

Harnessing ultra-intense x-rays for dynamic imaging



Linda Young 30th International Conference on Photonic, Electronic and Atomic Collisions Cairns, Queensland, Australia 26 Jul - 1 Aug 2017



Outline

- Birth of world's first hard x-ray FEL LCLS
- Non-resonant high intensity x-ray phenomena Atoms: Ne, Ar, Kr, Xe
- Resonant high intensity x-ray processes
 Rabi flopping, stimulated Raman
- Towards single particle imaging --- with hard x-rays & also water window
- New XFEL capabilities

Linac Coherent Light Source at SLAC X-FEL based on last 1-km of existing 3-km linac

Proposed by C. Pellegrini in 1992

Injector (35^o) at 2-km point

Argonne

UCLA

Existing 1/3 Linac (1 km)

New e⁻ Transfer Line (340 m)

(14-4.3 GeV)

1.5-15 Å

X-ray.Transport Line (200 m)

Undulator (130 m)

Near Experiment Hall





-

April 10, 2009: LCLS lases at 1.5Å



- Saturation after ~65 meters of undulator!
- Alignment req'd 5 microns over 100 m



Yes I Do Smile on Occasion





Project Status Update LCLS Science Advisory Committee

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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







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 10²² W/cm² - 1Å
- nonlinear x-ray processes
 role of coherence
- quantum control of inner-shell processes



3D reconstruction possible from many views 10 fs \Rightarrow 10²² W/cm²

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

LCLS Experiment 1 - Oct 1, 2009

Nature of the electronic response to

10⁵ x-rays/Å² 80 - 340 fs 800 - 2000 eV

$\sim 10^{18} \, W/cm^2$

Original single molecule imaging parameters, Neutze et al. Nature (2000) $3 \times 10^{12} x$ -rays/(100 nm)² = $3 \times 10^{6} x$ -rays/Å² 10 fs ~ $10^{22} W/cm^{2}$

Our approach to understanding ultraintense x-ray interactions

Start with a well-characterized target

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Binding energies in neutral neon

2p: ~21 eV

2s: ~48 eV

1s: ~870 eV

Inner-shell excitation

Auger yield 98%

Auger clock - \tau_{1s}: 2.4 fs
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 Probe changes in interaction from outer- to inner-shell between 800-2000 eV

Guided by theory

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



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

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Valence ionization, core ionization and Auger decay



Sequential single photon processes dominate the interaction

How does one arrive at a particular charge state?



- 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.

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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 $\theta = 90^{\circ}$

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



Summary: non-resonant ultraintense x-ray interactions

- Ultraintense x-ray interactions nonlinear multiphoton processes rule!
 - establish sequential single photon absorption as dominant ionization mechanism fully stripped neon: six-photon, ten-electron ($\sim 10^{12}/\mu m^2$)
 - multiple photon absorption probability high when fluence > 1/ σ
 - 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: $\sigma_{\rm scatt}/\sigma_{\rm abs}$ is increased
- Femtosecond time-scale atomic processes provide FEL diagnostics

Direct two-photon absorption cross section small He-like neon



G. Doumy et al., PRL 106, 083002 (2011). 21

Two-photon absorption cross-sections v Z



Maria Goeppert-Mayer



Ge: Tamasaku *et al.*, Nat Phot (2014) Zr: Ghimire *et al.*, PRA **94**, 043418 (2016)

Cu: Szchlactko et al, Sci Rep (2016)

doi:10.1038/nature10721

Atomic inner-shell X-ray laser at 1.46 nanometres pumped by an X-ray free-electron laser

Nina Rohringer¹[†], Duncan Ryan², Richard A. London¹, Michael Purvis², Felicie Albert¹, James Dunn¹, John D. Bozek³, Christoph Bostedt³, Alexander Graf¹, Randal Hill¹, Stefan P. Hau-Riege¹ & Jorge J. Rocca²



Gain medium: 500 torr neon, 1.5 cm, LCLS focused to ~1-2 μm

Stable wavelength, same divergence as XFEL pump, 10000x increase output for 2x increase pump power

Resonant x-ray processes at high intensity

LCLS Expt 5

Can we control inner-shell electron dynamics? "Rabi flopping" may inhibit Auger decay & x-ray damage.



But LCLS linewidth ~ 8 eV!



 $E_{Ne} \sim 6.3 \text{ a.u.}$ $I_{Ne} \sim 1.4 \times 10^{18} \text{ W/cm}^2$

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



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



Is the ¹D 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

Stimulated Raman scattering in Ne with SASE pulse





Small overlap of SASE pulse w/resonances Tail of SASE pulse used to stimulate Raman

SRS Signature: Stochastic scattering intensity below edge

> Weninger *et al*, PRL 111, 233902 (2013) Weninger & Rohringer, PRA **88**, 053421 (2013)

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Improving the X-ray Laser

Hard x-ray self-seeding proposed 2010





Diamond C(004): 100 μ m λ = 0.15 nm, $\theta_{\rm B}$ = 57°

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



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

GENESIS simulation



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Also arXiv Jun 2013: S. Serkez *et al.* 10 TW FEL, 10¹⁴ 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)





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 100 coefforting notion
- Set of 198 scattering patterns
 - 3D: reconstruction to 120 nm



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 fewfemtosecond pulses can probe an unperturbed electronic structure." - Keith Nugent



Beyond the sequential single photon ionization model



@ 480 eV

sequential single photon limit 10+ observe 13+

Schorb et al., PRL (2012)



sequential single photon limit 27+

observe 36+

@2000 eV

sequential single photon limit 32+ observe 32+





Resonance-enabled x-ray multiple ionization



Tracking electronic configurations during XFEL pulseincluding resonances! $\frac{\# \text{ of ECs with no RE}}{\text{Ar}}$ $\frac{\# \text{ of ECs with no RE}}{\text{Ar}}$ $\frac{\# \text{ of ECs with no RE}}{1.33 \times 10^3}$ $\frac{2.85 \times 10^{13}}{1.33 \times 10^3}$

Monte Carlo Rate Equation Approach

	# of ECs with no RE	# of ECs with RE
Ar	1.33×10^{3}	2.85×10^{13}
\mathbf{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) ⁴¹

X-ray diffraction image - electronic & structural damage



AMO approach to the ultraintense x-ray frontier

- Complete simulation of experimental observables w/ atomistic detail (MC/MD)
 - -Monte Carlo for quantum processes -Molecular dynamics to follow ions and electrons
- Expt'l AMO observables: Ion, photoelectron, Auger, fluorescence and x-ray diffraction pattern
- Computations on large scale systems







748kcores 786 TB memory 10 PetaFLOPS

Electronic damage: from atoms to complex systems

- Electronic damage effect on biological systems considered
 - H. Quiney & K. Nugent, NatPhot (2011) frozen lattice, no Compton
 - U. Lorenz et al., PRE (2012) frozen lattice, no Compton Scattering
 - J. Slowik et al., NJP (2014) Compton Scattering considered for carbon atom
 - O. Yu. Gorobtsov *et al.*, PRE (2015) w/ Compton Scattering on

27 nm, 200,000 non-hydrogen atoms

- Frozen lattice appx (typically assumed valid for pulses <5 fs)
- Compton scattering substantial contribution in hard x-ray region 1A
- Compton scattering limits resolution to 4 Å
- C. H. Yoon *et al.*, Sci. Rep. (2016) full start-to-end XFEL simulation

No damage



30 fs w/damage

5 x 10¹¹/pulse @ 5keV 250 x 160 nm² 64 kD protein 5.3 Å half-period resolution

Simulations on a non-biological, high-Z system 7-shell Ar cluster electron and ion dynamics 10¹⁴ photons/µm² @ 8 keV



P. Ho et al. PRA (2016)

Cluster expansion - but pulse weighted average



"Original" structure recoverable despite substantial atom movement Analogous to "self-terminating Bragg gates" in XFEL crystallography

Scattering patterns for 7-shell Ar 8 keV, 30 fs and 2 fs pulses



Three major contributions: nanoparticle, free electron & Compton scattering

Reconstruction in the face of radiation damage-ideal case



7-shell Ar cluster, ~ 5nm

1415 atoms , 25470 e-8 keV, 2 fs Ideal case reconstruction Complete Q-space info in 3D available 1-D: Qmax = 3.24 Å⁻¹, dQ = 0.065Å⁻¹ No noise

Atomic level info still visible @ $10^{14}/\mu m^2$

With Miklos Tegze & Gyula Faigel



Single particle imaging: bio v non-biological systems

Compton scattering plays a smaller role



- High-Z systems require shorter pulses Inner-shell lifetimes: Carbon ~10 fs, Argon ~1 fs
- Two effects analogous to serial femtosecond crystallography: Pulse-weighted scattering allows structure recovery despite atomic movement -- self-terminating Bragg gates Distribution of radiation damage favors larger samples

Why water window flash imaging at XFELs?

Optical microscopy: superresolution imaging



Chemical labelling

Soft x-ray tomography



Cryo fixation

XFEL flash imaging has the potential to interrogate cells, viruses .. in native state with nanometer resolution – Janos Hajdu



• Resonances just below the oxygen K-edge increase ionization & decrease scattering power

• Pulse duration dependence: 10x decrease scattering power between 2 and 180 fs.

Ultra-intense x-ray interactions in molecules - CH₃I hard x-rays at 10²⁰ W/cm²



- Maximum total charge 54+
- Greater than analogous atom Xe 48+
- Greater than analogous SACLA expt which obtains 22+ w/ 50x less pulse energy

Nature June 1, 2017 Artem Rudenko Sang-Kil Son

Charge rearrangement enhanced x-ray ionization of molecules: "CREXIM"



- Sequential single photon processes dominate now with nearby reservoir of electrons.
- XMOLECULE calculates the **molecular** electronic structure unlike XMDYN
- CREXIM mechanism may be more important in larger molecules and clearly important for radiation damage

A. Rudenko et al. Nature **546** 129 (2017)

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Expt'l Strategy: spectroscopy + imaging



Simultaneous ion yield and x-ray diffractive image

Ion & electron yields sensitive to pulse integral X-ray imaging probes only during the pulse Cluster size from small angle scattering Pulse intensity from integral of scattering



T. Gorkhover ... C. Bostedt, PRL 108, 245005 (2012) 54

X-ray pump/x-ray probe capabilities at LCLS - I





Fresh-slice multicolor (Lutman *et al.*, Nat. Photon. **10**, 745 (2016)) Dechirper First undulator section Second undulator section Magnetic Dump chicane corrector Pump and probe Electron Dechirper axis photon pulses bunch To electron-beam Orbit Orbit dump correcto corrector

Expt'l strategy II: x-ray pump / x-ray probe



Evolution of ultrafast x-ray pump/x-ray probe

- Two-color -> fresh slice
- Recoil ion -> PES

April 2014: XeF_2 Recoil Ion 690 eV, 683 eV ~30 µJ, 10 fs $\Delta t = 4, 29, 54$ fs

Nov 2016: CO Photoelectron Spec 535 eV, 525 eV ~500 μ J, pump 5-10 fs, probe 5 fs $\Delta t = -10, +10, +40$ fs



More XFEL capabilities becoming available



International hard X-ray FELs here and on the horizon

SACLA Image Gallery















Summary

- AMO physics expts & theory have established fundamental understanding of the response of matter to ultraintense XFEL irradiation
 - sequential single (multi) 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 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 theoretical & computational studies promise fuller understanding of radiation damage in extended systems

Future is bright with better-characterized ultraintense x-ray lasers, multiple pulse configurations, attosecond pulses, high repetition rates ...





Heroes at AMO Control

0.0

Acknowledgements:

Argonne AMO

Christoph Bostedt Gilles Doumy Bob Dunford Phay Ho Elliot Kanter Anne Marie March Antonio Picón Steve Southworth Linda Young Andre Al Haddad Max Bucher

SLAC

John Bozek Alberto Lutman Tim Maxwell Jacek Krzywinski Dipanwita Ray Timor Osipov Ago Marinelli Zhirong Huang Paul Emma David Reis Phil Bucksbaum Tais Gorkhover Ken Ferguson

Wigner Institute Gyula Faigel Miklos Tegze **CFEL/DESY Robin Santra** Sang-kil Son Ludger Inhester Nina Rohringer KSU Artem Rudenko **Daniel Rolles** UConn Nora Berrah OSU Lou DiMauro

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Christoph Bostedt, Gilles Doumy, Bob Dunford, Phay Ho, Elliot Kanter, Bertold Krässig, Anne Marie March, Antonio Picon, Steve Southworth, Linda Young, Max Bucher, Yuan Gao, Dooyshaye Moonshiram

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



XFELs 10⁸ "brighter" than HHG sources HHG pulses 100x shorter & self-synchronized w/pump laser

From: Miao, Ishikawa, Robinson, Murnane, Science **348**, 530 (2015)⁶⁴

Compare ultraintense optical and x-ray sources

Hign-intensity at optical wavelengths

- high harmonic generation
- tabletop coherent x-ray radiation



High-intensity at x-ray wavelengths



D. Moncton, George Brown

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



-> Look for Auger line broadening on resonance

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

XLEAP

SLAC



DT = ~0.5 fs FWHM, E_{pulse} ~ 20 to 50 uJ

- 10x shorter than fastest pulse measured at LCLS.

- 4 times shorter than typical cooperation length.

From R. Schoenlein – atto @ LCLS initiative



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