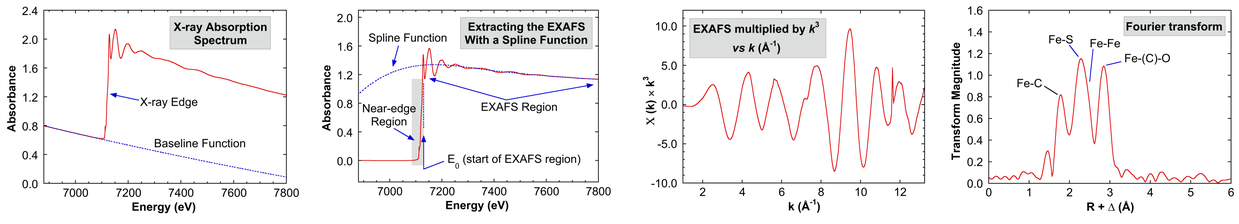
Laboratory-Based EXAFS Spectroscopy with Cryogenic Detectors

Simon J. George[1], Matthew H. Carpenter[1], Stephan Friedrich[2], Robin Cantor[1]
[1] STAR Cryoelectronics, Santa Fe NM 87508 USA
[2] Lawrence Livermore National Laboratory, Livermore CA 94550 USA

EXAFS Spectroscopy

What is EXAFS?

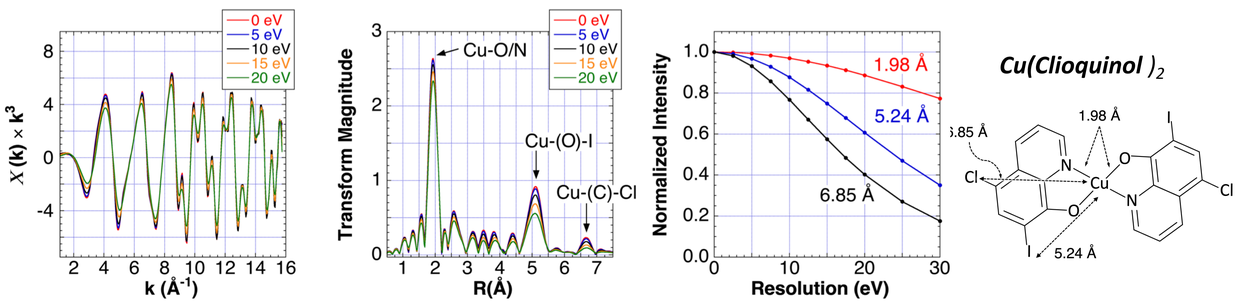


- EXAFS = Extended X-ray Absorption Fine Structure, a technique for determining atomic structure
- EXAFS oscillations are due to quantum interference between the photoelectron waves from X-ray emission and those backscattered from nearby atoms
- EXAFS gives numbers, elements, distances of neighboring atoms without the need for a crystalline sample
- Currently, EXAFS requires a synchrotron radiation light source with a tunable monochromatic beam

Laboratory EXAFS Spectroscopy

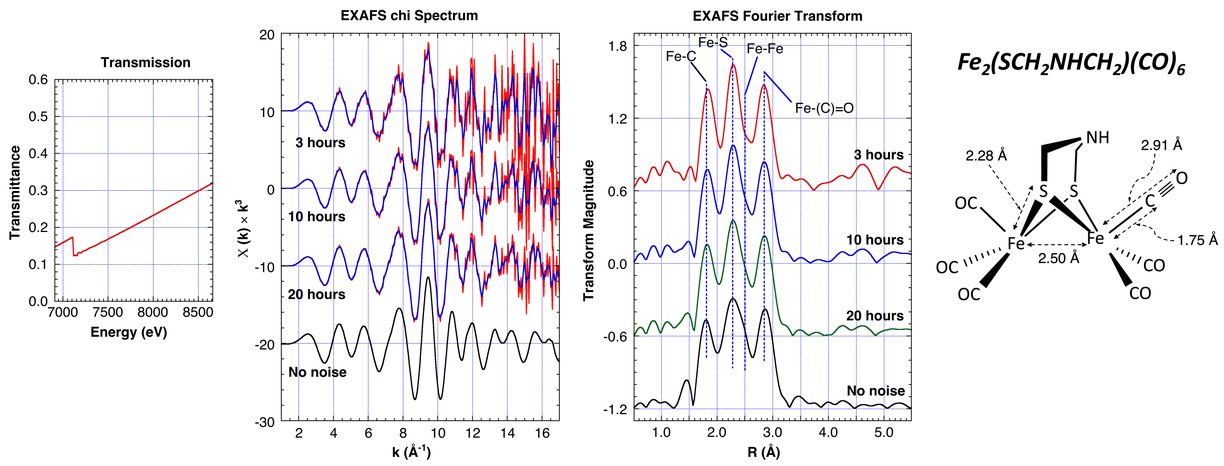
- A commercial laboratory EXAFS instrument would be a major advance, opening up the technique
- ***Problem***: EXAFS is limited by statistics at high energies above the X-ray edge, where oscillations are weak
- ***Question***: What are resolution and statistics requirements for a commercially viable EXAFS instrument?

Resolution Requirements for EXAFS



- Gaussian broadening of experimental EXAFS spectra deceases peak amplitudes in Fourier transform
- ***Conclusion***: ~20 eV FWHM is OK, < 15 eV is ideal

Statistics Requirements for EXAFS



- Adding Poissonian noise to 15 eV broadened EXAFS spectra degrades spatial resolution
- Estimate 50 mM Fe can be measured to k = 17 Å^{-1} in a few hours – assuming 100 counts/s/eV bandwidth
- **Conclusion:** Acquisition time of ~3-10 hours for a 50 mM sample requires 400,000 cts/s at 4 keV bandwidth

Comparison of Different Technologies

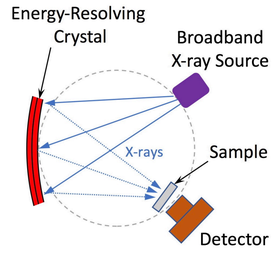
Detector	Resolution	Speed/Pixel	Stability	Energy Range
Ge, Si	~100-200 eV	~1000 counts/s	Good	~MeV
Crystals	~1 eV	Scanning	Difficult	~100 keV
TESs, MMCs	~2 eV	~10 counts/s	Good	~100 keV
STJs	~15 eV	~1000 counts/s	Good	~1keV

Conclusions

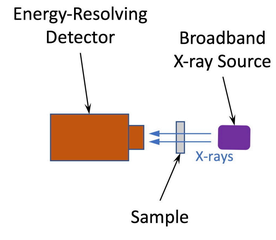
- STJ detectors offer the best prospects for a practical commercial laboratory EXAFS spectrometer
- Need to develop a 128-pixel hard X-ray STJ with:
  Energy Range: 1000 – 12,000 eV
  Resolution: < 20 eV FWHM
  Speed: ~ 2,000 – 5,000 counts/s per pixel

Laboratory EXAFS Using STJ Detectors

Laboratory X-ray Spectrometers with a Broadband Source

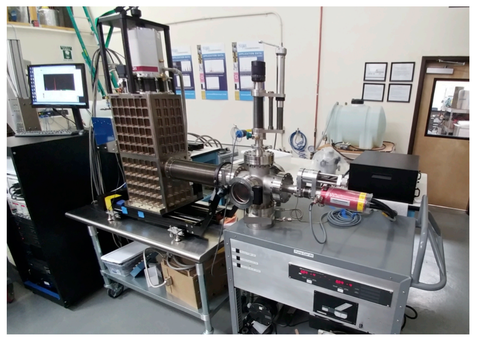


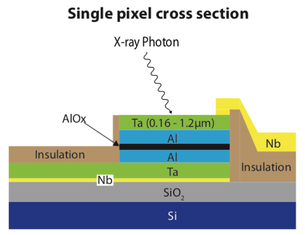
Option 1:
- Use a energy-resolving crystal
- Scan detector / crystal to measure spectrum

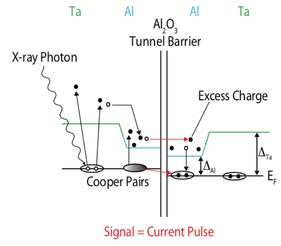


Option 2:
- Use energy resolving detector
- Collect the entire spectrum in a single exposure

A Superconducting Soft X-ray EXAFS Spectrometer

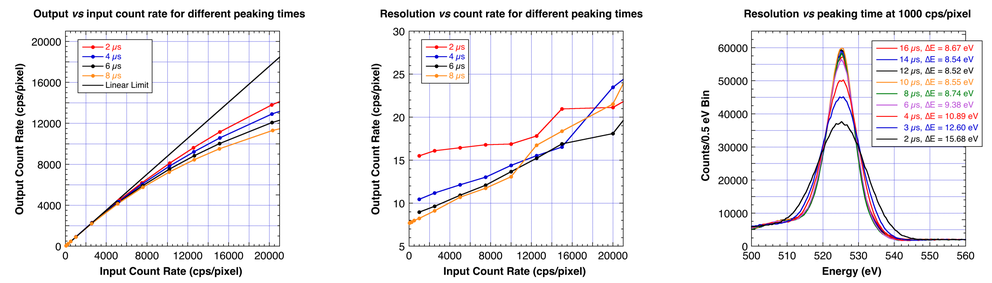






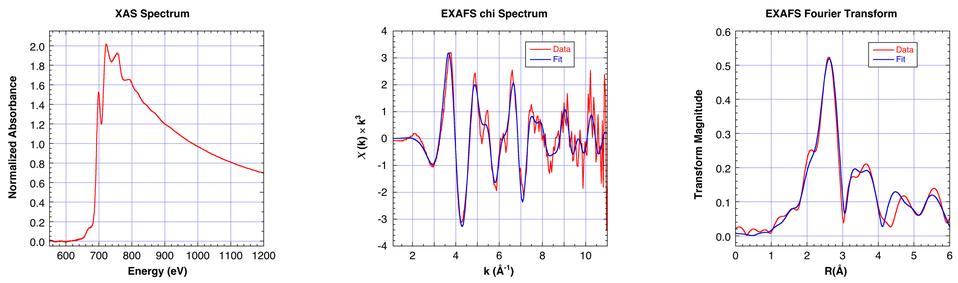
- Sample chamber with windowless broadband X-ray generator with Ag/Ti anode
- 32 pixel STJ detector array in liquid-cryogen-free two-stage ADR

Superconducting Tunnel Junction Detector Performance



- Cryogenic detector with high count rate performance at good resolution
- Linear response up to ~ 5,000 counts/s per pixel with < 5% deadtime
- Best compromise between count rate, resolution and linearity for 4 μs peaking time

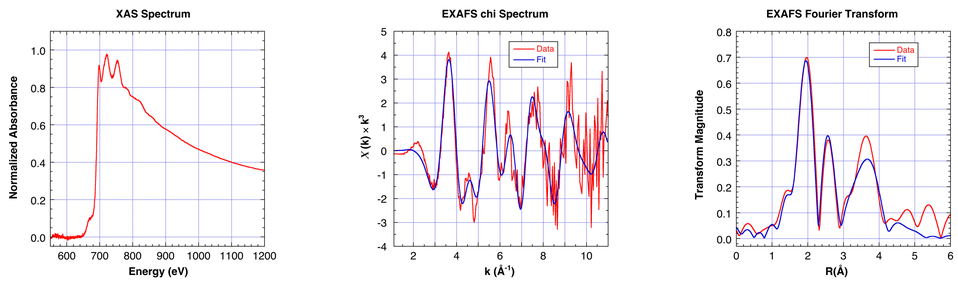
LiF EXAFS with STJs



LiF F-Kedge EXAFS Curve Fitting				
	N	R (Å)	σ² (Å²)	ΔE₀ (eV)
F-Li	3	1.980	0.018	-14.25
F-Li	1	1.717	0.017	
F-F	6	2.792	0.014	
F-Li	4	3.330	0.0078	
F-F (*)	3	4.029	0.014	
F-F	6	4.859	0.0087	
F-F (*)	6	5.791	0.0088	
(*) = multiple scattered interaction				

- Publication quality EXAFS spectrum of 0.7 μm LiF film on 0.1 μm parylene in 4 hours
- EXAFS dominated by F-F interactions. Curve fit consistent with expected cubic structure

MgF_2 EXAFS with STJs



MgF₂ F-Kedge EXAFS Curve Fitting				
	N	R (Å)	σ² (Å²)	ΔE₀ (eV)
F-Mg	3	1.983	0.0073	-10.66
F-Mg	0.4	1.773	0.0072	
F-F	8	2.763	0.011	
F-F (*)	4 (16)	3.284	0.011	
F-Mg (*)	2 (4)	3.411	0.010	
F-F (*)	2 (4)	3.531	0.010	
F-F (*)	4 (6)	3.972	0.012	
(*) = multiple scattered interaction				

- Publication quality EXAFS spectrum of 0.4 μm MgF_2 film on 0.1 μm parylene in 3 hours
- EXAFS contains F-Mg and F-F interactions. Curve fit consistent with expected cubic structure

25-A Bisbee Court, Santa Fe, NM 87508, USA Phone: 505.424.6454 www.starcryo.com Email: info@starcryo.com