

X-ray Absorption by Atomic and Multi-Particle Gases

AtomDB

2020

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Collaborators:

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Science:

Theoretical Atomic Physics

Atomic Data Production (e.g., XSTAR)

X-ray Spectral Models

Technologies:

Atomic R-matrix Method

Molecular R-matrix Method

Multiple Scattering Theory

(Atomic R-matrix + Fermi Model Perturber)

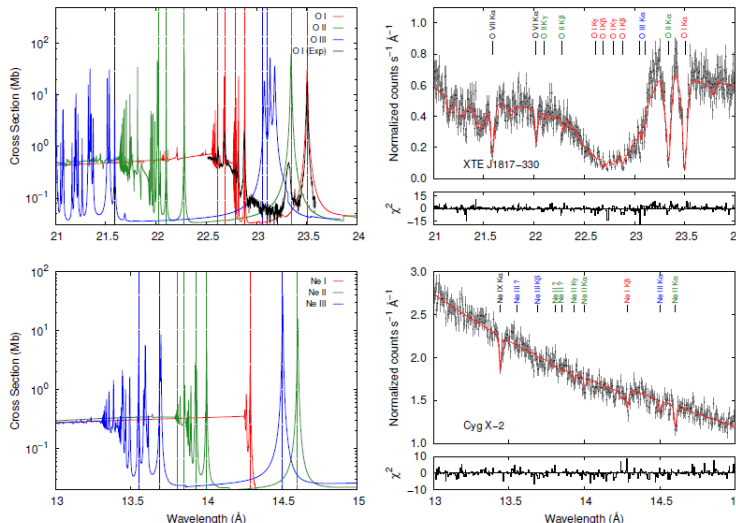
Science Objectives:

Atomic K-shell photoabsorption model for Si

Atomic L-shell photoabsorption model for Fe

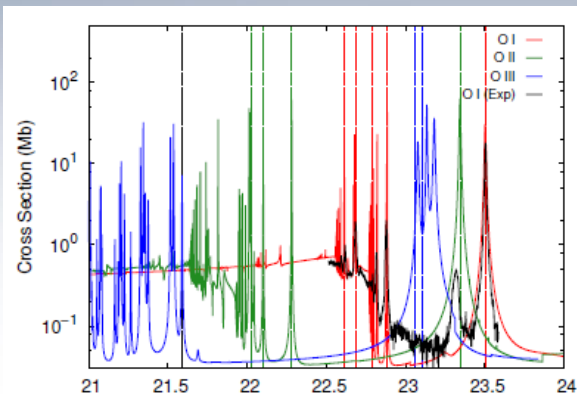
Molecular and solid-state photoabsorption models for O and Si K-edges, and Fe L-edge

Interpretation of X-ray (Chandra) Spectra

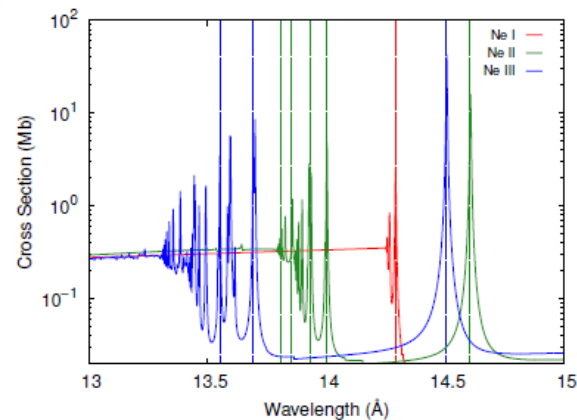


Atomic Cross Sections and X-Ray Spectral Models

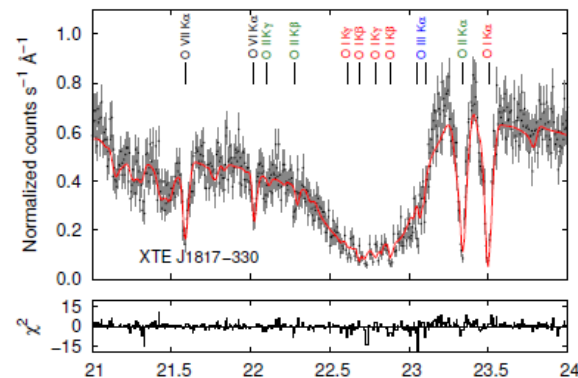
Oxygen Cross Sections



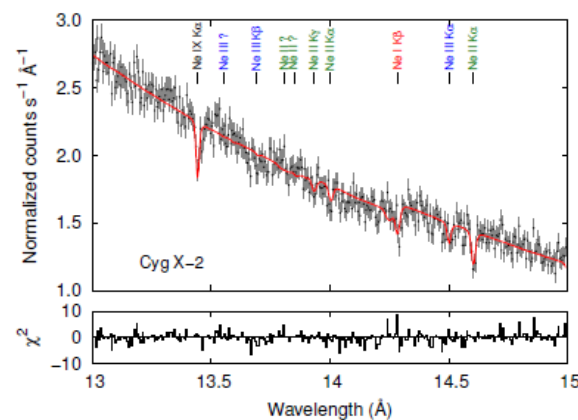
Neon Cross Sections



Oxygen X-ray Spectra



Neon X-ray Spectra



$$\sigma = \frac{4}{3} \pi^2 \alpha \omega |\langle \psi_i | r | \psi_f \rangle|^2$$

Goal: Compute σ

$$I = I_0 e^{-\sigma N}$$

I_0 = Source Intensity

I = Observed Intensity

N = Column Density

σ = Absorption Cross Section₂

Ne R-matrix, Experiment, QDT Analytical Formula

$$\sigma(E) = \frac{\pi(k_e e^2)h}{mc} \times \frac{df}{dE}$$

$$\frac{df}{dE} = \sum_n f_n \frac{\Gamma/2\pi}{(E - E_n)^2 + (\Gamma/2)^2}$$

$$E_n = E^{th} - \frac{Z^2 E_{au}}{2(n - \mu)^2}$$

$$f_n = \frac{f_0}{(n - \mu)^3}$$

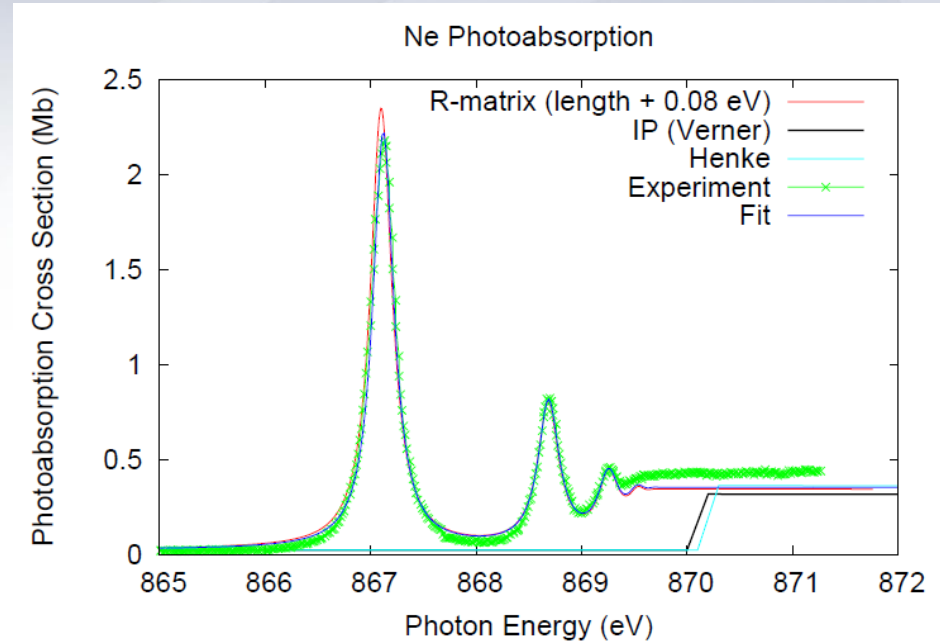
Four Physical Parameters:

E^{th} = Threshold Energy

μ = Quantum Defect

Γ = Core Auger Width

f_0 = Scaled Oscillator Strength



Consistency:

Continuity at ionization threshold

(Photoabsorption \rightarrow Photoionization)

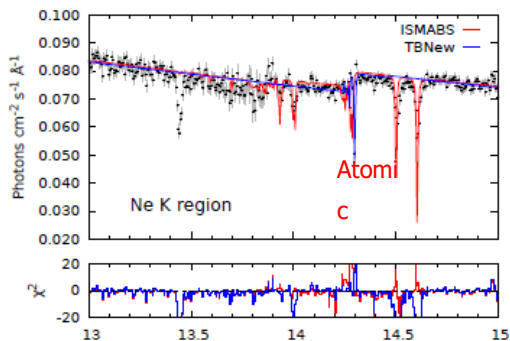
Continuity for \rightarrow atoms \rightarrow molecules \rightarrow solids

Thomas-Reiche-Kuhn Sum Rule:

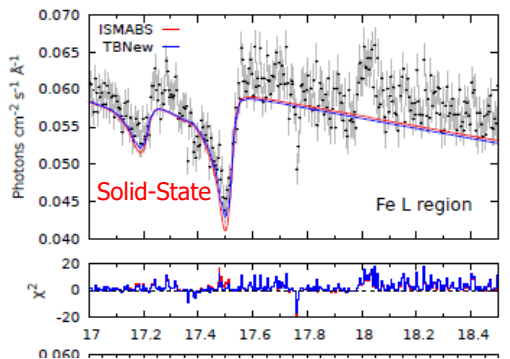
$$\sum_n f_n + \int dE \left(\frac{df}{dE} \right) = 1$$

X-ray Spectra for O and Ne (atomic?) and Si and Fe (solid?)

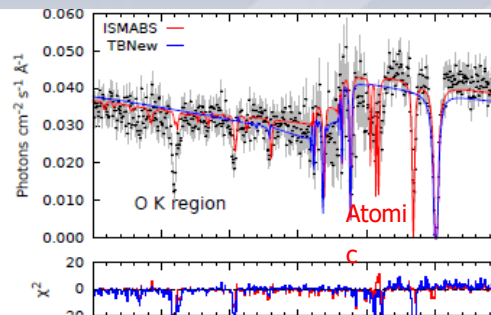
Ne



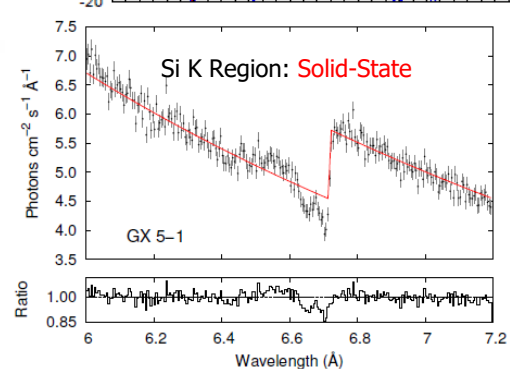
Fe



O



Si



Atomic absorption:
Rydberg series of symmetric Lorentzians

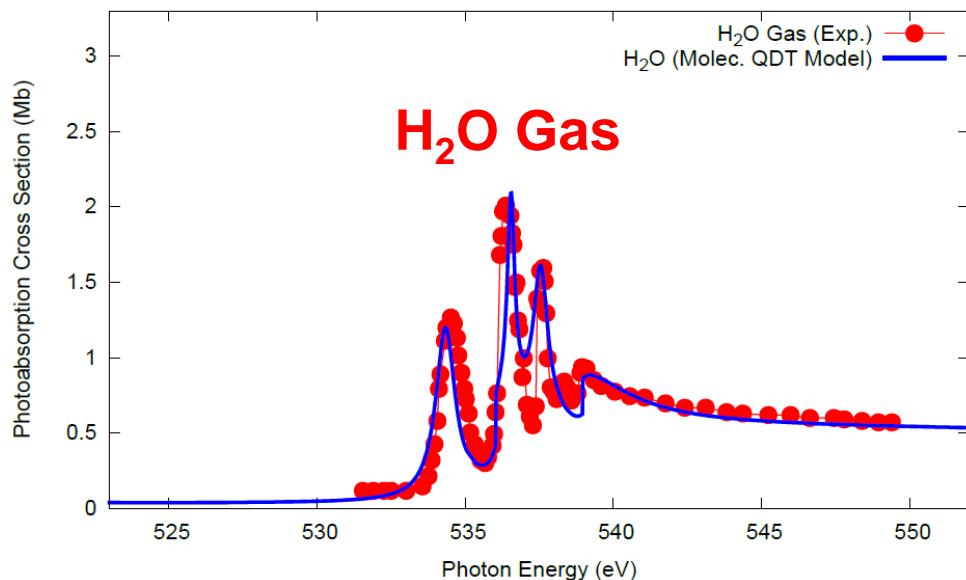
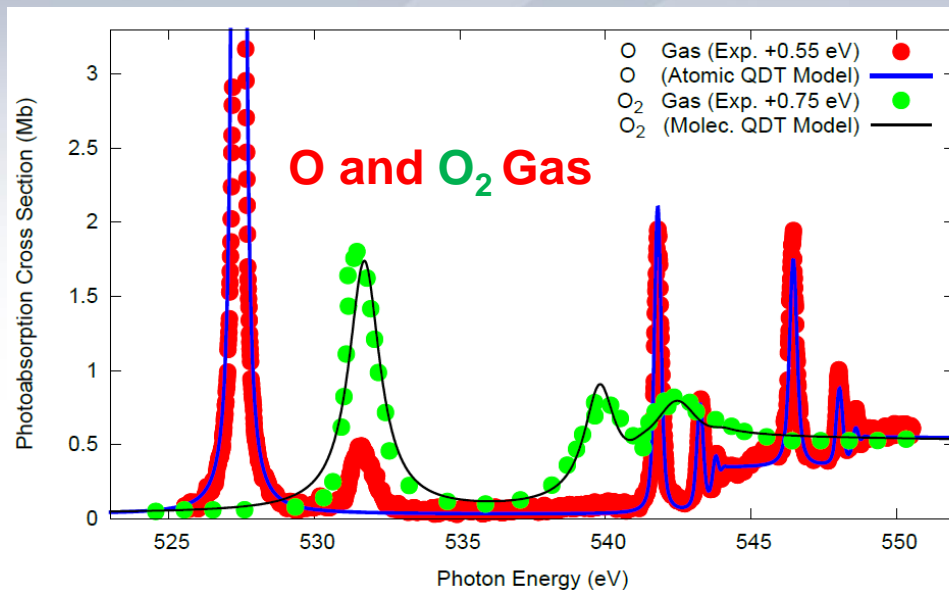
Molecular/Solid-state absorption:
Broad, oscillatory, asymmetric profile

Molecular/Solid-State O?
Atomic Si, Fe?

Differences between Atomic and Molecular Absorption Spectra

Experimental cross sections (originally arbitrary units and uncalibrated energies. Renormalized and shifted for consistency.

Analytical Fits (solid lines through experimental data):
Atomic Quantum Defect
Analytical formula for O,
Modified Rydberg Series and
Resonance formula for O₂
H₂O.



Molecular R-matrix Calculations with Quantemol (with Jonathan Tennyson, UCL)

AtomDB

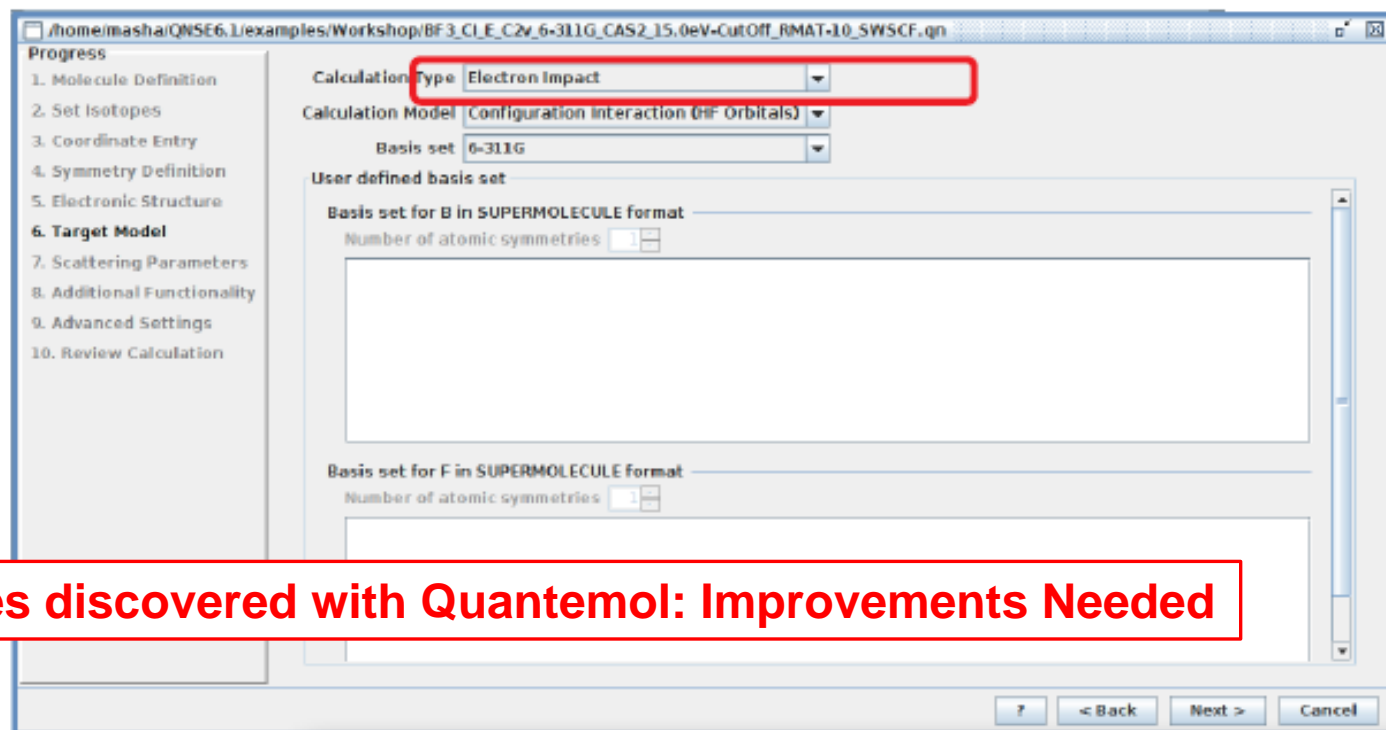
2020

Using Quantemol- N: Quantemol

6. Type & level of the calculation

2 types:

- Electron Scattering
- Photoionization



The screenshot shows the Quantemol software interface with the following settings:

- File path: /home/masha/QN5E6.1/examples/Workshop/BF3_Cl_E_C2v_6-311G_CAS2_15.0eV-Cutoff_RMAT-10_SWSCF.qn
- Progress list:
 1. Molecule Definition
 2. Set Isotopes
 3. Coordinate Entry
 4. Symmetry Definition
 5. Electronic Structure
 6. Target Model
 7. Scattering Parameters
 8. Additional Functionality
 9. Advanced Settings
 10. Review Calculation
- Calculation Type: Electron Impact (highlighted with a red box)
- Calculation Model: Configuration Interaction (HF Orbitals)
- Basis set: 6-311G
- User defined basis set:
 - Basis set for B in SUPERMOLECULE format
 - Number of atomic symmetries: 1
- Basis set for F in SUPERMOLECULE format
- Number of atomic symmetries: 1

Buttons at the bottom: ? < Back Next > Cancel

Inner-shell difficulties discovered with Quantemol: Improvements Needed

September 14, London

Atomic R-matrix for Fe (2017) vs. Solid Experiment

Large $2p^{-1}_{3/2} - 2p^{-1}_{1/2}$ fine structure splitting large (720-707 ~ 13 eV)

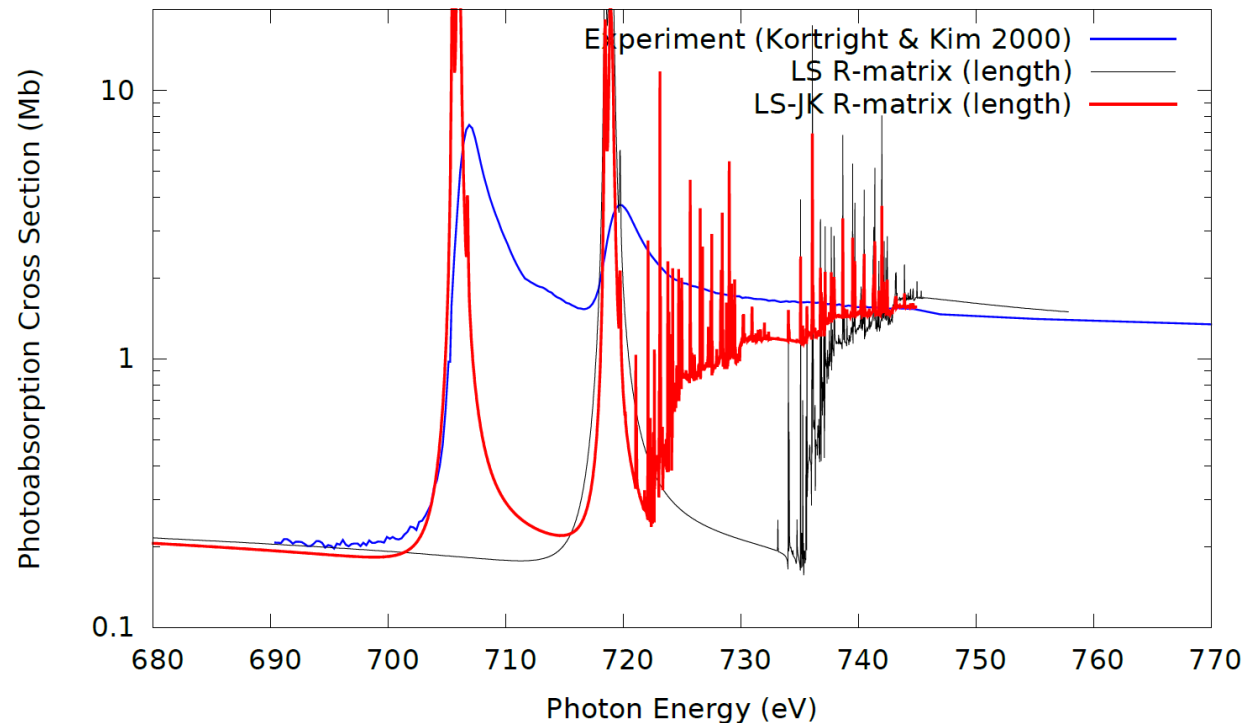
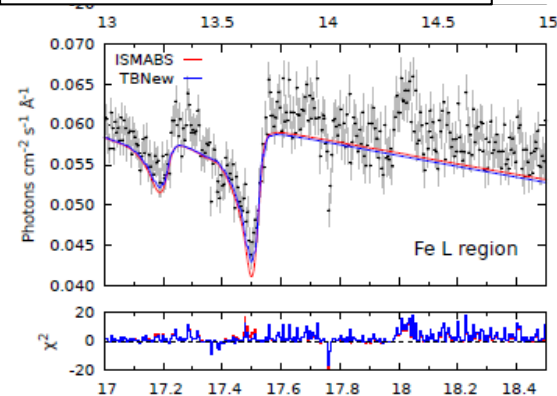
Requiring relativistic treatment (spin-orbit+)

$2p^{-1}$ and $3d^6$ open shells require numerous configurations even in LS lowest order CI

Full Breit-Pauli, large CI R-matrix calculation is ultimately required

Cruder Fano-Transformation (Spin-Orbit series splitting) used

Chandra Data vs.
Model using Solid Fe
Experimental Data



Atomic R-matrix for Fe (2017) vs. Solid Experiment

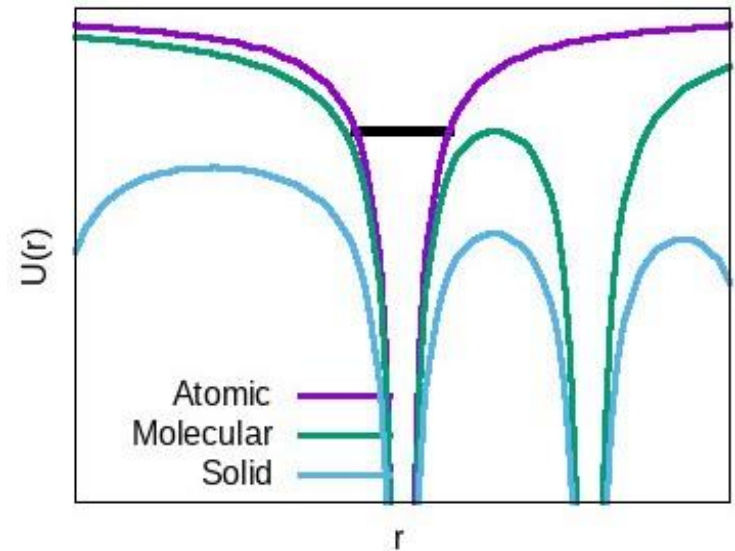
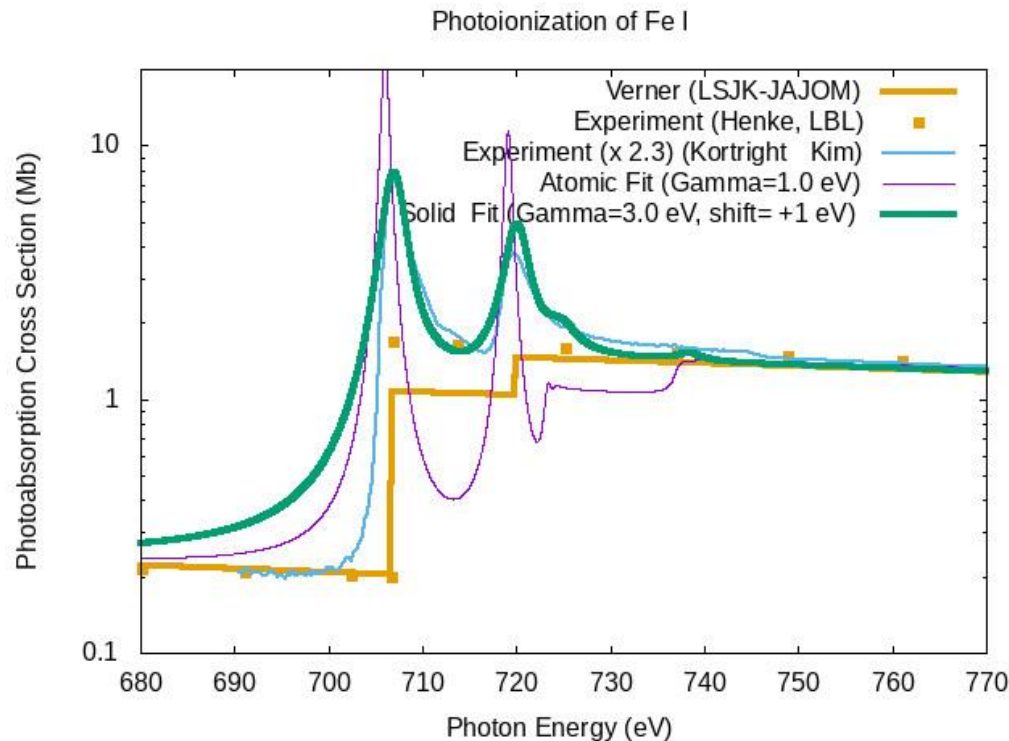
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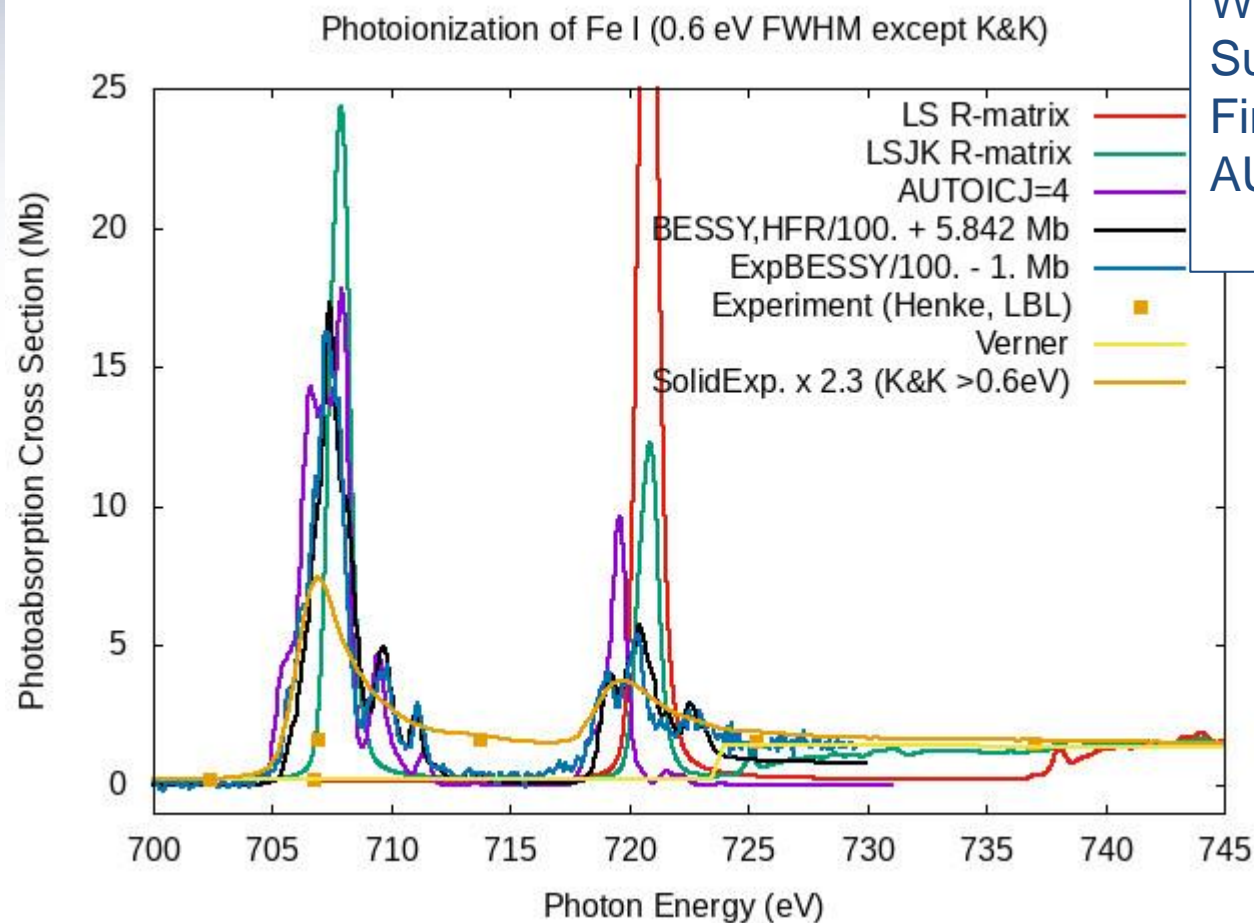
Full Breit-Pauli, large CI R-matrix calculation is ultimately required (unfinished)

Cruder Fano-Transformation (Spin-Orbit series splitting) used



Fe L-Edge: R-matrix and AUTOSTRUCTURE Calculations

WMU Visit by N. R. Badnell
 Summer 2019
 Fine-Structure Splitting Needed
 AUTOSTRUCTURE Easier



Comparisons with Theoretical and Experimental Data

Experiment: Initial State Resolved

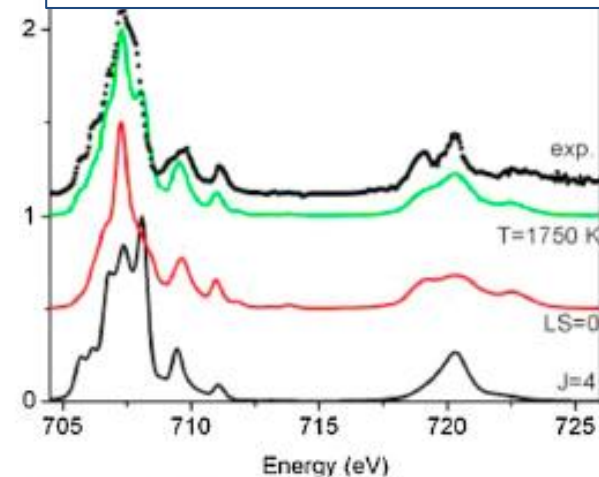


Fig. 2. The atomic multiplet calculations for 5D_5 ground state (bottom), the averaged spectrum for the five J -states (middle) and the spectrum calculated at 1750 K using a Boltzmann distribution (top, solid). The 1750 K spectrum is compared with the experiment 2p XAS spectrum of an iron atom. Digitized from [196].

“Solid” asymmetric features might be nonuniform fine structure broadening

Si K-edge Models:

Atomic R-matrix (LS)
Corrales+ 2019 Model
LLNL Solid Experiment

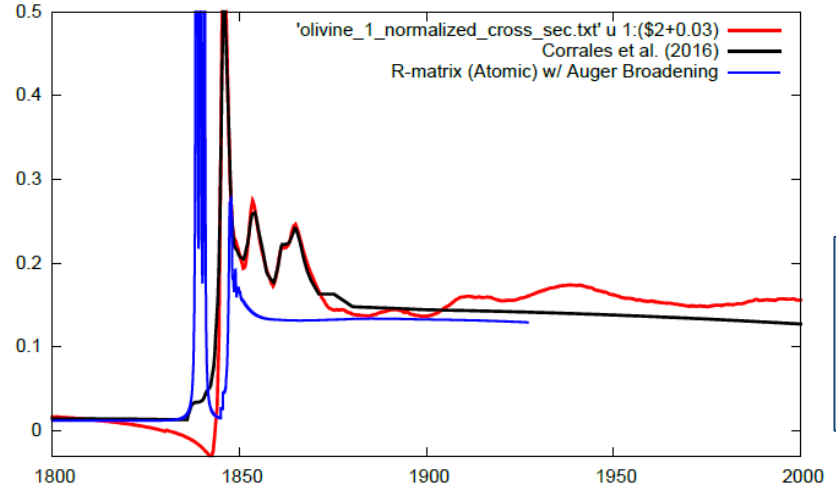
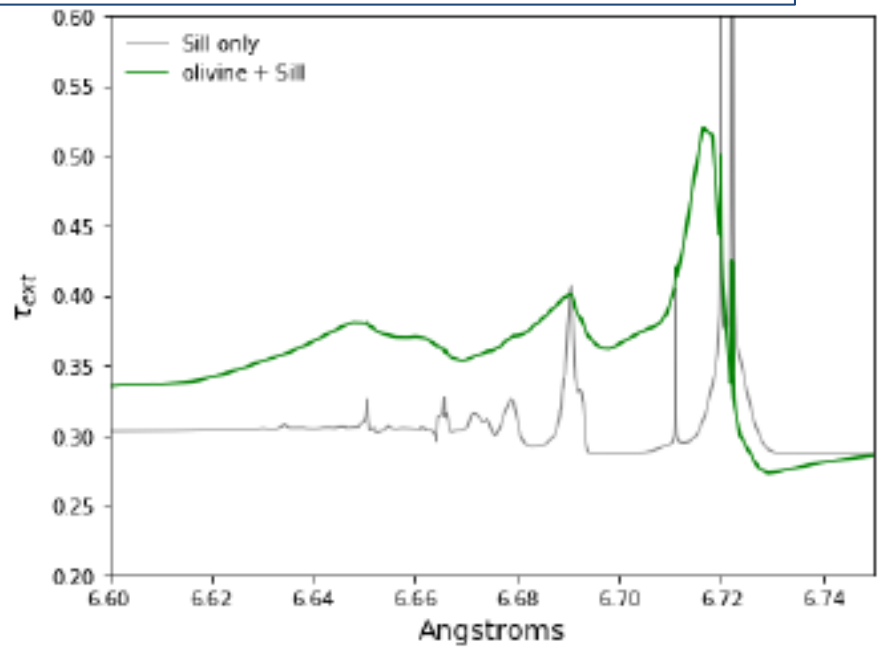
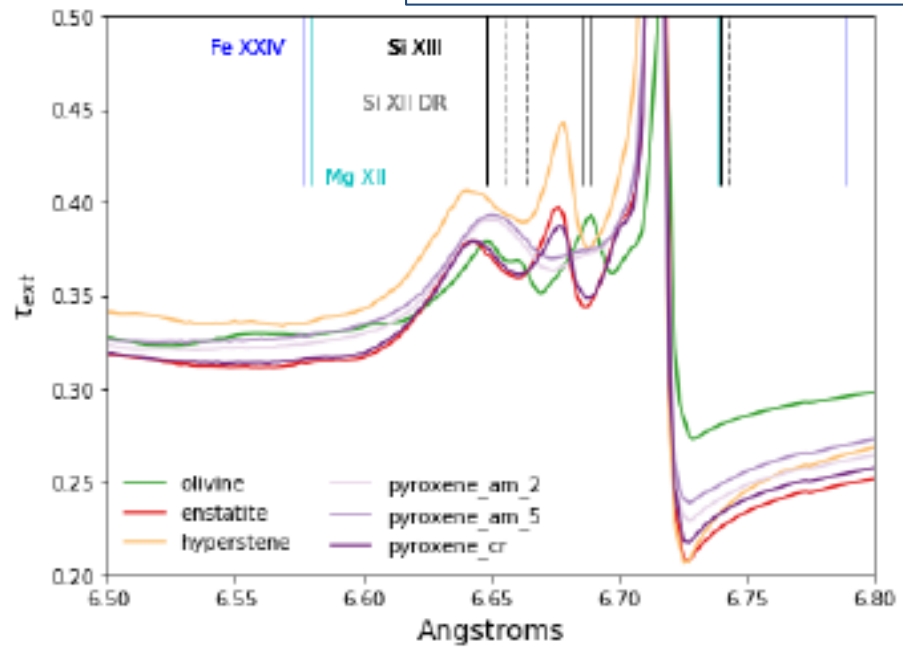


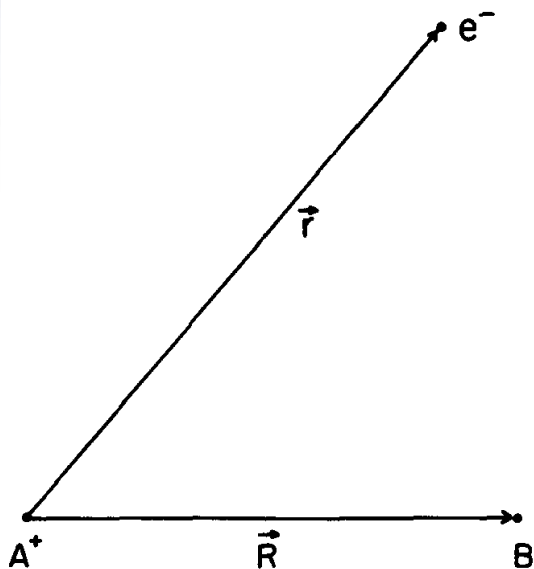
Fig. 3.— Present R-matrix atomic cross section compared to the experimental olivine measurements (arbitrary units, rescaled?) and the Chandra Si absorption model (Corrales et al. 2016).

Various Solid Si X-ray Absorption Measurements



Multiple Scattering vi Fermi Model

⁹E. Fermi, *Nuovo Cimento* **11**, 157 (1934).



sis. In this model the Rydberg electron is perturbed by a neutral ground state rare gas atom B in the form of a Dirac-delta function:

$$V_{e-B} = V_0 \delta(\mathbf{r} - \mathbf{R}). \quad (4)$$

Collaboration with Purdue University:

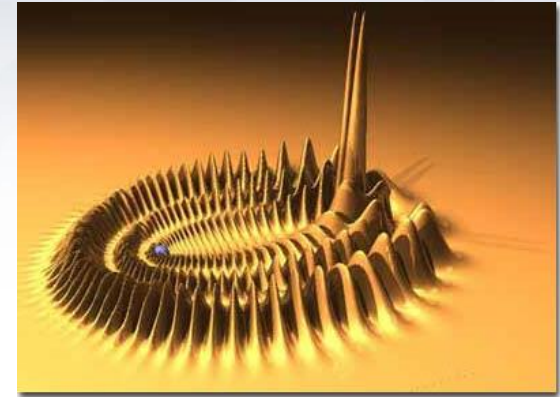
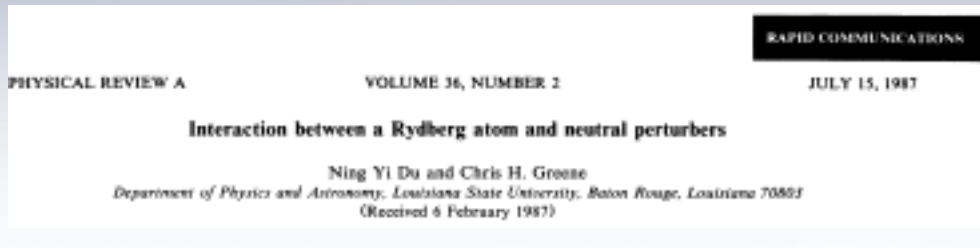
Professor Chris Greene

Undergraduate Student Praneeth Medepali

FIG. 1. Three-body model of a diatomic molecule A^*B , with the coordinate system centered on the nucleus of atom A.

Additional atoms in molecule or solid are treated as scattering sites and approximated by delta-function s-wave scatterers.

Photoabsorption of H (plus displaced δ -function perturber)



$$\gamma = 1/(1 - \langle v|w \rangle).$$

Using Eq. 31 at $r = R$, we have

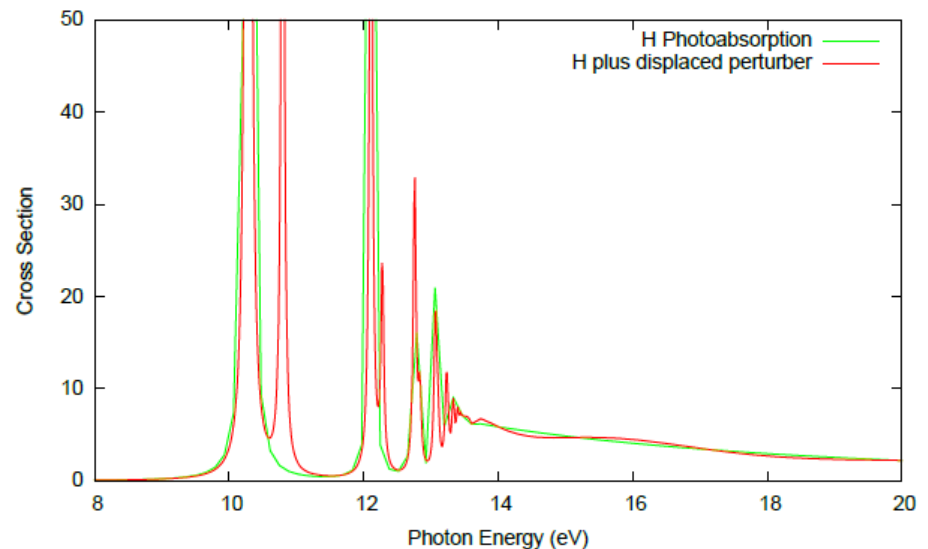
$$S(R) + C(R)K(\vec{R}) = S(R) + C(R) \left[\frac{2V_0}{R^2} S(R) X(\hat{R}) \{ S(R) + C(R)K(\vec{R}) \} \right], \quad (35)$$

which can be solved to get

$$\begin{aligned} K(\vec{R}) &= \left[1 - \frac{2V_0}{R^2} S(R) X(\hat{R}) C(R) \right]^{-1} \left[\frac{2V_0}{R^2} S(R) X(\hat{R}) S(R) \right] \\ &= \left[1 - \vec{v}(\vec{R})^* \vec{w}(\vec{R})^T \right]^{-1} \vec{v}(\vec{R})^* \vec{v}(\vec{R})^T \\ &= \gamma \vec{v} \vec{v}^\dagger. \end{aligned} \quad (36)$$

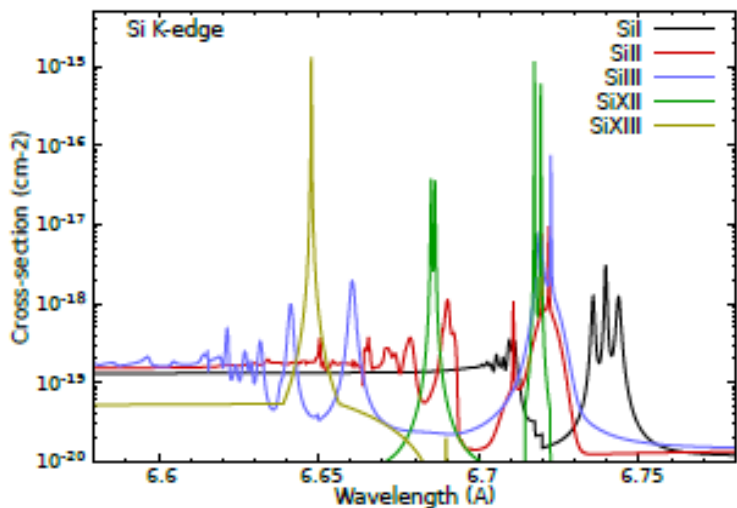
Analytic K-matrix addition
Rydberg series split into two series
Second series shifted by
a quantum defect

XAFS-like oscillations above threshold



Silicon ISM X-ray absorption: the gaseous component

Gatuzz, et al., MNRAS, July 2020

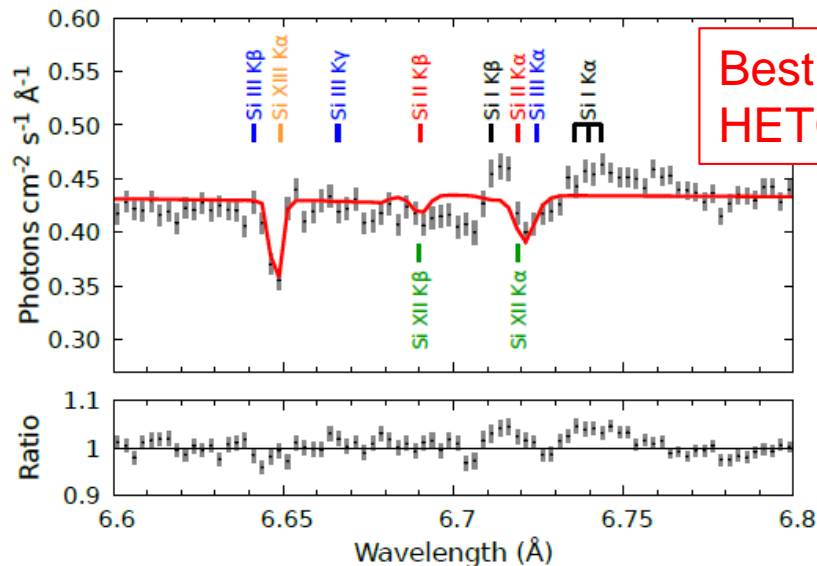


R-matrix
cross sections

Although the solid absorption analysis is beyond the scope of this work, we emphasize that the gaseous component must be carefully modeled before measuring the solid absorption features to estimate accurately the depletion factor of atomic silicon.

Chandra HETG data and ISMabs model

Figure 1. Si I, Si II, Si III, Si XII and Si XIII photoabsorption cross sections included in the model.



Best Fit for all
HETG data

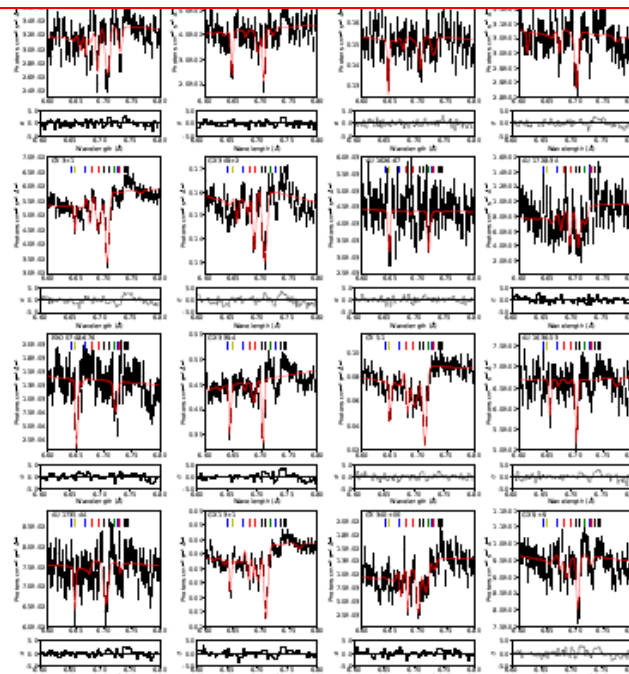


Figure 3. Best fit results using Chandra HETG data for the Si K-edge wavelength region. For each source the observations have been corrected for illustrative purposes. The main resonances for the Si I (black), Si II (red), Si III (blue), Si XII (green) and Si XIII (yellow) are indicated.

Atomic, Molecular, Solid-State Findings

- C, O, Ne predominantly gaseous atomic for K-edge models
- Fe calculations and model fit including giant 2p- \rightarrow 3d resonance shows significant ~ 13 eV core fine-structure splitting AND ~ 1 eV individual 5D J-splitting: atomic fine structure needs refinement
- Atomic Si I, Si II, Si III, Si XII, and Si XIII model Chandra HETG data
- Theory: global cross section including asymptotic limits, absolute cross section, but resonance positions are least accurate
- Experiment: relative resonance spacing accurate, global energy calibrated using other experiments
- X-ray Spectra (e.g., Chandra) and Spectral Models (e.g., XSTAR/ISMdabs) have best energy calibration
- (e.g., OI K α 1s-2p EBIT experimental confirmation)

Thank You!



Gaetan VanGysegghem, Ph. D. December 2016

An R-Matrix, Quantum Defect Theoretical Approach for Photoionization of Diatomic Molecules: Application to N_2

Jianqing Yang, M.S. December 2016

X-ray Absorption in Polyatomic Systems using Multiple Scattering Theory

Jagjit Kaur, Ph.D. December 2017

Near-Threshold Dielectronic Recombination of Si-Like Ions: Perturbative, R-matrix, and MCHF Methodologies **Madhushani**

Wickramarathna, M.S. December 2017

Double Photoionization of Multi-Electron Atoms using an R-matrix with Pseudostates Approach