Soft X-ray scattering instrumentation at LCLS-II

IXS2019 Meeting

June 26th 2019

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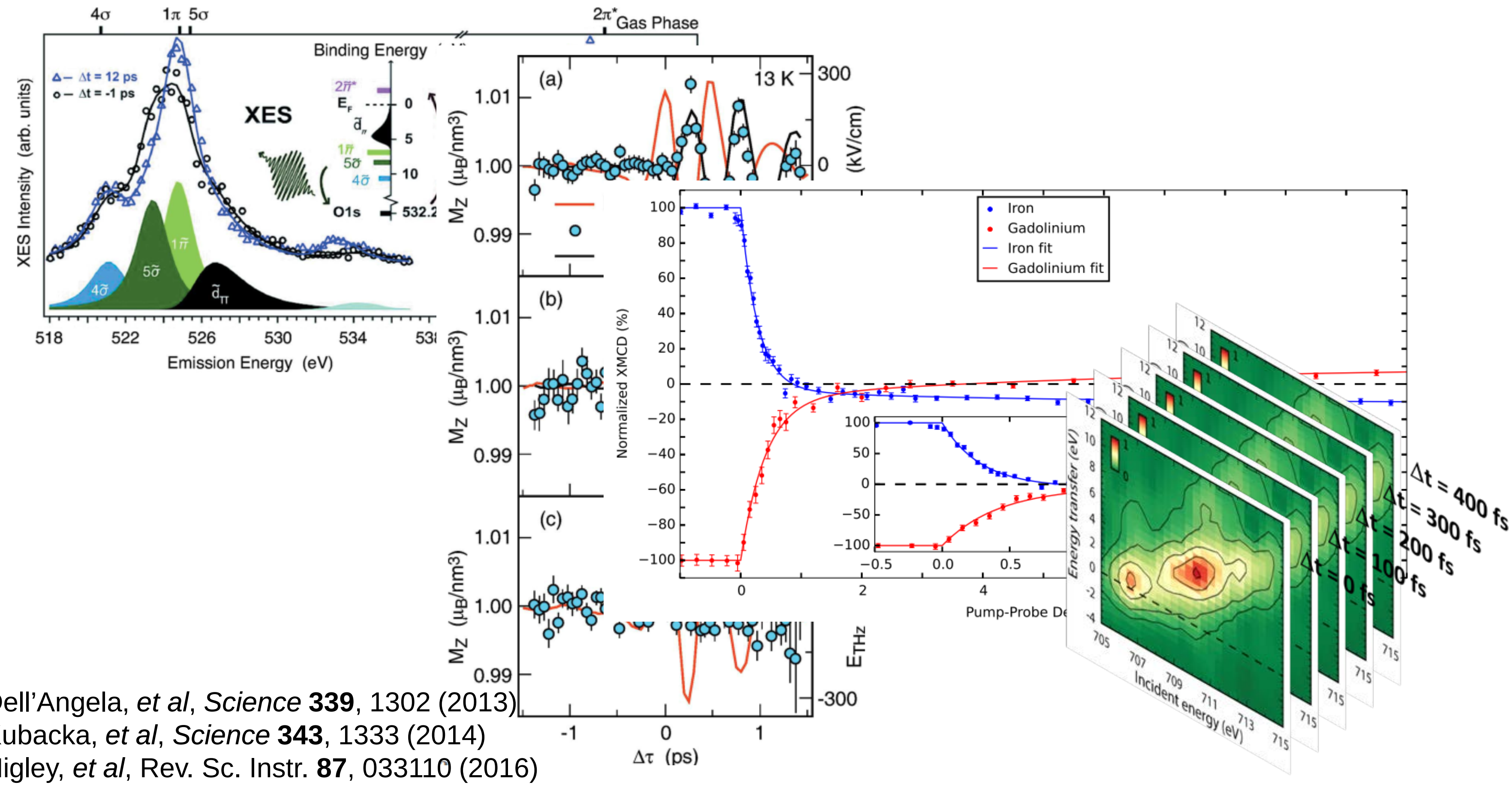
U.S. DEPARTMENT OF ENERGY
Office of Science

SLAC NATIONAL ACCELERATOR LABORATORY

Outline

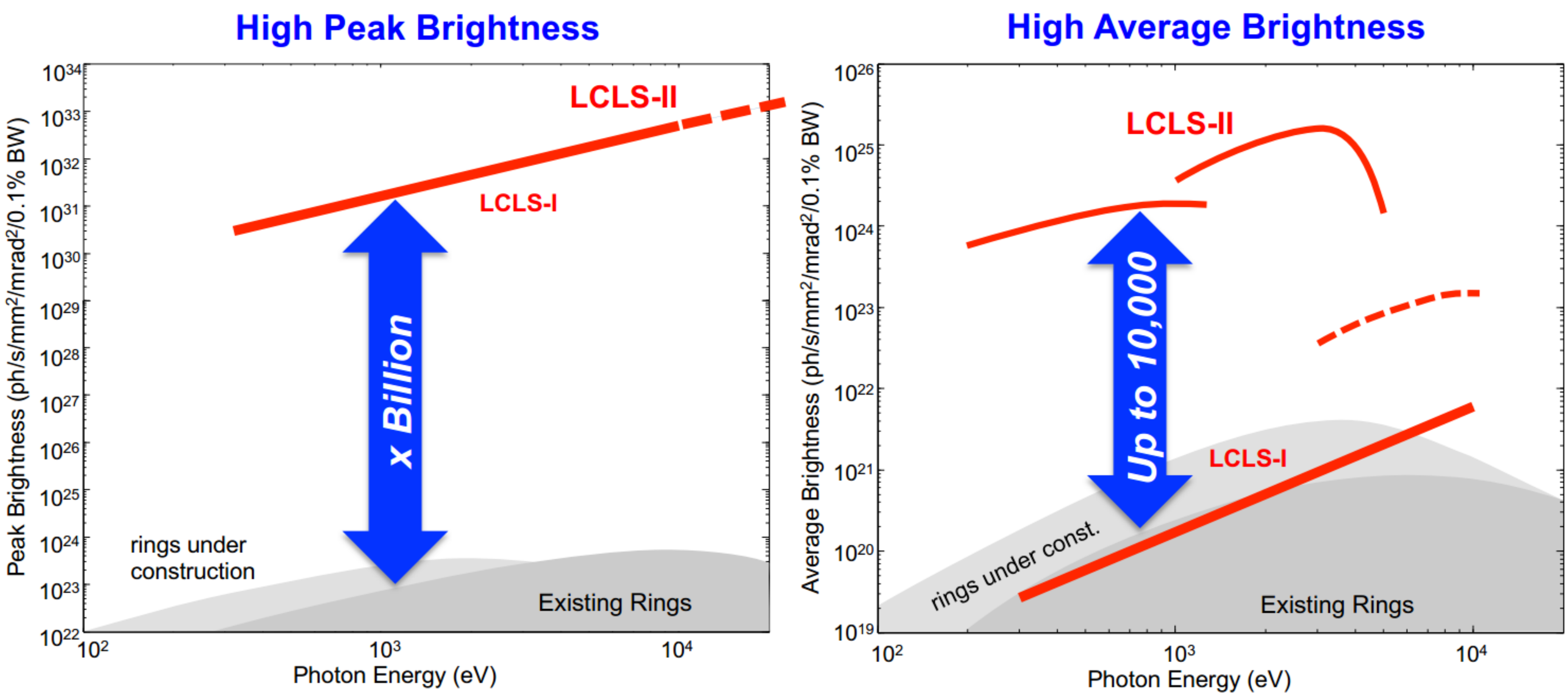
- Update on LCLS-II
- chemRIXS: spectroscopy of samples in solution phase
- qRIXS: high-resolution spectroscopy of correlated materials
- PAX

Highlights from SXR @ LCLS



Dell'Angela, *et al*, *Science* **339**, 1302 (2013)
Kubacka, *et al*, *Science* **343**, 1333 (2014)
Higley, *et al*, Rev. Sc. Instr. **87**, 033110 (2016)
Wernet *et al*, *Nature* **520**, 78 (2015)

LCLS-II



LCLS-II provides:
- **Repetition rate**
- **Stability**

Deciphering the intramolecular electron transport on the femtosecond timescale

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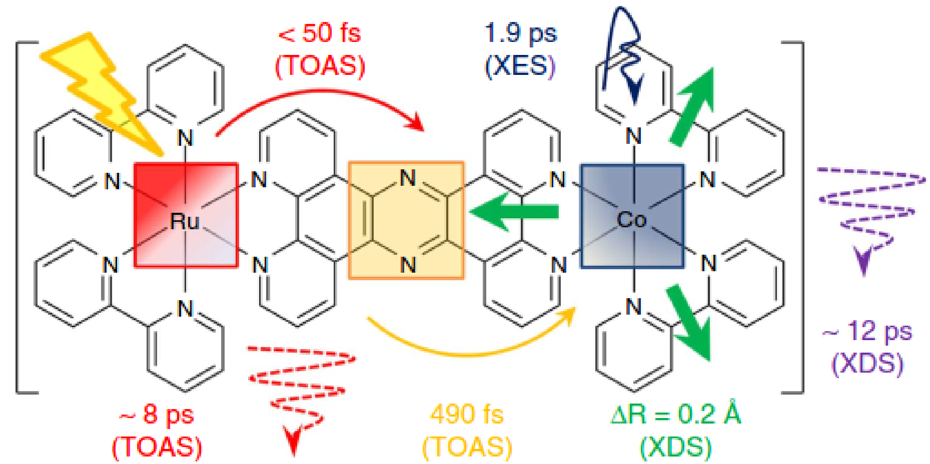
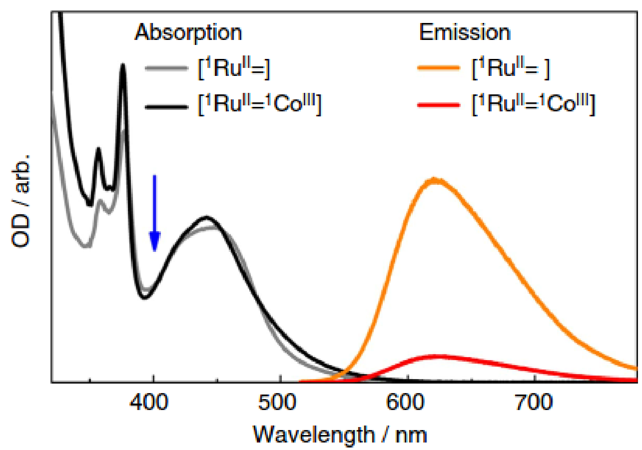
- **Key steps**:

1. Selective photoexcitation at the Ru site
2. XAS, XES and RIXS at Ru, N and Co to map (un)occupied DOS





S. Canton, *et. al* Nat. Comm. 6:6359, 2015

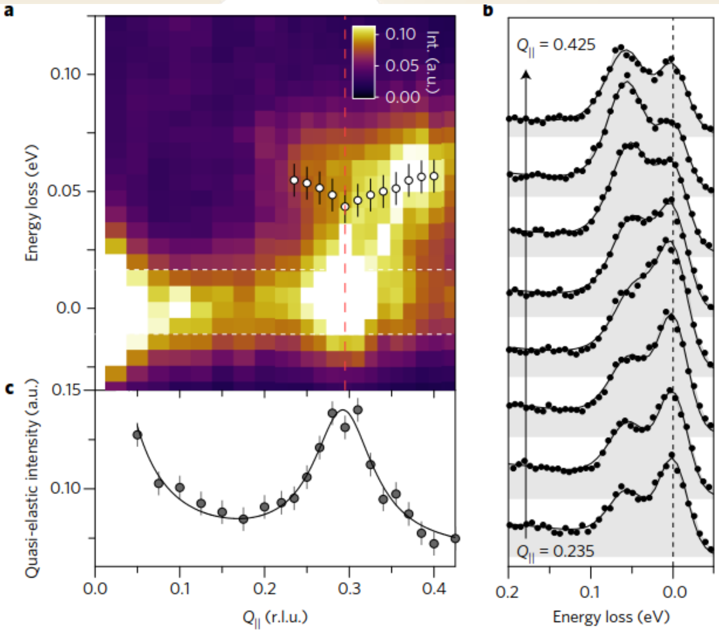
- **Key requirements**:

1. High throughput moderate resolution RIXS spectrometer
2. Tunable visible excitation
3. Fourier-transform-limited optical and x-ray pulses

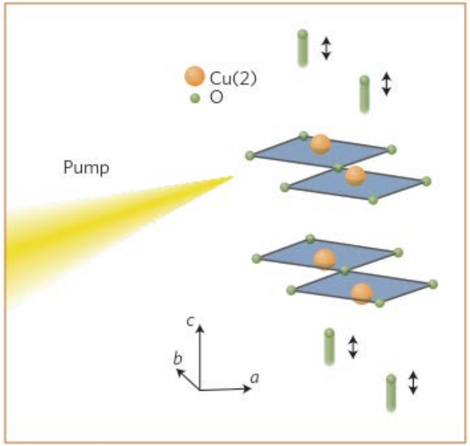
What is the dynamics of light-enhanced coherent transport in cuprates?

- **Key steps**:

1. Characterize the spectrum of excitations at different temperature
2. Optically drive lattice distortion



L. Chaix, *et. al*, Nat. Phys. 2017

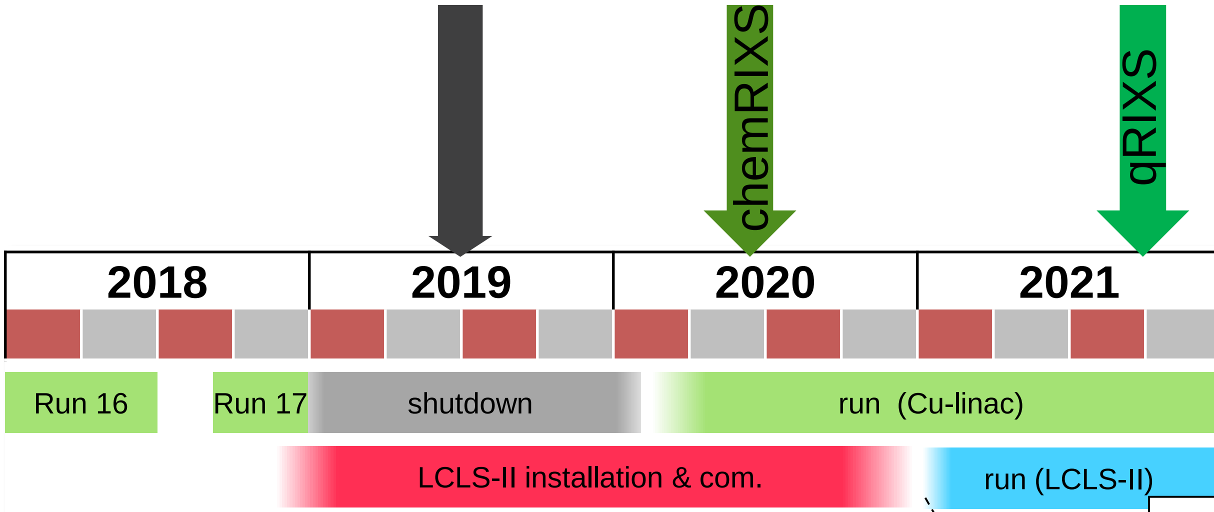


N. P. Armitage, Nat. Mater. 13, 665

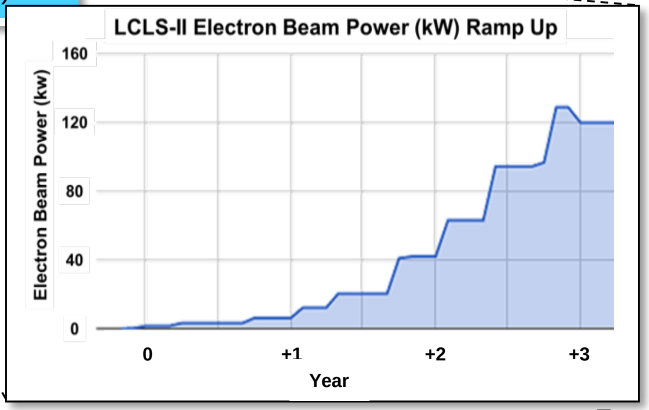
- **Key requirements**:

1. High energy resolution *q*-dependent RIXS spectrometer
2. Tunable mid-IR excitation
3. Fourier-transform-limited optical and x-ray pulses

LCLS-II – Notional Timeline



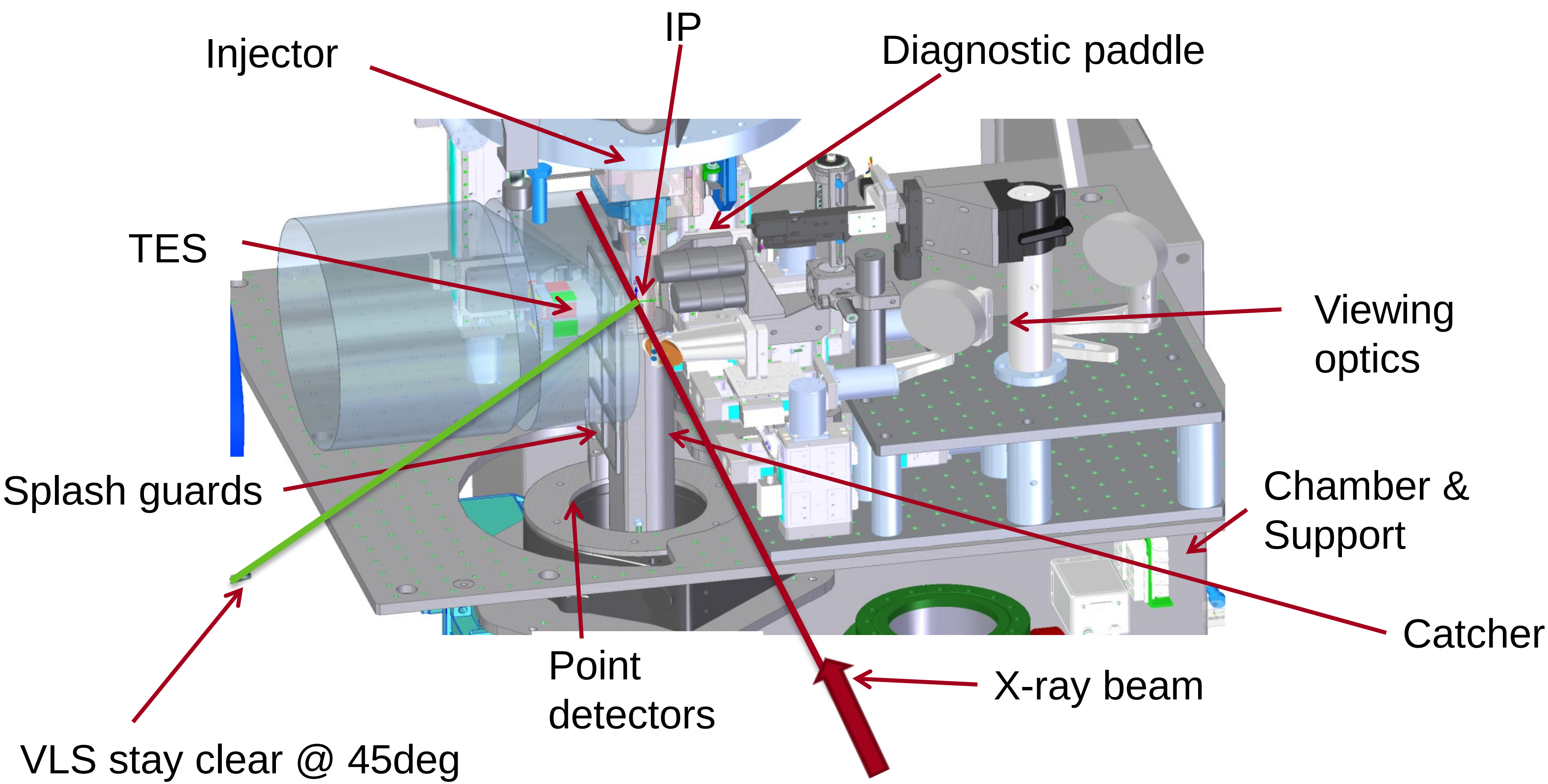
LCLS-II first light *ca.* early 2021



e-beam power ramp-up…

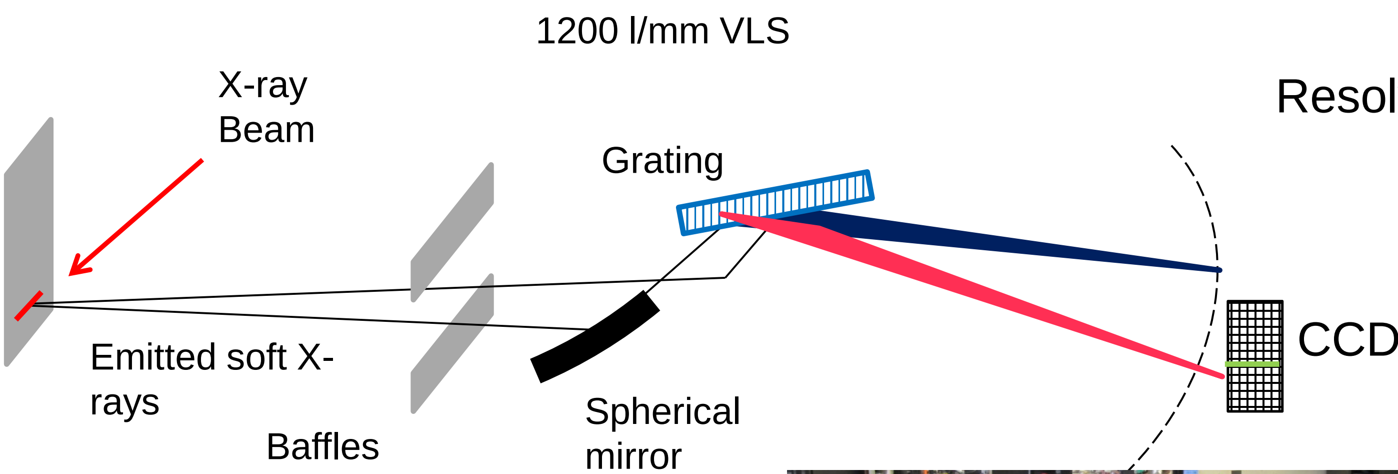
$\Rightarrow$ ramp-up in X-ray rep. rate and/or X-ray pulse energy (i.e. average X-ray power)

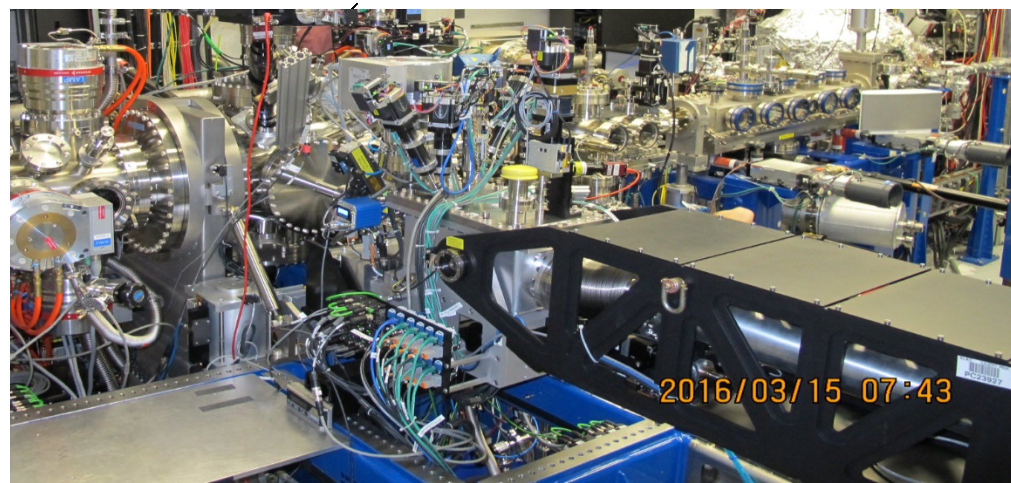
chemRIXS: major components



chemRIXS: portable RIXS spectrometer

Resolving power ~2000

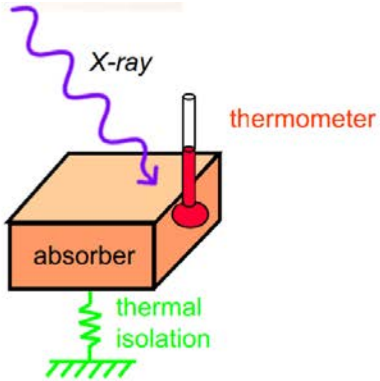


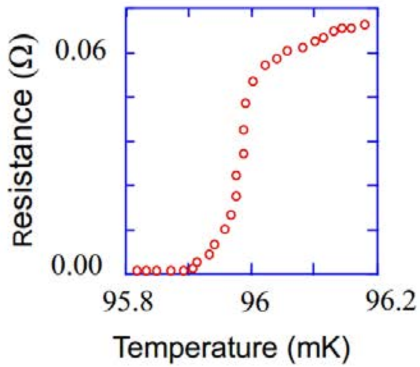


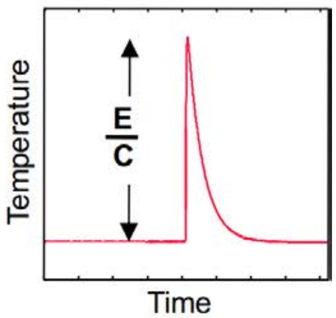
Acknowledgments:

- Project lead: Bill Schlotter
- Design: LBNL -> Yi-De Chuang, Zahid Hussain
- Design SLAC -> Daniele Cocco, Michael Holmes (CAM)
- Funding: **Nora Berrah**

Transition Edge Sensor Spectrometer

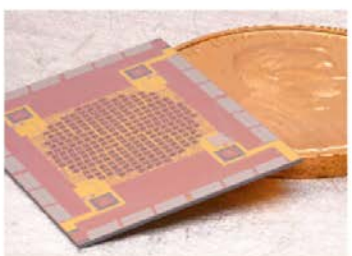






Capabilities:

- Large collection efficiency
- Fast readout
- 1 eV resolution demonstrated
- 0.5 eV targeted



TES spectrometers provide a unique combination of spectral resolution, efficiency, and broadband coverage

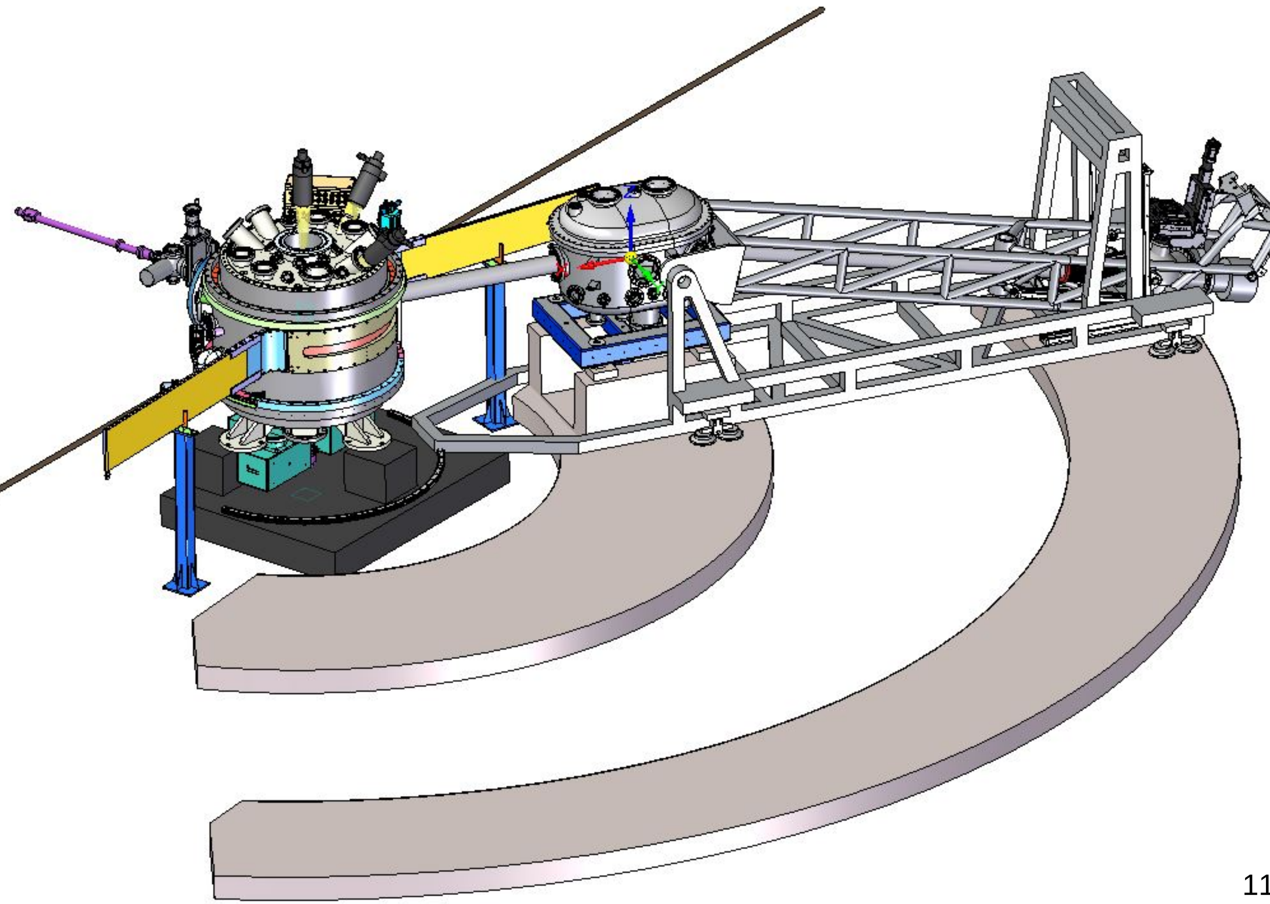
CDMS heritage

$$\Delta E \propto \sqrt{k_B T E_{max}}$$

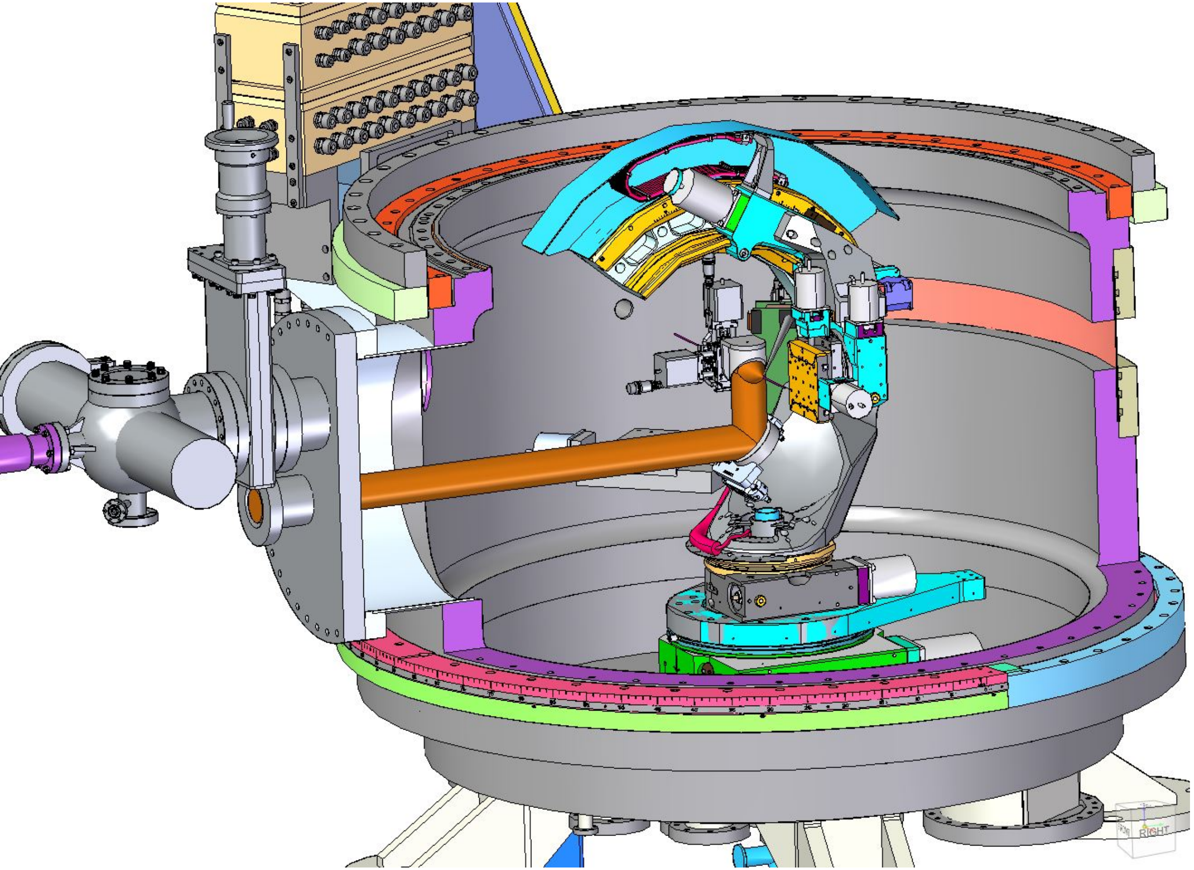
qRIXS: spectrometer arm

Capabilities:

- Scattering angle 40 – 150 deg
- 1 chamber for parabolic mirror and gratings
- Downward deflection
- Polarimeter
- Current collaboration with BNL team to define the optical layout



qRIXS: sample chamber



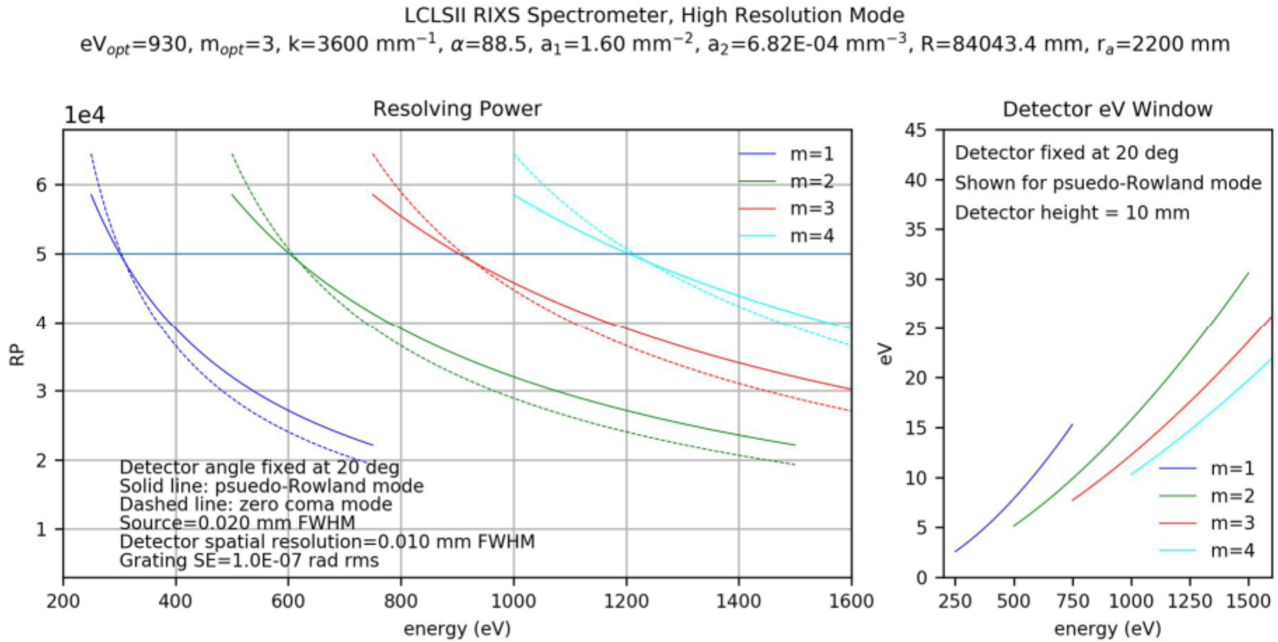
Capabilities:

- Cryostat, 6 DOF, 20 K
- Flexible long-wavelength in-coupling
- In-vacuum detectors for peak finding and XAS in fluorescence yield mode

Parameter optimization

$r_a = 2200$ mm
α(psuedo-Rowland) = 88.5°
$k_0 = 3600$ mm^{-1}
$a_1 = 1.6003$ mm^{-2}
$a_2 = 6.818\times10^{-4}$ mm^{-3}
$a_3 = 2.70\times10^{-7}$ mm^{-4}
R = 84043.4 mm
optimized at 930 eV, m=+3, r_b(930 eV) = 3469.03 mm, blaze angle = 4.2°

Courtesy of Joe Dvorak



XPCS X-ray Photon Correlation Spectroscopy

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LCLS-II Science Opportunity:

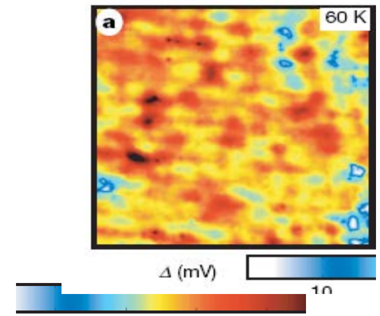
- Connect spontaneous fluctuations, dynamics and heterogeneities on multiple length- and time-scales to material properties

Significance & Impact

- Electronic structure dynamics
- Chemical heterogeneity/dynamics
- Phase transitions

LCLS-II Strengths & Challenges

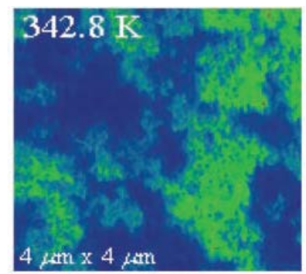
- High rep rate
- Coherence (energy resolution) near FT limit
- Sub-Nanosecond 2-pulse technique (FEL)



SC Gap in BSCCO

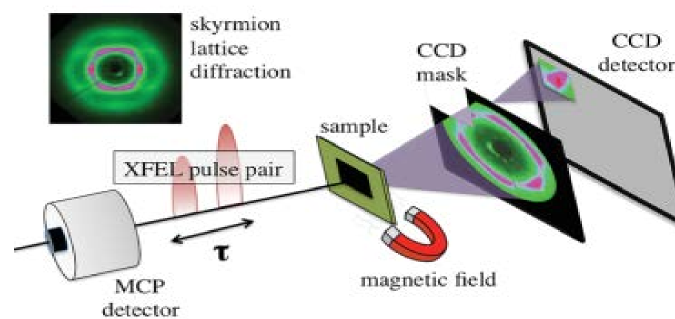
Gomes et al.,
Nature **447**, 569 (2007)
(Yazdani Group, Princeton)

Δ(meV)



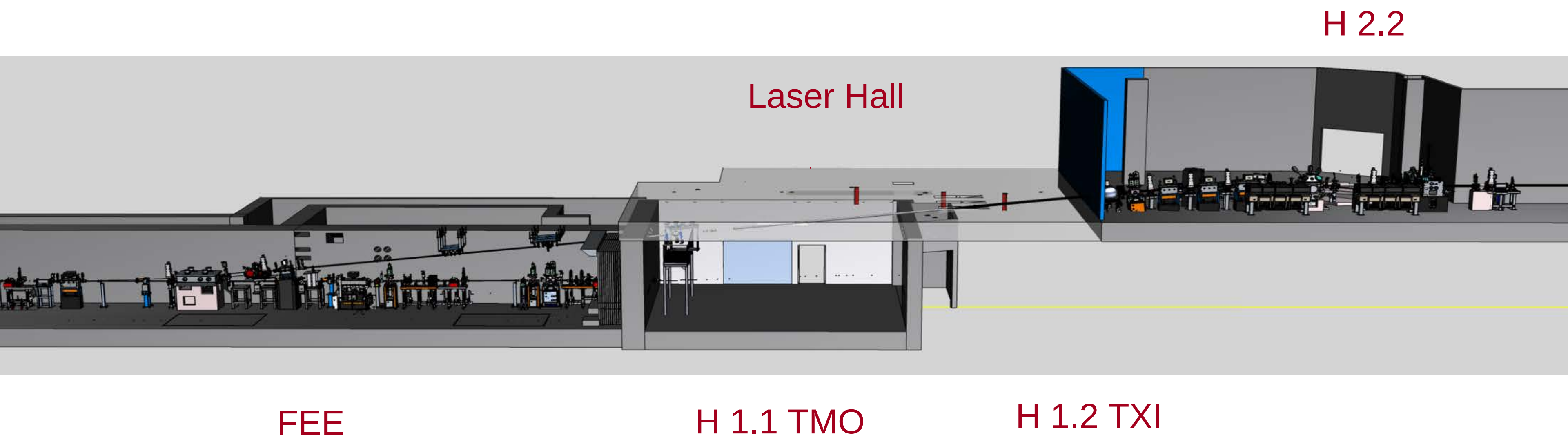
Metal-Insulator Transition in VO_2

Qazilbash et al.,
Science **318**, 1750 (2007)
(Basov Group, UCSD)



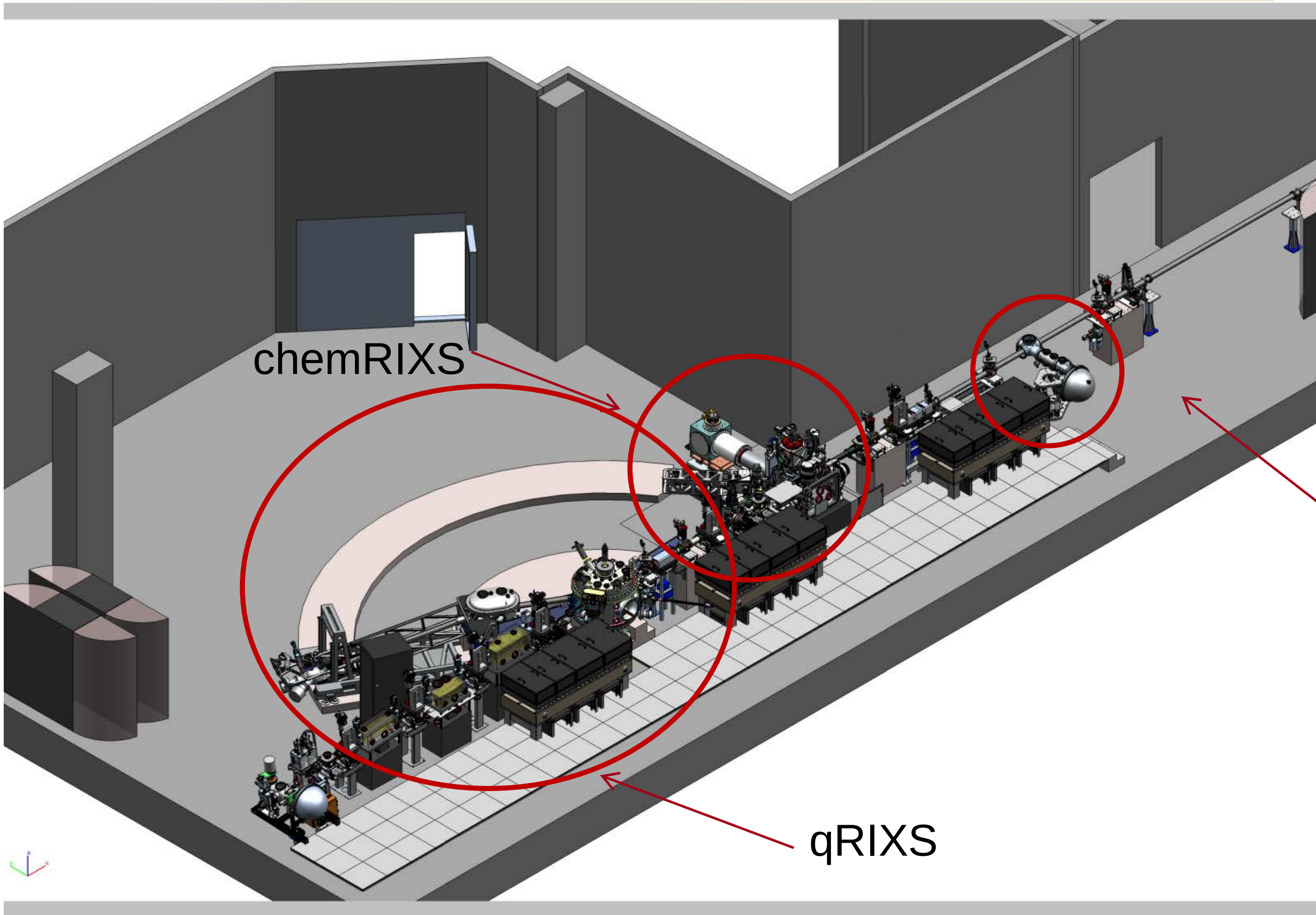
Seaberg *et al*, *PRL* **119**, 067403 (2017)

NEH 2.2 Beamline layout



Front End Enclosure & Near Experimental Hall

NEH 2.2 Hutch complete, FY 2023



Challenges of high resolution soft x-ray RIXS

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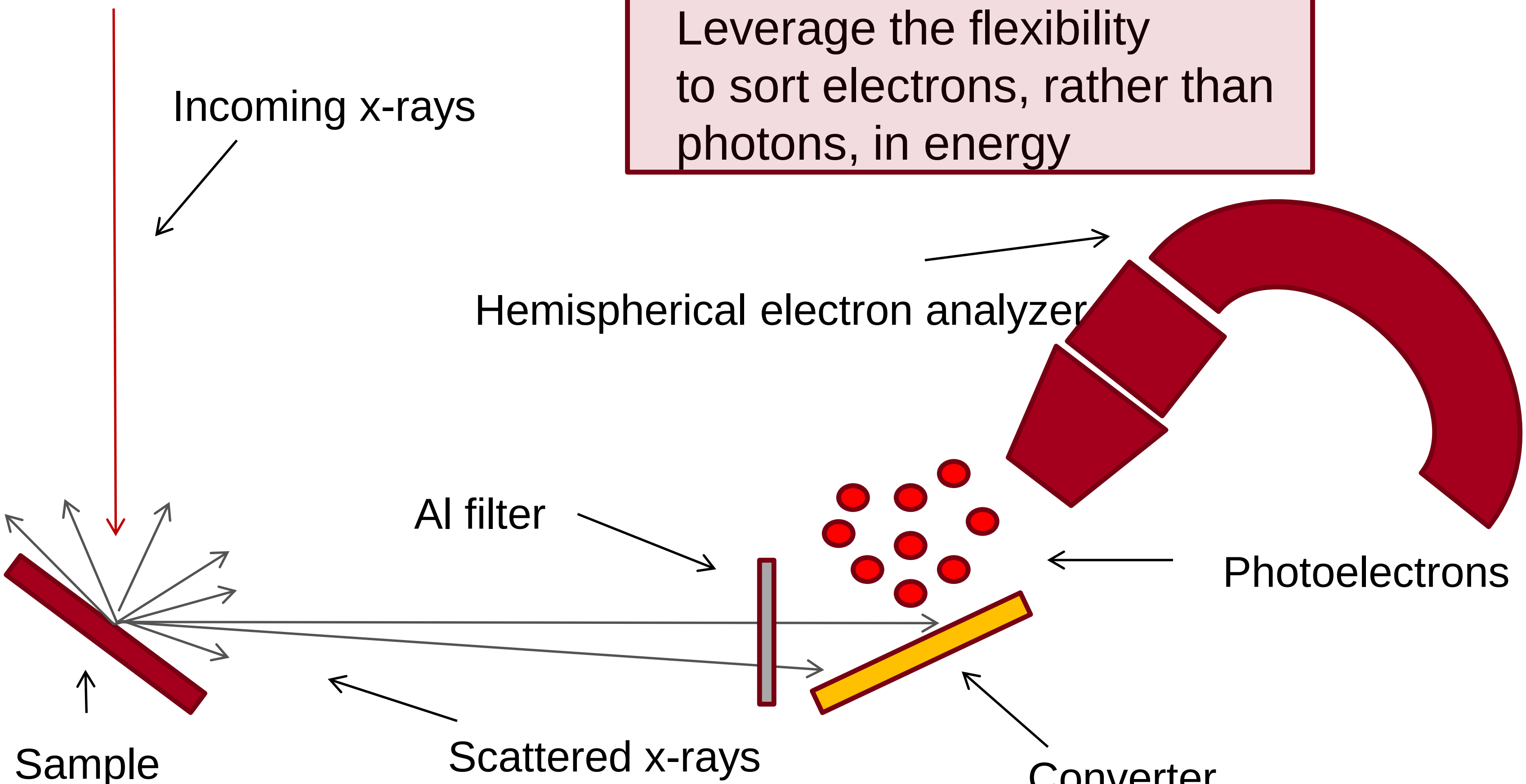
Source requirements	Engineering challenges	Collection efficiency
• Small x-ray spot, not good for FELs • Limited flux at 3rd generation light sources	• Grating with excellent quality • Long detector arm • Very high detector spatial resolution • Excellent stability	• Low fluorescence yield, ~ 10^{-3} • Very low collection efficiency, ~ 10^{-5}

Efforts to improve the throughput/resolution

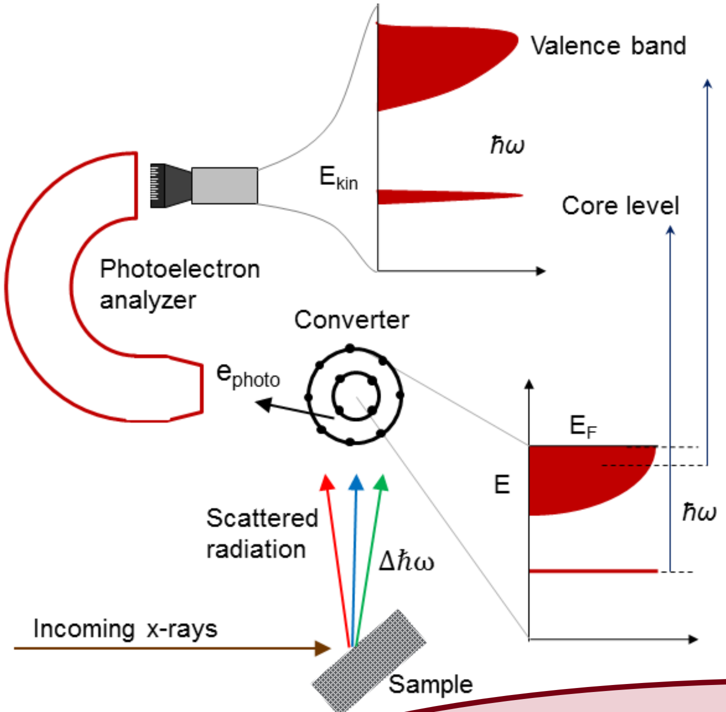
Source requirements	Engineering challenges	Collection efficiency
• Small x-ray spot, not good for FELs • Limited flux at 3rd generation light sources	• Grating with excellent quality • Long detector arm • Very high detector spatial resolution • Excellent stability • **Small pixel, counting centroiding**	• Low fluorescence yield, ~ 10^{-3} • Very low collection efficiency, ~ 10^{-5} • **Stimulated emission** • **Transition Edge Sensors**

Alternative: convert photons to photoelectrons

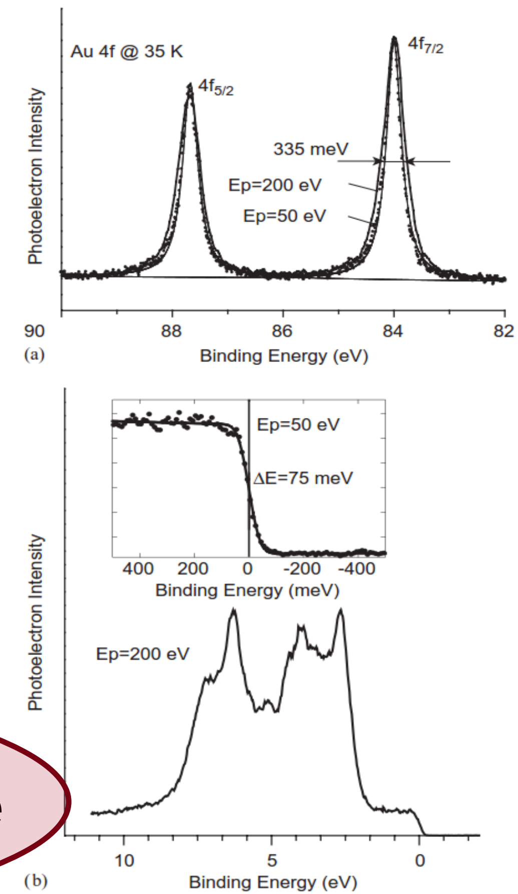
Leverage the flexibility to sort electrons, rather than photons, in energy



Proposed solution: convert photons to photoelectrons!



Example: Monochromatic beam, PES from Au



The PES spectrum is a convolution of the unknown RIXS spectrum with the response of the material

Y. Takata et al. / Nuclear Instruments and Methods in Physics Research A 547 (2005) 50–55

Basis and characteristics of method

The principle of the method is simple and consists of converting the x-rays under study into photoelectrons which are then analyzed in a suitable instrument according to energies and flux.

Given a known energy level E_B of some converter atom, free or bound, the energy $h\nu$ of the photon is obtained from the kinetic energy $E_{e,kin}$ of the photoelectron according to

$$h\nu = E_B + E_{e,kin} \quad (1)$$

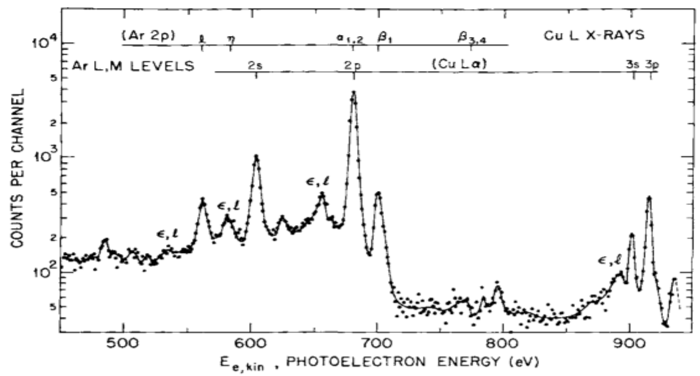
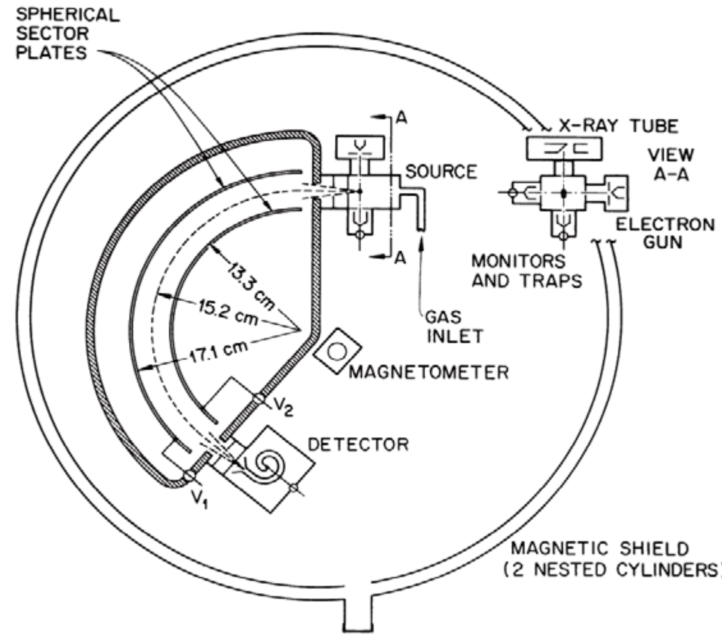


Fig. 9. The general photoelectron spectrum, exemplified by Ar L,M(CuL). The spectrum can be interpreted as an atomic-level spectrum, Ar L,M(Cu $L\alpha$), or as an x-ray spectrum, Ar 2p(Cu L). Peaks ϵ,l arise from excitation of a 3p electron concomitant with the ionization of an L or M electron.

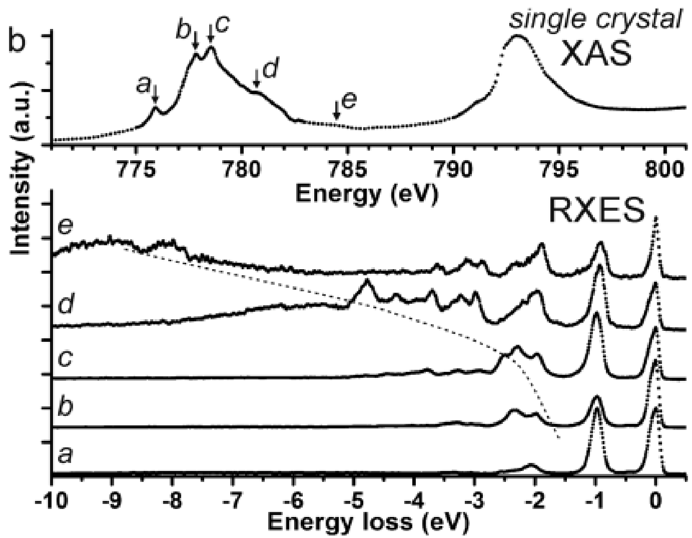


X-Ray Analysis by Photoelectron Spectrometry*

Manfred O. Krause and François Wuilleumier†

Transuranium Research Laboratory
Oak Ridge National Laboratory
Oak Ridge, Tennessee 37830

Test experiment: CoO



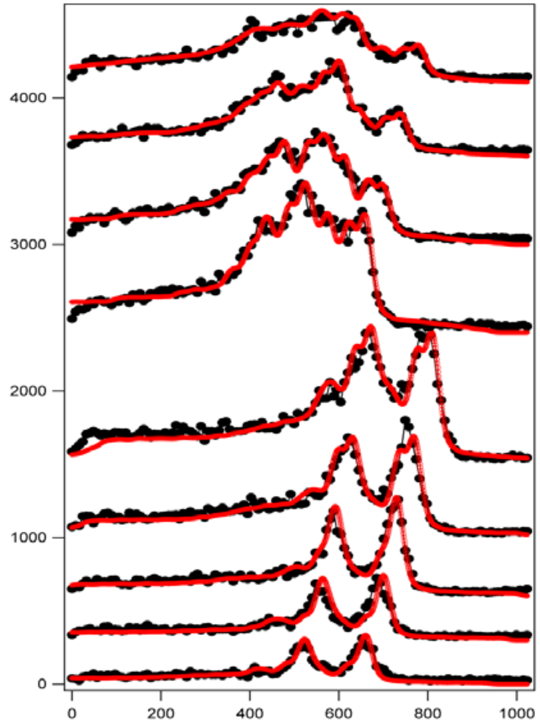
Electronic Structure of CoO Nanocrystals and a Single Crystal Probed by Resonant X-ray Emission Spectroscopy

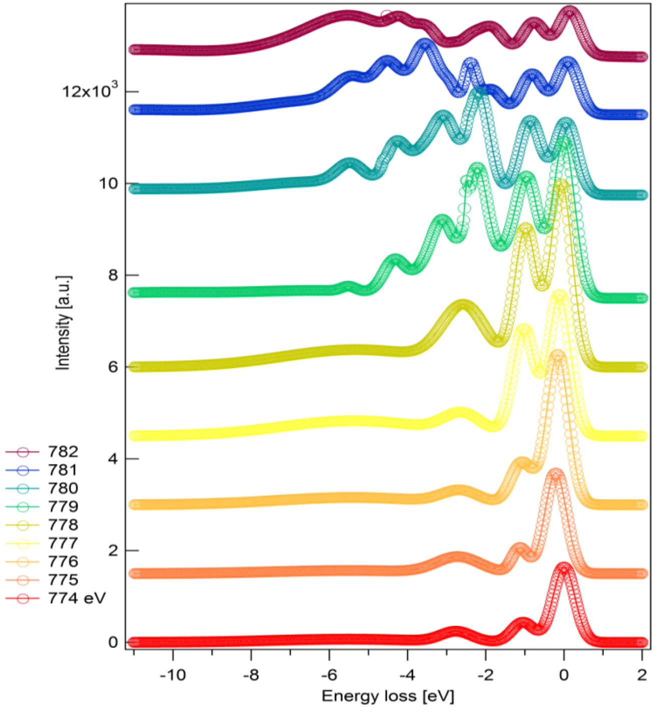
Matti M. van Schooneveld,*,† Reshmi Kurian,† Amélie Juhin,†,‡ Kejin Zhou,‡ Justine Schlappa,‡ Vladimir N. Strocov,‡ Thorsten Schmitt,‡ and Frank M. F. de Groot*,†

J. Phys. Chem. C 2012, 116, 15218–15230

Pilot study: Cobalt oxide
Step 2: PES from CoO and RIXS map retrieval

PES spectra when varying incident *E*





- We simulated the unknown RIXS spectrum as a sum of 12 Gaussians, and
- Convolved it with the analyzer response (the *4f* peaks): very good fit

Comparison of first test to grating VLS spectrometer

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Comparison (per shot) between grating spectrometer and PAX

Parameter	VLS	PAX
# photons/shot	2×10^{10}	2×10^{10}
Fluence mJ/cm^2	25	0.25
F yield	10^{-3}	10^{-3}
Al foil transmission	0.8	0.8
Collection efficiency	10^{-6}	2.5×10^{-2}
Diffraction efficiency	2.5×10^{-2}	---
Conversion to e^-	-	3×10^{-2}
Detection efficiency	0.8	0.8
Analyzer transmission	---	10^{-5}
Estimated ph/e^-	0.32	0.096
Measured ph/e^-	0.15	0.05

- Big gain for PAX
- Similar
- Target for improvement

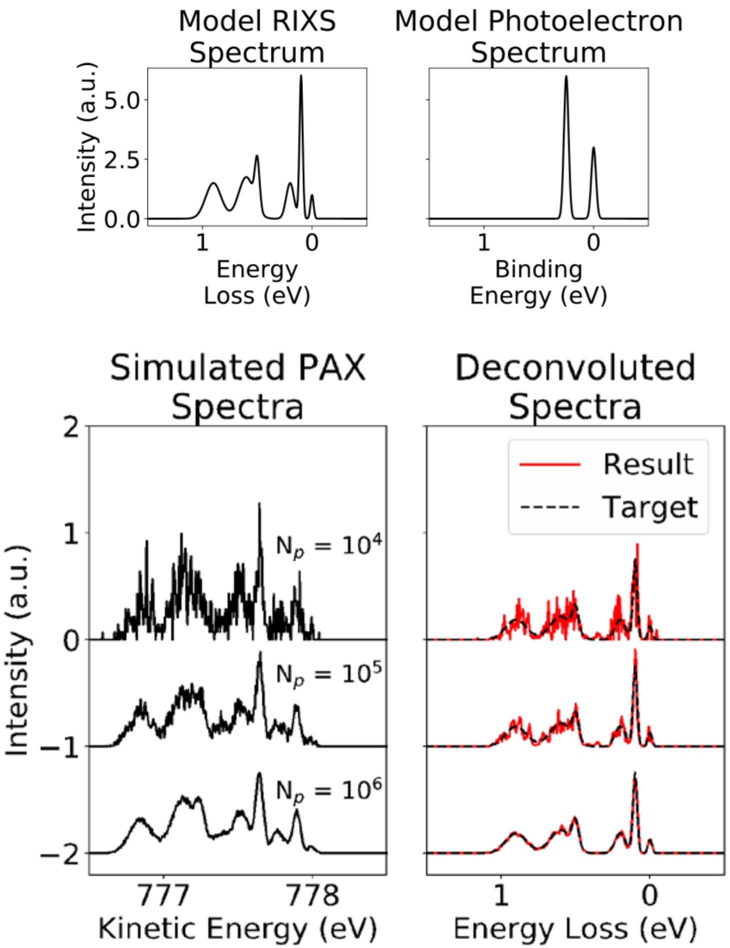
VLS signal rate is 3 times higher than PAX (same # photons)

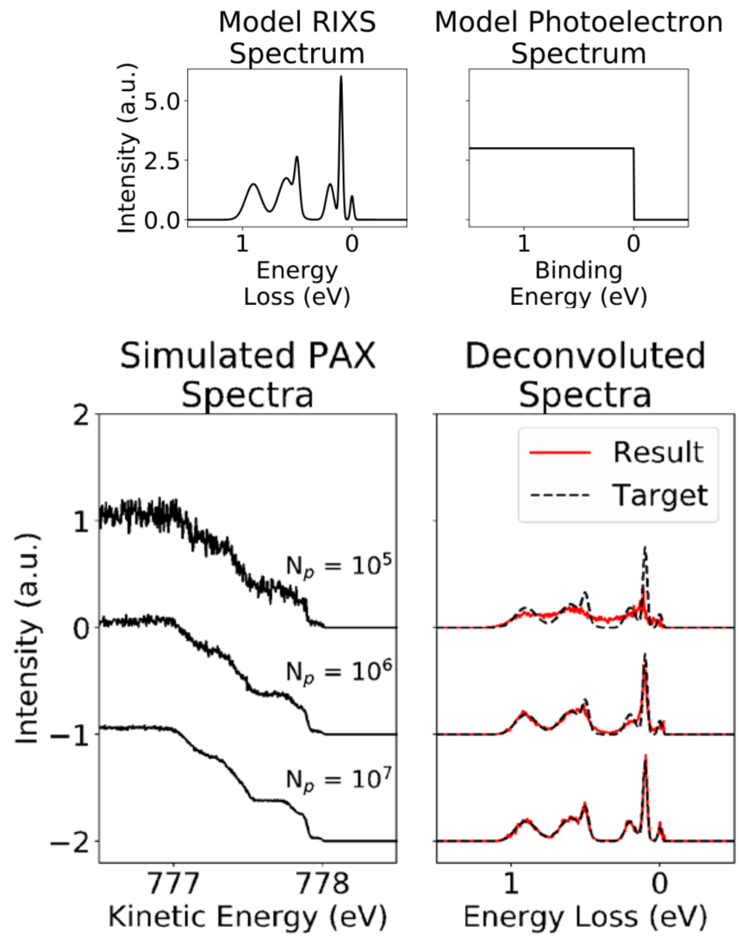
PAX signal rate is 30 times higher than VLS (same fluence)

Advantages

Source requirements	Engineering challenges	Collection efficiency
• Small x-ray spot, not good for FELs • Limited flux at 3rd generation light sources • No need b/c not imaging the spot	• ~~Grating with excellent quality~~ • ~~Long detector arm~~ • Very high detector spatial resolution • ~~Excellent stability~~ • Commercial detectors • Insensitivity to vibrations	• Low fluorescence yield, ~ 10^{-3} • Very low collection efficiency, ~ 10^{-5} • High collection efficiency
• Flexibility of manipulating e⁻ e.g. high energy resolution	• Straightforward extension to tender and hard x-rays	• Opportunity for angle-resolved acquisition

Simulations





Can PAX help RIXS?

- Signal rate is already comparable to grating-based RIXS
- Various improvement schemes can boost signal rate much higher: fresh Au evaporation, electrical bias
- Path towards efficient RIXS with ~200 meV looks good
- Path towards very high resolution possible, needs testing, possibly at BL 13-1 at SSRL in the fall
- Opportunity to do RIXS at any edge appears possible with no modification of the spectrometer

Acknowledgements

- Daniele Cocco
- Frank O'Dowd
- Joe Dvorak
- Scott Coburn
- Serge Guillet
- Ted Osier
- Hengzi Wang
- Jim Defever

- and many others

- Ming-Fu Lin
- Dan Damiani
- Bill Schlotter
- Josh Turner
- Dennis Nordlund
- Hirohito Ogasawara

THANK YOU !

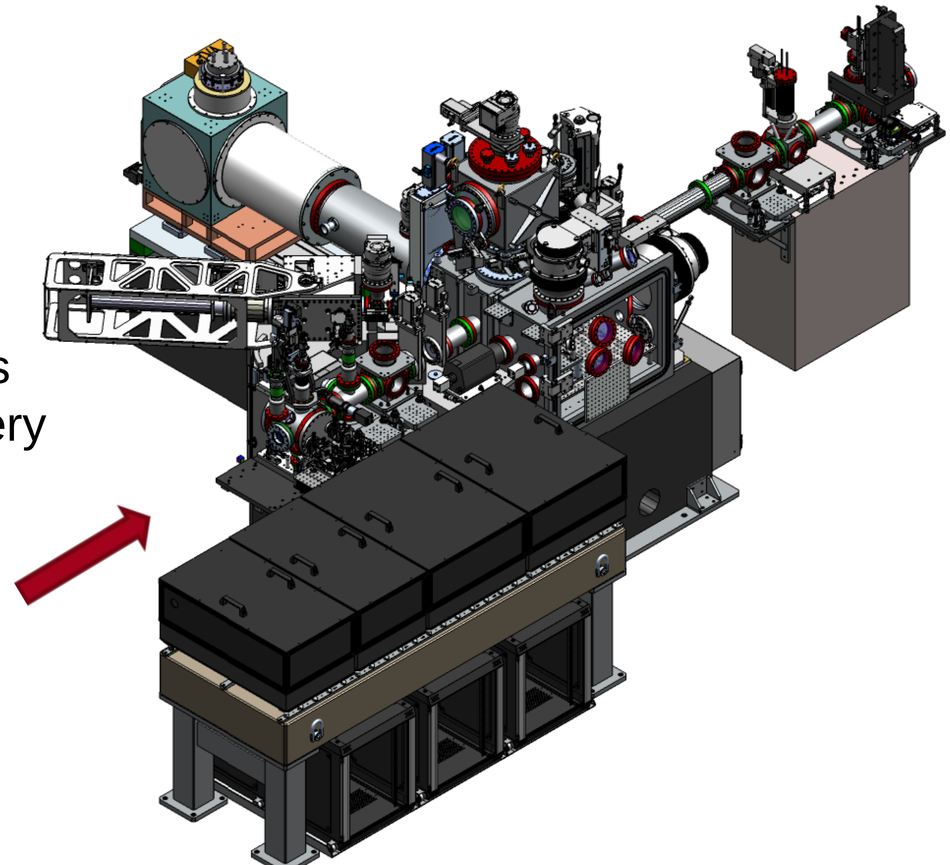
Key Beamline Performance Requirements

Parameter	Range	Comment
Photon Energy Range SXU [eV]	250-1600	Cover the C, N, O *K*-edge, L-edge of 3*d* transition metals, *M*-edge of rare earths
Bandwidth Control [RP, resolving power]	50,000; 10,000 5,000	Required for RIXS: 2 x FT limit Required for all other techniques: < 2 x FT limit
Experimental Spot Size Range a. Horizontal [µm] b. Vertical [µm]	 3-1000 3-1000	Spot size adjustable to the interaction point at each endstation
Lasers	Visible to THz Visible to 2.4 µm	For correlated materials For chemistry
Synchronization [fs]	<10	

chemRIXS endstation

Capabilities:

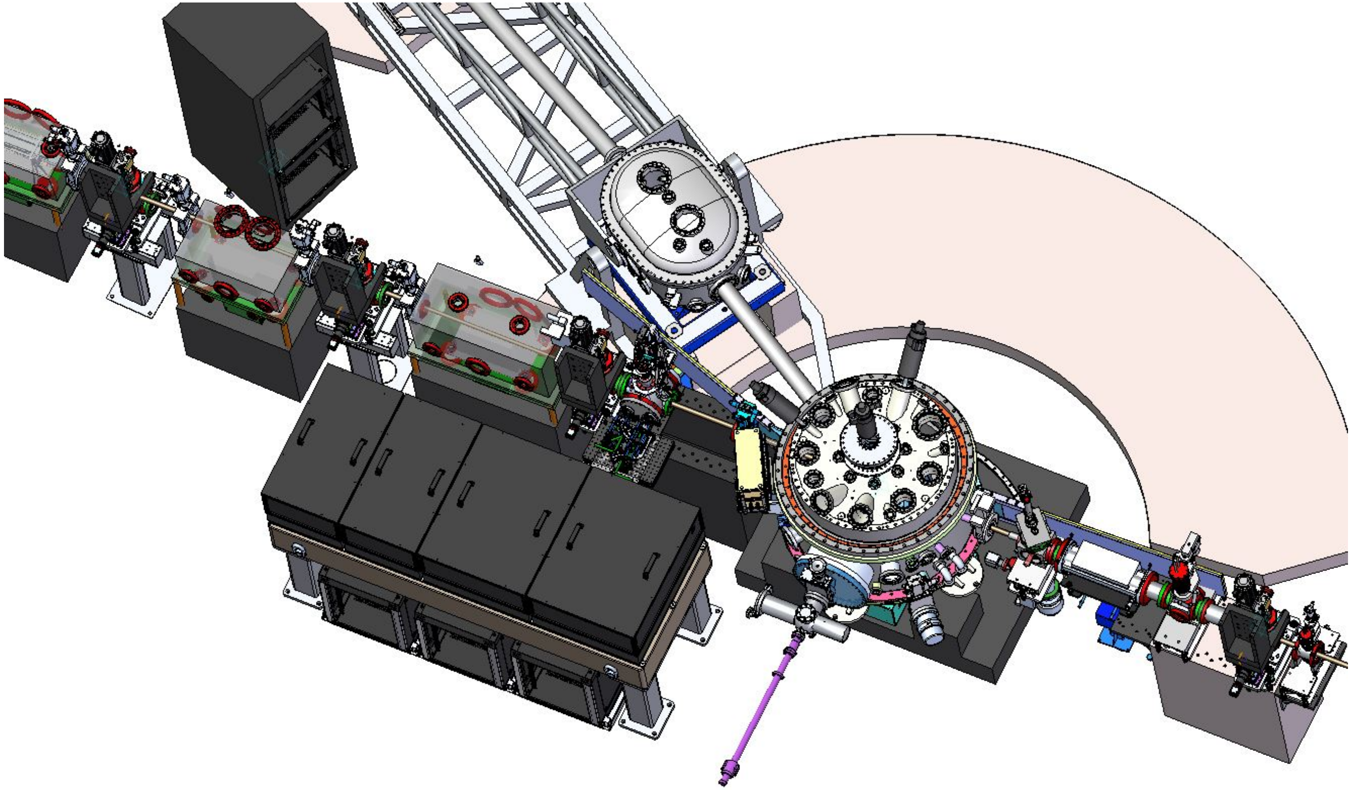
- Variable x-ray spot
- Wide range of laser wavelengths
- (Semi) Automated sample delivery
- Arrival Time Monitor



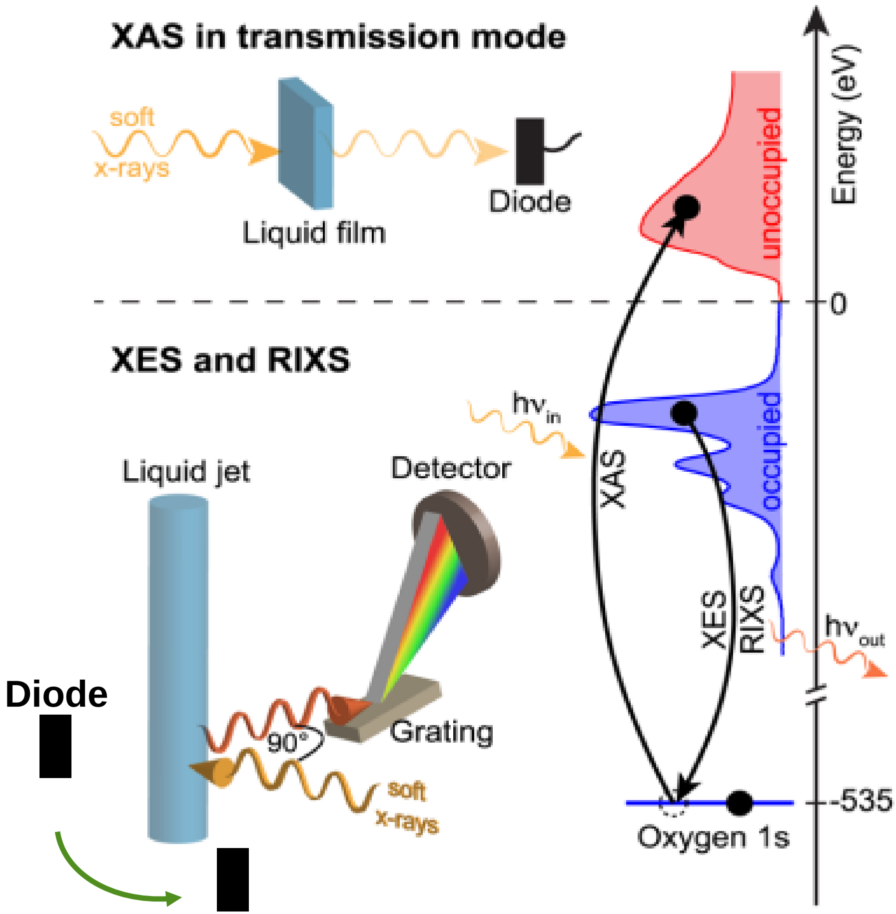
qRIXS endstation

Capabilities:

- Variable x-ray spot
- Wide range of laser wavelengths
- Automated sample delivery

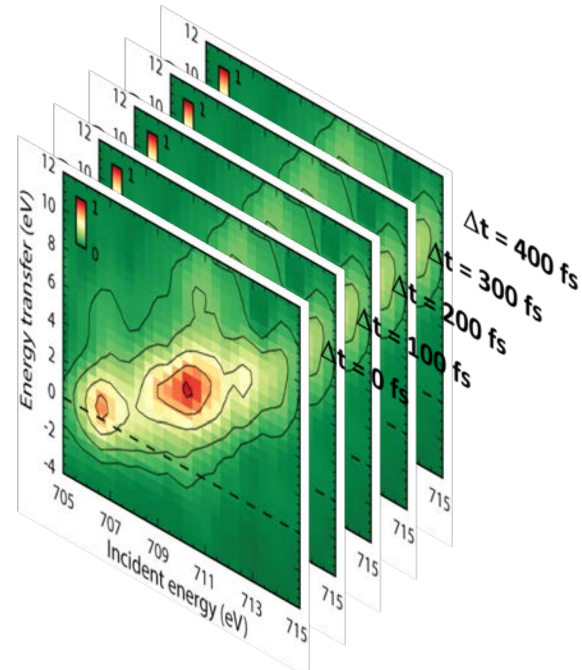


Experiment schematic



S. Schreck, *et al.*, Struct. Dyn., **1**, 054901, 2014

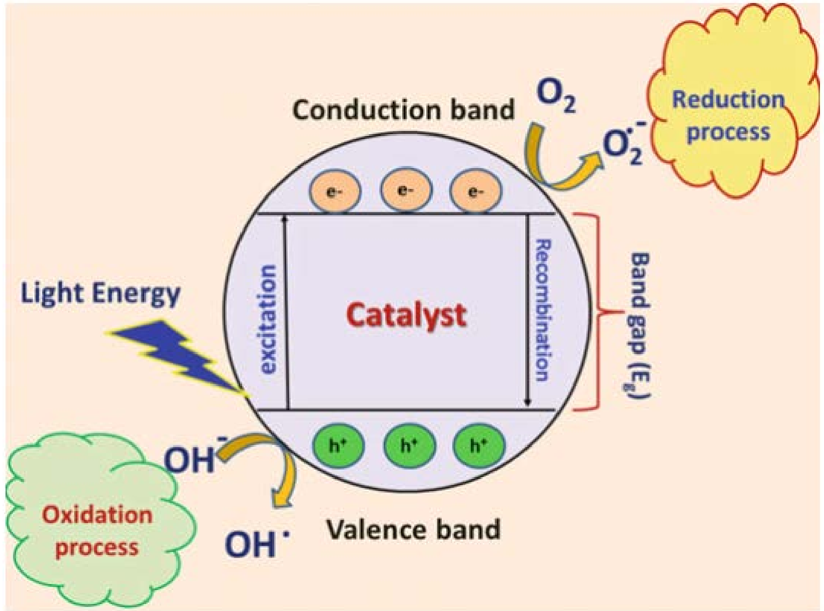
XAS: X-ray Absorption Spectroscopy
XES: X-ray Emission Spectroscopy
RIXS: Resonant Inelastic X-ray Scattering



Wernet *et al*, *Nature* **520**, 78 (2015)

Science motivation: Photocatalysis

- **Key questions**:
  - How do photogenerated carriers catalyze chemical reactions?
  - How does charge separation, transport and localization occur on ultrafast time scales?
  - For systems in a photoexcited state, how does the nuclear structure and local defects influence the catalytic process?

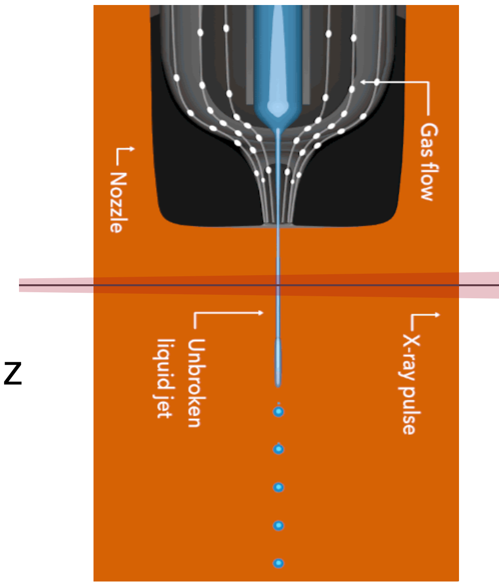


R. Saravanan, Francisco Gracia and A. Stephen, Springer, *Nanocomposites for Visible Light-induced Photocatalysis*

chemRIXS Requirements

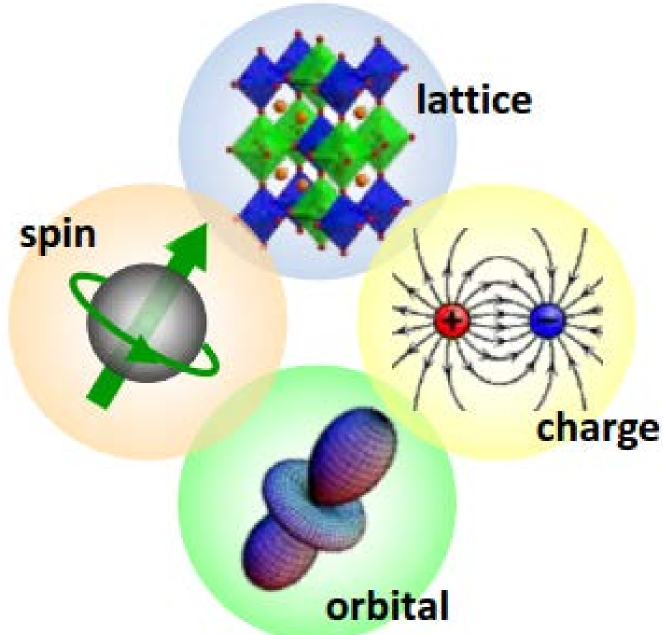
Key requirements:

- Vacuum: 10^{-4} Torr (jets) to <10^{-8} Torr (solids)
- Spectrometers: grating and transition-edge sensor, 2000 RP
- Point detectors: avalanche photodiode and microchannel plate: 1 MHz readout rate
- Sample Viewing: on-axis & perpendicular ; infrared illumination
- **Develop large-area sheet jets to best utilize x-rays**
- **Automated delivery**

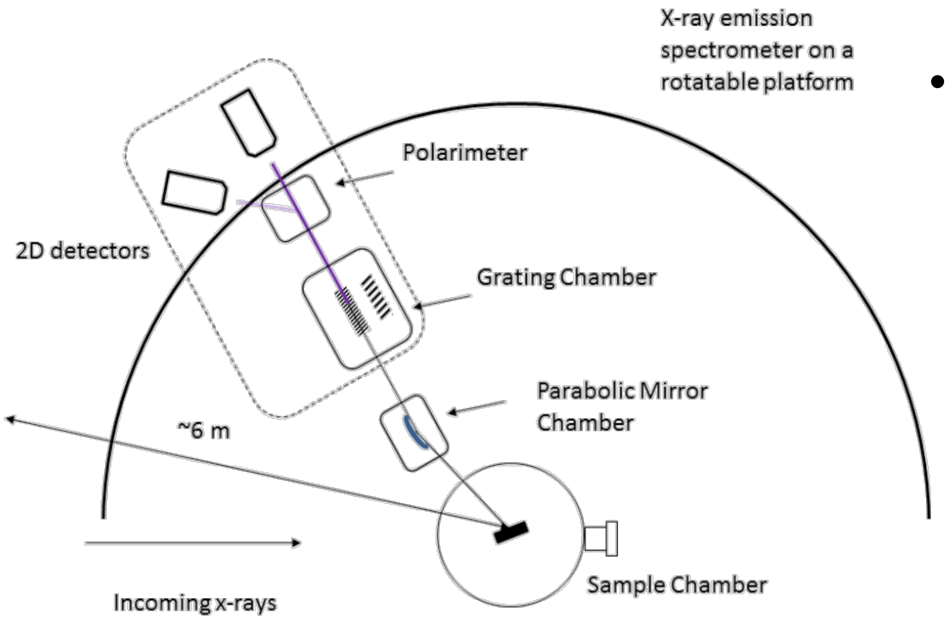


Science motivation: Quantum Materials

- **Key questions**:
  - How does the interplay between constituent entities lead to emergent behavior?
  - What is the spectrum of the collective excitations and how do they evolve with temperature, strain, magnetic field, tailored optical excitations, etc.?
  - Can we understand and control the properties of these materials?
  - How do spontaneous fluctuations and heterogeneities affect macroscopic behavior?



Spectrometer schematic

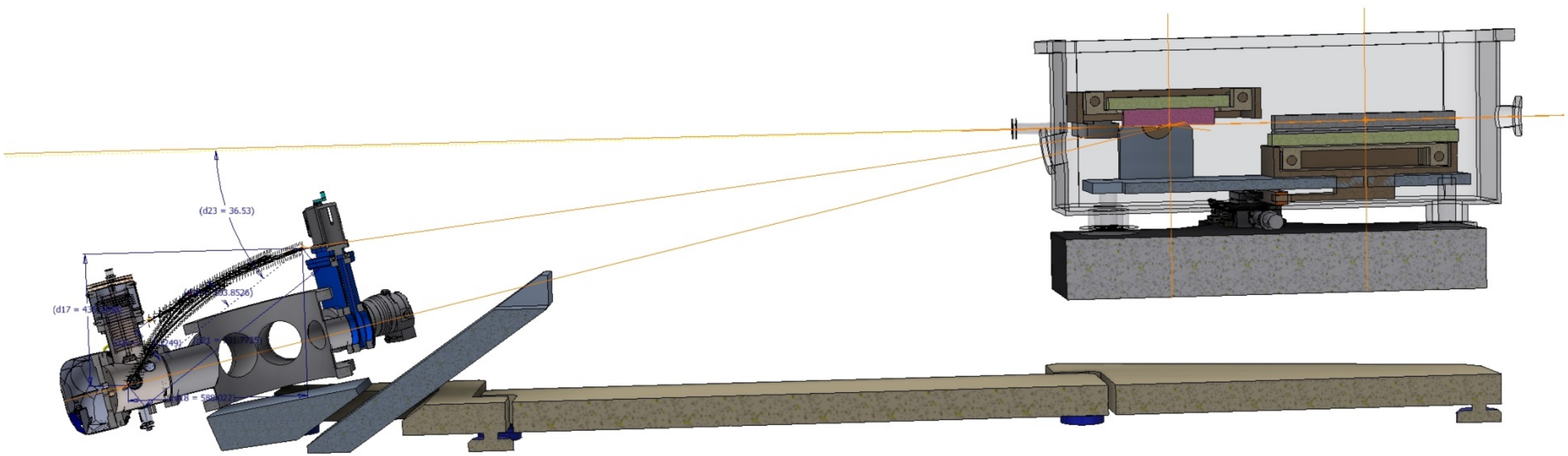


- An extra **parabolic mirror** is added to collimate horizontally the beam
  - Increase the detector acceptance
  - Reduce/eliminate inter-plane aberrations (smile profiles)
  - Make use of Multilayers for polarimetry studies feasible

qRIXS Requirements

- Sample chamber capable of supporting XAS, REXS
- UHV (<3 x 10^{-9} Torr), Low Temperature (20 K)
- Laser in-coupling: support of wavelengths from visible to THz
- Continuous rotation of spectrometer arm: 40 – 150 degrees
- Overall arm length: maximum length is fixed to 6000 mm
- Reach RP of 50,000 & 1 keV, option for ~10,000 (second grating)
- Polarization analysis
- Provisions for XPCS
- Design guideline
  - Make the spectrometer as simple as possible to operate with the minimum number of actuators (ideally one) to change the energy

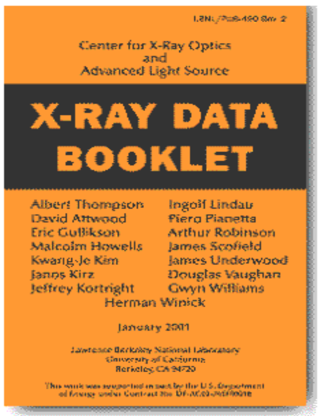
Travel range

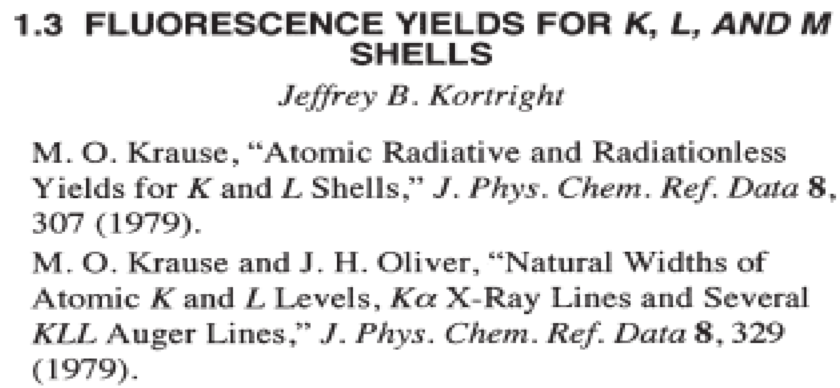


What is PAX?

- PAX stands for “Photoelectron spectrometry for Analysis of X-rays”

- Introduced by Manfred O. Krause, Oak Ridge National Laboratory





1.3 FLUORESCENCE YIELDS FOR *K, L, AND M* SHELLS

Jeffrey B. Kortright

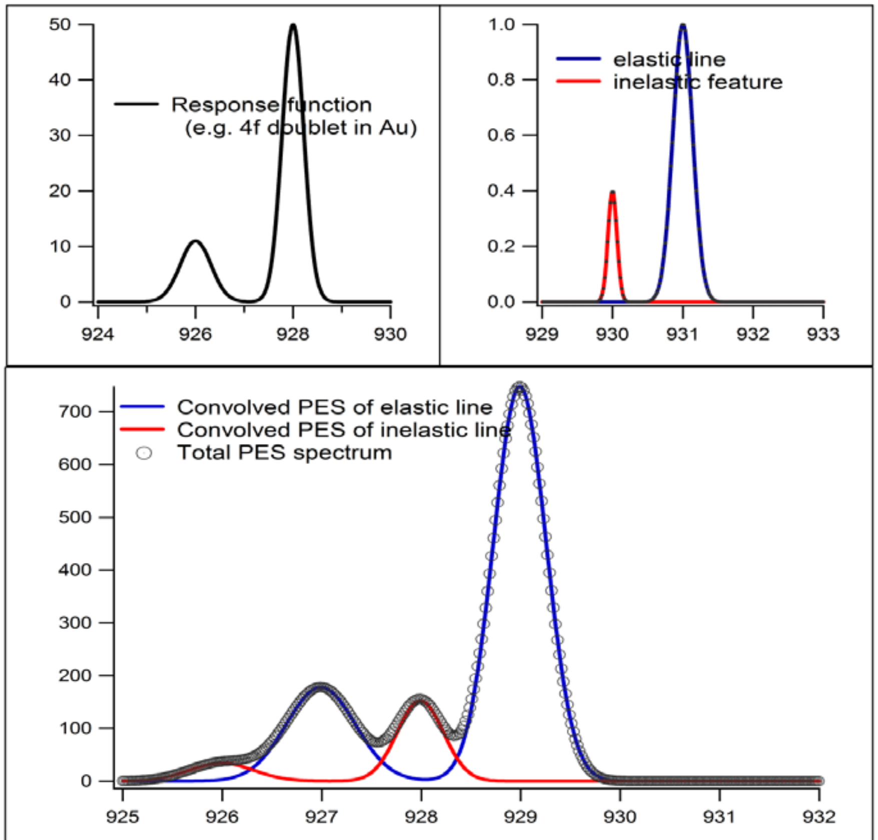
M. O. Krause, “Atomic Radiative and Radiationless Yields for *K* and *L* Shells,” *J. Phys. Chem. Ref. Data* **8**, 307 (1979).

M. O. Krause and J. H. Oliver, “Natural Widths of Atomic *K* and *L* Levels, *Kα* X-Ray Lines and Several *KLL* Auger Lines,” *J. Phys. Chem. Ref. Data* **8**, 329 (1979).

What is the difference between Spectrometry and Spectroscopy?

- Spectroscopy is the science of studying the interaction between matter and radiated energy while spectrometry is the method used to acquire a quantitative measurement of the spectrum.

How to retrieve the RIXS spectrum with the *4f* doublet?



Retrieve the RIXS spectrum via de-convolution of the PES spectrum

- Using e.g. *4f* of Au should be trivial: 2 Gaussians (appr.)
- Using the Fermi edge needs more study: numerical simulation to investigate signal-to-noise ratio

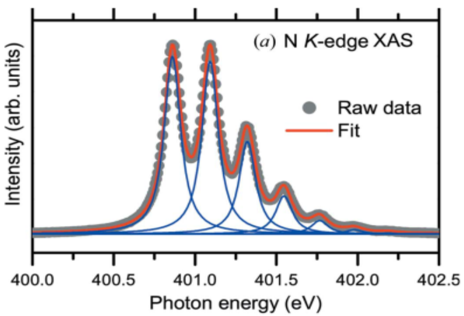
The problem of de-convolving a spectrum from a known response function is well-known and various stable iterative algorithms exist.

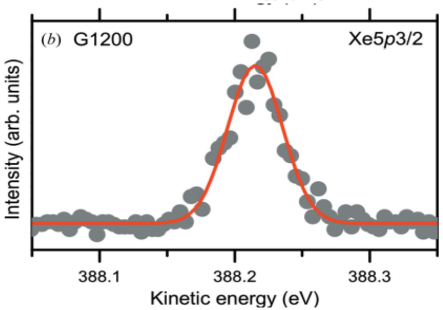
The concept has been used…

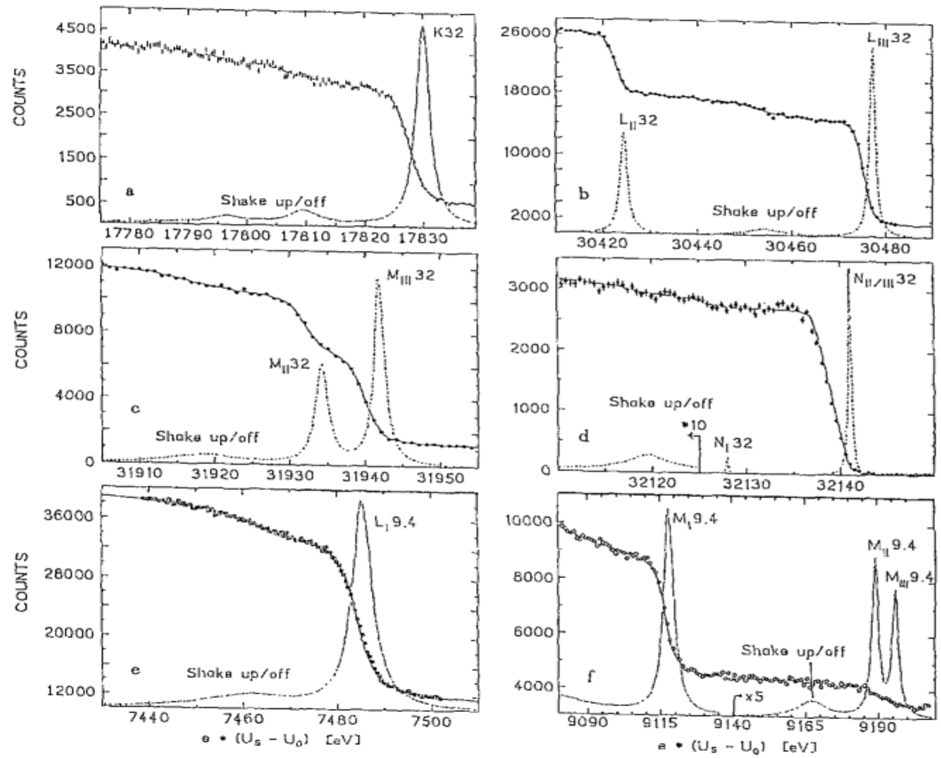
New soft X-ray beamline BL07LSU at SPring-8

Susumu Yamamoto,[a,b] Yasunori Senba,[c] Takashi Tanaka,[d] Haruhiko Ohashi,[c] Toko Hirono,[c] Hiroaki Kimura,[c] Masami Fujisawa,[a,b] Jun Miyawaki,[a,b] Ayumi Harasawa,[a,b] Takamitsu Seike,[c] Sunao Takahashi,[c] Nobuteru Nariyama,[c] Tomohiro Matsushita,[c] Masao Takeuchi,[c] Toru Ohata,[c] Yukito Furukawa,[c] Kunikazu Takeshita,[c] Shunji Goto,[c] Yoshihisa Harada,[a,b] Shik Shin,[a,b] Hideo Kitamura,[d] Akito Kakizaki,[a,b] Masaharu Oshima[b,e] and Iwao Matsuda[a,b]*

J. Synchrotron Rad. (2014). **21**, 352–365



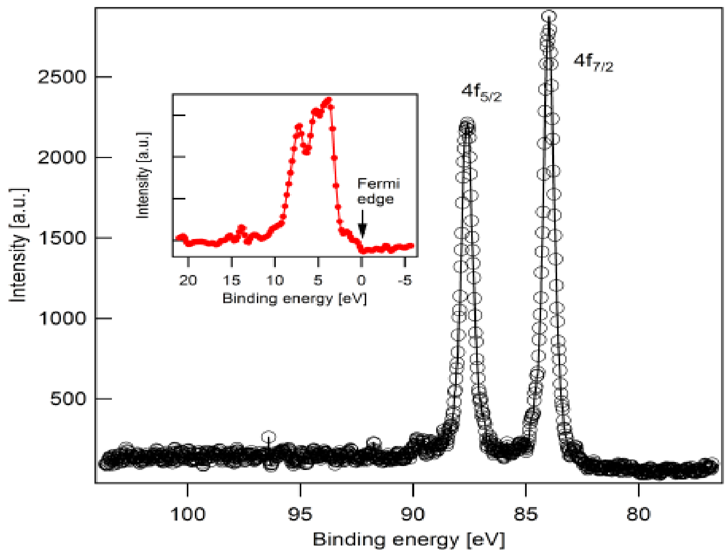




Precision measurement of the conversion electron spectrum of ^{83m}Kr with a solenoid retarding spectrometer*

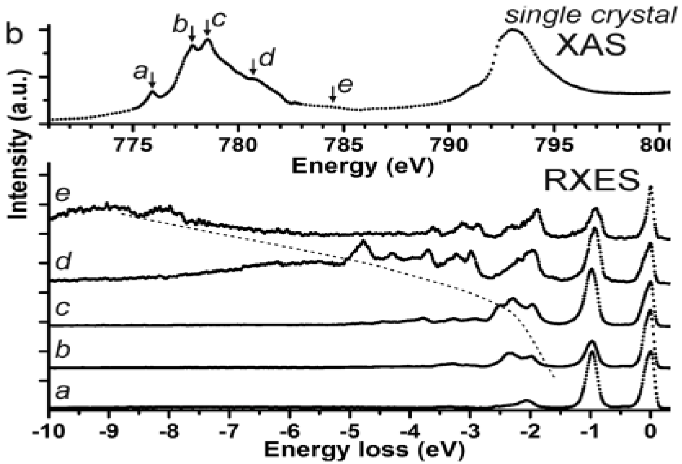
Z. Phys. A – Hadrons and Nuclei 342, 71–78 (1992)

Pilot study: Cobalt oxide
Step 1: Au characterization with direct beam



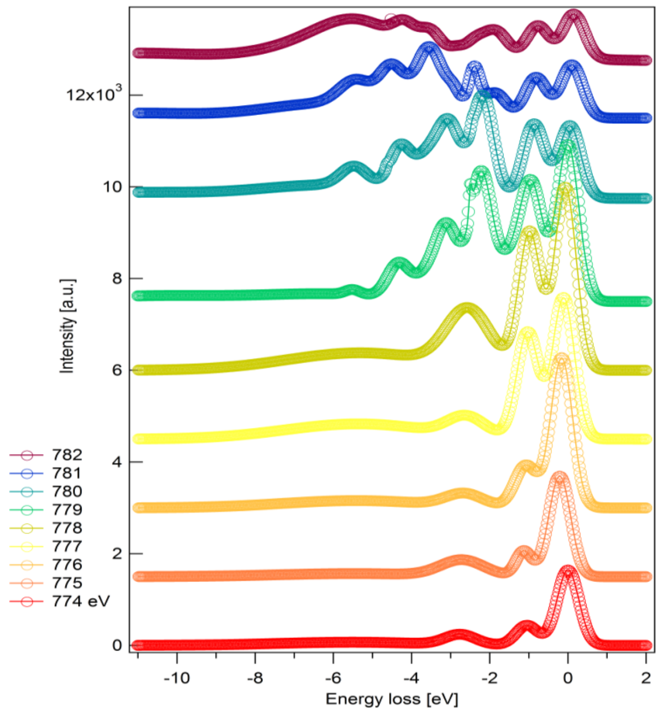
$4f$ peaks in Au. Inset: valence band spectrum. The sharp $4f$ resonances represent the response function of the converter-analyzer system and are used to encode the spectrum of the inelastically scattered x-rays. If very high resolution is desired the region in the vicnity of the Fermi edge could be used instead.

Comparison to grating-based RIXS

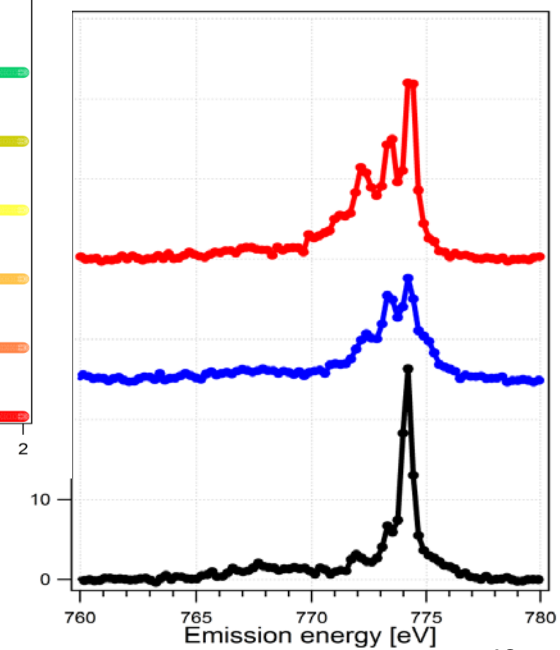


High-resolution SLS

PAX



Our own VLS



Electronic Structure of CoO Nanocrystals and a Single Crystal Probed by Resonant X-ray Emission Spectroscopy

Matti M. van Schooneveld,*,† Reshmi Kurian,† Amélie Juhin,†,‡ Kejin Zhou,‡ Justine Schlappa,‡ Vladimir N. Strocov,‡ Thorsten Schmitt,‡ and Frank M. F. de Groot*,†

PAX and M. O. Krause

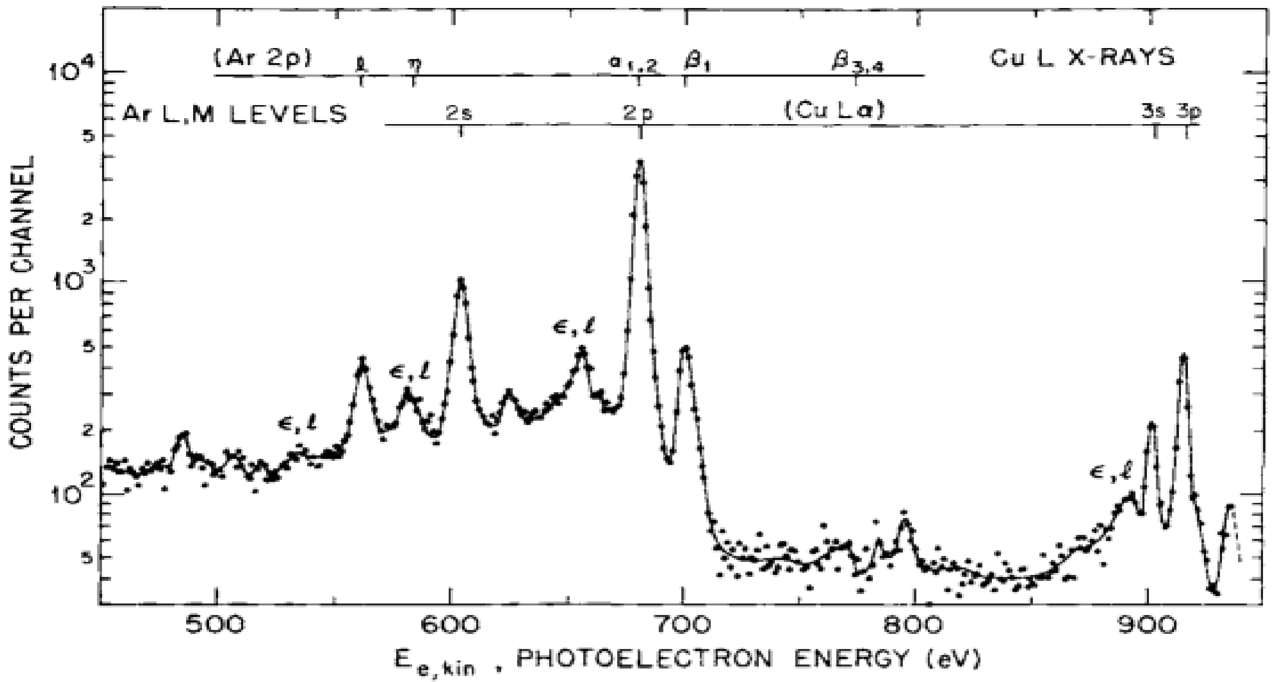


Fig. 9. The general photoelectron spectrum, exemplified by Ar L,M(CuL). The spectrum can be interpreted as an atomic-level spectrum, Ar L,M(Cu $L\alpha$), or as an x-ray spectrum, Ar 2p(Cu L). Peaks ϵ,l arise from excitation of a 3p electron concomitant with the ionization of an L or M electron.

$L_{\alpha 1,2}$ = 929 eV
$L_{\beta 1}$ = 949
$L_{\beta 3,\,4}$ = 1022
L_{l} = 810
L_{η} = 830

Conclusions

PAX is demonstrated. Multiple improvement options possible:

Maximize throughput for medium resolution (~0.5 eV) experiments using the *4f* peaks of Au

Maximize throughput for high resolution (30 meV) experiments using the Fermi edge in Au/Pt

Variable, user-defined

$$\Delta E_{res} = \sqrt{\Delta E_{mono}{}^2 + \Delta E_{atom}{}^2 + \Delta E_{det}{}^2}$$