science from emission spectroscopy: Spin Cross-Over (SCO)

- ultrafast x-ray sources at large facilities (x-fels, sliced sources) have recently enabled study of the dynamics of SCO
- why x-rays? magnetic moment of valence electrons perturbs Kβ (and also Kα) transitions

Zhang et al, Nature 509 (2014) 345
science from emission spectroscopy: Spin Cross-Over (SCO)

- Use TES x-ray spectrometer to measure Fe emission from high and low spin iron compounds

**TES array**

![Graph showing Fe Kβ’ and Kβ₁,₃](image)

**xtal**

![Graph showing normalized intensity vs energy](image)

*low and high spin iron compounds distinguished using a TES spectrometer*

Y. Joe et al, J Phys B, submitted
x-ray scattering

Bragg Condition for constructive interference:

\[ 2d \sin \theta = n\lambda \]

scattering intensity determined by the density of electrons in the molecule

x-ray wavelengths: sensitive to nm-scale structure

x-ray scattering probes molecular spatial structure
resonant soft x-ray scattering (RSXS)

tune incident beam to excite electron from core to valence state
electron decays back to core level and emits an x-ray with no change in phase or energy


RSXS probes electronic structure and spatial information in the valence band

see poster by Dan Swetz for more details
charge stripes in high Tc superconductors; universal?

Spatial charge order detected in LaBaCuO using RSXS

La$_{2-x}$Ba$_x$CuO$_4$ (LBCO)

Are ‘charge stripes’ present in all HiTc materials? unknown

Are charge stripes present but disordered and heterogeneous?

Disordered stripes will produce a weak, broad scattering signal

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Valence charge density

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The large fluorescence background is a problem in RSXS need a spectrometer capable of separating signal/background
240 TES spectrometer at APS for elastic scattering

Measurements at the NSLS demonstrating ability of TESs to separate elastically scattered beam from low-Z fluorescence

240 TES instrument installed at APS in 2014. Scattering chamber functional ~ fall 2015
conclusions and future directions

- TES spectrometers are proliferating and there is interest from new communities
- new applications in both laboratory-scale experiments and at large facilities
  - absorption spectroscopy (O’Neil)
  - emission spectroscopy
  - x-ray scattering
- still, the technology has shortcomings. Fortunately, these are not fundamental. Solutions are in sight.
- higher system count rates desirable: more and faster pixels
  - physics and engineering details matter in large arrays at high count rates (B-field sensitivity, x-talk). Want to routinely achieve 1-2 eV resolutions at ≥ 100 cps/sensor
  - x-ray optics (polycapillary, HOPG, multilayer, ...) can help reject out-of-band photons
  - more mature multiplexing, especially microwave (Mates, Kempf/Wegner, Kohjiro)
  - algorithms for piled-up (Fowler, Alpert) and non-linear events (Moseley)

see also “review of superconducting transition-edge sensors for x-ray and gamma-ray spectroscopy” Supercond. Sci. Technol. 28 (2015) 084003
thank you!

+ Jim Beall, Joe Fowler, Jiansong Gao, James Hays-Wehle, Hannes Hubmayr, Young Il Joe, Jeff van Lanen
Optical pump, x-ray probe technique

Ti:Sapphire Laser system
20mJ/pulse @ 1kHz, 40fs

Delay Stage

X-ray Optic

Water Jet

Microcal array detector

Liquid jet
Sample

Chopper

BBO

expect 2 picosecond time resolution, limited by path variation in x-ray optic
Microcalorimeter arrays

- 2.5 eV fwhm resolution at 6 keV possible
- 80%+ quantum efficiency 6-8 keV
- Large collection area

- 240 pixel array
- 23.4 mm$^2$ active area
- 30% fill fraction
Lanthanide x-rays among ruler lines

- calibration lines establish a “ruler” that is used to locate less well known lines
- only one spectrum is shown above but we have a spectrum from each of the 94 sensors
Low spin (LS) and high spin (HS) Fe compounds; equivalent to 100% excitation fraction
- 5.5 eV resolution, 4 hrs per sample
- Comparison to Vanko, JPCB 110, 11647 (2006) shows all expected LS <-> HS features
- Count rate about 1 cps per pixel, limited by exciting plasma source. Strong motivation for brighter source.
charge: shift the edge and add pre-edge features
x-ray absorption fine structure (XAFS)

number of atoms affects fringe intensity
x-ray absorption fine structure (XAFS)

bond length shifts fringe location
bond lengths can be measured with .001-.01 Å accuracy
Demonstration of fluorescence mode XAS with TESs

NEXAFS spectrum measured at NSLS

- better than MultiLayer Mirror (MLM) spectrum of same sample
- Unlike MLM, also works at N, O, ... all other edges