Using Photoelectron Spectroscopy to Measure Resonant Inelastic X-Ray Scattering Spectra

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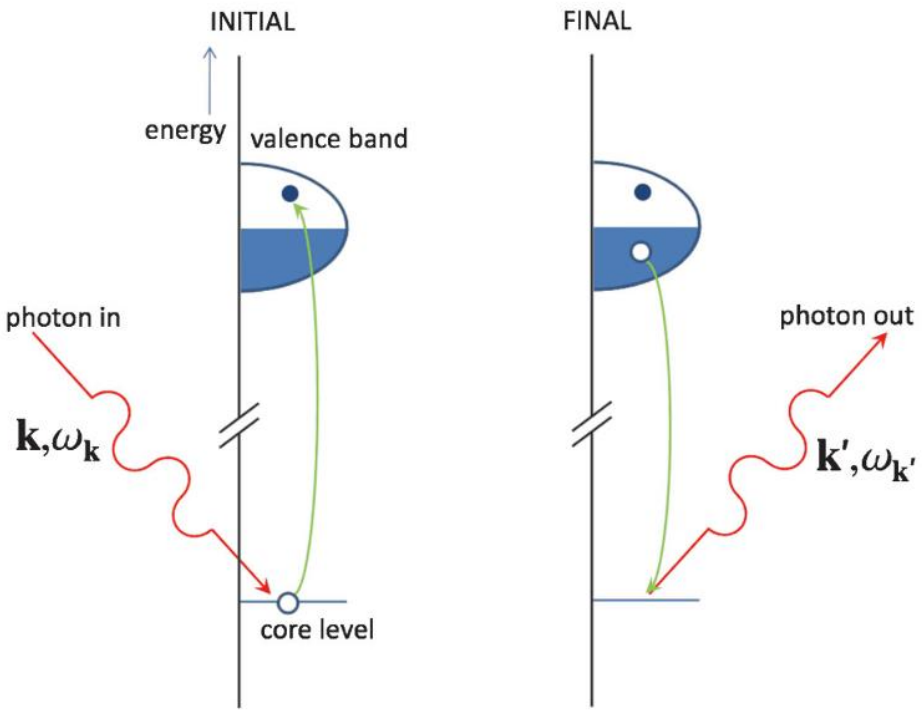
U.S. DEPARTMENT OF ENERGY
Office of Science

SLAC NATIONAL ACCELERATOR LABORATORY

Resonant Inelastic X-Ray Scattering (RIXS)

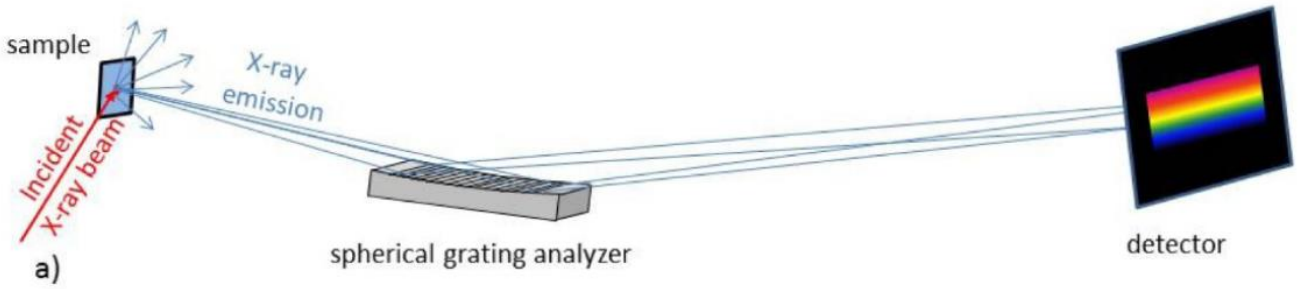


- Probe elementary excitations of matter with elemental specificity.
- Has become a powerful and widely used technique to study materials and molecules.

L. J. P. Ament *et al*. (2011)

Challenges of Soft X-Ray RIXS

Schematic of grating-based RIXS spectrometer from F. Marschall *et al.* (2017)



X-ray Source

- Limited flux at 3rd generation light sources
- Small x-ray spot, not good for FELs

Engineering challenges

- Grating with high quality
- Detector spatial resolution
- Stability
- Instrumentation not commercially available

Collection efficiency

- Low fluorescence yield, ~ 10^{-3}
- Very low collection efficiency, ~ 10^{-5}

Efforts to Improve the Throughput of RIXS

SLAC

Source requirements

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Small pixels, centroiding

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Stimulated RIXS

Transition edge sensors

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PAX (this talk)

Outline of Rest of Talk

- Concept of Photoelectron Spectrometry for Analysis of X-rays (PAX) applied to RIXS
- Analyzing PAX data with a deconvolution algorithm
- Simulated PAX performance
- Ongoing experimental work

Objective

Measure RIXS with

- High resolution,
- High efficiency and
- With a simple implementation.

Concept of Photoelectron Spectrometry for Analysis of X-Rays (PAX)

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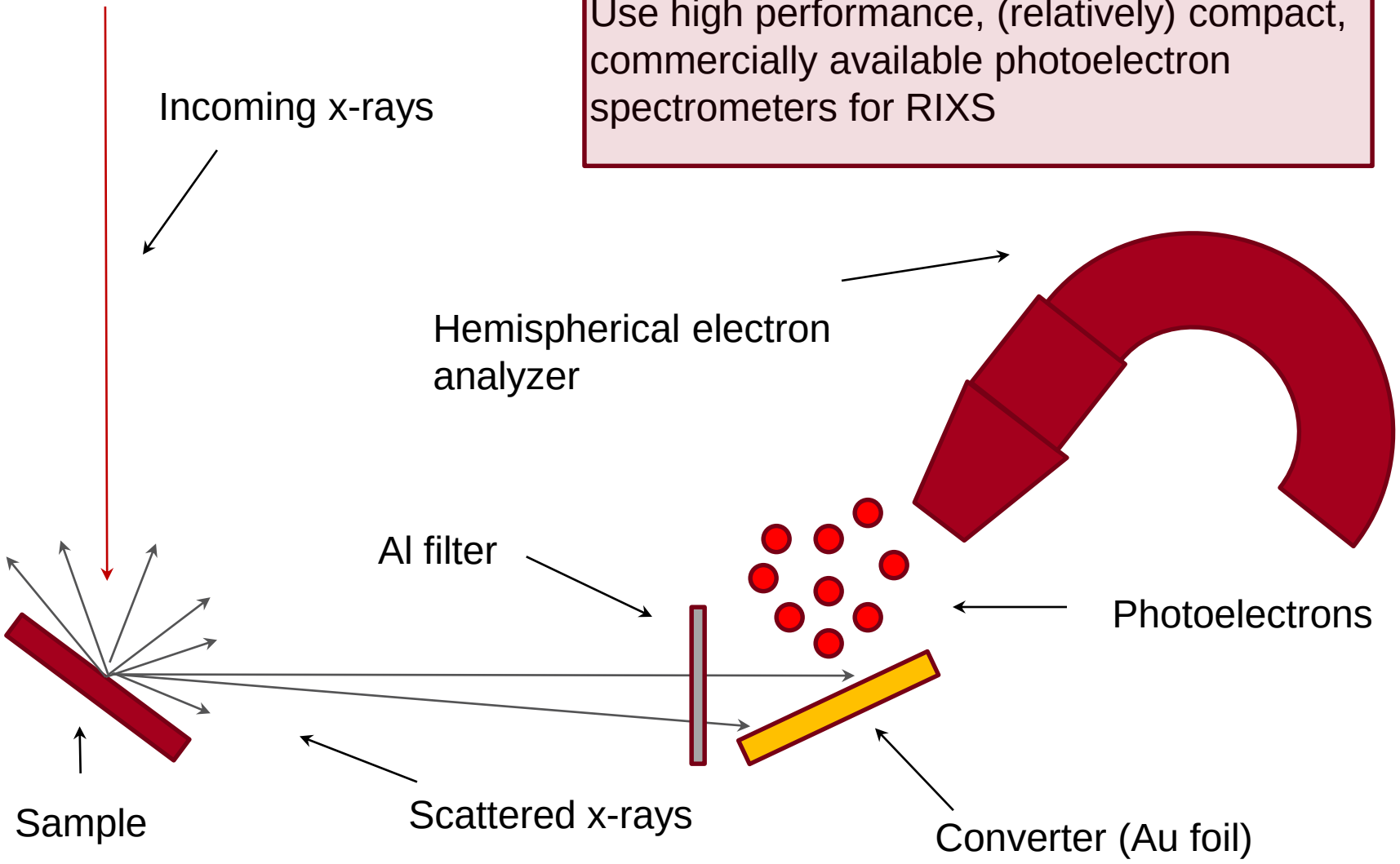
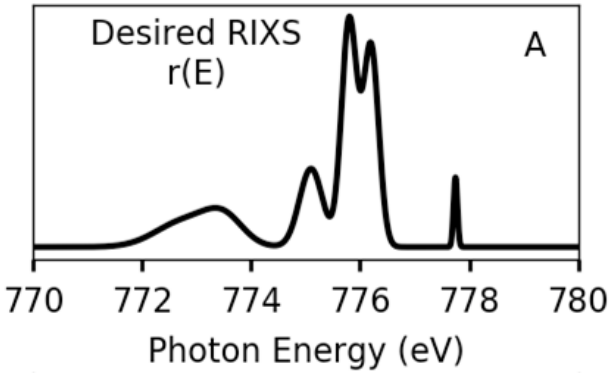
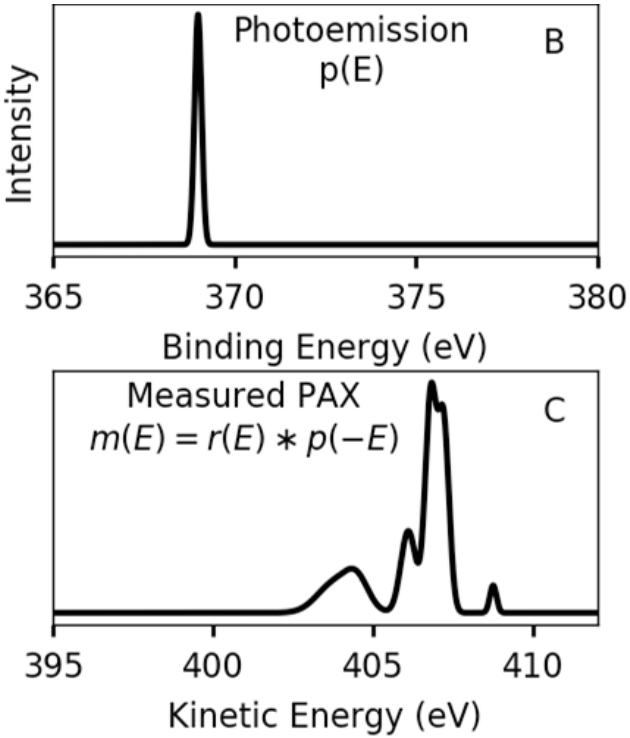
Use high performance, (relatively) compact, commercially available photoelectron spectrometers for RIXS



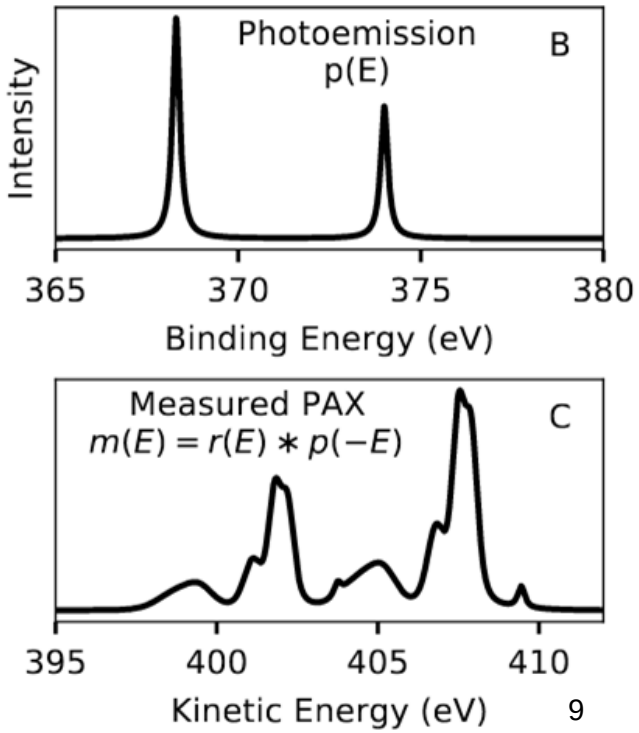
Example PAX Spectra



Isolated Photoemission Peak



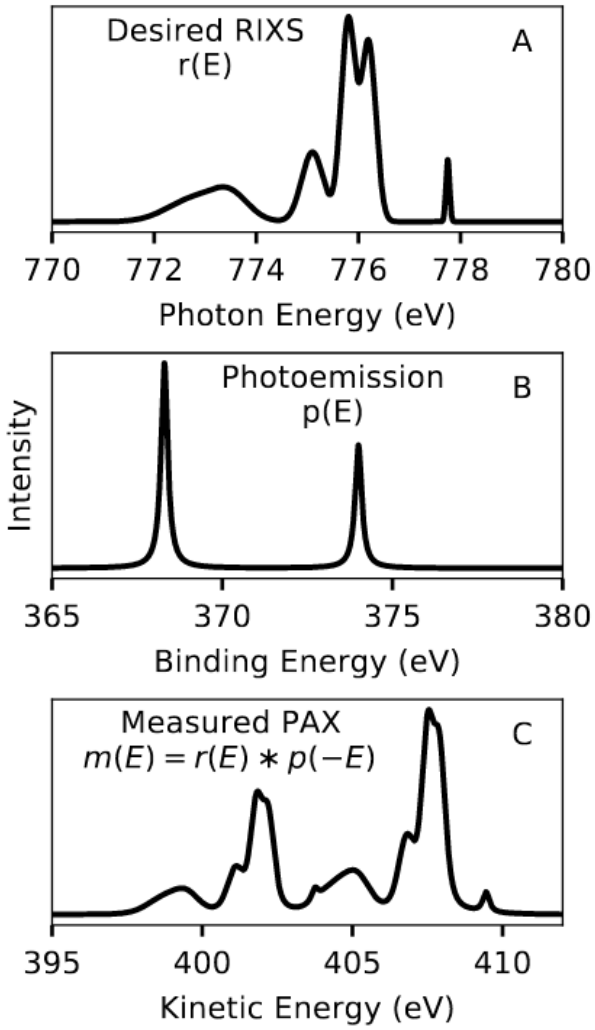
Two Photoemission Peaks (Ag 3d)



Model RIXS from
J. Schlappa *et al.* (2012)

How do we Analyze and Understand a PAX spectrum?

- Assume we have a separate measurement of the converter material photoemission spectrum
- One option: Fit PAX spectrum to a sum of RIXS features (e.g. Lorentzians) convolved with the converter material photoemission spectrum
- Option explored in this talk: estimate desired RIXS spectrum from measured PAX spectrum



Lucy-Richardson Deconvolution for Analysis of PAX Spectra

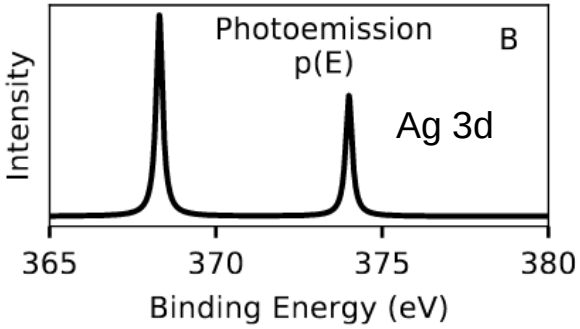
The RIXS spectrum can be estimated using the Lucy-Richardson deconvolution Algorithm.

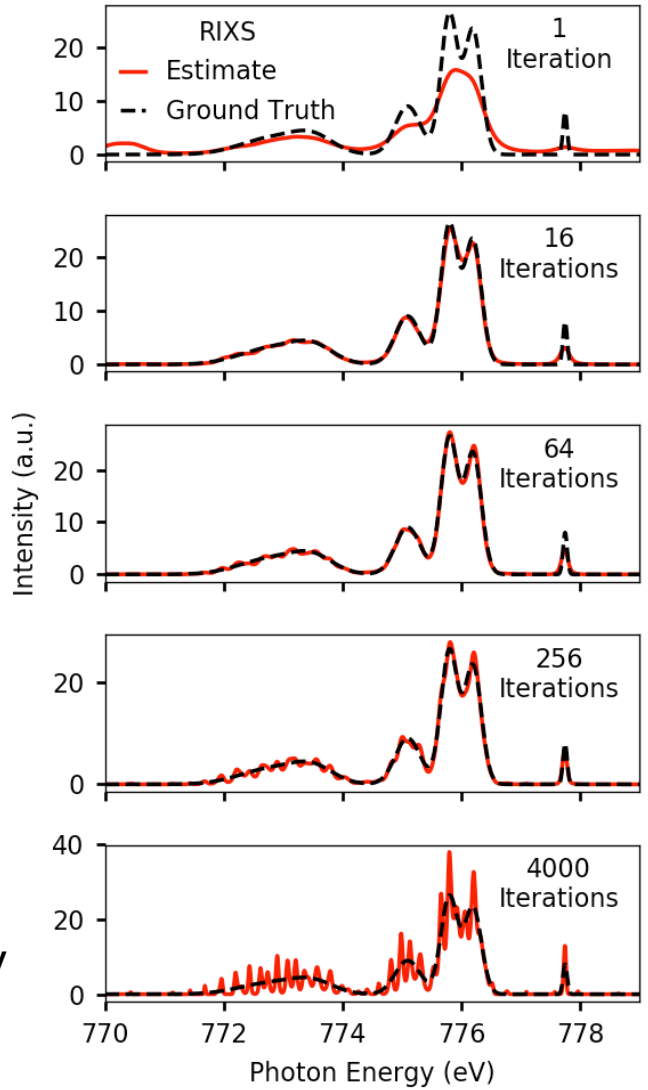
$$\hat{s}^{(n+1)} = \hat{s}^{(n)} \left(p^T * \frac{m}{p * \hat{s}^{(n)}} \right)$$

$\hat{s}^{(n)}$ Estimate of unknown RIXS spectrum after n iterations

m Measured PAX spectrum

p : Converter material photoemission spectrum



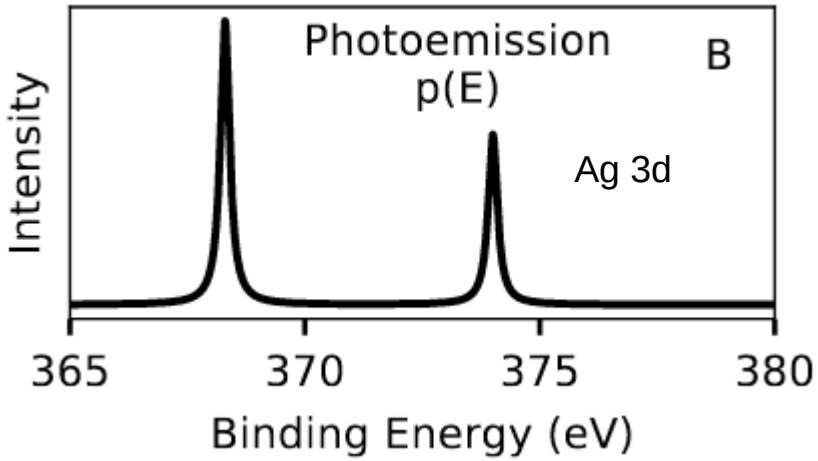


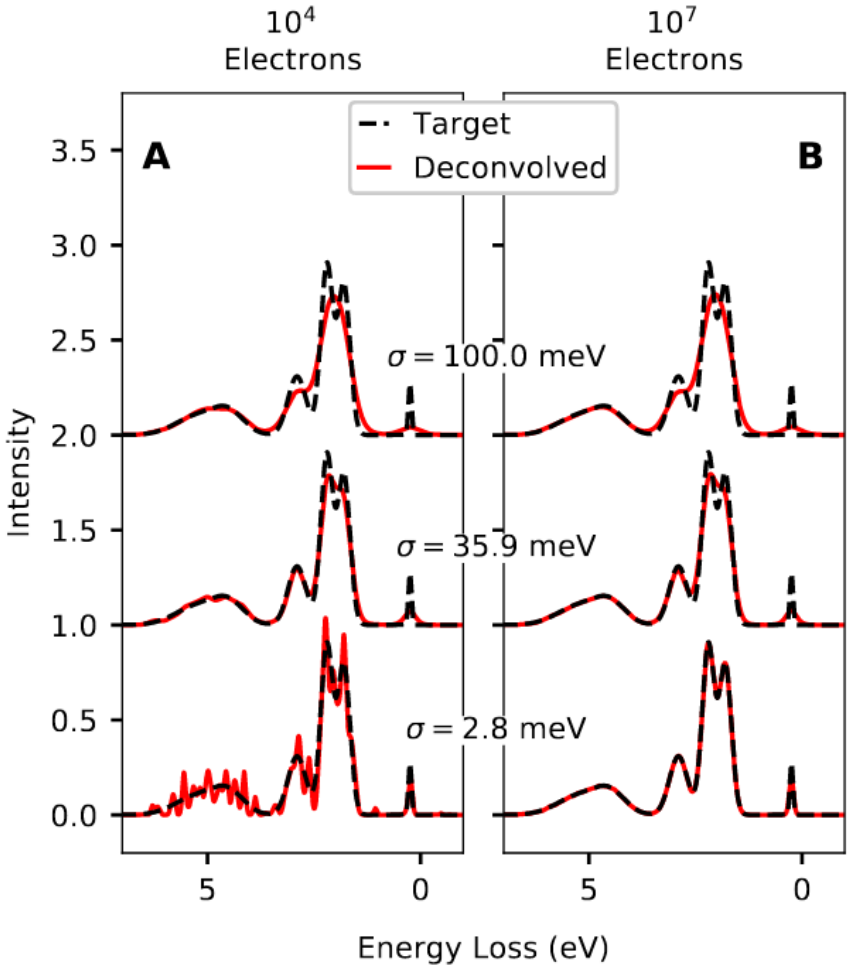
But this algorithm can amplify high frequency noise with high numbers of iterations.

W. H. Richardson (1972), L. B. Lucy (1974)

Avoiding High Frequency Noise Amplification

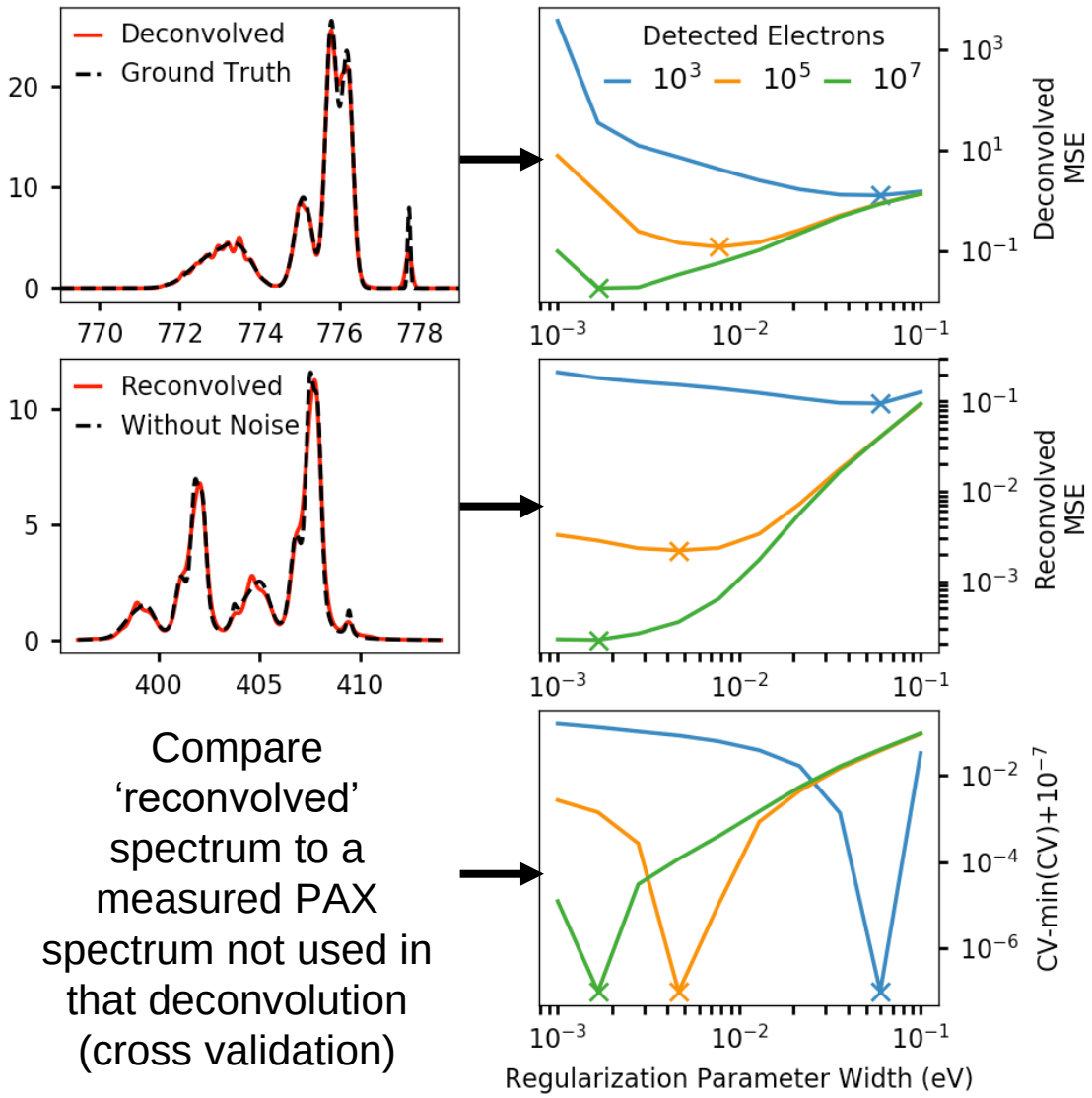
- One solution: convolve with a Gaussian after each iteration.
- How do we set the width of the Gaussian (degree of smoothness)?





T. T. Fister *et al.* (2007)

Estimating a Good Degree of Sharpness (Regularization Hyperparameter)



Compare ‘reconvolved’ spectrum to a measured PAX spectrum not used in that deconvolution (cross validation)

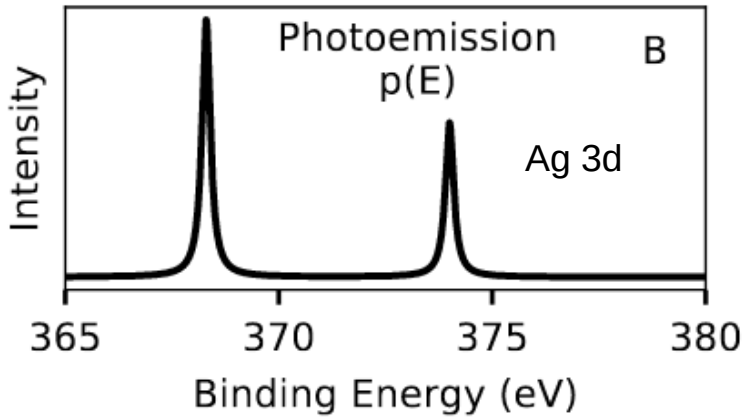
Want, but not experimentally accessible

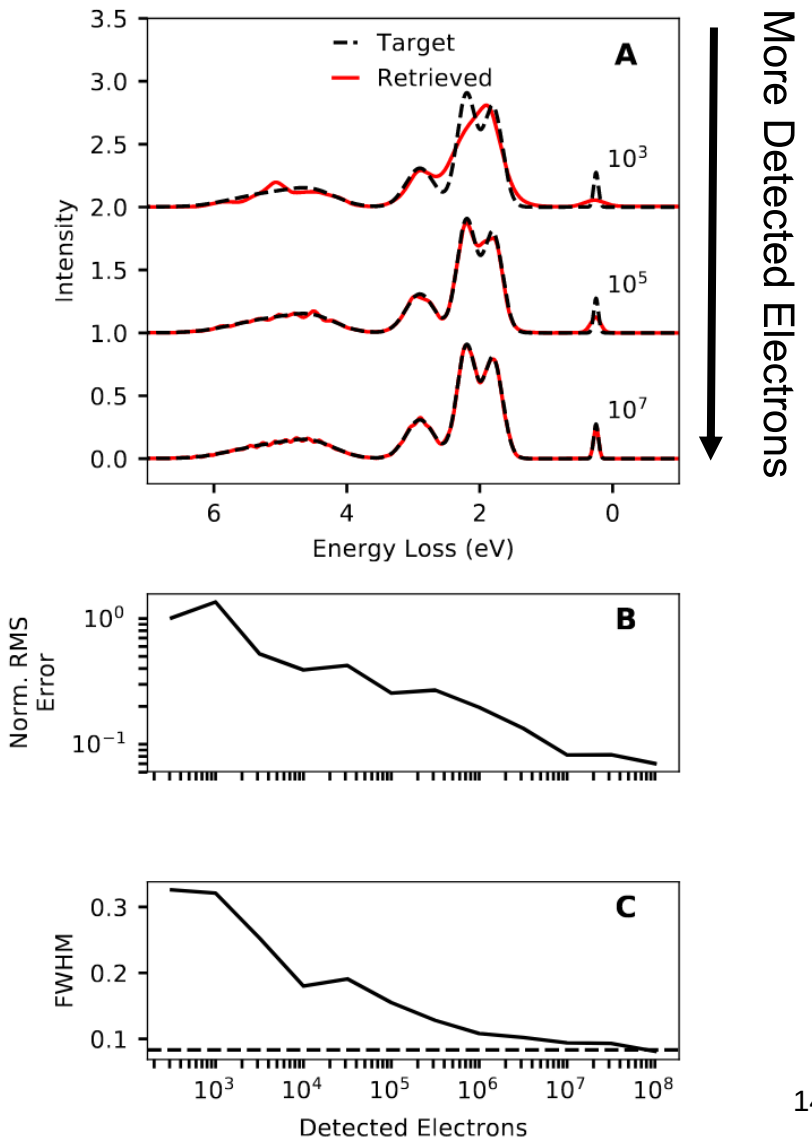
Works, but also not experimentally accessible

Works, and experimentally accesible!

Simulated Performance of PAX in Measurement of a Model RIXS Spectrum

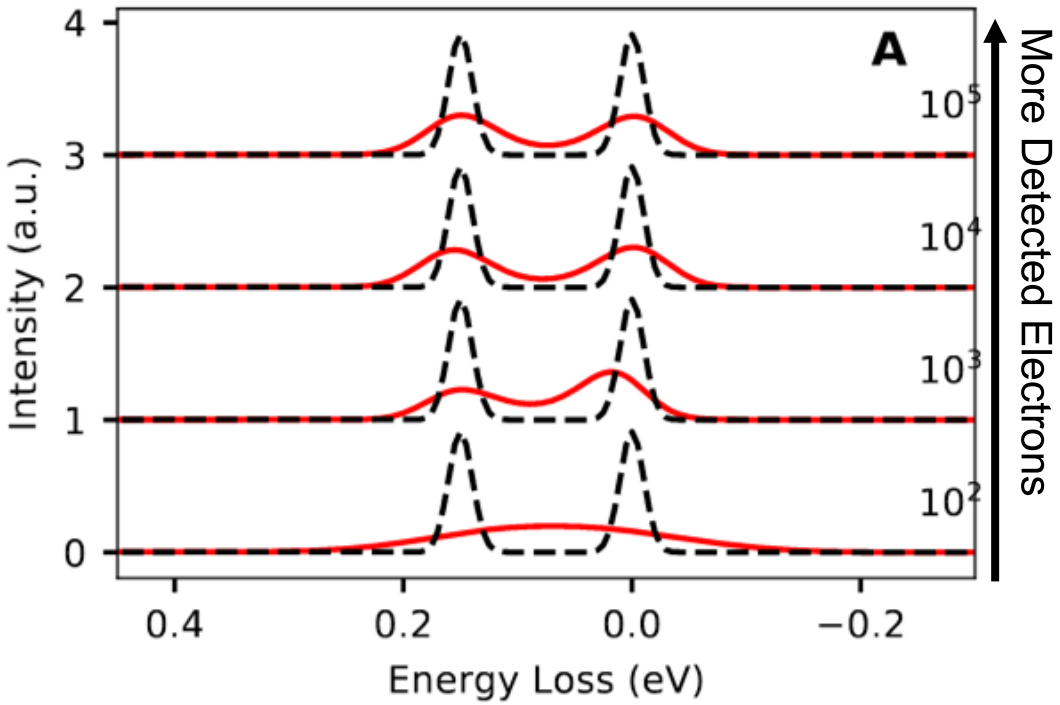
- Mean squared error decreases with increasing number of detected electrons.
- Sharpness of estimated RIXS spectrum increases with increasing number of detected electrons.

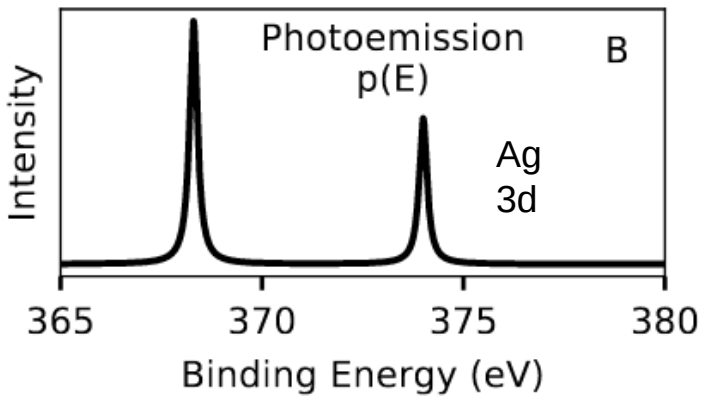




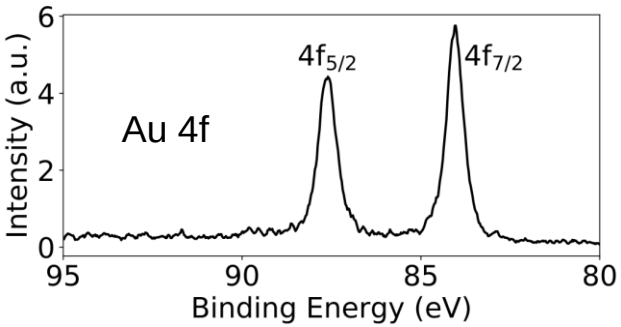
Simulated Performance of PAX in Measurement of a Doublet

- With increasing numbers of detected electrons, the two peaks of the doublet become more clearly separated.



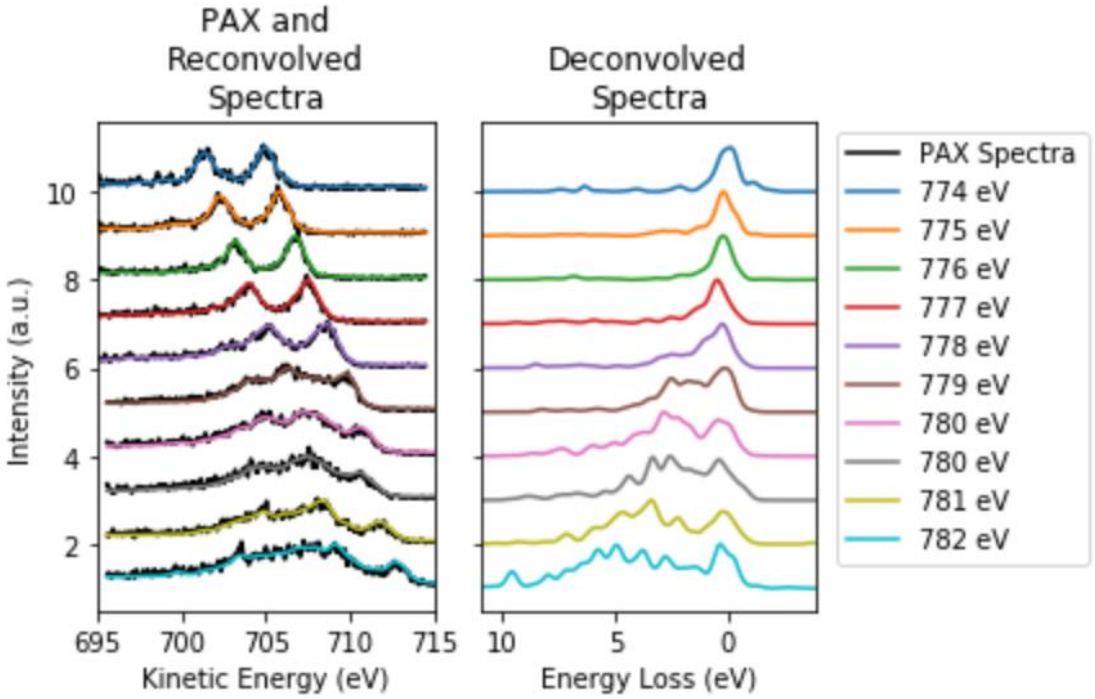


Pilot Study at LCLS Successfully Demonstrated Concept



~550 meV Resolution

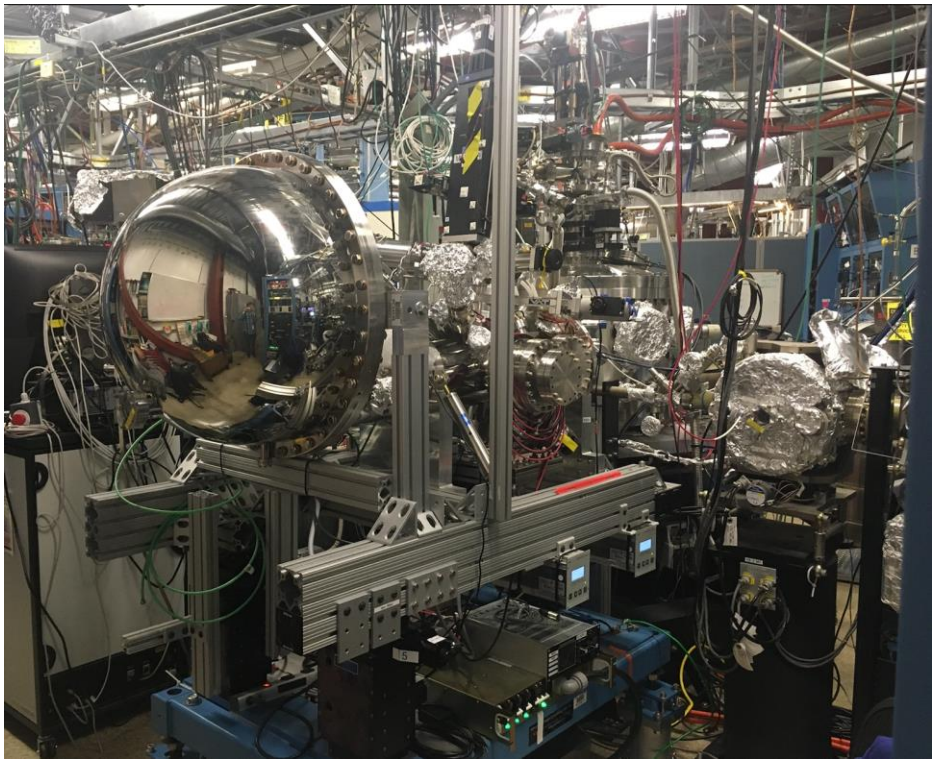
CoO Sample Results:



Raw data from G. L. Dakovski *et al.* (2017)

Ongoing Experimental Efforts

- We have assembled and tested an endstation at SSRL, a synchrotron radiation light source at SLAC.
- We just moved the endstation to an undulator beamline to perform PAX experiments in the next few months.



Summary

- It is possible to accurately estimate an unknown RIXS spectrum from the corresponding PAX spectrum.
- Simulations show high potential of PAX for moderate resolution experiments (characterizing features with 100s of meV widths).
- PAX concept has been demonstrated with low resolution, experimental work is ongoing.

Acknowledgements

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