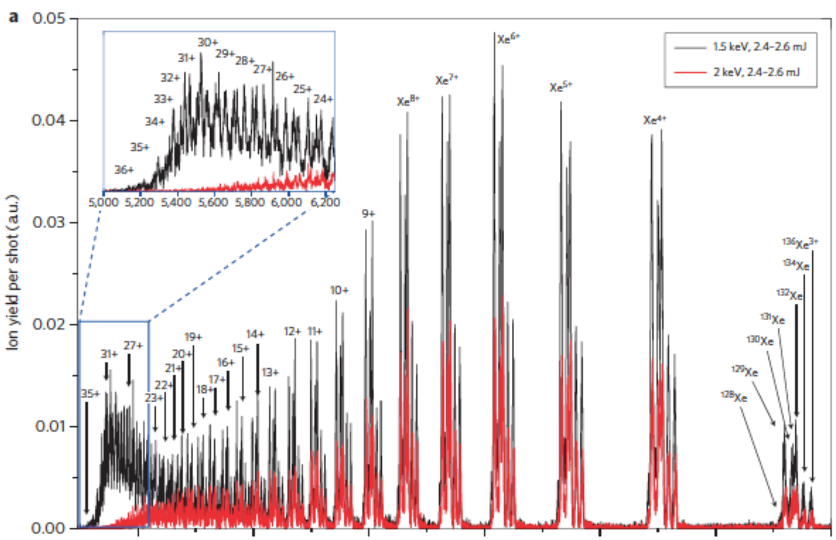
Argon



@ 480 eV

- sequential single photon limit 10+
- observe 13+
- Schorb et al., PRL (2012)

Xenon



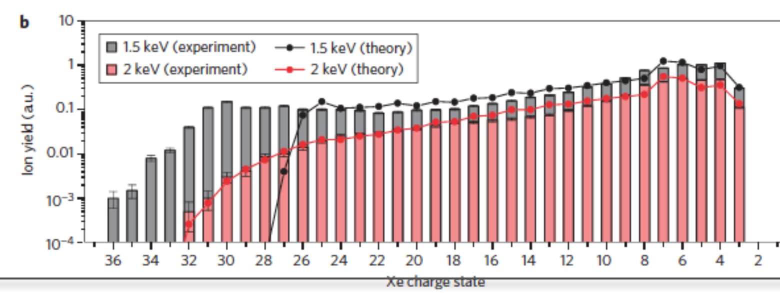
@1500 eV

- sequential single photon limit 27+
- observe 36+

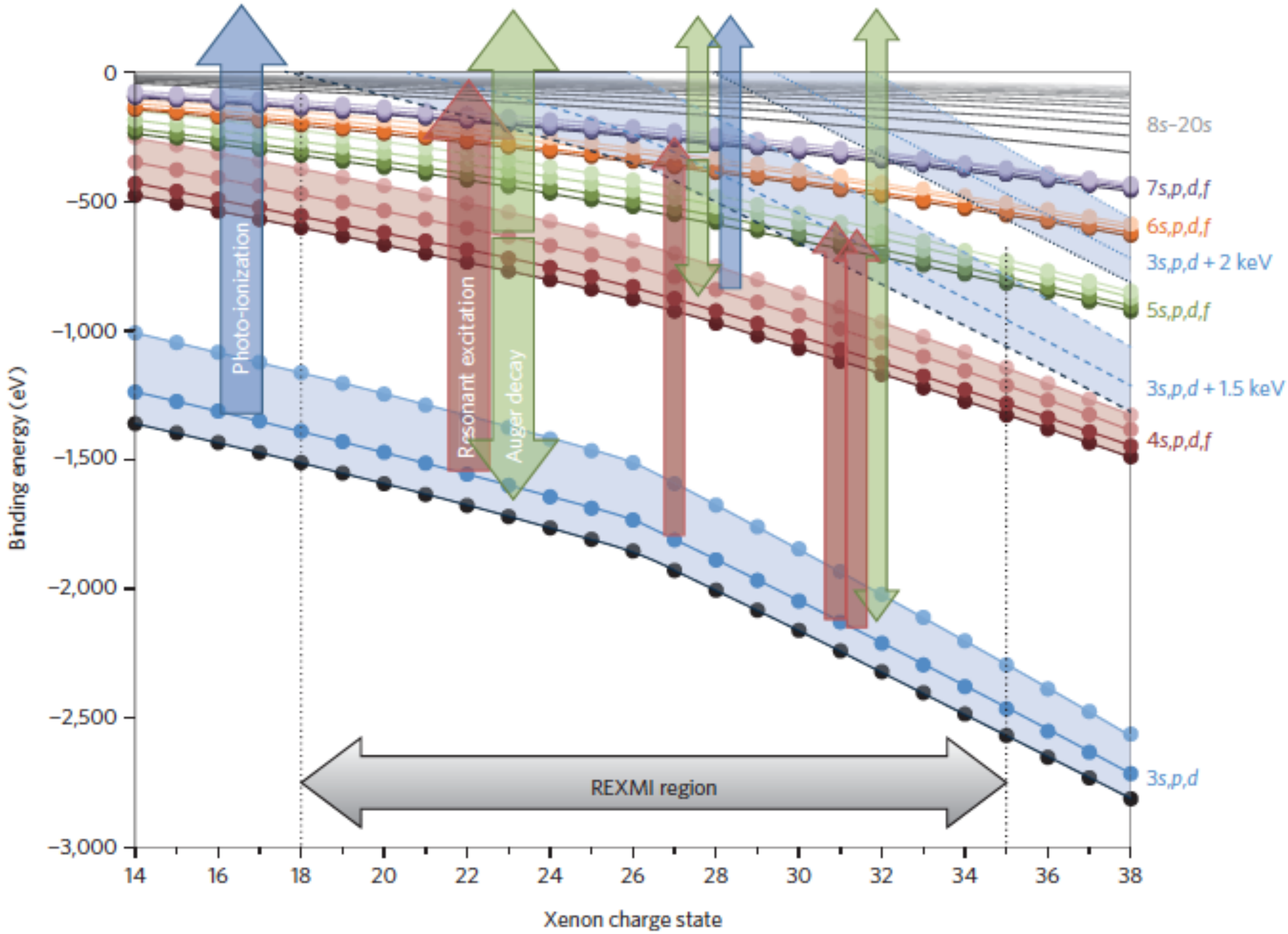
@2000 eV

- sequential single photon limit 32+
- observe 32+

Rudek et al., Nat. Pho. (2012)



Resonance-enabled x-ray multiple ionization

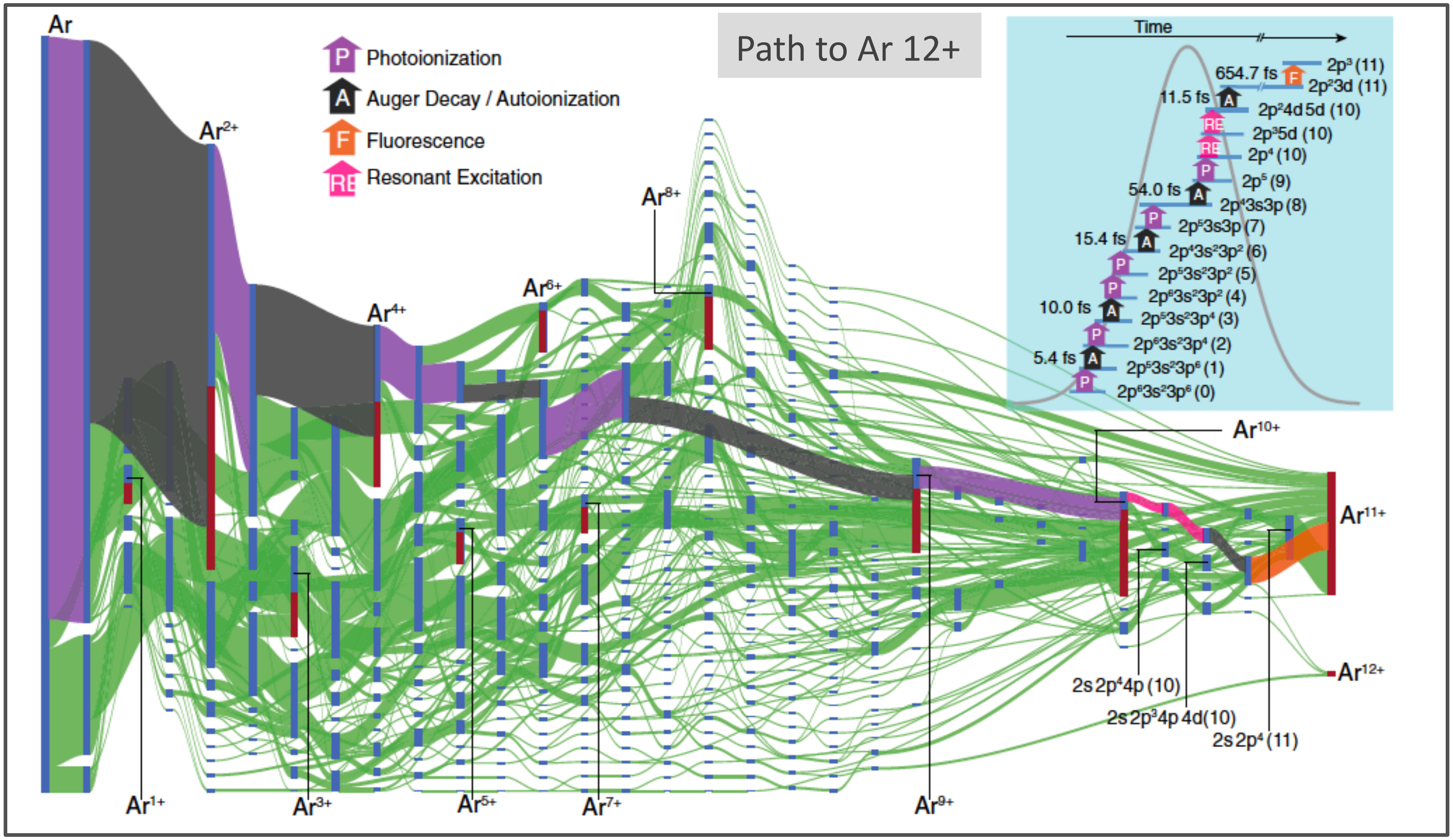


B. Rudek *et al.*, Nat. Phot. (2012)

Tracking electronic configurations during XFEL pulse including resonances!

Monte Carlo Rate Equation Approach

	# of ECs with no RE	# of ECs with RE
Ar	1.33×10^3	2.85×10^{13}
Kr	3.05×10^5	2.08×10^{19}
Xe	7.06×10^7	9.05×10^{22}



P. Ho, C. Bostedt, S. Schorb, L. Young, PRL **113**, 253001 (2014)

Ultraintense hard x-ray interactions: seeded v SASE

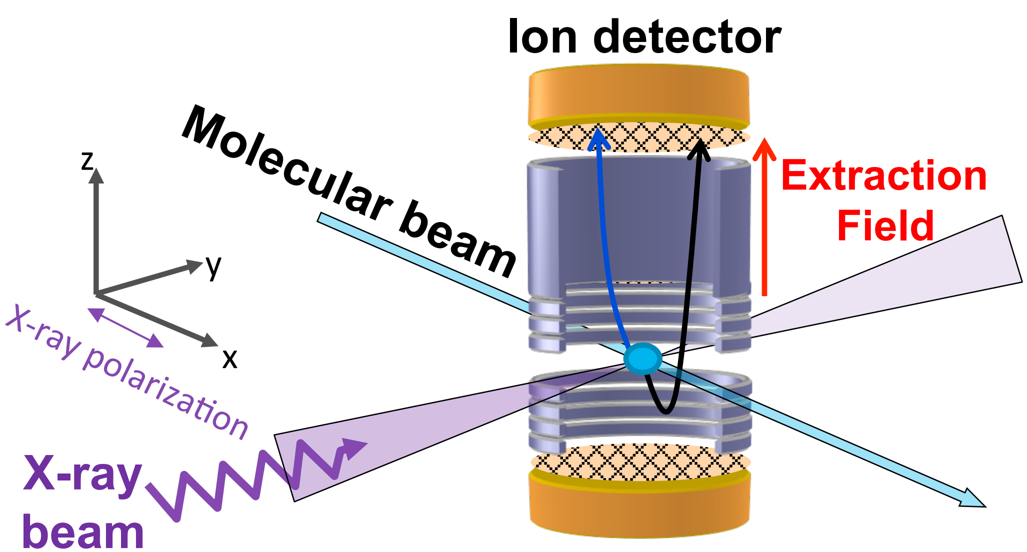
CXI endstation experiment Apr 2014: I >10^{20} W/cm^2

5.5 – 8.3 keV, 2 mJ, 30-40 fs
"100 nm" focus
10^5 x-rays/Å^2

...but no shot-by-shot spectral monitor

Observations:

- **Ar^{18+}:** fully stripped
- **Kr^{34+}:** two 1s electrons
- **Xe^{48+}:** 6 electrons
- Largely consistent with sequential single photon
- **CH_3I:** sum of charges 55+

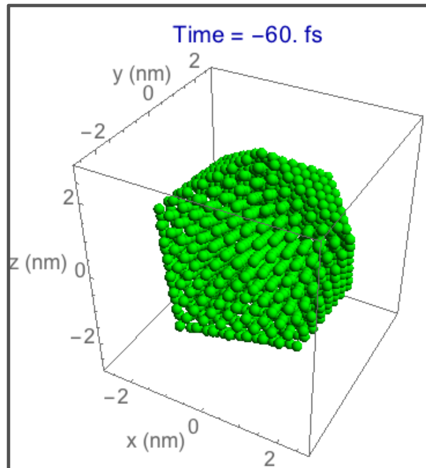


Calculations by Phay Ho:
Identified photon energies where seeded pulses reduce absorption & radiation damage relative to SASE pulses.

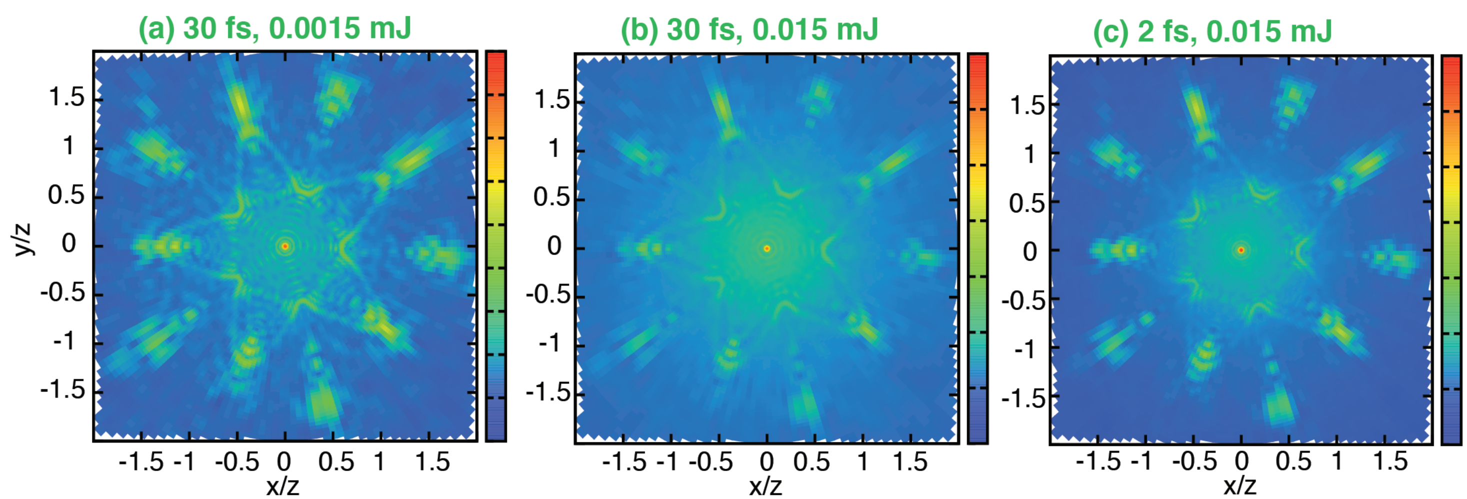
Joined/Artem Rudenko, Daniel Rolles & team

High Intensity X-ray Imaging of Nanosystems

- Intense x-ray pulses lead to efficient stripping of e^- from atoms/ions and significant displacement of e^-, ions/atoms from equilibrium during the pulse – nanoplasma
- Our MC/MD model includes: Elastic Scattering, Photoionization, Auger, Fluorescence, Resonant excitation, Three-body recombination, Electron impact ionization, Lattice dynamics
- Investigate the impact of electronic damage on various observables (ion, photoelectron, Auger and fluorescence spectra, x-ray diffraction)
- Recorded diffraction patterns depend strongly on pulse parameters.



7-shell Ar cluster (~5 nm)
1415 nuclei, 25470 e-
8 keV x-ray pulses



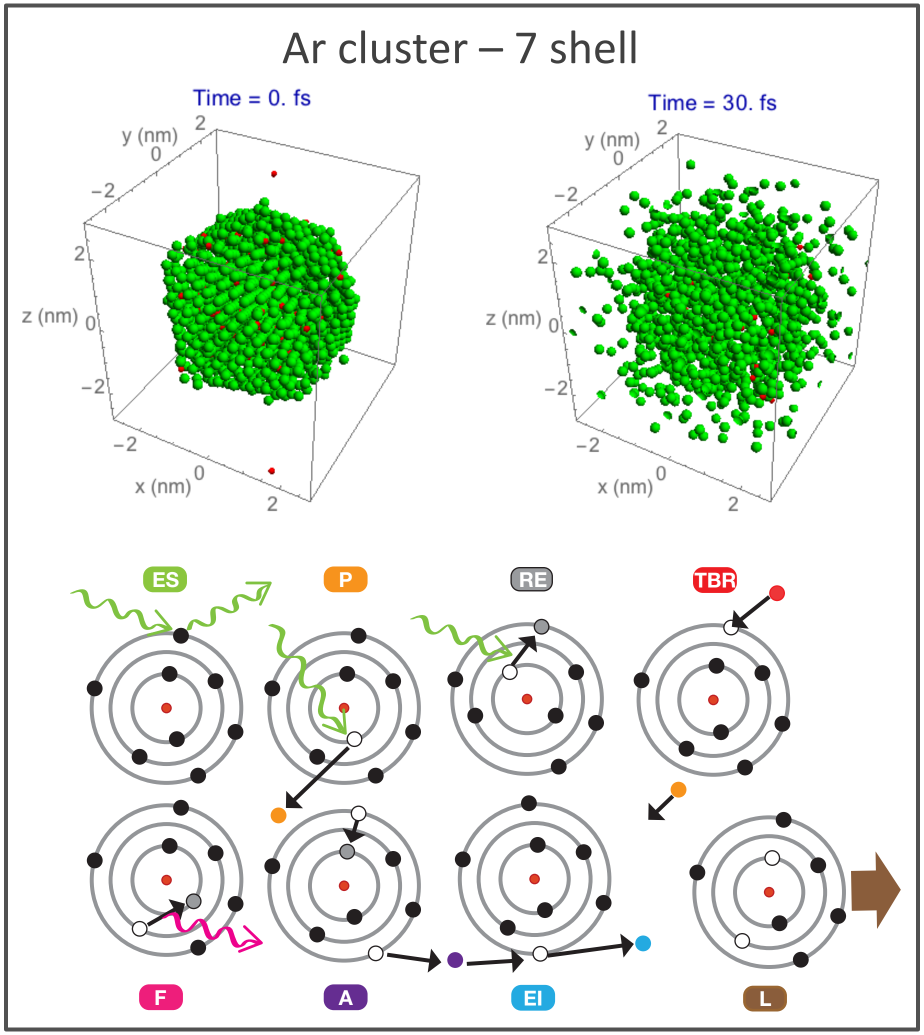
AMO "solutions" at the ultraintense x-ray frontier

- Fundamental nature of x-ray damage at high intensity
  - -Coulomb explosion
  - -electronic damage
  - -behavior at 10^{22} W/cm^2 – 1Å
  - -nanoplasma formation

- Expt'l AMO observables:
  - Ion, photoelectron, Auger, fluorescence and …
  - x-ray diffraction pattern

- Computational studies on large scale systems

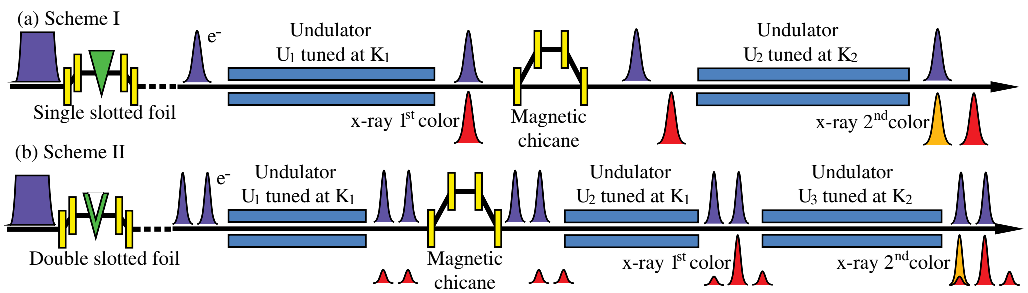
Phay Ho
Chris Knight



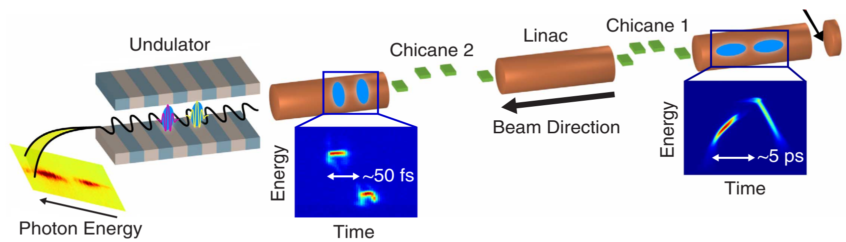
Atomistic computational studies based upon Monte Carlo/Molecular Dynamics

New XFEL capabilities becoming available

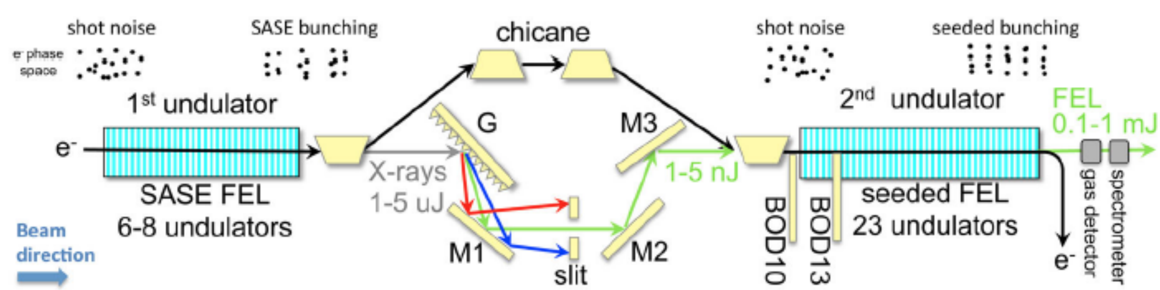
One bunch – two pulse – two color (Lutman *et al.*, PRL **110**, 134801 (2013))



Two bunch – two pulse – two color (Marinelli *et al.*, Nat. Comm. **6**, 6369 (2015))



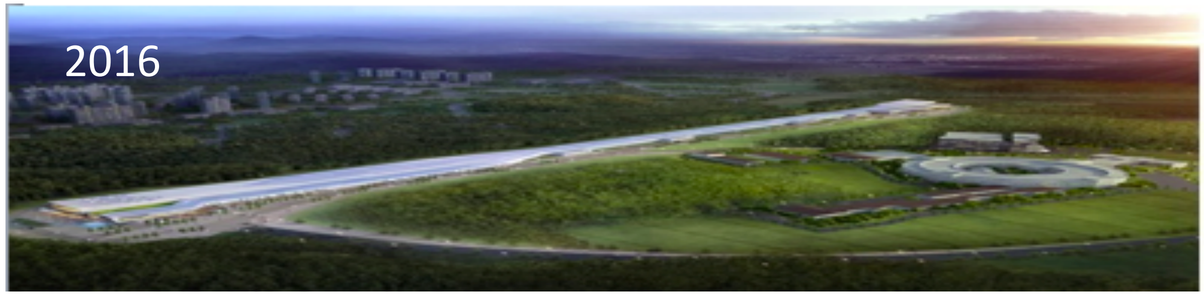
Seeded soft x-ray beam (Ratner *et al.*, PRL **114**, 054801 (2015))



International hard X-ray FELs here and on the horizon

SACLA
Image Gallery

PAL POHANG ACCELERATOR LABORATORY



SwissFEL

European XFEL

Summary

- Our atomic physics expts have established fundamental understanding of the response of matter to ultraintense XFEL irradiation

  - sequential single photon ionization dominates

  - intensity-induced x-ray transparency (frustrated absorption)

  - intense x-rays can "control" inner-shell electron dynamics

  - resonances can be critical in XFEL interactions

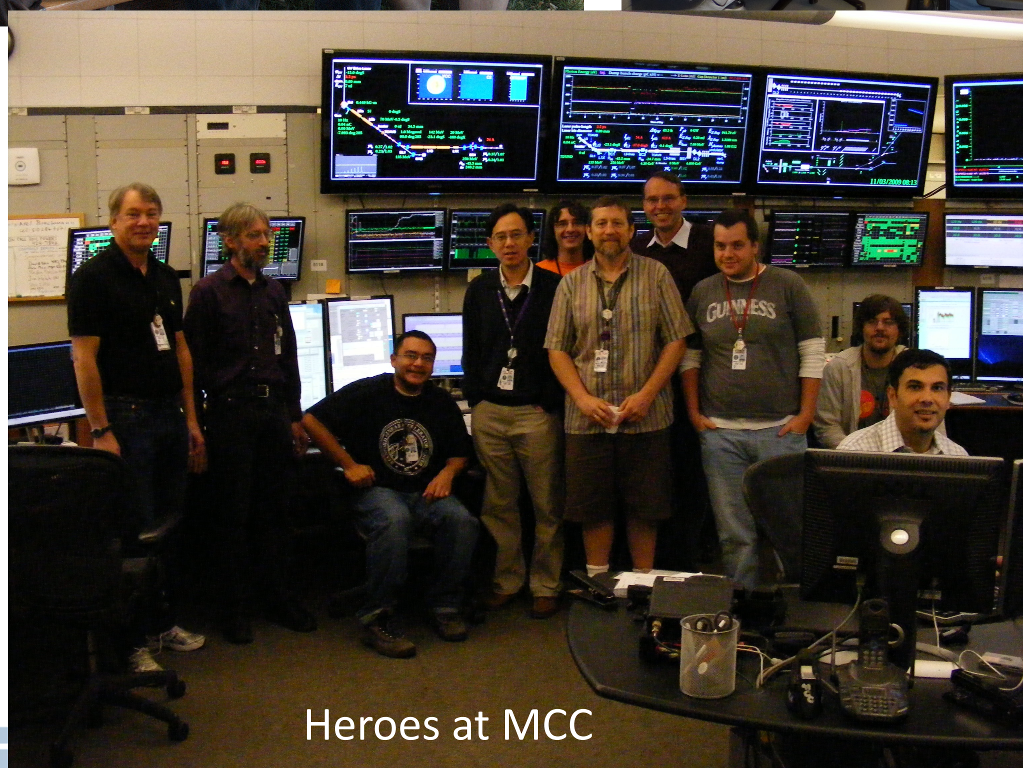
- This fundamental understanding will aid in the quest for single molecule imaging and other applications, e.g. high energy density matter

  - AMO methods (ion, electron, photon) in concert with computational studies promise fuller understanding of radiation damage in extended systems

- Future is bright with well-characterized ultraintense x-ray lasers, multiple pulse configuraitons … with 1000x higher intensity.

Argonne AMO group Oct 2009

Heroes at AMO Control



Heroes at MCC