

AMO Adventures at the LCLS

The Argonne Atomic, Molecular, and Optical Physics Group's experiments utilizing the Linac Coherent Light Source (LCLS) – the first X-ray Free-Electron Laser

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Thanks to 33 Undulators from the APS



X-Ray Flux: Per Pulse versus Per Second

LCLS: 2×10^{12} photons per pulse at 9 keV;

2×10^{13} photons per pulse at 750 eV;

≈ 70 -300 femtoseconds pulse length

(≈ 1 -3 femtoseconds, if the bunch charge is lowered from 250pC to 20pC)

currently 30 Hz repetition rate, 120 Hz maximum:

$\leq 2.4 \times 10^{15}$ photons per second

For comparison, APS 14-ID (2 undulators):

10^{10} photons per pulse at 12 keV;

≈ 100 picoseconds pulse length;

6.5 MHz, mechanically limited to ≤ 1 kHz:

$\leq 6.5 \times 10^{16}$ photons per second



High Intensity X-rays

- 2×10^{13} photons
- 750 eV photon energy
- 200 fs pulse length
- $1 \mu\text{m}^2$ focal diameter
- $1.2 \times 10^{18} \text{ W/cm}^2$

- Pulses are spiky → peak intensities are higher than the average
- Individual atoms absorb multiple photons during a single pulse

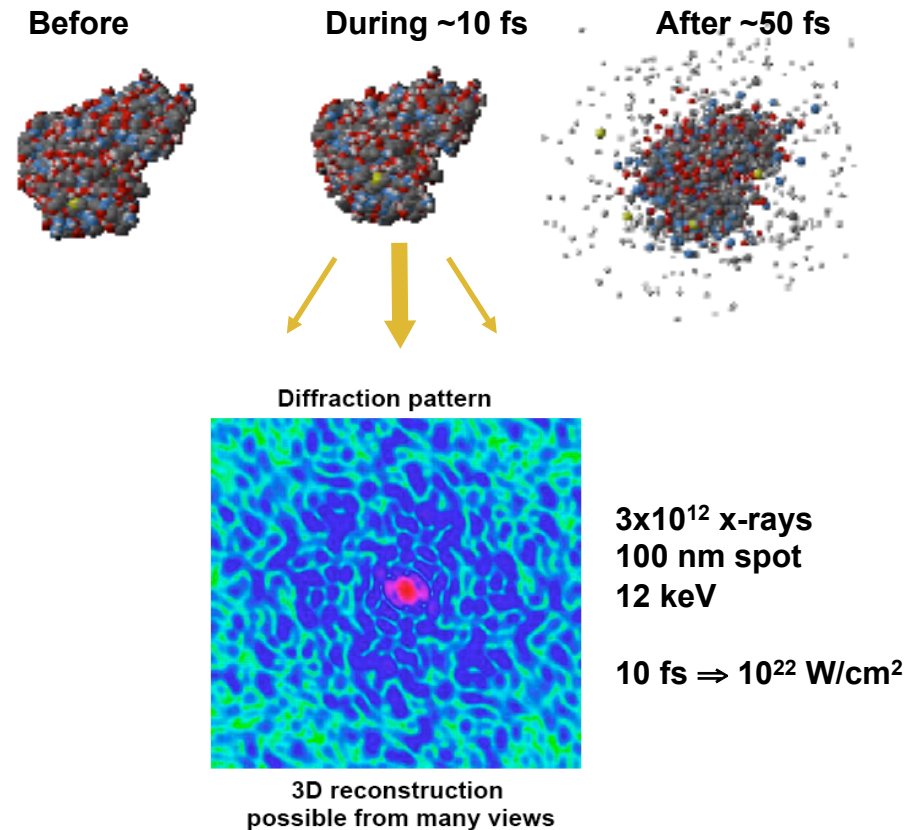


“Unlocking the World of the Ultra Small and the Ultra Fast”

AMO questions

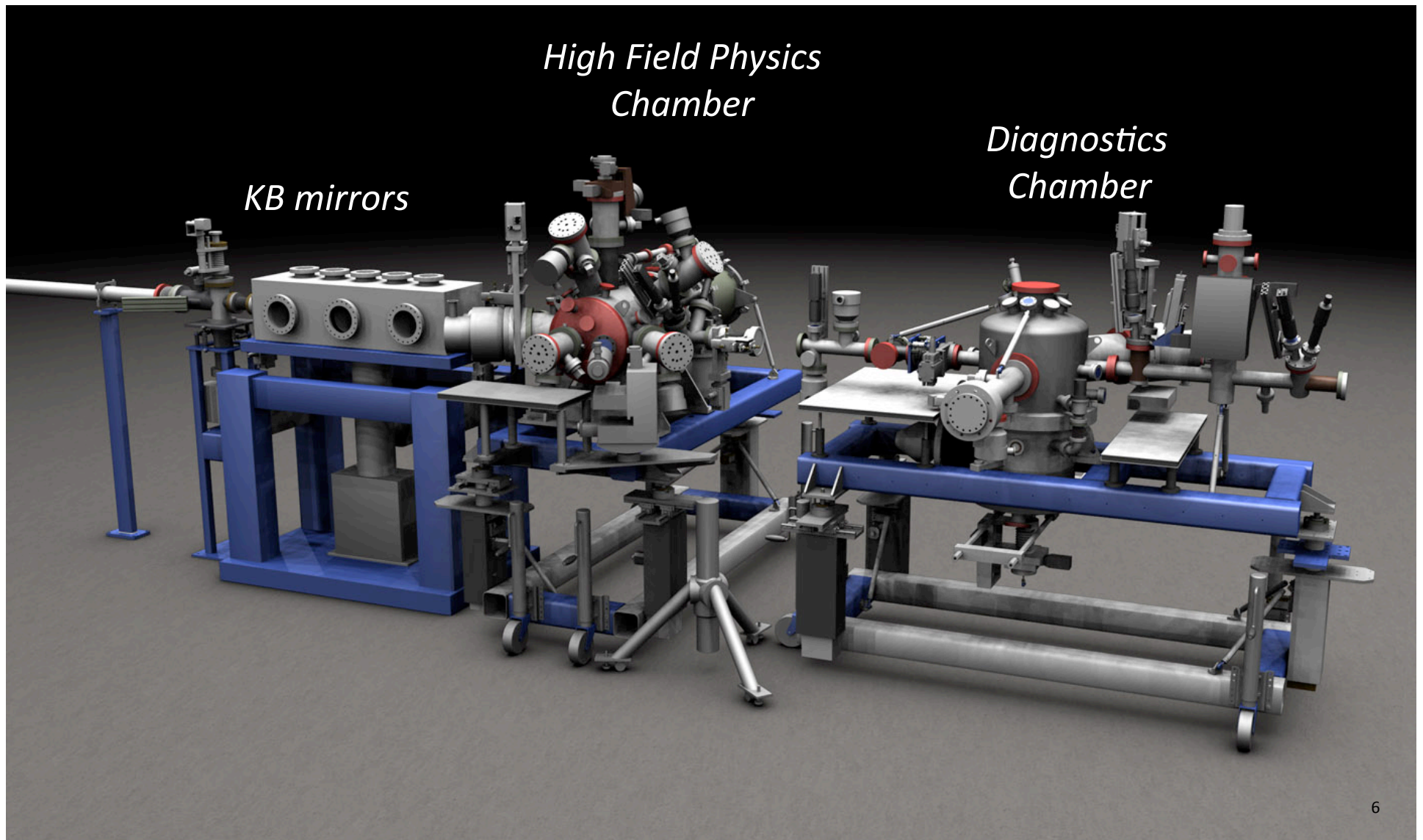
- fundamental nature of x-ray damage at high intensity
 - electronic damage
 - Coulomb explosion timescale
 - behavior at $>10^{18}$ W/cm² – 15Å
- nonlinear x-ray processes
- role of coherence
- quantum control of inner-shell processes

The LCLS Poster Child

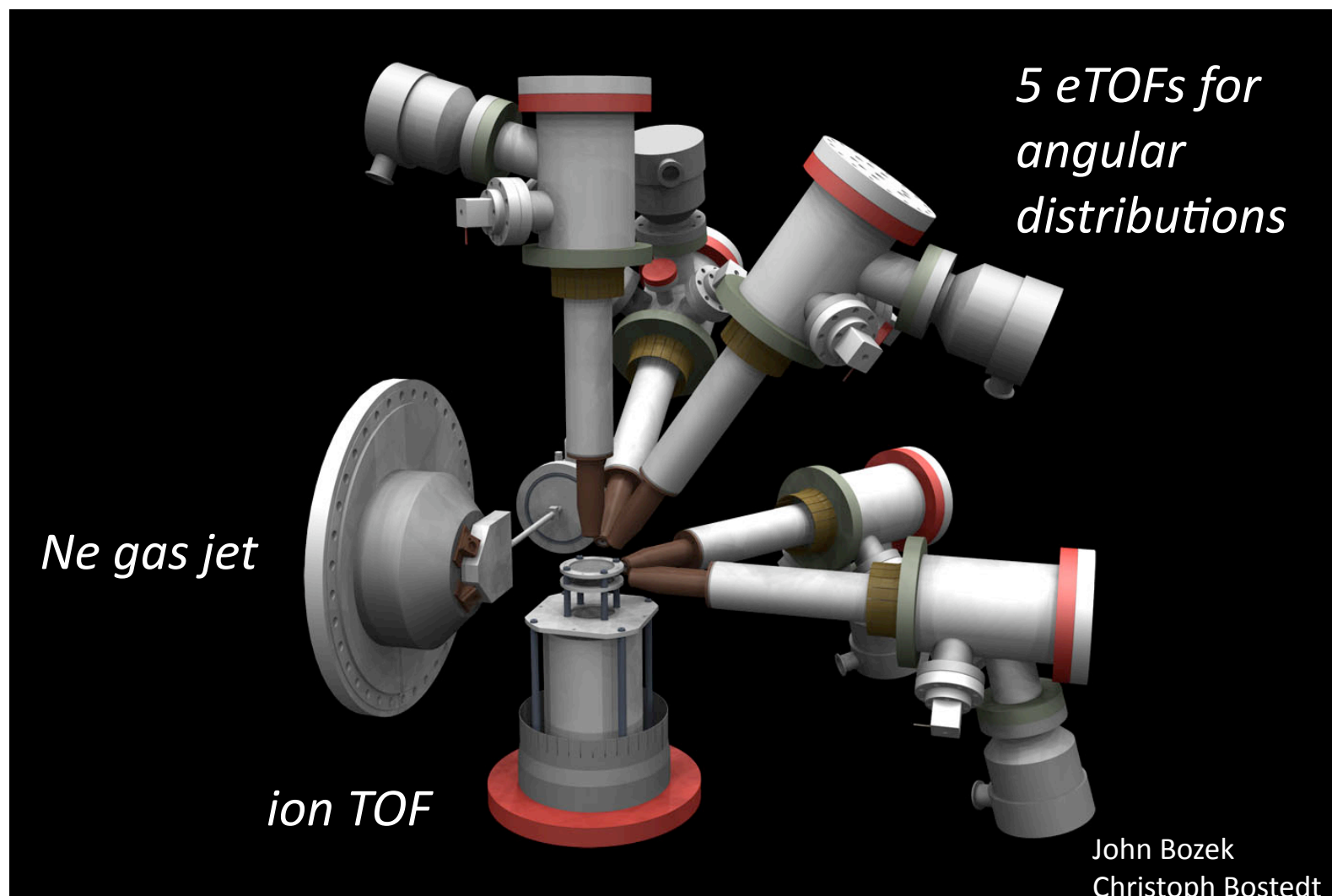


Neutze, Wouts, van der Spoel, Weckert, Hajdu
Nature **406**, 752 (2000)

The First Endstation: AMO Physics



The Components of the High Field Chamber





The First User Experiment , October 1–5, 2009

Tracking transient atomic states produced by ultraintense x-ray pulses

The Team

Yuelin Li, Elliot Kanter, Bertold Krässig, Anne Marie March, Steve Pratt,
Robin Santra, Steve Southworth, Linda Young

Argonne

John Bozek, Christoph Bostedt, Mark Messerschmidt

LCLS

Lou DiMauro, Gilles Doumy, Chris Roedig

Ohio State University

Nora Berrah, Li Fang, Matthias Höner

Western Michigan University

Phil Bucksbaum, James Cryan, Mike Glownia, David Reis

Pulse Center, Stanford



LCLS Launches User Science Today

SLAC

today



Neon, the Prototypical Atom

10 electrons, ground state configuration
 $1s^2 2s^2 2p^6$

Binding energies in neutral neon:

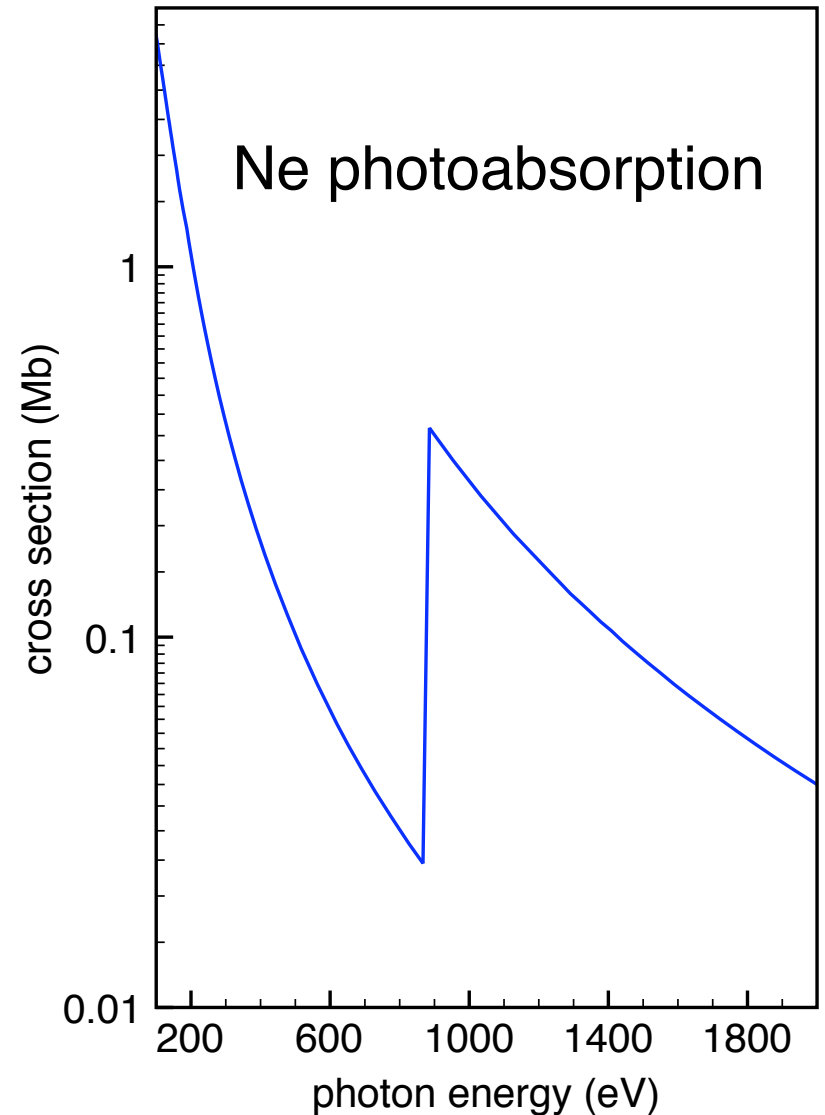
2p : 21.6 eV

2s : 48.5 eV

1s : 870.2 eV

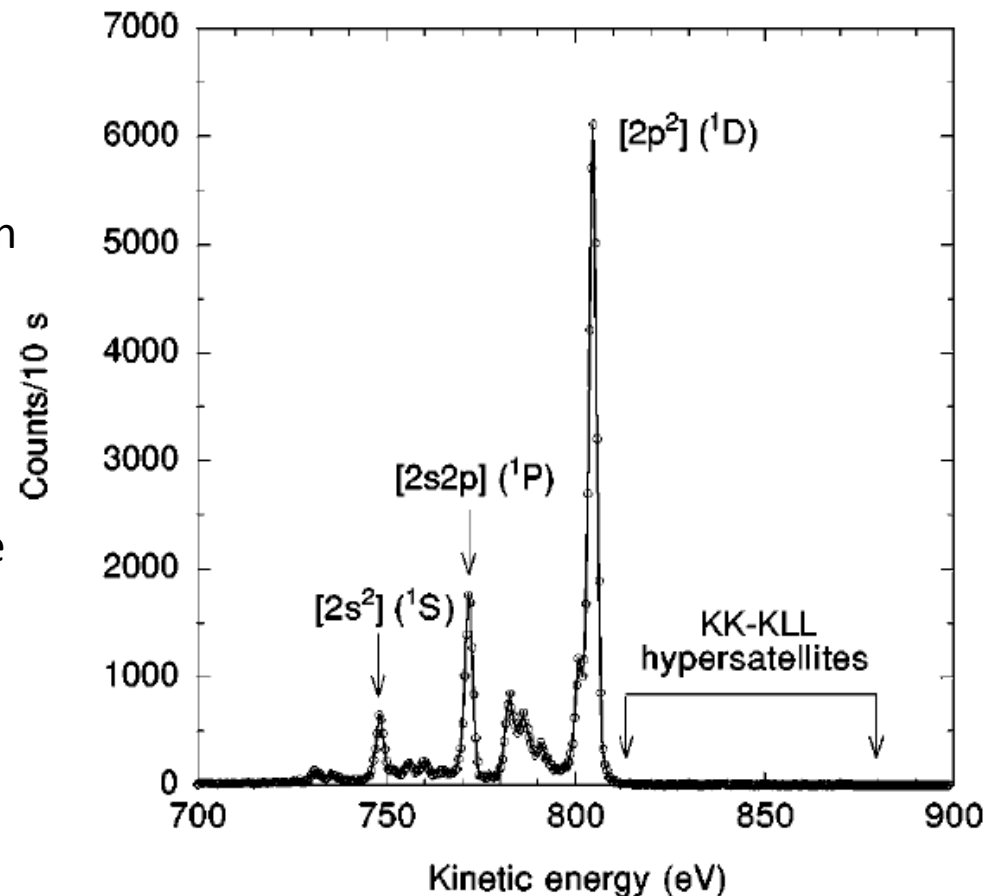
The higher the charge state, the higher the binding energies.

At threshold, the photoabsorption cross section for the 1s shell is 15 times larger than for the valence shell.



K-Shell Ionization (870 eV and above) Auger Decay

- K-hole state lifetime $\tau = 2.4$ fs
- Predominantly Auger decay (98.2%)
- K-shell hole is filled by a valence electron and a second valence electron is emitted (Auger electron)
- Auger electron energy is the binding energy difference between the inner and valence shells
- Double K-holes (hollow atom) give rise to hypersatellite transitions
- Double-K/Single-K = 0.32(4) % (for 5 keV x-rays, single-photon absorption, 12-ID)



Southworth et al., *Phys. Rev A* **67** (2003)

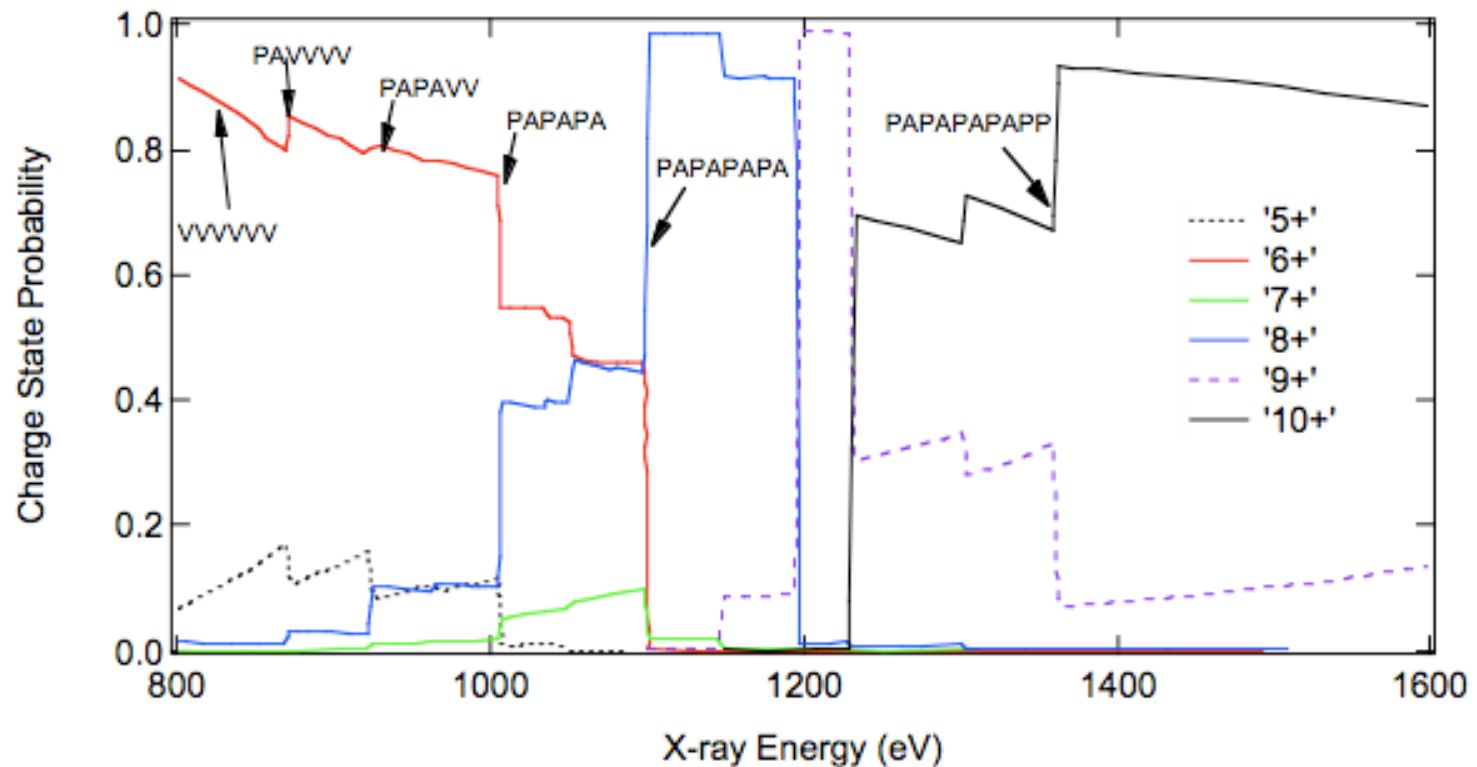


Theory: Nina Rohringer and Robin Santra

Phys. Rev. A 76, 033416 (2007)

- **P**: K-shell photoionization
- **A**: Auger electron emission
- **V**: Valence ionization
- Sequential ionization PAVVVV ...

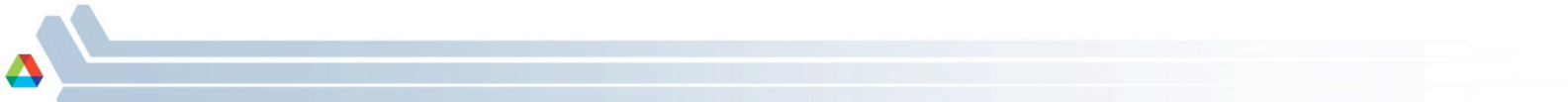
10^{13} x-rays
230 fs
1 μm spot






The “First Glimpse at the Experimental Data” will be posted after publication of this work

Thank you for your understanding.





The Fifth Experiment at LCLS, Oct 30– Nov 3, 2009

Resonant nonlinear x-ray processes at high x-ray intensity

The Same Team

Yuelin Li, Elliot Kanter, Bertold Krässig, Anne Marie March, Steve Pratt,
Robin Santra, Steve Southworth, Linda Young

Argonne

John Bozek, Christoph Bostedt

LCLS

Lou DiMauro, Gilles Doumy, Chris Roedig

Ohio State University

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Western Michigan University

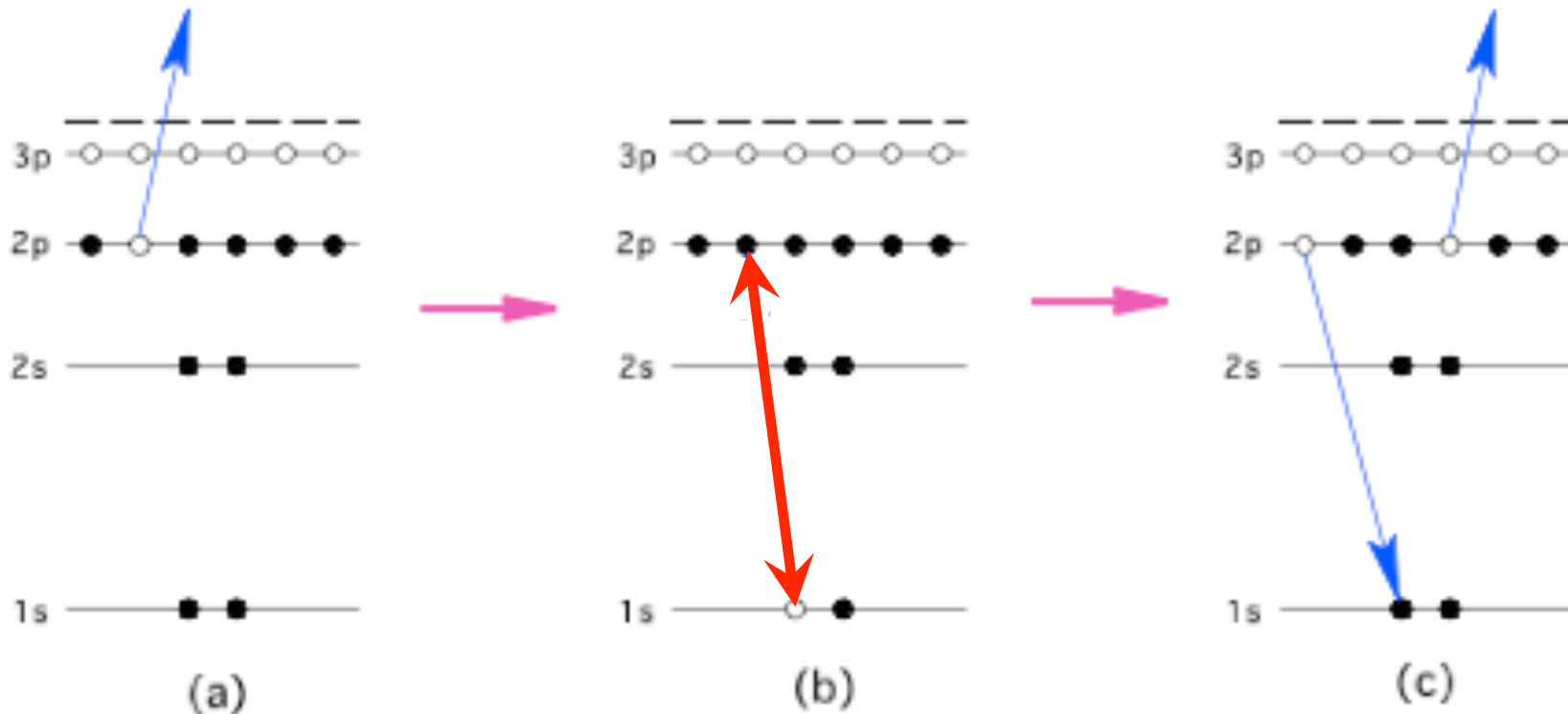
Phil Bucksbaum, James Cryan, Mike Glownia, David Reis

Pulse Center, Stanford

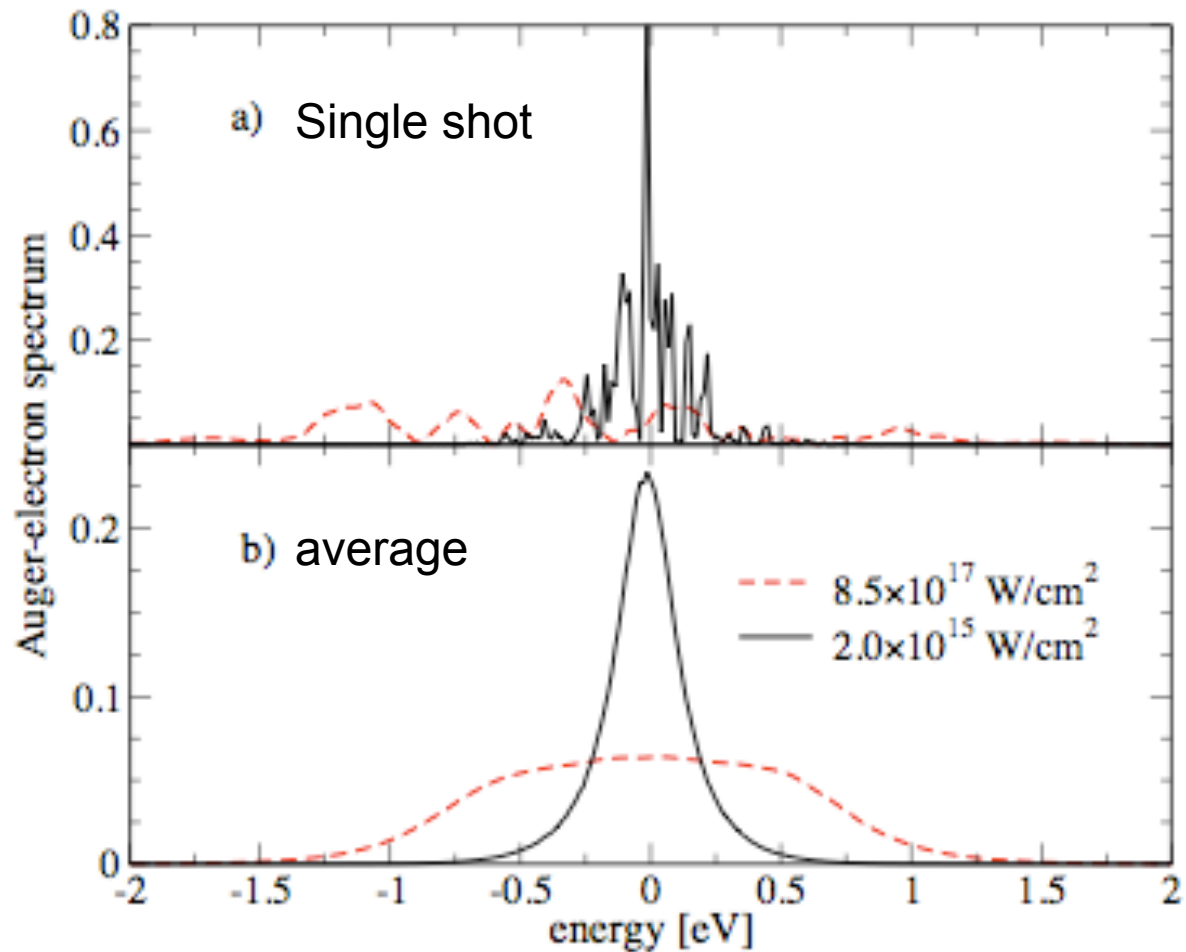


Schematic of the Two-photon 1s-2p Resonance Excitation

- a) Valence ionization by first absorbed x-ray photon (or IR laser)
- b) 1s-2p excitation at 848.6 eV by second absorbed photon;
resonant absorption/stimulated emission cycling at high intensity
(Rabi Flopping)
- c) Auger decay



Theory: Nina Rohringer and Robin Santra, Phys. Rev. A 77, 053404 (2008)



Summary and Outlook

- The LCLS is up and running.
- The Argonne AMO group had two successful experiments at the LCLS in October 2009.
- The data show the signatures of
 - sequential multiphoton absorption,
 - double K-hole (hollow neon) production,
 - intensity-induced transparency/saturable absorption,
 - 1s-2p resonant absorption.
- Data analysis is ongoing.
- Our next run: August 1-5, 2010.

Femtosecond x-ray two-photon photoelectron spectroscopy of organic molecules



Heroes at the Main Control Center

