Streamlining qIXS: *q*-dependent Inelastic X-ray Spectroscopy

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Motivation

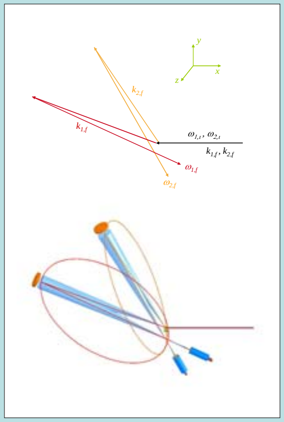
Non-resonant inelastic x-ray scattering (IXS) from core electrons, or non-resonant x-ray Raman scattering (XRS), is a proven method for measuring the electronic states and local structure of low-*Z* materials. XRS is a bulk probe and can operate without a vacuum, unlike many soft x-ray experiments. For $qr \ll 1$, XRS is essentially identical to x-ray absorption spectroscopy (XAS), as seen in its double differential cross section:

$$\frac{d^2\sigma}{d\Omega d\omega} = \left(\frac{d\sigma}{d\omega}\right)_{Th} S(q,\omega) = r_0^2(\varepsilon_1\cdot\varepsilon_2)^2 \frac{\omega_2}{\omega_1}\sum_f \left|\left\langle f\,|\sum_j e^{iq\cdot r_j}|\,0\right\rangle\right|^2 \cdot \delta(E_f - E_i - \hbar\omega)$$

$$\xrightarrow[qr\ll 1]{} \sum_{f,j}\left|\left\langle f\,|\,iq\cdot r_j\,|\,0\right\rangle\right|^2 \delta(E_f - E_0 - \hbar\omega)$$

$$\propto \mu(E) \quad \text{for } q \leftrightarrow \varepsilon$$

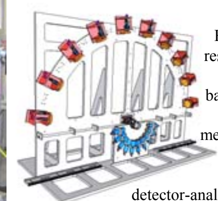
Unlike XAS, it is often possible to raise *q* beyond the limitations of the dipole limit, allowing for multipole transitions. In particular, q-dependent excitons can provide new insight on the local symmetries of materials [1, 2]. New *ab-initio* theories can incorporate this parameter into existing models, adding to the information already present in XRS.



XRS measurements are limited by its low cross section. This fact is compounded when trying to measure spectra for multiple values of *q*. To address this problem, we have built at multicrystal spectrometer that measures multiple momentum transfers at once. The basic idea is shown on the right. Here, the incoming radiation is scattered to our analyzers in the vertical *x-y* plane. These analyzers Bragg-reflect photons of energy $\hbar\omega_f$ to detectors *located outside of the original scattering plane*. As indicated in the drawing on the right, each Rowland circle is perpendicular to the vertical scattering plane with the detectors *behind* the sample. This is a good arrangement for the polarization of the incoming beam and provides ample room for controlling the sample environment with a furnace, cryostat, diamond anvil cell, etc. While other XRS instruments using multiple analyzers generally focus x-rays back to a single detector, there is only one analyzer for each detector in this setup. Our design allows for 19 analyzers and detectors to *simultaneously* measure IXS at different values of *q*. By eliminating much of the time devoted to calibrating and measuring IXS at different momentum transfers, we have greatly improved the efficiency of a qIXS experiment.

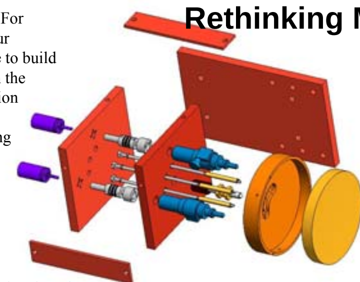
The “Beast”

The photograph on the left was taken during our August run and the drawing below was part of the design for the apparatus. The focused x-ray beam comes through a beam-pipe from the right and scatters off the sample, located three inches from the large aluminum frame. In the vertical plane, x-rays travel to the analyzer crystals which Bragg-reflect them to their respective detectors, located about six inches from the back frame. While we have space for simultaneous measurement of 19 values of *q*, our preliminary setup involved only six detector-analyzer pairs. The rigidity of the entire setup was intentional: following the alignment of each crystal, only the sample's position changed.



Rethinking Mirror Tilts

Homemade Actuators: For adequate resolution in our measurements, we chose to build our own actuators. With the current setup, our precision is 4 mdeg with excellent reproducibility. Including parts and labor, each module cost less than $1000!



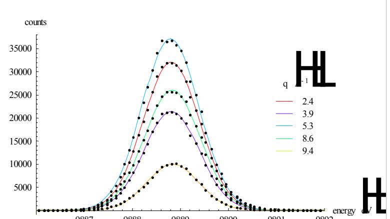
4 inch analyzers: Each silicon <111> analyzer was made by NJXRS-TECH. New bonding technology minimizes bending aberrations toward the edge, dramatically improving both the solid angle and the energy resolution.

Modular design: The box itself is also rigidly designed and can be pinned in one of 19 values of *q* on the back frame.

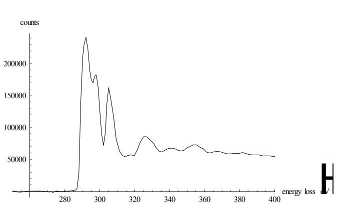
Initial Data

For our trial run, we ran with five analyzers ranging from $q = 2.37 - 9.36$ Å^{-1}. We found that we were able to rapidly collect high-quality XRS data and easily exchange samples without retuning the analyzers. The shape and width of the elastic peak for each analyzer, shown on the right, indicates the overall quality of the spherically bent crystal. The measured FWHM of 1.3 eV is the theoretical limit for the <111> monochrometer at sector 20. Diamond and lithium fluoride, both XRS standards, are shown below.

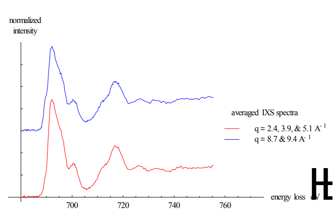
Elastic scattering from LiF (all analyzers)



Diamond XRS
(summed over q = 2.4 and 3.9 Å^{-1} spectra)

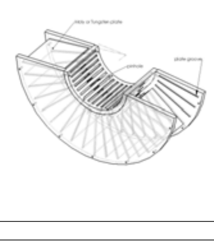


LiF XRS for two ranges of *q* (fluorine K edge)



Future Improvements

Initial results during the August run were quite promising. We only needed a few hours to simultaneously measure the inelastic spectrum for several momentum transfers and new samples did not require any retuning.



Nonetheless, there is still considerable room for improvement before our next run in December. We will eliminate more stray scattering with a rigid helium box, replacing the Mylar balloon used this summer. Tighter baffling around the sample will separate scattered photons from the incident beam. Spatial filtering will be added to the pinholes to further isolate each detector. These modifications can be seen on the left. Sample alignment will also be streamlined by adding motorized control to all three translational axes with one rotational axis for single-crystal studies. The pinhole and sample regions will also be flushed with helium, although the sample chamber will be isolated from the helium box, acting as a load-lock chamber. Most importantly, we will also be working with ten analyzers and detectors and a <311> monochrometer for better overall energy resolution. Eventually, we hope to work with all 19 analyzers and detectors to fully realize the potential of qIXS spectroscopy.

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References:

1. Yejun Feng, G.T. Seidler, J.O. Cross, A.T. Macrander, J.J. Rehr, Phys. Rev. B **69**, 125402 (2004).
2. K. Hamalainen, S. Galambosi, J.A. Soininen, Eric L. Shirley, J. P. Rueff, A. Shukla, Phys. Rev B. **65**, 155111 (2002).