UK X-FEL Science Case: Workshop on Chemical Dynamics and Energy
G.003, Urban Sciences Building, Newcastle University, December 11, 2019

Non-Linear X-ray Emission Spectroscopy on 3d Transition Metals

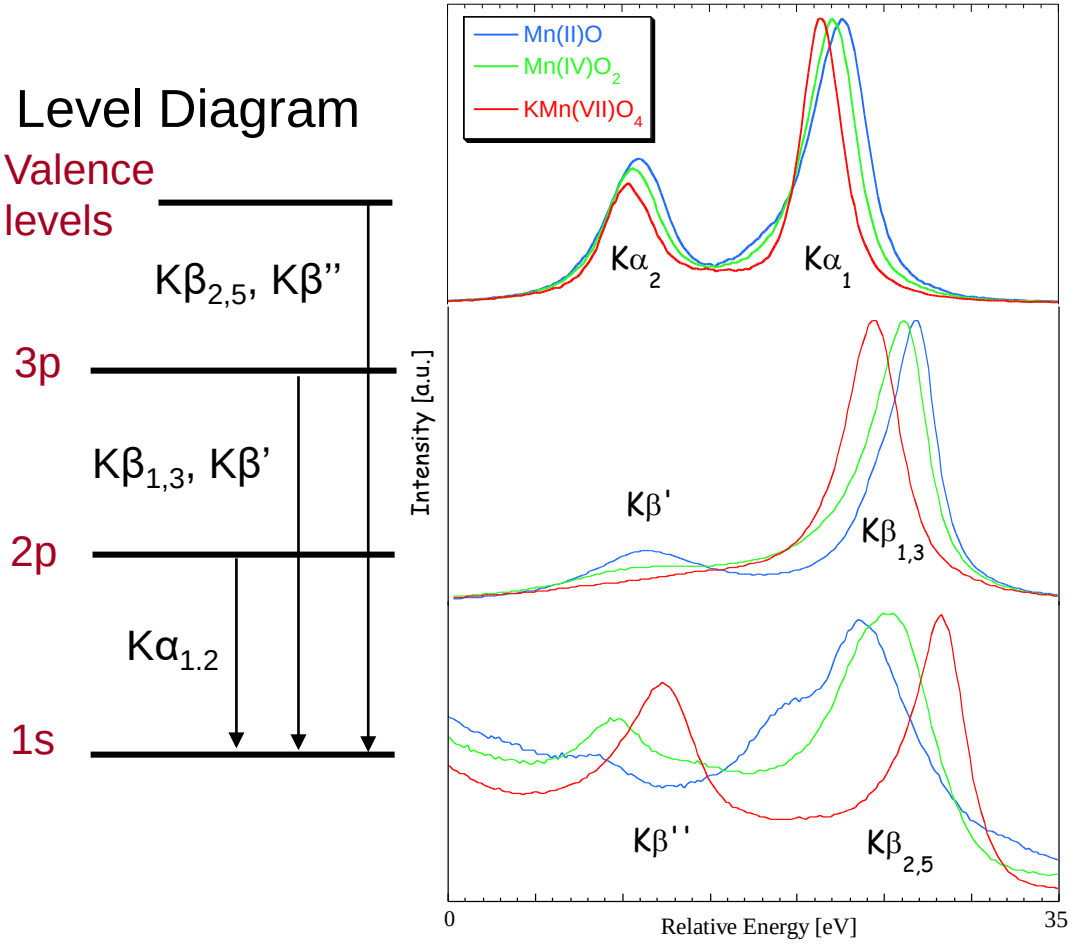
Uwe Bergmann

SLAC National Accelerator Laboratory

U.S. DEPARTMENT OF ENERGY
Office of Science

SLAC NATIONAL ACCELERATOR LABORATORY

X-Ray Emission Spectroscopy



Spin/oxidation state of transition metals

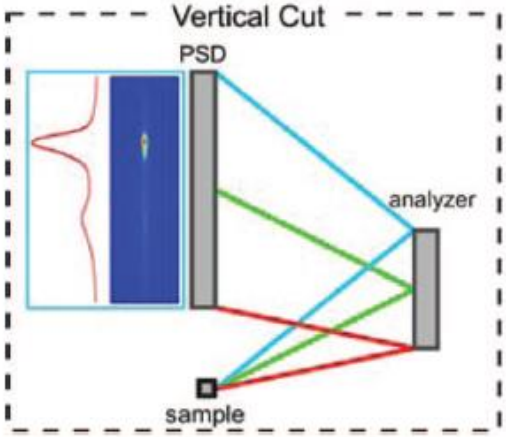
Valence orbitals: ligand type, structure, covalency, ligand protonation, etc

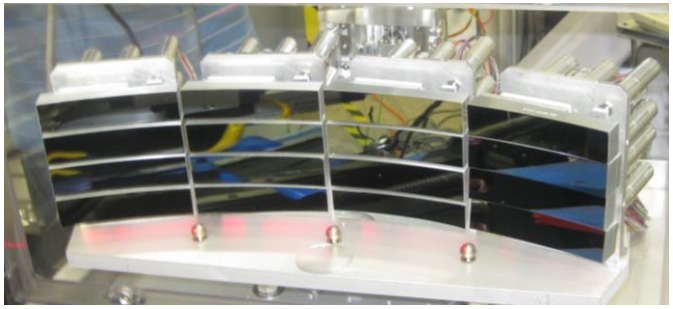
UB et al, *J Synchr Rad*. 8, 199 (2001)

For reviews see e.g.: Glatzel & UB, *Coord. Chem. Rev.*, **249**, 65-95, (2005)
Pollock & DeBeer, *Accounts of Chemical Research* (2015)

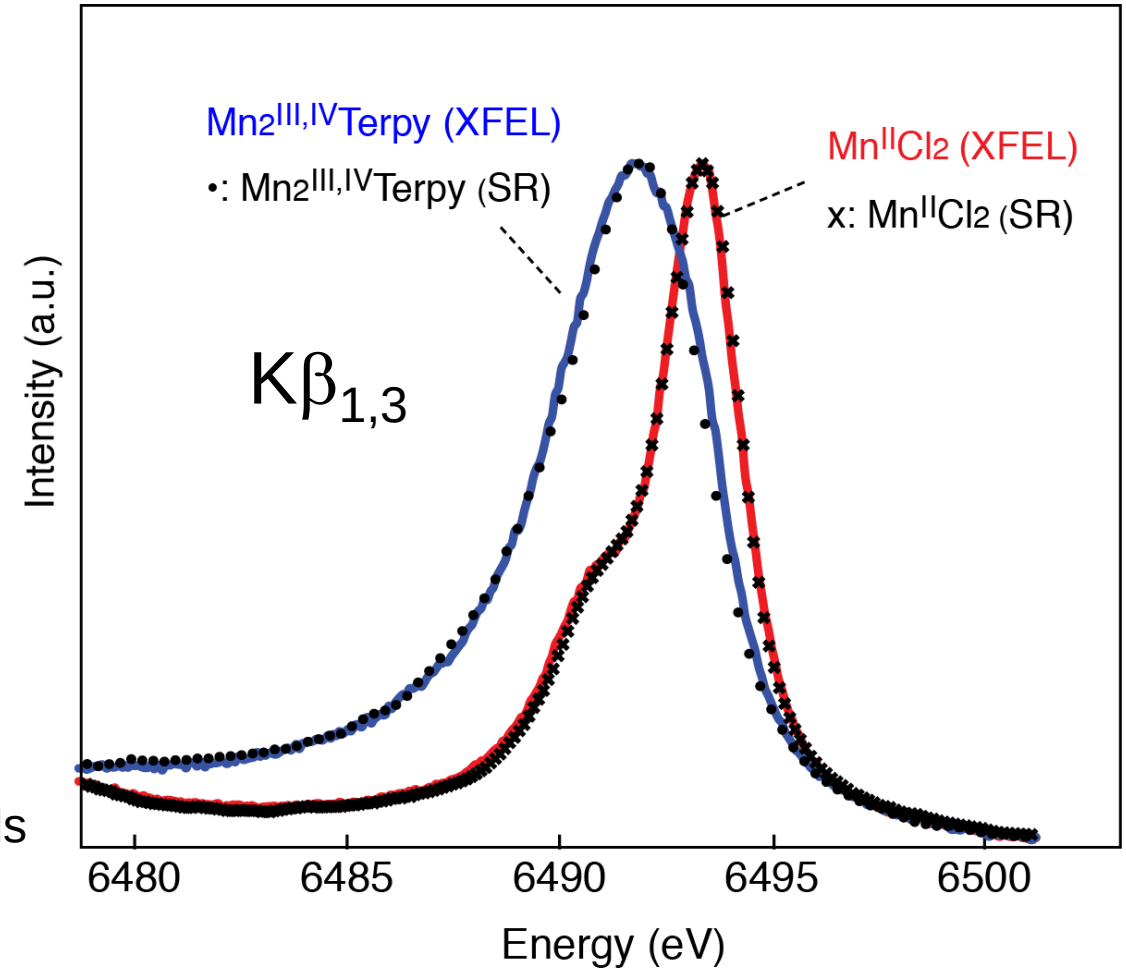
X-ray Emission Spectroscopy at an XFEL Works

SLAC





- cylindrically curved Si/Ge crystals
- 500 mm radius of curvature
- ~ 0.5 eV resolution



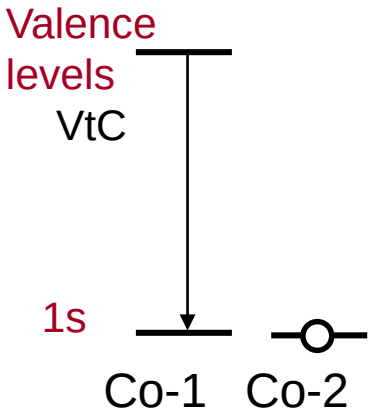
Alonso-Mori et al,
Rev. Sci. Inst., **83**, 073114 (2012)

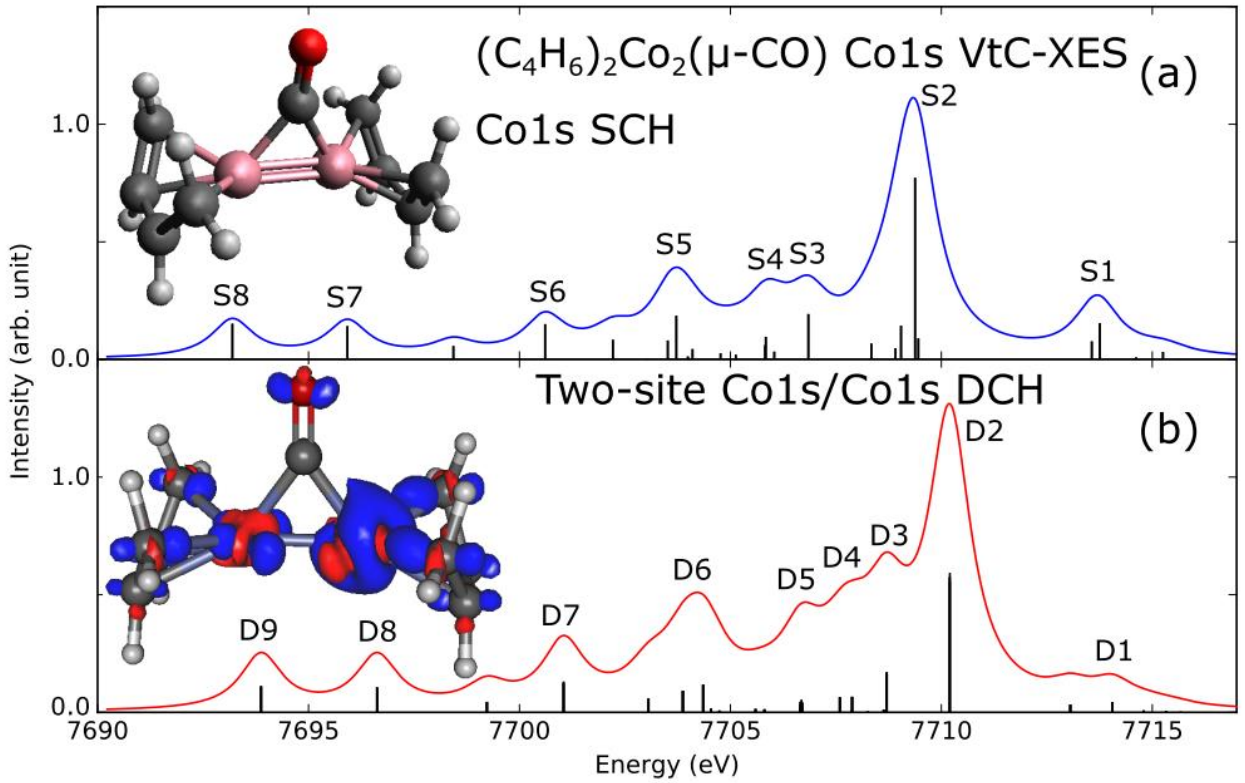
Alonso-Mori et al, *PNAS*, **109**, 19103 (2012)

Calculated Co VtC Spectra

SLAC

Level Diagram

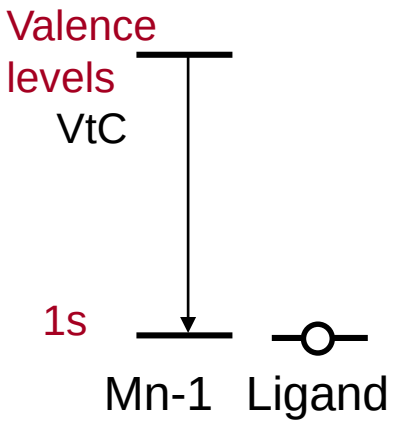


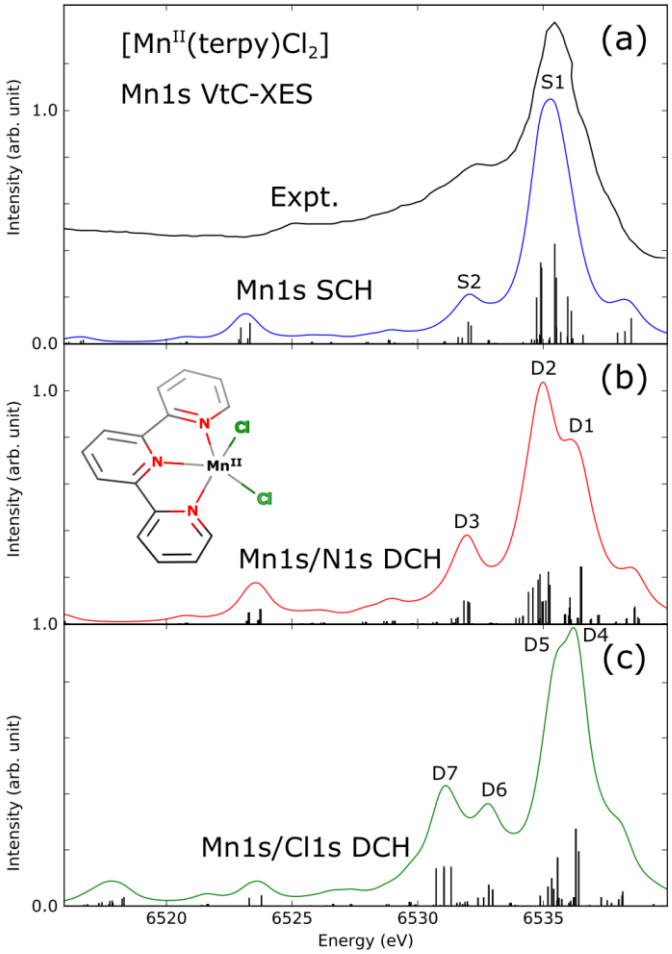


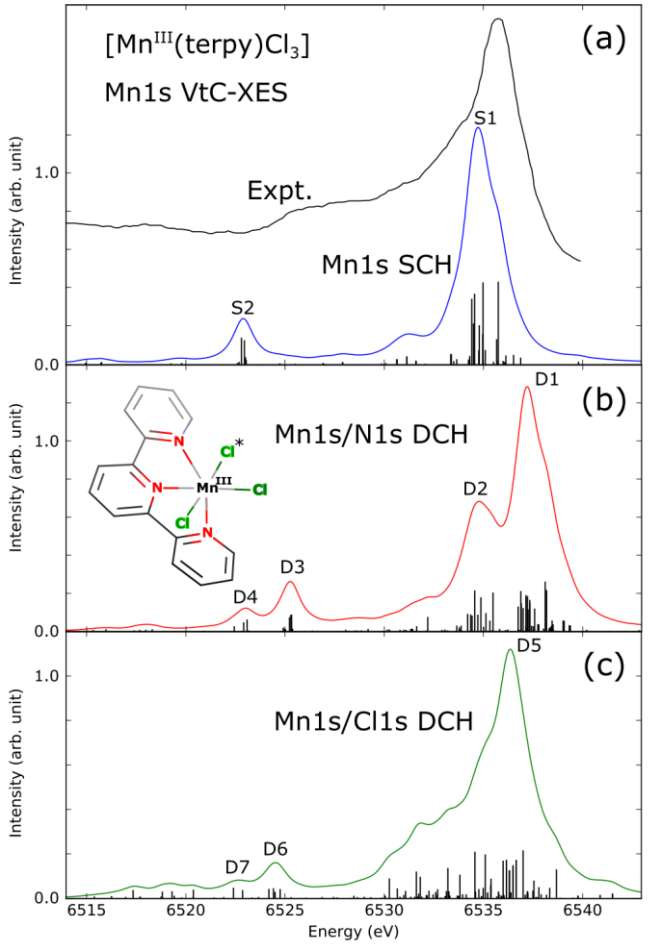
Zhang et al, *J. Chem. Phys*. **151**, 144114 (2019)

Mn Double-Core-Hole VtC Transitions

Level Diagram





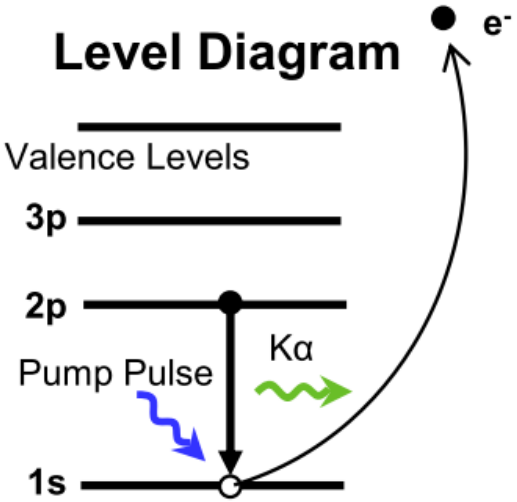


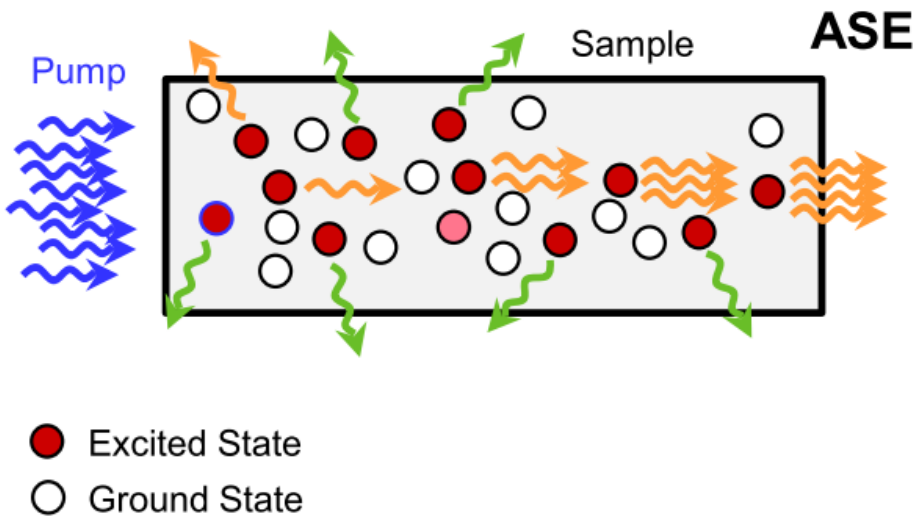
Two-color X-ray pulses would help!

Zhang et al,
J. Chem. Phys. **151**, 144114 (2019)

Schematics of Stimulated X-ray Emission

SLAC



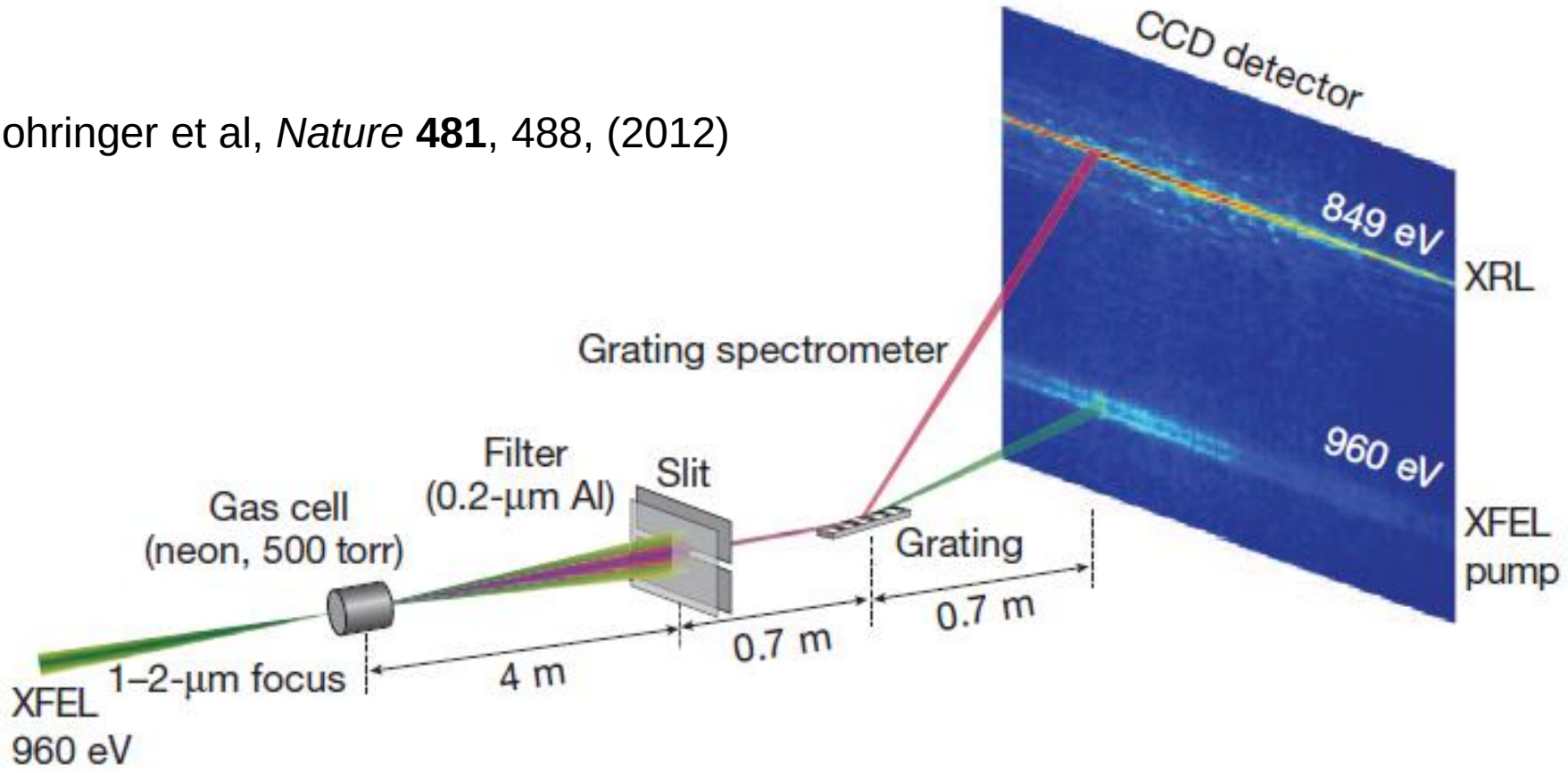


ASE: Amplified Spontaneous Emission

Stimulated Kα Emission from Neon Gas

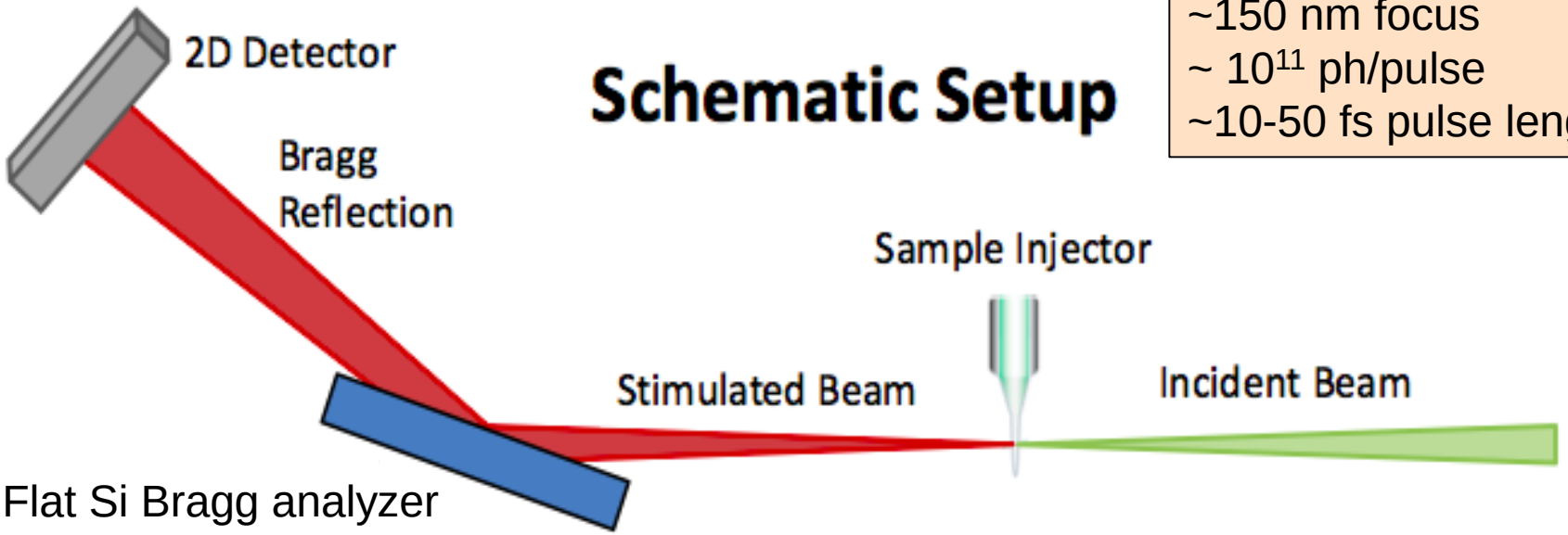
SLAC

Rohringer et al, *Nature* **481**, 488, (2012)



Hard X-rays Cu foil Yoneda et al, *Nature* **524**, 446 (2015)

Experimental Parameters and Setup



~150 nm focus
~ 10^{11} ph/pulse
~10-50 fs pulse length

Collect 100% of emission in forward direction

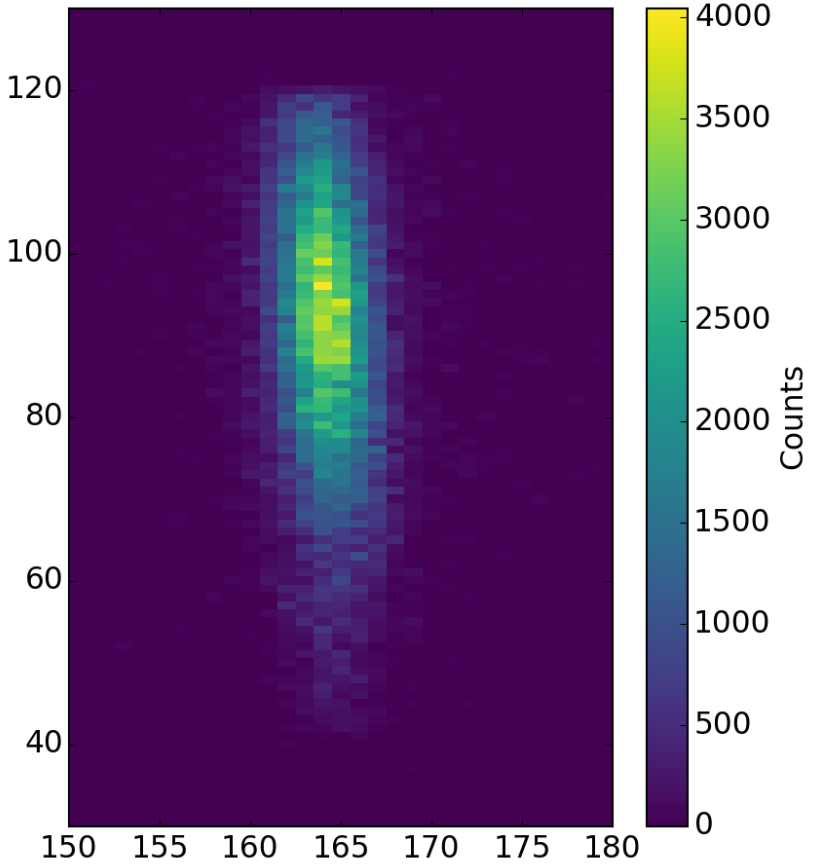
Use flat analyzer crystal – high efficiency

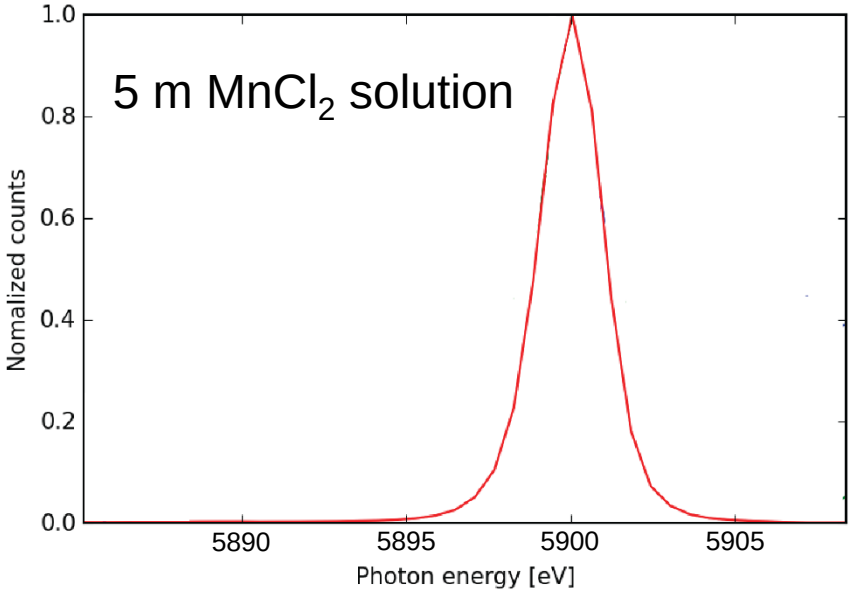
Experiments performed at LCLS CXI instrument and SACLA nanofocus instrument

Observation of Strong Lasing at 5.9 keV $K\alpha_1$ XES

SLAC

Single shot



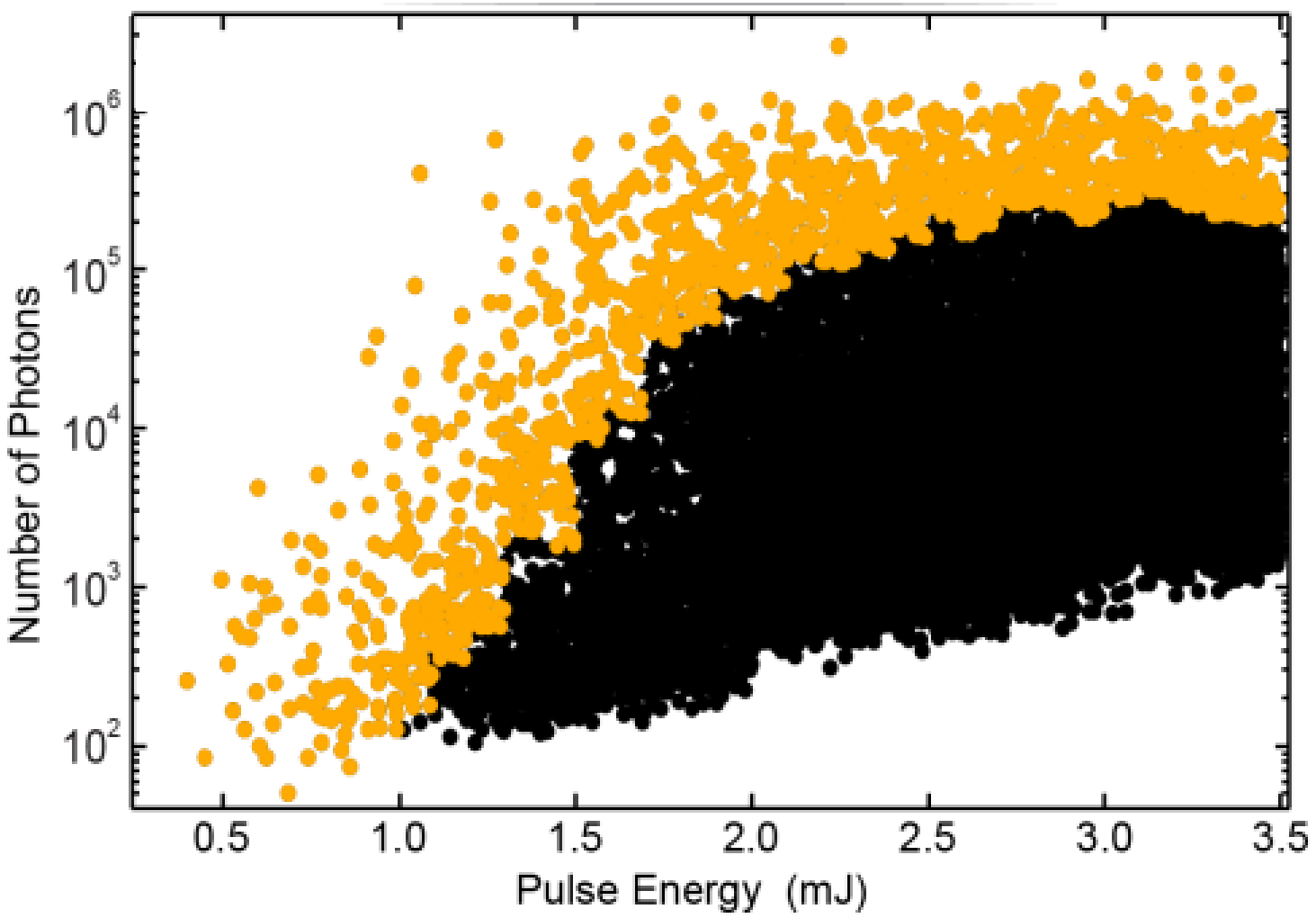


Strong gain narrowing

~10^6 Mn Kα photons

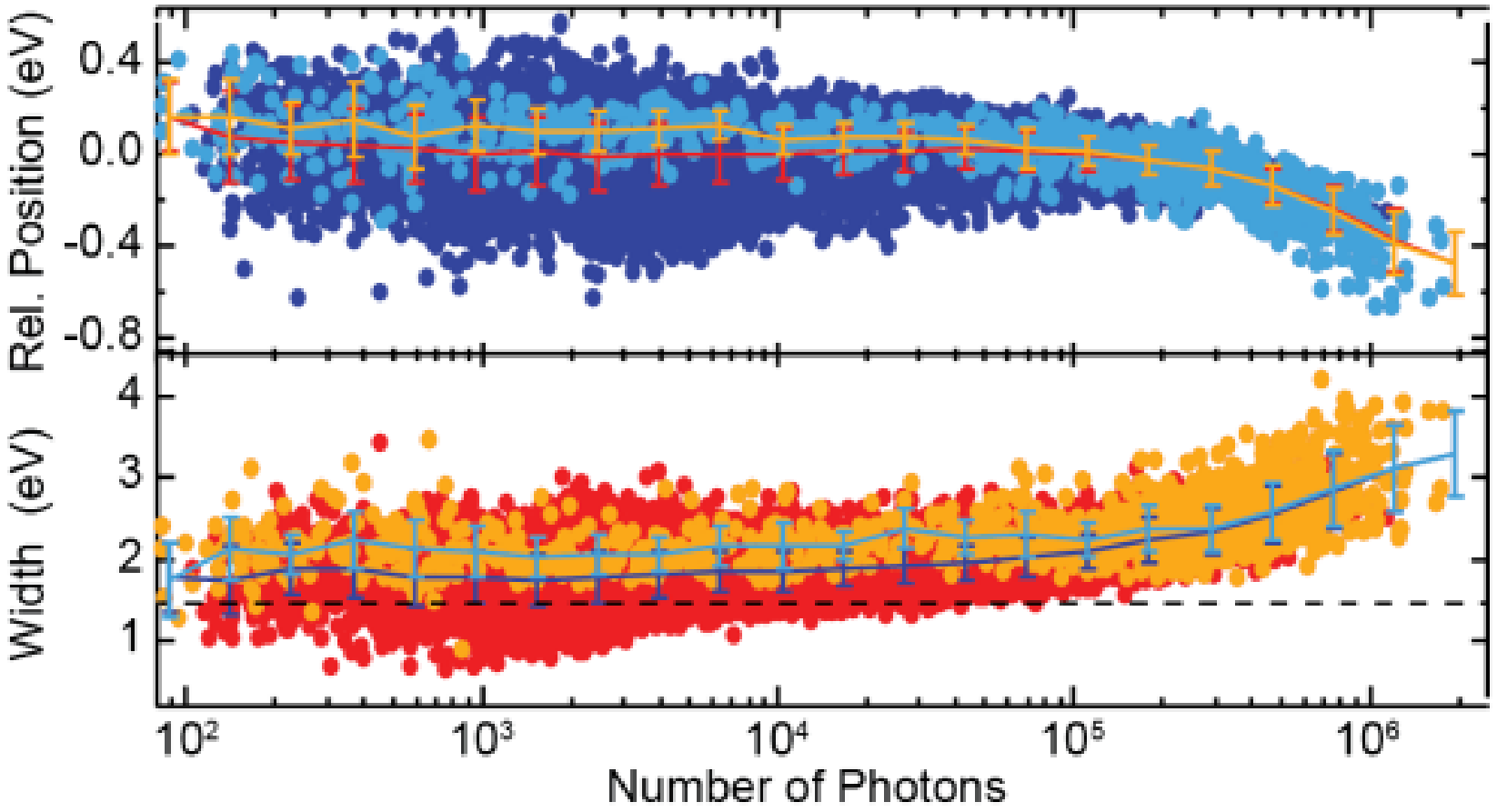
Stimulated X-Ray Emission Spectroscopy in $MnCl_2$ Solution

SLAC



Kroll et al, *Phys. Rev. Lett.*, **120**, 133203 (2018)

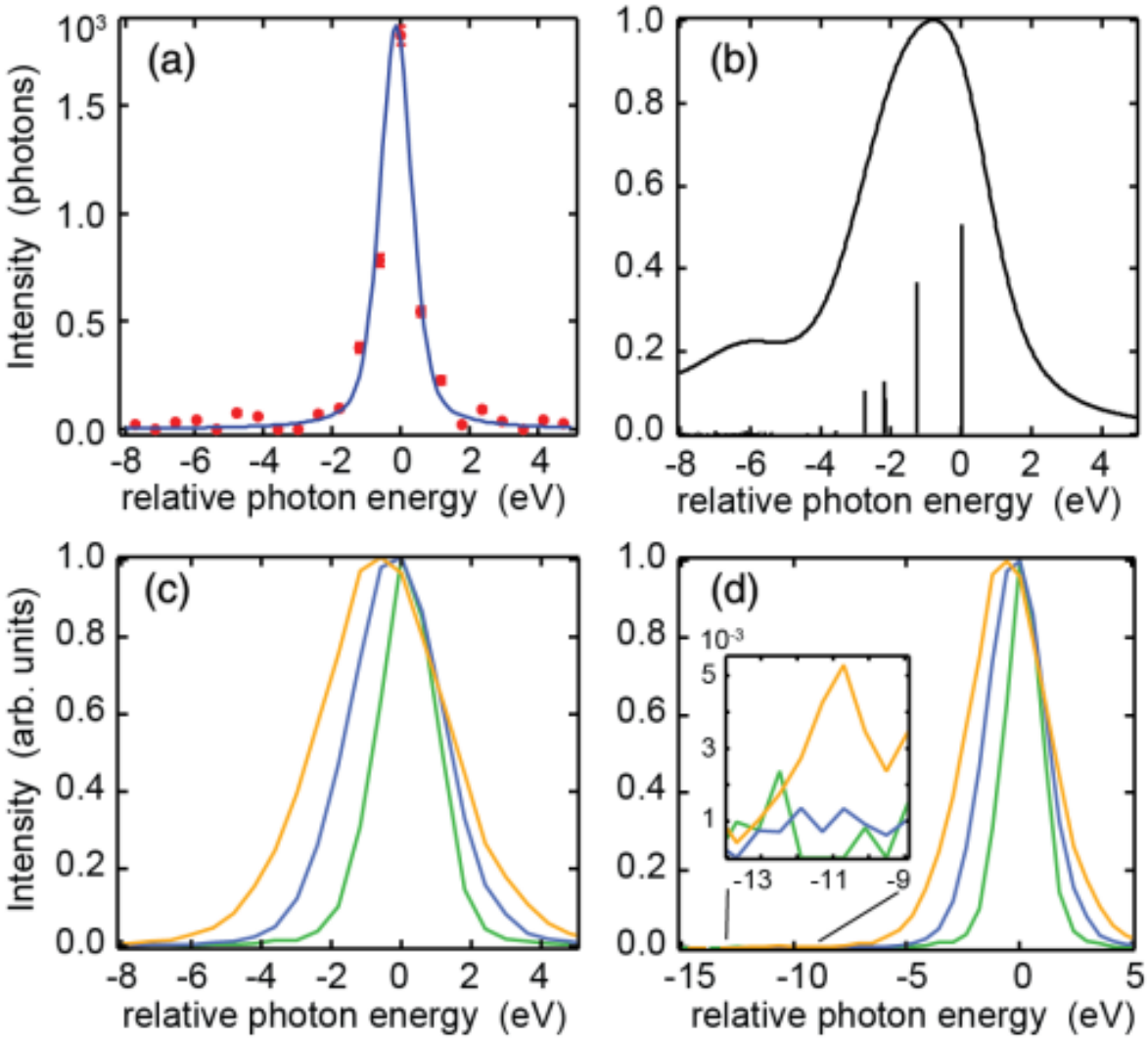
Stimulated X-Ray Emission Spectroscopy in $MnCl_2$ Solution



Kroll et al, *Phys. Rev. Lett.*, **120**, 133203 (2018)

Stimulated Kα Emission in $MnCl_2$ Solution

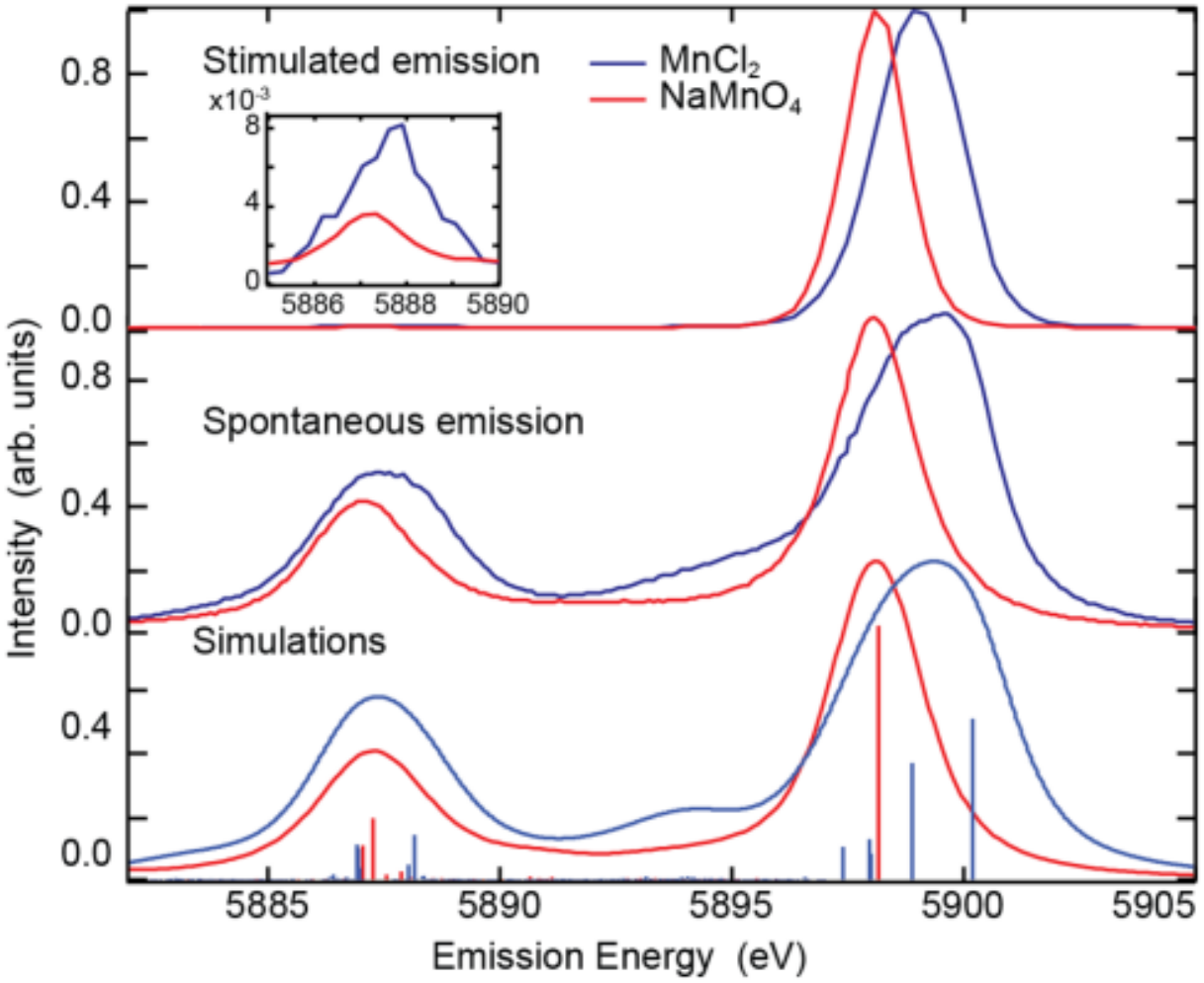
SLAC



Kroll et al, *Phys. Rev. Lett.*, **120**, 133203 (2018)

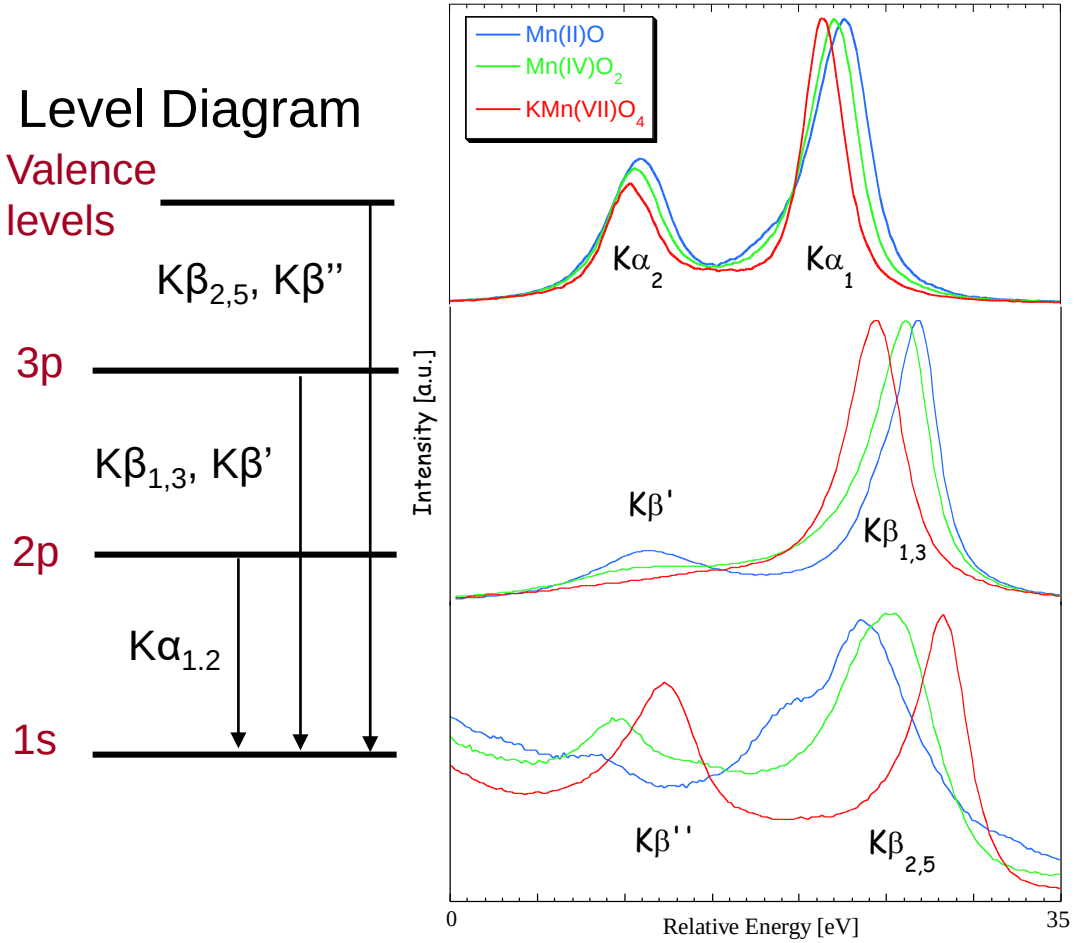
$Mn(II)Cl_2$ vs $NaMn(VII)O_4$

SLAC



Kroll et al, *Phys. Rev. Lett.*, **120**, 133203 (2018)

X-Ray Emission Spectroscopy



Spin/oxidation state of transition metals

Valence orbitals: ligand type, structure, covalency, ligand protonation, etc

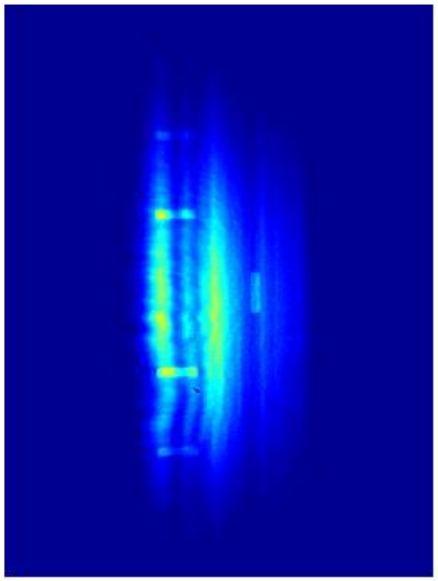
UB et al, *J Synchr Rad*. 8, 199 (2001)

For reviews see e.g.: Glatzel & UB, *Coord. Chem. Rev.*, **249**, 65-95, (2005)
Pollock & DeBeer, *Accounts of Chemical Research* (2015)

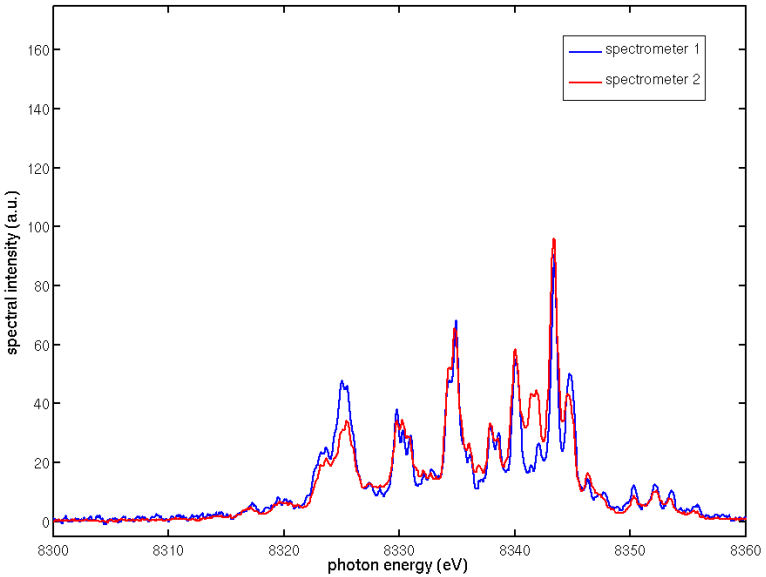
SASE Pulse Fluctuations

SLAC

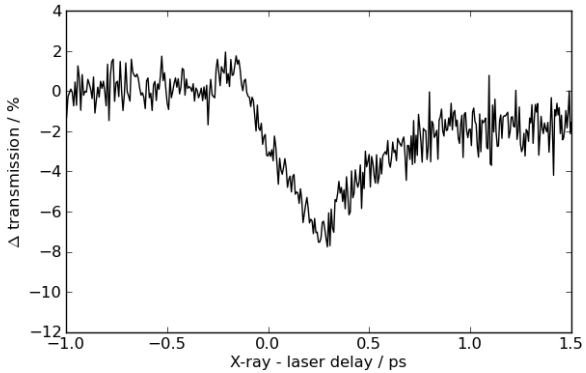
Spatial



Spectral



Temporal



XFEL experiments need to be designed to address these fluctuations

- Per pulse readout of detectors and diagnostics
- Appropriate X-ray optics and spectroscopy methods

An X-ray Laser Oscillator

SLAC

Design and characteristics of an X-ray Laser Oscillator

A. Halavanau,[1] A. Benediktovitch,[2] A.A. Lutman,[1] D. DePonte,[1]
D. Cocco,[3] N. Rohringer,[4,5] U. Bergmann,[1] and C. Pellegrini[1]

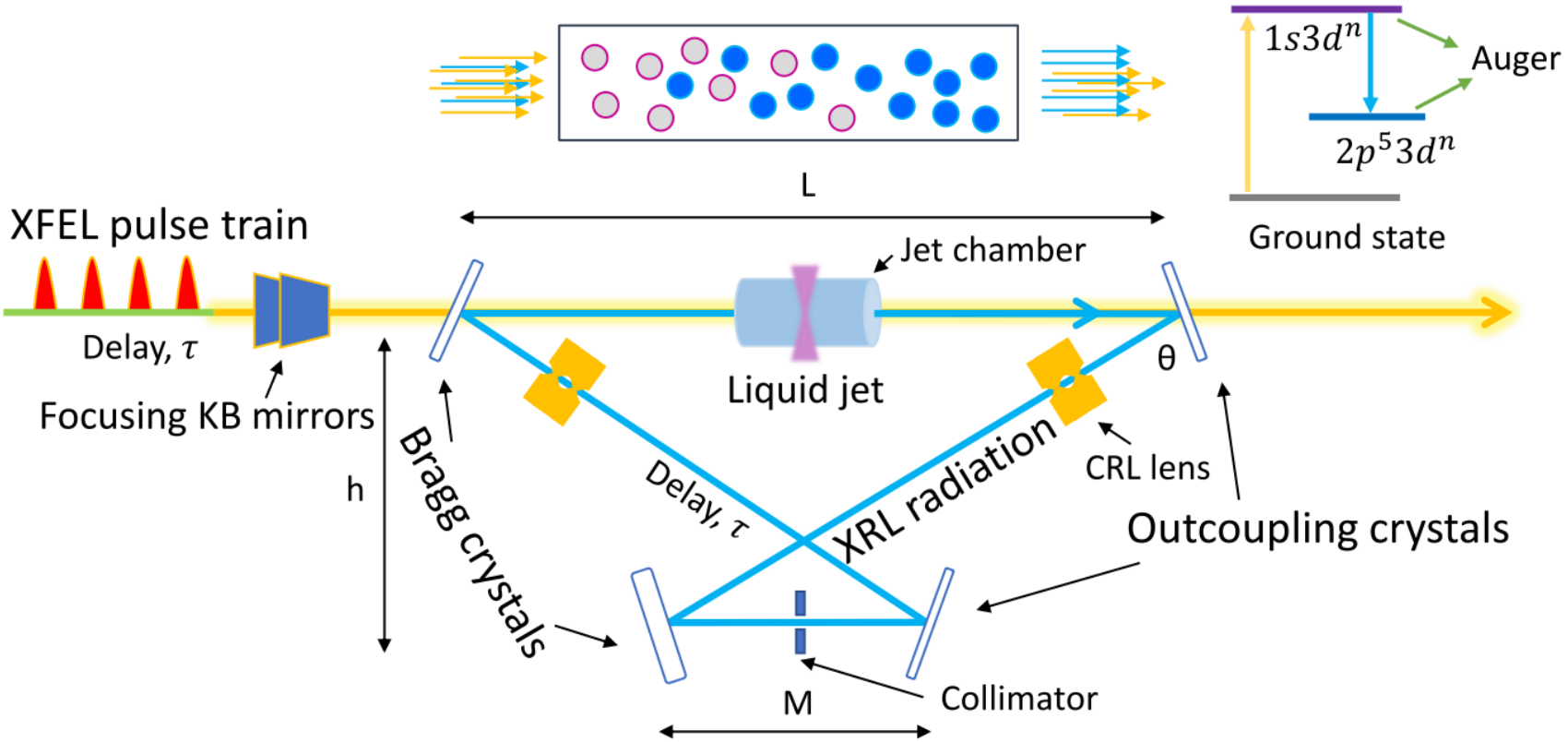
[1] *SLAC National Accelerator Laboratory, Menlo Park, CA 94025, USA*
[2] *Center for Free Electron Laser Science, DESY, Hamburg 22761, Germany*
[3] *Lawrence Berkeley National Laboratory, Berkeley, CA 94720, USA*
[4] *Center for Free Electron Laser Science, DESY, Hamburg, 22607, Germany*
[5] *Department of Physics, Universität Hamburg, Hamburg 22761, Germany*

Manuscript submitted

ArXiv link: https://arxiv.org/abs/1912.03554

An X-ray Laser Oscillator

SLAC

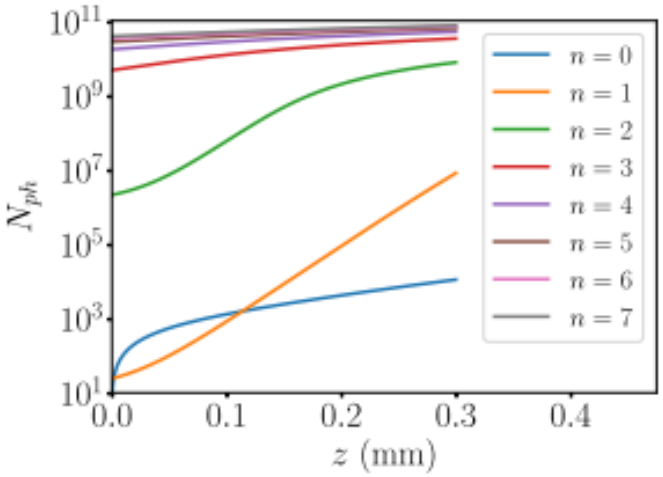


ArXiv link: https://arxiv.org/abs/1912.03554

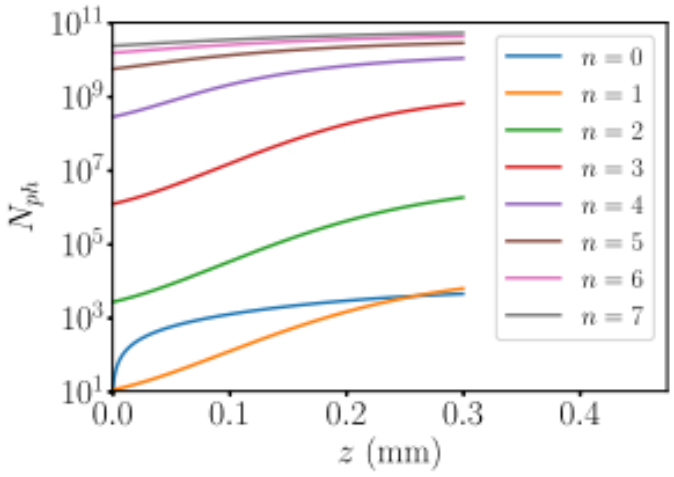
An X-ray Laser Oscillator

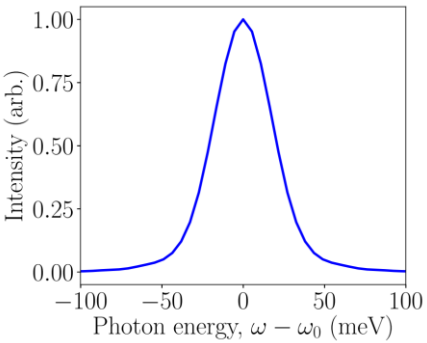
SLAC

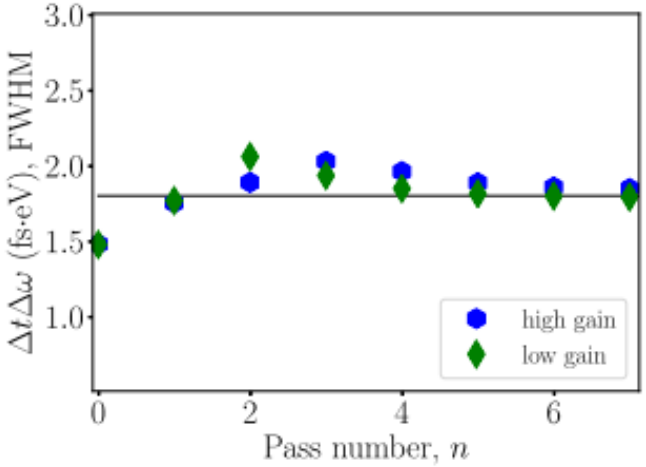
High Gain Regime



Low Gain Regime







- Transform limited Gaussian pulses
- ~5*10^{10} photons/pulse @ 8 keV
- 50 meV bandwidth at 36 fs pulse length (corresponding to 1.8 eV*fs)

ArXiv link: https://arxiv.org/abs/1912.03554

Wish List for the Future

High peak power, stability

Need to know as much as possible about incoming pulse

- Monochromatic seed beam ideally with self-seeding
- Shot-by-shot upstream spectrometers for seed pulse (and eventually for pump pulse)
- Intensity monitor to know photon # after KB mirror (non-destructive)
- Shot-by-shot beam profile, wave front sensor for focus , temporal diagnostics
- Angular streaking to measure both spectral and temporal pulse (‘cookie box’ for soft x-rays)
- ‘Fast’ switching from seeding to non-seeding (minutes instead of hours)

Collaborators

SLAC

SLAC
Roberto Alonso-Mori
Franklin Fuller
Marc Guetg (now Eu-XFEL)
Aliaksei Halavanau
Thomas Kroll
Alberto Lutman
Agostino Marinelli
Claudio Pellegrini
Bob Schoenlein
Dimosthenis Sokaras
Clemens Weninger
Yu Zhang

CXI Instrument at LCLS
Andy Aquila
Sébastien Boutet
Dan DePonte
Jason Koglin
Jake Koralek
Mengning Liang

SACLA
Ichiro Inoue
Yuichi Inubushi
Tetsuo Katayama
Taito Osaka
Kensuke Tono
Makina Yabashi

U Wahsington
Munira Khalil

PNNL
Niranjan Govind

Lawrence Berkeley Lab
Jan Kern
Vittal Yachandra
Junko Yano

Max-Planck Hamburg
Andrei Benediktovitch
Laurent Mercadier
Nina Rohringer

Tokyo
Yurina Michine
Hitoki Yoneda

Rice University
Pulickel M. Ajayan
Babu Ganguli
Devashish Salpekar
Farheen N. Sayed

Thanks to SLAC and SACLA staff, DOE for funding