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Impact of the Dynamic Structure Factor on Doppler Broadening for ^{238}U in UO_2

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Acknowledgements

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Objective

- Determining the self component of the dynamic structure factor (i.e. thermal scattering law, TSL) for U in UO_2 and then perform Doppler broadening including structure impacts.

Outline

- Fundamental definitions for Doppler broadening
- Predictive model for uranium in UO_2
- Generation of Doppler broadened cross sections
  - Free gas
  - Impacts of the UO_2 density of states
- Non-cubic representation of the self component of the dynamic structure factor

Doppler Broadening

- Free Gas
  - Assumes a Maxwellian velocity distribution

$$\sigma^{FG}(E) = \int_0^\infty dE'\, S^{FG}(E', E)\,\sigma(E')$$

$$S^{FG}(E', E) = \frac{1}{\Delta\sqrt{\pi}}\sqrt{\frac{E'}{E}}\exp\left[\frac{-(E'-E)^2}{\Delta^2}\right]$$

$$T_{eff} = \bar{\varepsilon} = \frac{1}{2k_B}\int_0^\infty \rho(\omega)\,\omega\coth(\omega/2k_BT)\,d\omega$$

Doppler Broadening

- Crystal Lattice
  - Compound nucleus effects separated from lattice effects
  - Transition probability

$$\sigma(E) = \frac{\sigma_0 \Gamma^2}{4} \int_{-\infty}^{\infty} d\beta \, \frac{S_s(\alpha, \beta)}{\left(E - E_0 - \beta k_B T\right)^2 + \left(\Gamma / 2\right)^2}$$

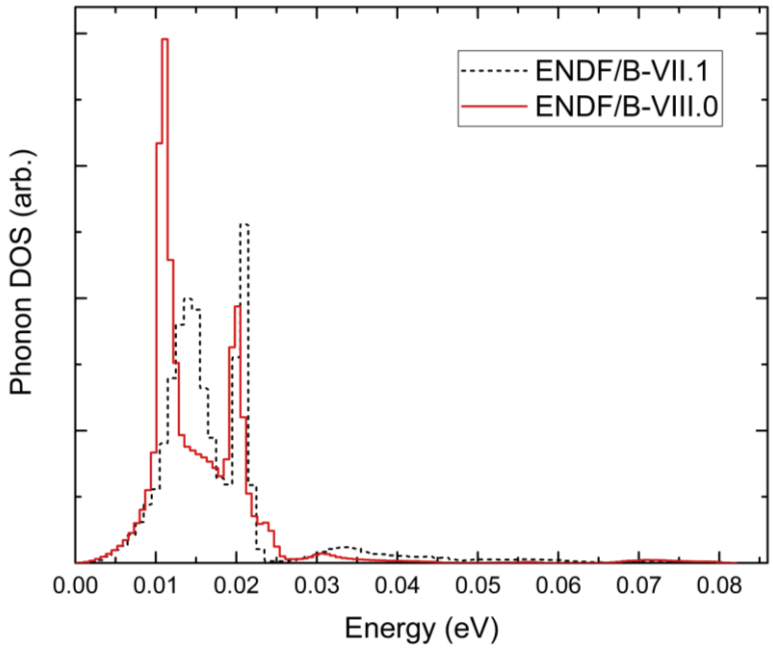
  - Self Scattering Law
    - Identical to that used in thermal scattering
    - Describes the energy-momentum phase space of a material
    - Currently implemented using the cubic approximation

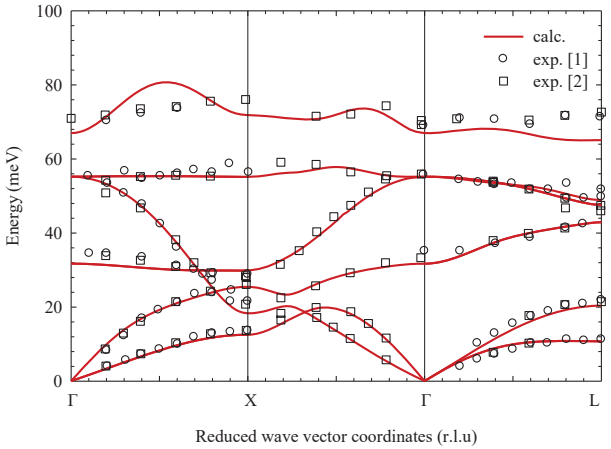
$$S(\alpha, \beta) = \frac{1}{2\pi} \int_{-\infty}^{\infty} e^{i\beta t} e^{-\gamma(t)} dt$$

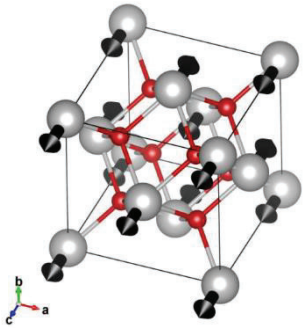
$$\gamma(t) = \alpha \int_{-\infty}^{\infty} \frac{\rho(\beta)}{2\beta \sinh(\beta / 2)} \left[1 - e^{-i\beta t}\right] e^{-\beta/2} d\beta$$

Uranium Dioxide

- *Ab initio* lattice dynamics
  - Predictive density of states (DOS)
  - Current DOS implemented in the ENDF/B-VIII.0 cross section library for U in UO_2







- Fluorite Structure
- 2x2x2 supercell
- GGA-PBE+U

Doppler Broadened Cross Sections

- Free Gas Model
  - Effective Temperature
    - Used to correct the free gas model
    - Calculated using the ENDF/B-VII.1 and ENDF/B-VIII.0 density of states
    - At lower temperatures, larger difference between actual and effective temperature

	ENDF/B-VII.1 DOS	ENDF/B-VIII.0 DOS
23.6 K	99.11 K	91.07 K
293.7 K	308.98 K	305.22 K

Doppler Broadened Cross Sections

- Experimental Data

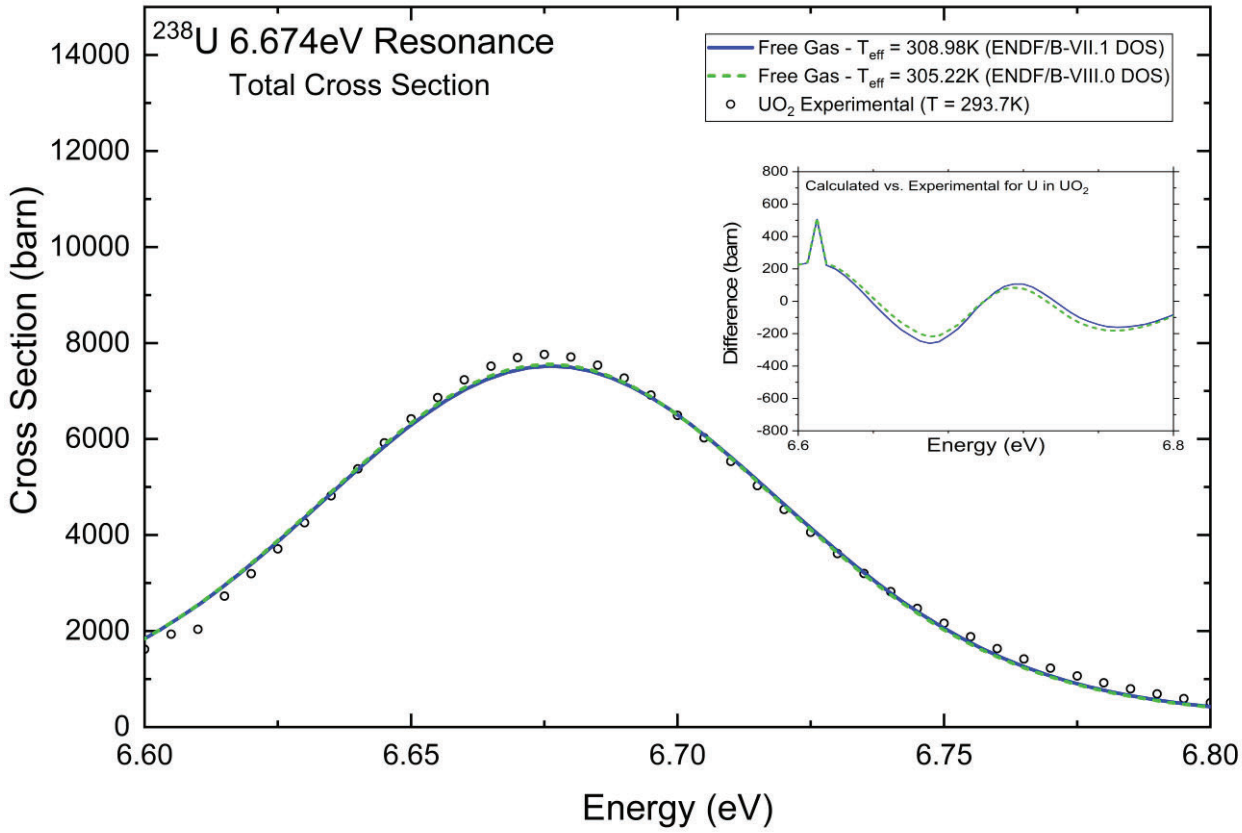
Energy (eV)	6.674
$g\Gamma_n$ (meV)	1.4923 ± 0.0011
Γ_γ (meV)	22.711 ± 0.019
Γ_f (μeV)	0.00988 ± 0.00039

- Resonance Parameters from Analysis

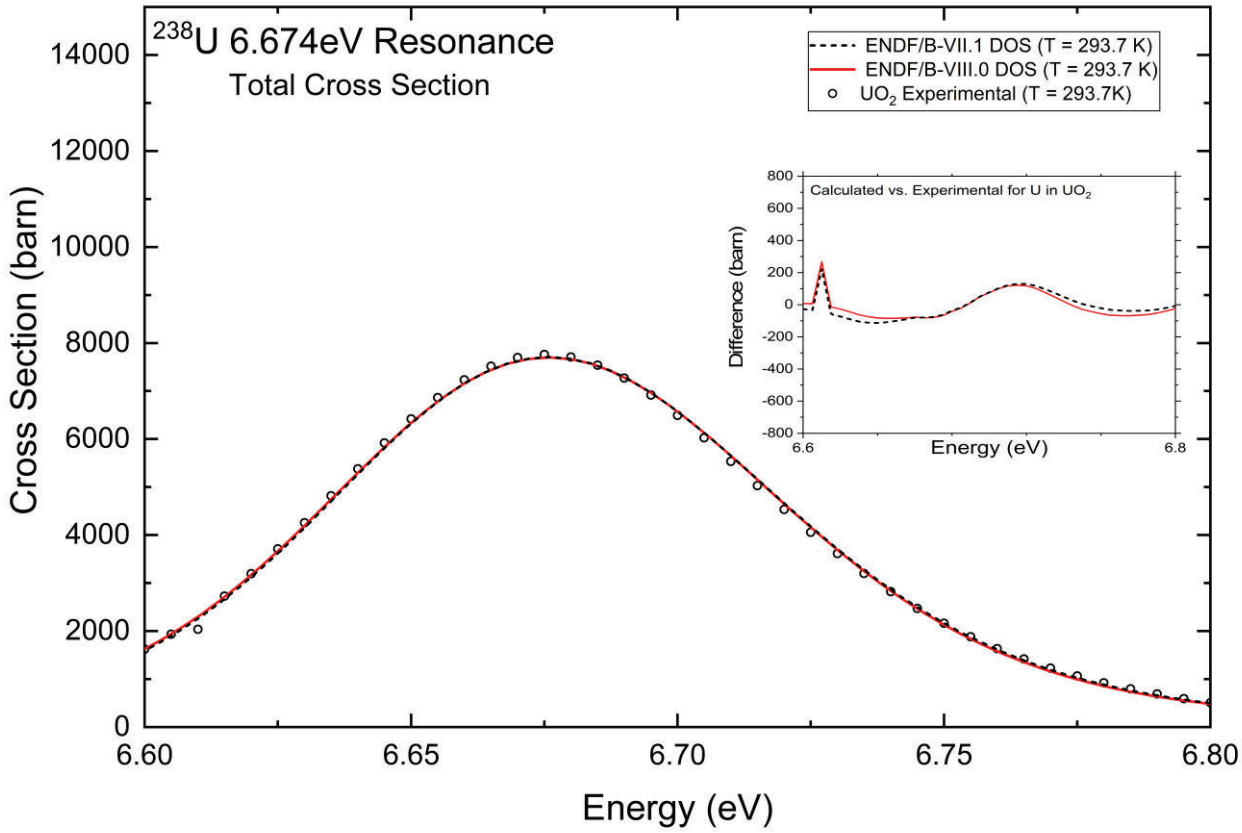
Energy (eV)	Source	$g\Gamma_n$ (meV)
6.674	Experiment *	1.4923 ± 0.0011
6.678151	ENDF/B-VIII.0 CLM	1.49239733
6.678691	ENDF/B-VII.1 CLM	1.49245769
6.675936	ENDF/B-VIII.0 FGM	1.49214982
6.676205	ENDF/B-VII.1 FGM	1.49217989

* H.I. KIM, et al., "Neutron Capture Cross Section Measurements for ^{238}U in the Resonance Region at GELINA," *Eur. Phys. J. A,* **52**, 170 (2016).

Doppler Broadened Cross Sections



Doppler Broadened Cross Sections



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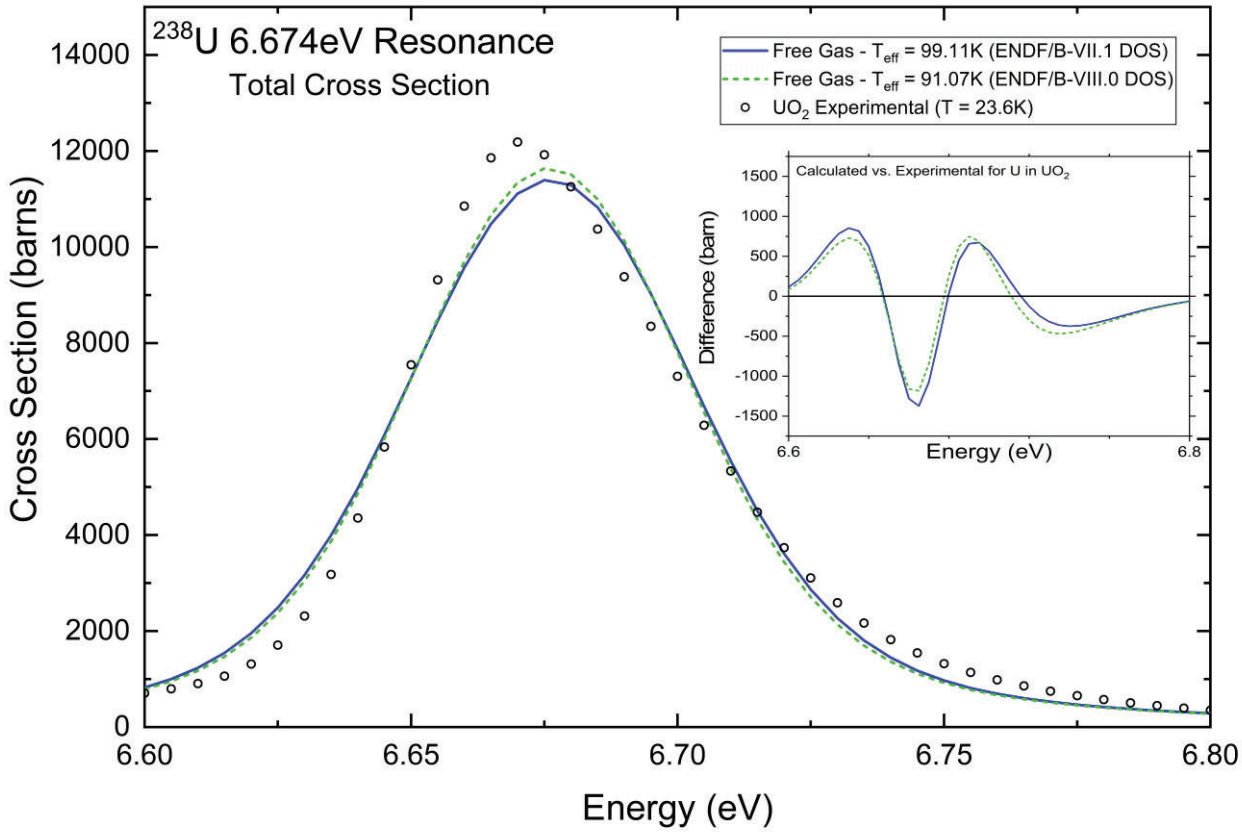

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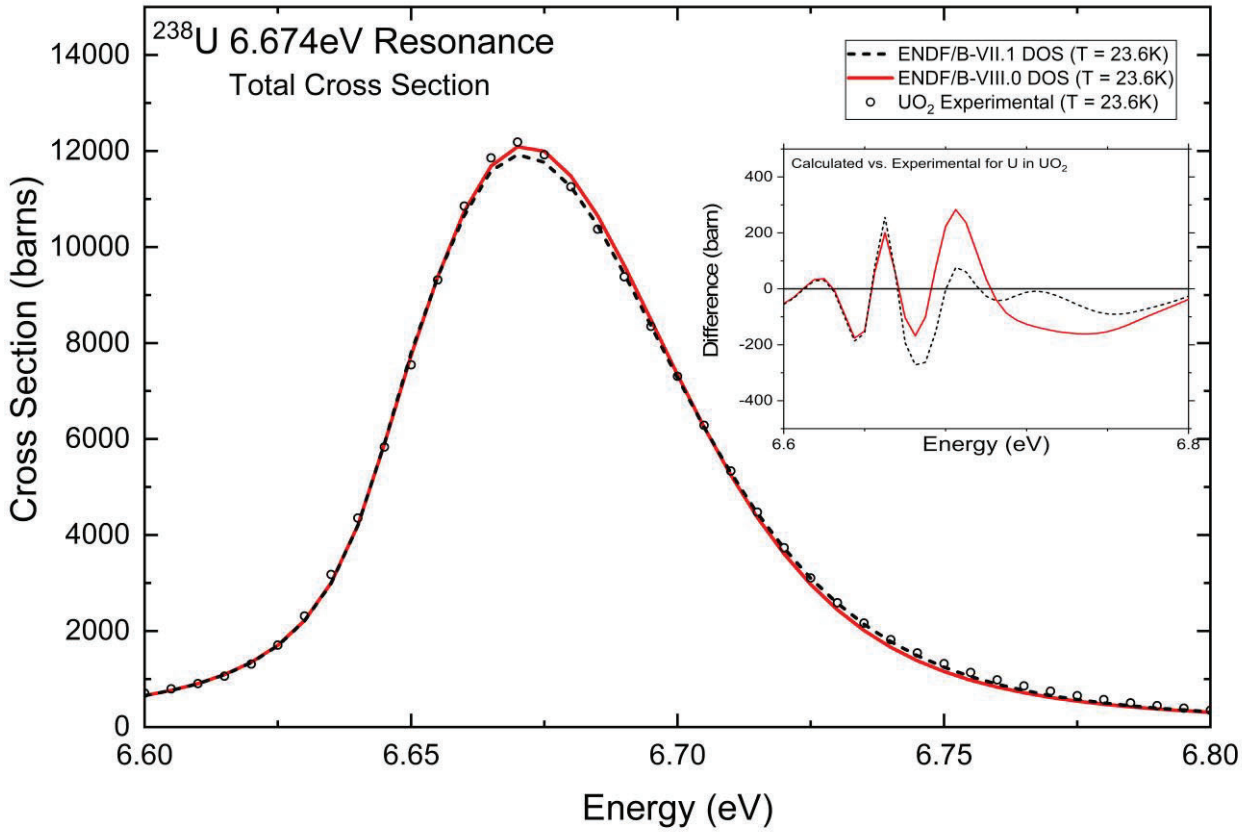
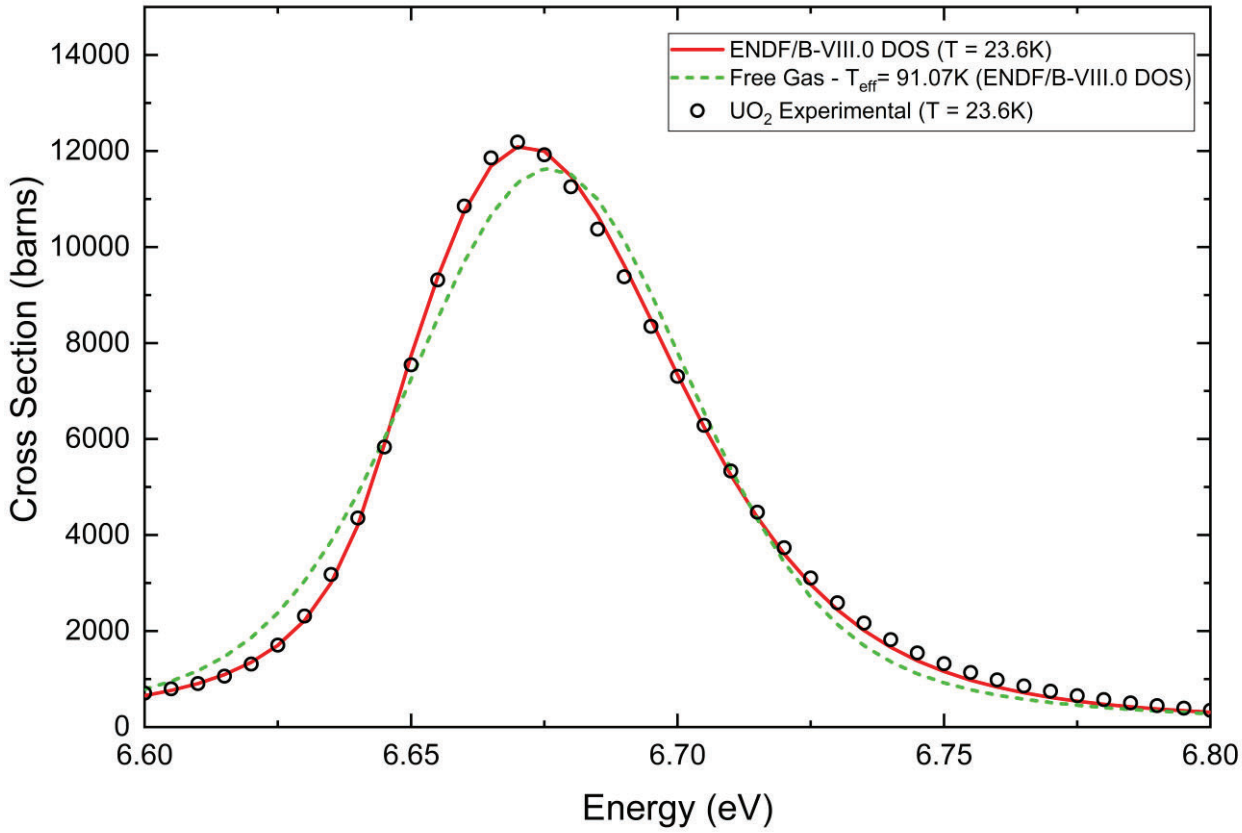
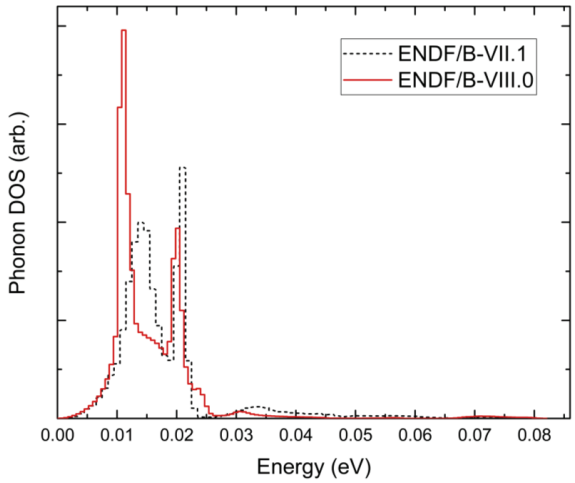


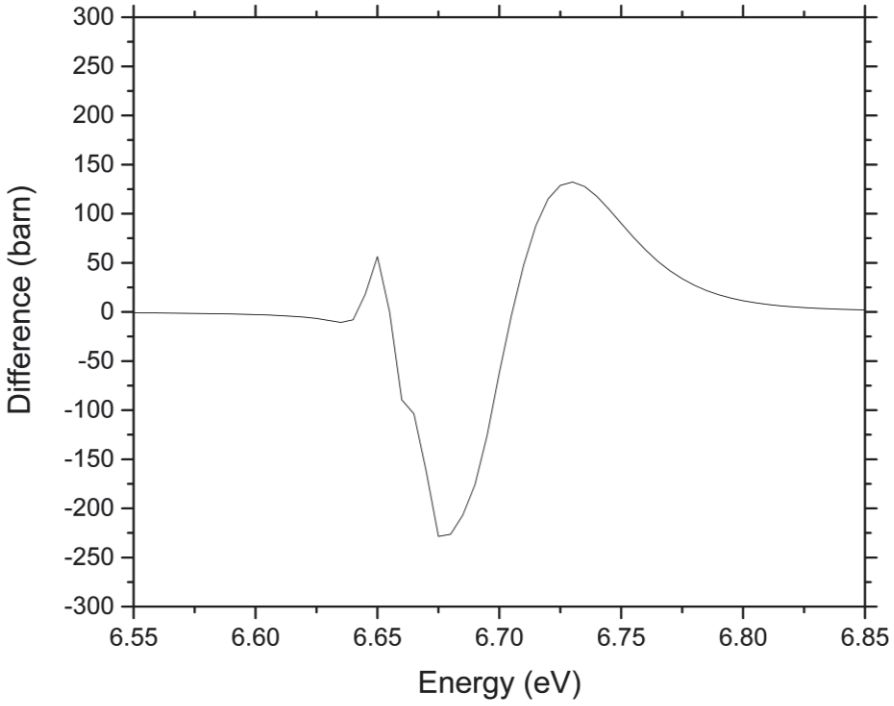
Doppler Broadened Cross Sections



Doppler Broadened Cross Sections

- ENDF/B-VII.1 compared with ENDF/B-VIII.0 evaluation
  - Crystal Lattice Calculation
    - Calculation at 23.6K
    - Differences up to 200 barns





Dynamic Structure Factor

Full Equation

$$S_s(k,\omega) = \int e^{-i\omega t} I_s(k,t)\,dt$$

$$= \int e^{-i\omega t} e^{\langle U^2 \rangle + \langle UV_0 \rangle}\,dt$$

$$\langle n_s + 1 \rangle = \frac{e^{\frac{\hbar\omega_s}{k_B T}}}{e^{\frac{\hbar\omega_s}{k_B T}} - 1}$$

$$\langle U^2 \rangle = -2W = -\frac{\hbar}{2N}\sum_s \frac{1}{\omega_s}\sum_d \left|\frac{(\kappa \cdot e_{sd})}{\sqrt{M_d}}\right|^2 \coth\left(\frac{\hbar\omega_s}{2k_B T}\right)$$

Exponential for
Debye Waller

$$\langle UV_0 \rangle = \frac{\hbar}{2N}\sum_s \sum_d \left|\frac{(\kappa \cdot e_{sd})}{\sqrt{M_d}}\right|^2 \frac{\left[e^{i\omega_s t}\langle n_s + 1 \rangle + e^{-i\omega_s t}\langle n_s \rangle\right]}{\omega_s}$$

$$\langle n_s \rangle = \frac{1}{e^{\frac{\hbar\omega_s}{k_B T}} - 1}$$

- U and V_0 are functions of the polarization vector and dispersion relations

Removing the Cubic Approximation

$$\sum_s (\boldsymbol{k}\cdot\boldsymbol{e}_s)^2 = \frac{1}{3}k^2$$

Full Equation

$$\langle U^2\rangle = -2W = -\frac{\hbar}{2N}\sum_s \frac{1}{\omega_s}\sum_d \left|\frac{(\kappa\cdot e_{sd})}{\sqrt{M_d}}\right|^2 \coth\left(\frac{\hbar\omega_s}{2k_BT}\right)$$

$$\langle UV_0\rangle = \frac{-\hbar^2}{2Nk_BT}\sum_s\sum_d \left|\left(\frac{\kappa\cdot e_{sd}}{\sqrt{M_d}}\right)\right|^2 \left[\frac{e^{-\beta_s}e^{i\omega_s t}+e^{-i\omega_s t}}{\beta_s\left(e^{-\beta_s}-1\right)}\right]$$

$$S_s(k,\omega) = \int e^{-i\omega t} I_s(k,t)dt$$
$$= \int e^{-i\omega t} e^{\langle U^2\rangle + \langle UV_0\rangle} dt$$

- U and V_0 are functions of the polarization vector and dispersion relations

Cubic Approximation

$$\langle U^2\rangle = -\frac{\hbar k^2}{2M}\int_0^{\omega_m}\frac{\rho(\omega)}{\omega}\coth\left(\frac{\hbar\omega_s}{2k_BT}\right)d\omega$$

$$\langle UV_0\rangle = \frac{\hbar k^2}{2M}\int_0^{\omega_m}\frac{\rho(\omega)}{\omega}\left[\coth\left({}^{\hbar\omega}/_{2k_BT}\right)\cos(\omega t)+i\sin(\omega t)\right]d\omega$$

$$S_s(k,\omega) = \int e^{-i\omega t} I_s(k,t)dt$$
$$= \int e^{-i\omega t} e^{-k^2\gamma(t)} dt$$

- γ(t) is the width function and a function of the density of states ρ(ω)

Summary

- Predictive AILD techniques used to model U in UO_2
- Doppler broadening is impacted by the density of states
  - Crystal lattice calculations demonstrate improved agreement with experimental data
  - Evaluations consistent now with the ENDF/B-VIII.0 libraries
- Differences between the broadening based on the ENDF/B-VII.1 and ENDF/B-VIII.0 evaluation of UO_2
- Initial work to incorporate the non-cubic representation of the dynamic structure factor underway

Thank you!

Questions?