X-Ray Spectroscopy

WHY ARE X-RAY SPECTROSCOPIES USEFUL?

X-ray spectroscopies can not only determine elemental makeup, but also local molecular structure, and are able to detect geometric, chemical, and electronic properties of samples.

Some examples…

- Catalysis
- Coatings
- Food safety
- Microelectronics

HOW DOES X-RAY SPECTROSCOPY WORK?

Primarily, an intense beam of x-rays excites core electrons in a sample and is then simply scattered and absorbed, bringing down other electrons.

Many types of cascading events can take place, and the measurement of specific types of events is what makes up X-ray spectroscopy.

XPS

X-rays excite bound core electrons to become free photoelectrons, which are detected

Screens the core electron's binding energy

XAS

X-rays excite core electrons to unoccupied shells above the Fermi level, detected by the following cascading events as electrons fall to fill the core hole

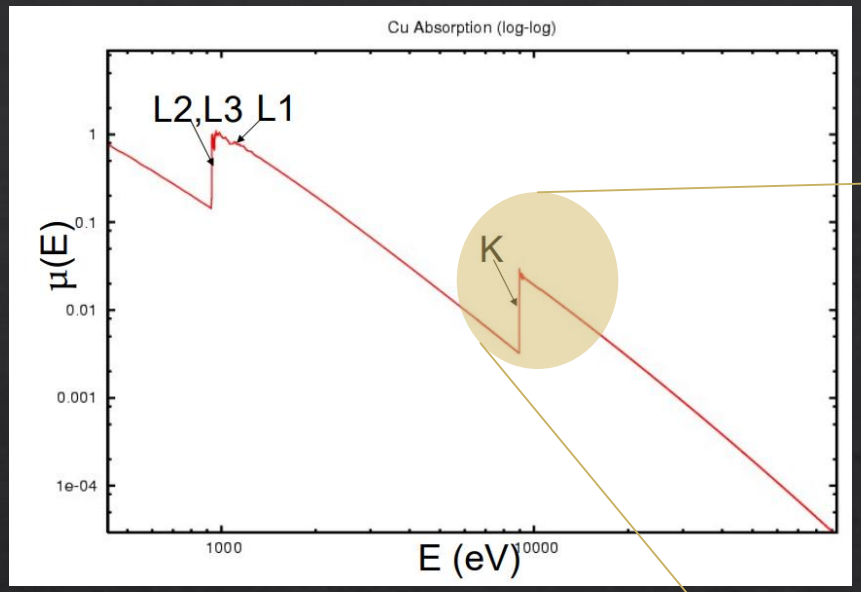
-XANES

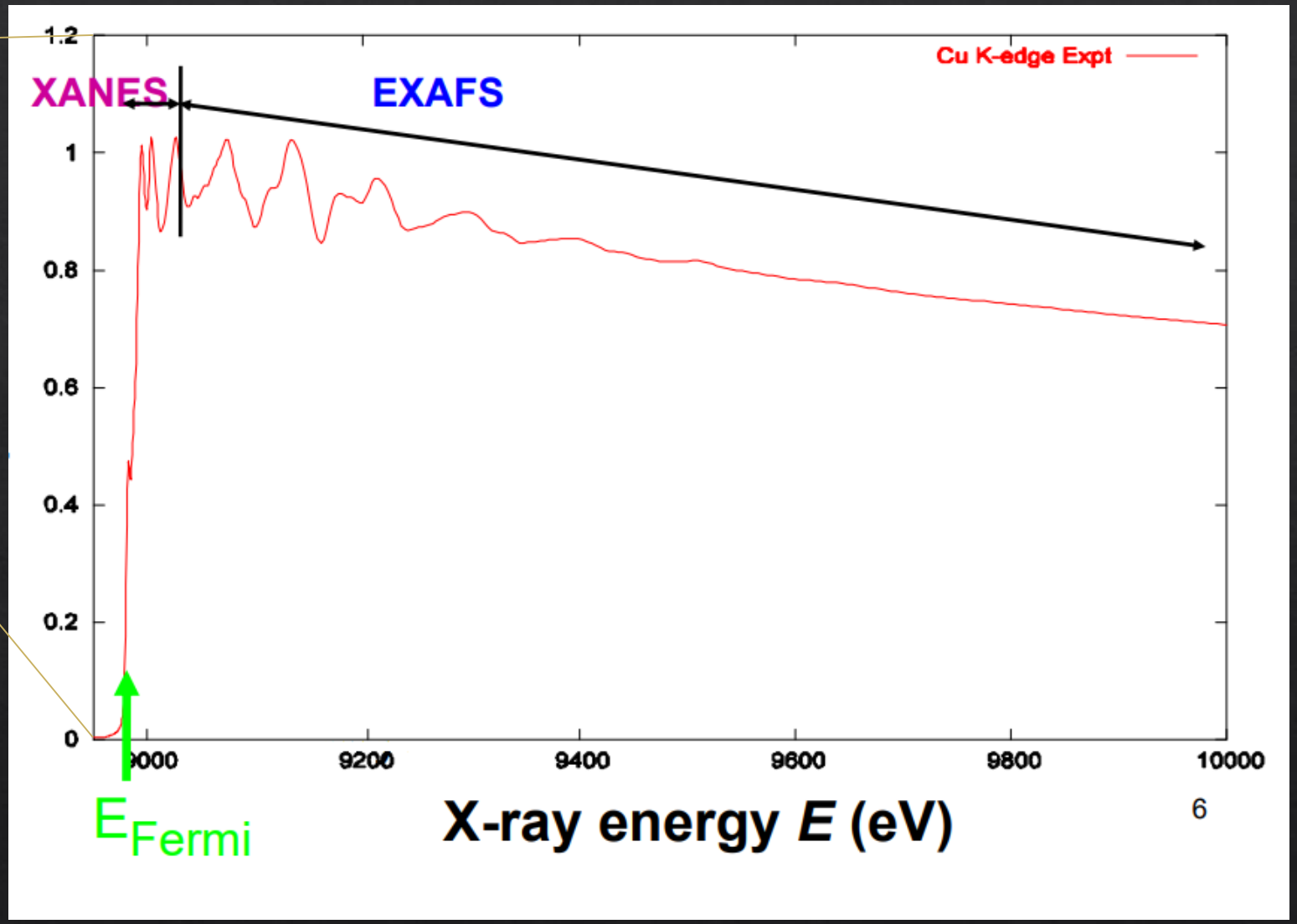
-EXAFS

XES

X-rays create core hole that is filled by valence electrons, shooting out an x-ray of equal energy difference

Similar to XAS, but only valence (sub-Fermi) electron decays are measured


Cu Absorption (log-log)
L2,L3 L1
K
μ(E)
E (eV)
1
0.1
0.01
0.001
1e-04
1000
10000


Cu K-edge Expt
XANES
EXAFS
1.2
1
0.8
0.6
0.4
0.2
0
9200
9400
9600
9800
10000
E Fermi
X-ray energy E (eV)

ANALYSIS AND PREDICTION OF X-RAY SPECTRA

X-ray spectra are not straightforward to analyze, and have to be understood mostly on a case-by-case basis.

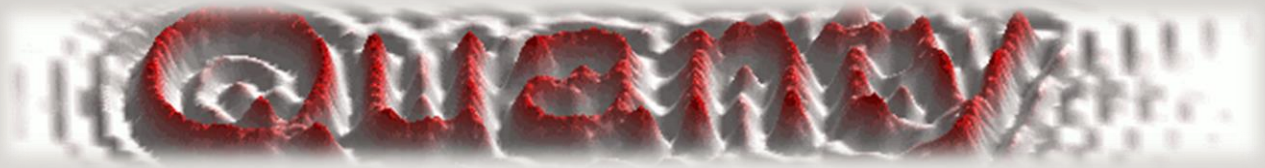
However, by building models that can calculate the spectra, one could use that to both analyze and predict spectra for a large variety of samples.

Beginning 20 years ago, my group began writing code for calculating different types of x-ray spectra. This ongoing project culminated in FEFF, an internationally recognized program for the ab-initio calculation of an assortment of x-ray spectra.

FEFF 9.6.4

Can calculate:

- EXAFS – Extended X-ray Absorption Fine Structure
- XANES – X-ray Absorption Near-Edge Structure
- XNCD & XMCD – X-ray Natural & Magnetic Circular Dichroism
- SPXAS & SPEXAFS – Spin Polarized X-ray Absorption Spectra
- XES – non-resonant X-ray Emission Spectra
- Compton scattering
- NRIXS – Non-Resonant Inelastic X-ray Scattering
- EELS – relativistic Electron Energy Loss Spectroscopy



MY GOAL: INCORPORATE MULTIPLET EFFECTS USING QUANTY

QUANTY is a quantum many-body script language that solves quantum mechanical problems formulated in second quantization.

My goal was to write scripts for QUANTY to calculate simple XAS and/or XPS spectra, and attempt to include atomic multiplet effects in the spectra.

SECOND QUANTIZATION AND FOCK SPACE

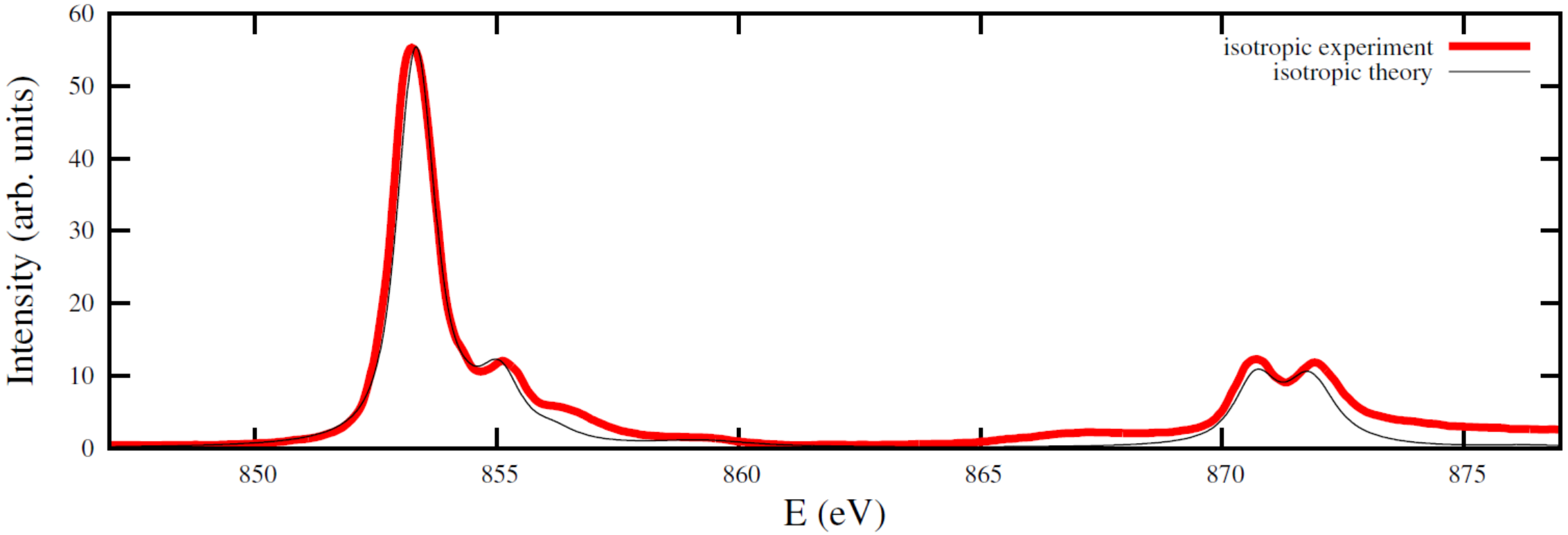
Essentially, a Fock space is a space of quantum states identified by parameters (e.g. quantum numbers).

Working in this space allows one to rewrite any operator as a linear combination of products of creation and annihilation operators.

This is used in the second quantization formalism for many-body systems.

$$F_\nu(H) = \bigoplus_{n=0}^{\infty} S_\nu H^{\otimes n} = \mathbb{C} \oplus H \oplus (S_\nu (H \otimes H)) \oplus (S_\nu (H \otimes H \otimes H)) \oplus \dots$$

QUANTY - NiO XAS Spectrum



FUTURE WORK

- Solve XPS spectra for simple TMOs
- Include multiplet effects for XAS spectra

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Thank you for the REU

Thank you for the money