Thermal Neutron Scattering Law Methods and Evaluations

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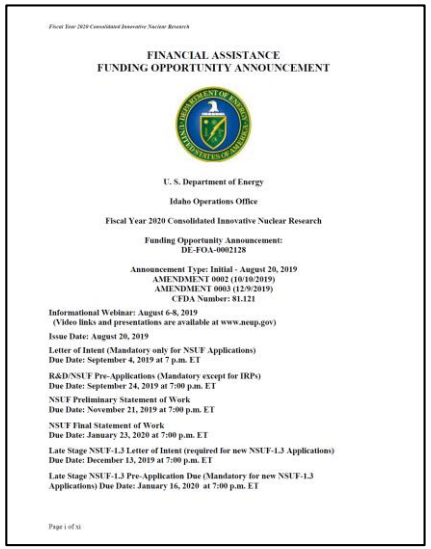
Workshop for Applied Nuclear Data Activities (WANDA)
March 3 – 5, 2020 • Washington, DC, USA

Acknowledgment

- DOE NE through the Nuclear Energy University Program (NEUP)
- NNSA Nuclear Criticality Safety Program (NCSP)
  - in collaboration with LLNL
- Naval Nuclear Propulsion Program (NNPP)



Fiscal Year 2020 Consolidated Innovative Nuclear Research

FINANCIAL ASSISTANCE
FUNDING OPPORTUNITY ANNOUNCEMENT

U. S. Department of Energy

Idaho Operations Office

Fiscal Year 2020 Consolidated Innovative Nuclear Research

Funding Opportunity Announcement:
DE-FOA-0002128

Announcement Type: Initial - August 20, 2019
AMENDMENT 0002 (10/10/2019)
AMENDMENT 0003 (12/9/2019)
CFDA Number: 81.121

Informational Webinar: August 6-8, 2019
(Video links and presentations are available at www.neup.gov)

Issue Date: August 20, 2019

Letter of Intent (Mandatory only for NSUF Applications)
Due Date: September 4, 2019 at 7 p.m. ET

R&D/NSUF Pre-Applications (Mandatory except for IRPs)
Due Date: September 24, 2019 at 7:00 p.m. ET

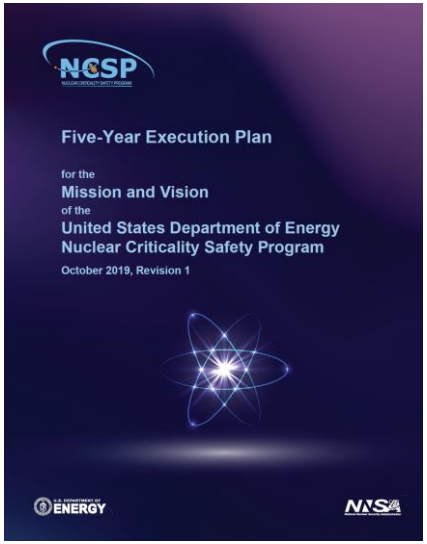
NSUF Preliminary Statement of Work
Due Date: November 21, 2019 at 7:00 p.m. ET

NSUF Final Statement of Work
Due Date: January 23, 2020 at 7:00 p.m. ET

Late Stage NSUF-1.3 Letter of Intent (required for new NSUF-1.3 Applications)
Due Date: December 13, 2019 at 7:00 p.m. ET

Late Stage NSUF-1.3 Pre-Application Due (Mandatory for new NSUF-1.3 Applications) Due Date: January 16, 2020 at 7:00 p.m. ET

Page i of xi



NCSP

Five-Year Execution Plan

for the
Mission and Vision
of the
United States Department of Energy
Nuclear Criticality Safety Program

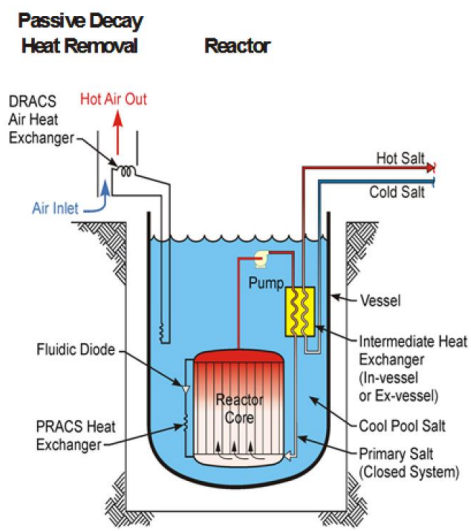
October 2019, Revision 1

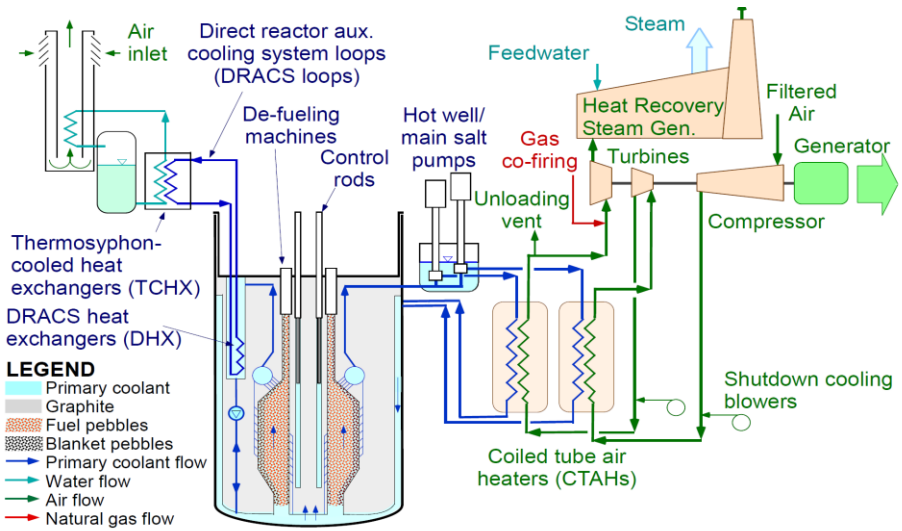
U.S. DEPARTMENT OF ENERGY

NNSA

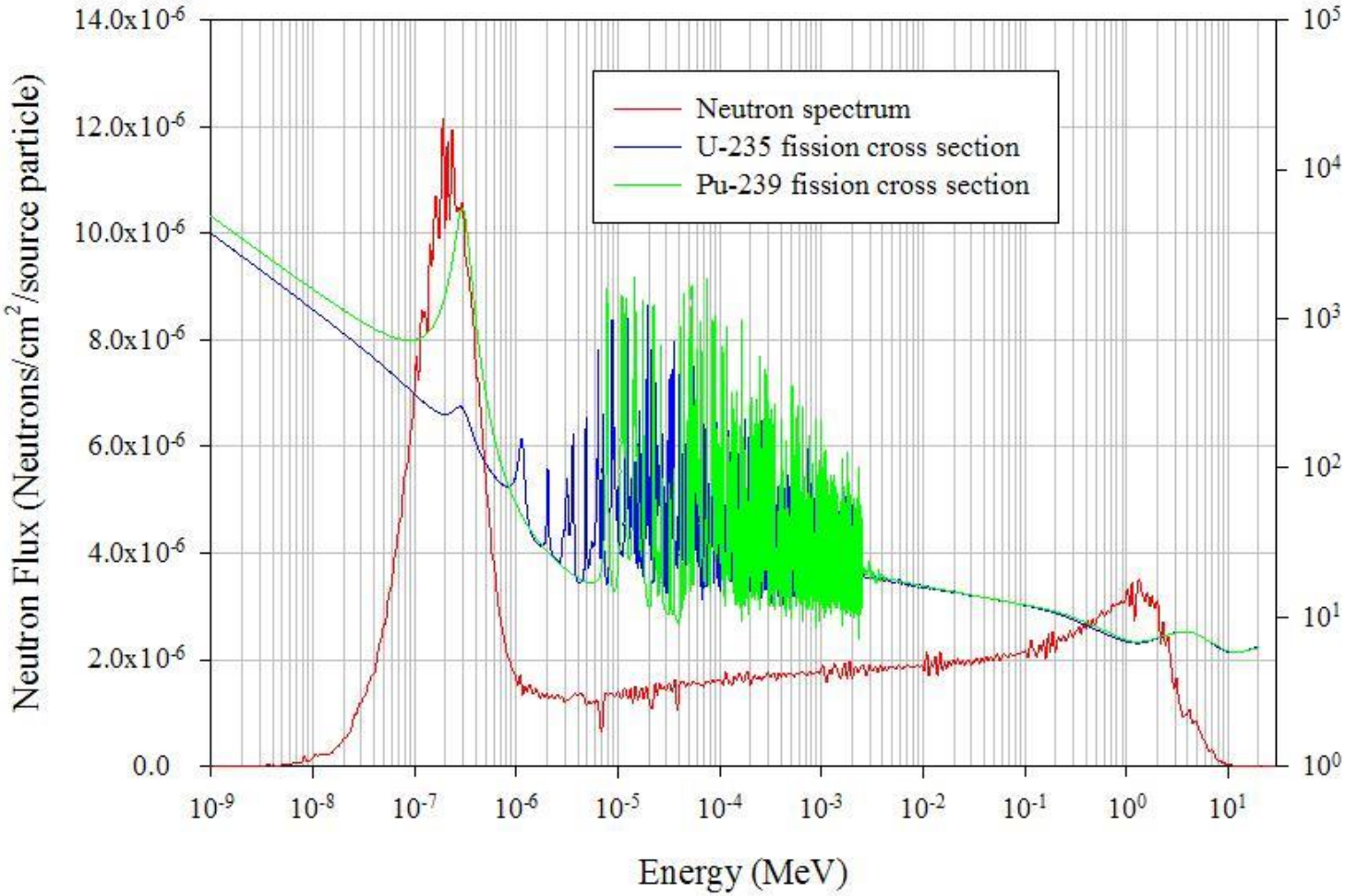
Motivation – Neutronic Systems

- Several concepts of advanced thermal reactors have been proposed including the LWR, AHTR and FHR
  - The core is dominated by light water, graphite and molten salt moderators

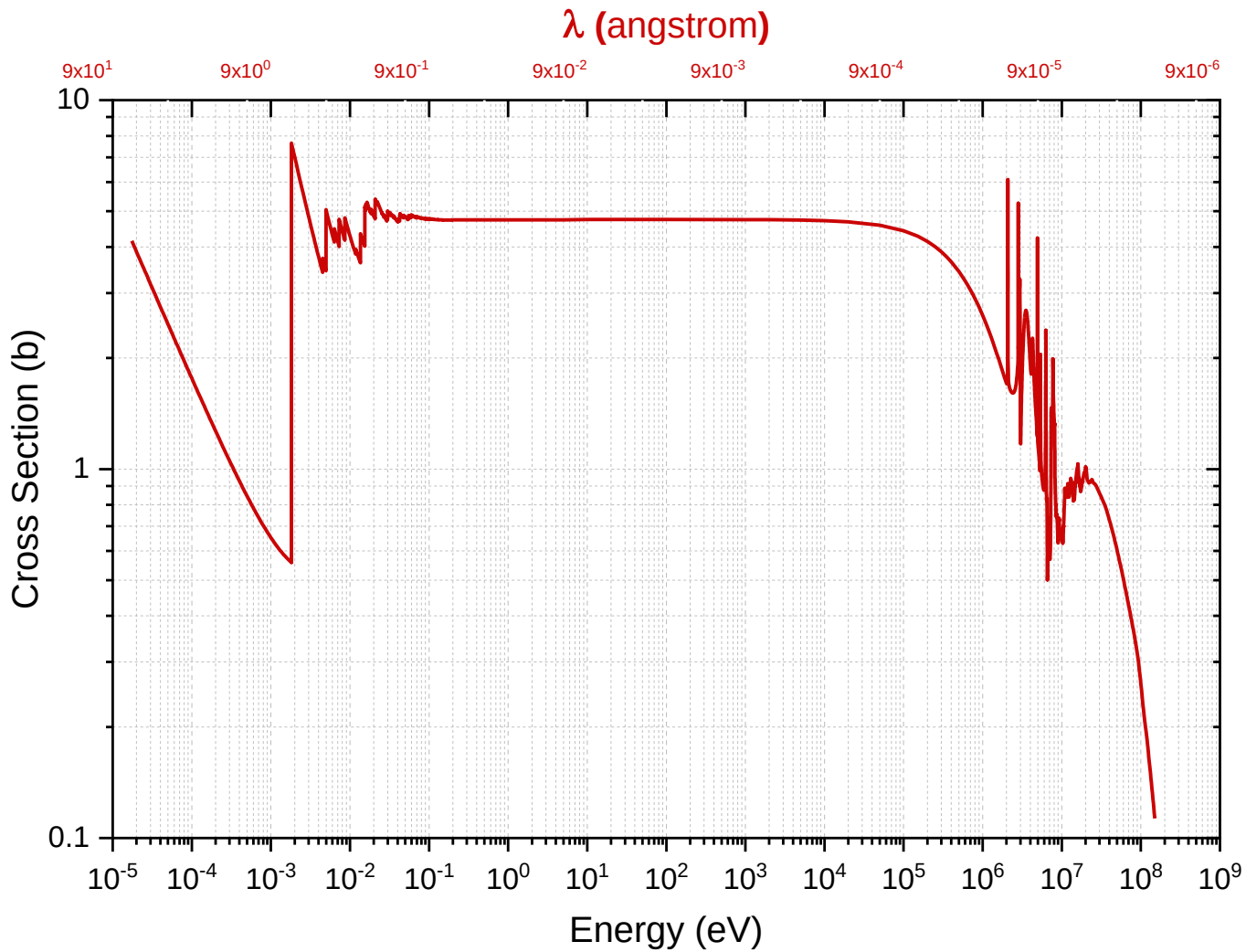




Reactor Characteristics



Neutron Interactions – Carbon



Neutron Thermalization

Using first Born approximation combined with Fermi pseudopotential, it can be shown that the double differential scattering cross section has the form

$$\frac{d^2\sigma}{d\Omega dE'} = \frac{1}{4\pi}\sqrt{\frac{E'}{E}}\left\{\sigma_{coh}S(\vec{\kappa},\omega) + \sigma_{incoh}S_s(\vec{\kappa},\omega)\right\}$$

The scattering law $S(\vec{\kappa},\omega)$ is composed of two parts

$$S(\vec{\kappa},\omega) = S_s(\vec{\kappa},\omega) + S_d(\vec{\kappa},\omega)$$

Van Hove's space-time formulation

$$I(\vec{\kappa},t) = \int G(\vec{r},t)\exp(i\vec{\kappa}\cdot\vec{r})d\vec{r}$$

$$S(\vec{\kappa},\omega) = \frac{1}{2\pi\hbar}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty} G(\vec{r},t)e^{i(\vec{\kappa}\cdot\vec{r}-\omega t)}d\vec{r}dt$$

where $G(\vec{r},t)$ is the *dynamic pair correlation function* and can be expressed in terms of time dependent atomic positions.

Since 1960s
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NJOY/LEAPR
INCOHERENT APPROXIMATION

$$S_s(\alpha,\beta) = k_B T \cdot S_s(\vec{\kappa},\omega)$$

$$\left.\frac{d^2\sigma}{d\Omega dE'}\right|_{inelastic} = \frac{\sigma}{2k_B T}\sqrt{\frac{E'}{E}}S_s(\alpha,\beta)$$

$\beta = \frac{E - E'}{k_B T}$ Energy transfer

$\alpha = \frac{(E + E' - 2\sqrt{EE'}\cos\theta)}{k_B T}$ Momentum transfer

The scattering law (TSL) is the Fourier transform of a Gaussian correlation function

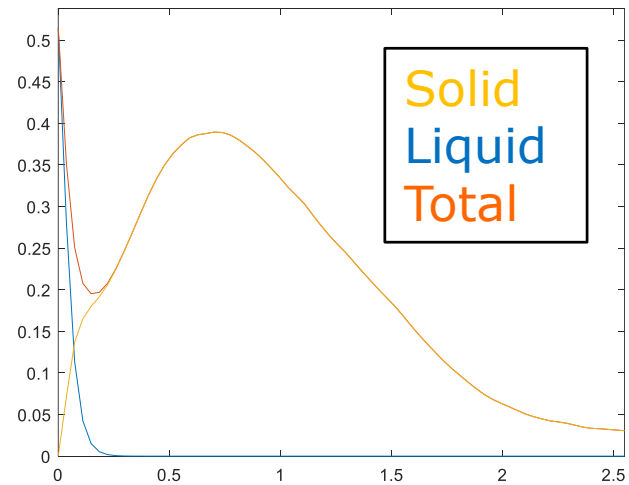
$$S_s(\alpha,\beta) = \frac{1}{2\pi}\int_{-\infty}^{\infty} e^{-i\beta t} e^{-\gamma(t)} dt$$

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

$\rho(\beta)$ – density of states (e.g., phonon frequency distribution)

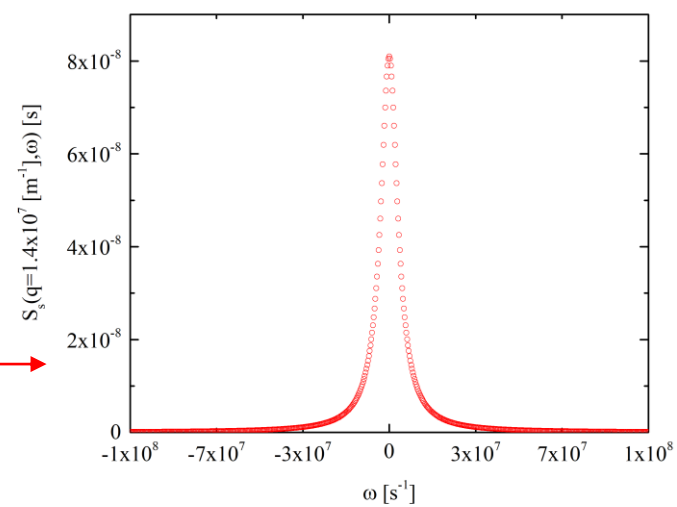
Thermalization in Liquids

- **Separation** of the diffusive DOS from the continuous (solid) DOS



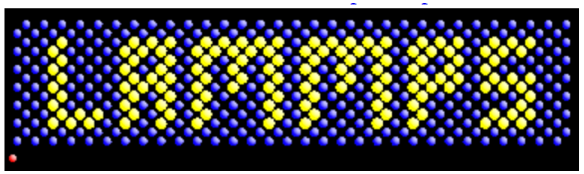
- **Convolution** of the solid and liquid TSL components

$$S_{total}(\alpha,\beta) = \left(S_{diff.}(\alpha,\beta) * S_{cont.}(\alpha,\beta)\right)$$



Thermal Scattering Law Analysis

- Key development in the last 20 years is the use of atomistic simulations methods to support the evaluation process
  - Produce data necessary to calculate the TSL including
    - DOS for evaluation of TSL
    - Direct access to TSL using correlation analysis
    - Support computational and experimental analysis



Thermal Scattering Law Analysis

2000 - Present

PHYSOR 2004 -The Physics of Fuel Cycles and Advanced Nuclear Systems: Global Developments
Chicago, Illinois, April 25-29, 2004, on CD-ROM, American Nuclear Society, Lagrange Park, IL (2004)

Ab Initio Generation of Thermal Neutron Scattering Cross Sections

A. I. Hawari, I. I. Al-Qasir, V. H. Gillette, B. W. Wehring, T. Zhou
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Quantum mechanical ab initio (i.e., first principle) methods are applied in generating the thermal neutron scattering cross sections of moderators and reflectors that are of interest in nuclear technology. Specifically, this work focuses on graphite and beryllium. In both cases, the ab initio code VASP and the lattice dynamics code PHONON were used to generate the dispersion relations, and the phonon frequency distributions (density of states). This information was then utilized in the LEAPR module of the NJOY code to calculate the thermal neutron scattering cross sections at various temperatures. The use of the ab initio approach represents a major departure from previously applied methods, which depended mainly on fitting simpler dynamical models to experimental data to arrive at the phonon frequency distributions. In this case, much more complicated models of the atomic system of interest can be set up, which allows the establishment of a more complete dynamical matrix. As opposed to the semi-empirical methods used previously, this method represents a fundamental and predictive approach for estimating materials' properties including ones that are of interest in nuclear reactor design.

KEYWORDS: neutron, thermal neutron, slow neutron, ab initio, VASP, graphite, beryllium, moderator, phonon frequency distribution, thermal neutron scattering cross section, nuclear reactor

1. Introduction

Due to advances in computational power, the possibility now exists to perform detailed quantum mechanical ab initio (i.e., first principle) simulations of atomic systems. These simulations are currently used in fields such as physics, chemistry, and materials science to characterize and predict the behavior of new and exotic materials [1]. Using this approach, it is possible to establish the equilibrium atomic positions of a given material and predict the various properties of the material starting from such basic information as the coordinates of the atoms. Consequently, ab initio simulations seek to gain insight into the bonding forces in the material, which are usually variations of the Coulomb force that result in the formation of ionic, covalent, molecular, and van der Waals bonds.

In nuclear reactor design, the effect of atomic and/or molecular bonding becomes important as the neutrons slow down and enter the thermal (or slow) region (neutron energy ≤ 1 eV). The microscopic interaction (i.e., absorption, scattering, etc.) of slow neutrons within the reactor core defines the thermal neutron energy spectrum, which affects several global (macroscopic) properties such as criticality, and safety and feedback response. Therefore, the accuracy of the thermal neutron scattering cross sections that are used in reactor core design calculations are important for operating the reactor in an optimized and safe manner.

In the past, the thermal neutron scattering cross sections were derived from structure dynamics models that were fitted to experimental data in order to quantify the forces between the atoms and calculate the required excitation density of states [2]. However, by using the ab initio approach, the ability now exists to treat much larger systems of atoms, and arrive at more accurate and complete dynamical models from

Available online at www.sciencedirect.com
ScienceDirect
Nuclear Data Sheets 118 (2014) 172–175
Nuclear Data Sheets
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Modern Techniques for Inelastic Thermal Neutron Scattering Analysis

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A predictive approach based on ab initio quantum mechanics and/or classical molecular dynamics simulations has been formulated to calculate the scattering law, $S(\vec{\kappa},\omega)$, and the thermal neutron scattering cross sections of materials. In principle, these atomistic methods make it possible to generate the inelastic thermal neutron scattering cross sections of any material and to accurately reflect the physical conditions of the medium (i.e, temperature, pressure, etc.). In addition, the generated cross sections are free from assumptions such as the incoherent approximation of scattering theory and, in the case of solids, crystalline perfection. As a result, new and improved thermal neutron scattering data libraries have been generated for a variety of materials. Among these are materials used for reactor moderators and reflectors such as reactor-grade graphite and beryllium (including the coherent inelastic scattering component), silicon carbide, cold neutron media such as solid methane, and neutron beam filters such as sapphire and bismuth. Consequently, it is anticipated that the above approach will play a major role in providing the nuclear science and engineering community with its needs of thermal neutron scattering data especially when considering new materials where experimental information may be scarce or nonexistent.

I. INTRODUCTION

Low energy or "thermal" neutrons are characterized by energies that are on the order of the excitation (vibration, rotation etc.) energy in the medium in which they interact. Furthermore, their de Broglie wavelength is on the order of the separation distance in solids. Consequently, such neutrons are highly sensitized to the atomic binding details of the system that surrounds them including its structure and dynamics. In fact, the structural and dynamic properties of the atomic system are sampled through scattering interactions between the system's atoms and molecules and the neutrons. The scattering of low energy neutrons in an atomic system is generally described using thermal neutron scattering cross sections. Traditionally, the cross sections are quantified based on Born scattering theory combined with Fermi's Golden rule and the assumption of an extremely short range (delta function) nuclear potential [1]. The outcome of this approach, is an expression for the double differential scattering cross section given by

$$\frac{d^2\sigma}{d\Omega dE} = \frac{1}{4\pi}\frac{k'}{k}\left(\sigma_{coh}S\left(\vec{\kappa},\omega\right) + \sigma_{inc}S_s\left(\vec{\kappa},\omega\right)\right), \quad (1)$$

where $S(\vec{\kappa},\omega)$ is known as the scattering law, $\vec{\kappa}$ is the scattering vector, ω is the frequency, k and k' represent the magnitude of the wave vector of the incident and scattered neutron respectively, σ_{coh} is the bound atom coherent scattering cross section, and σ_{inc} is the bound atom incoherent scattering cross section. In general, S is composed of two terms as follows

$$S\left(\vec{\kappa},\omega\right) = S_s\left(\vec{\kappa},\omega\right) + S_d\left(\vec{\kappa},\omega\right), \quad (2)$$

where S_s is known as the self-scattering law, which accounts for non-interference (incoherent) effects, while S_d is the distinct scattering law and accounts for interference (coherent) effects. Examination of Eq. 1 shows that the thermal neutron scattering cross section depends on two factors: first, the neutron-nucleus interaction as represented by the bound atom cross sections, and second, a factor that represents the dynamics of the scattering system (i.e., the collection of atoms) as represented by the scattering law.

Frequently, the calculations of the thermal scattering cross section invoke the incoherent approximation where S_d is set equal to zero in Eq. 2. Based on this assumption, Eq. 1 is developed to give (e.g., see Ref. [2])

$$\frac{d^2\sigma}{d\Omega dE} = \frac{1}{4\pi}\frac{k'}{k}\left[\sigma_{coh} + \sigma_{inc}\right] S_s\left(\vec{\kappa},\omega\right). \quad (3)$$

However, for some important neutronic materials such as graphite and beryllium, this assumption can introduce

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http://dx.doi.org/10.1016/j.nds.2014.04.029
0090-3752/2014 Published by Elsevier B.V.

Annals of Nuclear Energy 135 (2020) 106640
Contents lists available at ScienceDirect
Annals of Nuclear Energy
journal homepage: www.elsevier.com/locate/anucene
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On a measurement approach to support evaluation of thermal scattering law data

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ARTICLE INFO
Article history:
Received 31 January 2019
Received in revised form 13 July 2019
Accepted 17 July 2019

Keywords:
Neutron
Thermal scattering law
Cross section
Measurement
ENDF

ABSTRACT
Inelastic thermal neutron scattering in materials that act as neutron moderators, reflectors, and filters results in shaping the neutron spectrum at low energies. This phenomenon is described using differential scattering cross sections calculated from three components including the bound atom (i.e., nuclear) scattering cross section of the neutron, the ratio of the outgoing and incoming neutron energy, and the thermal scattering law (TSL), i.e., $S(\alpha,\beta)$, where α and β represent dimensionless momentum and energy exchange variables, respectively. To date, no TSL libraries are generated using measured data. However, valuable information may be derived from measurements and "targeted" experiments that can validate TSL data and the related inelastic scattering cross sections. As a demonstration, a suite of coordinated measurements and experiments is described that was designed and used to support the evaluation of the TSL for "nuclear" graphite. This experimental suite includes neutron powder diffraction (for structure analysis), positron annihilation (for nano porosity assessment), inelastic neutron scattering measurements using a chopper spectrometer, transmission experiments using neutrons with energy below the Bragg cutoff thereby accessing the total (inelastic) cross section, and a slowing-down-time experiment to observe the developing neutron spectrum in the material. This experimental suite was key to understanding the difference in TSL between "nuclear" and "ideal" graphite and for the inclusion of "nuclear" graphite in the ENDF/B-VIII.0 nuclear data library release.

1. Introduction

Over the past 15 years, a general methodology has been developed to generate thermal neutron scattering law (TSL) data, and calculate the inelastic thermal neutron scattering cross sections, for materials under various conditions and while relaxing many of the traditional approximations (e.g., the incoherent approximation) (Hawari, 2014, 2004). The methodology is based on using molecular dynamics (MD) and density functional theory (DFT) atomistic simulation methods to derive the fundamental input needed for TSL calculations, such as the atomic and molecular system's excitation density of states. Alternatively, the TSL may be directly accessed from the atomistic simulations and the resulting atomic correlations. In this case, corrections are needed to account for missing quantum effects such as detailed balance (Hehe, 2010). This methodology has resulted in the largest contribution (in the last 50 years) of TSL data to the recently released ENDF/B-VIII.0 nuclear data libraries (Brown, 2018).

In general, the formulation that would be the subject of the evaluation process originates from the fundamental equations for the double differential thermal scattering cross section. This formulation is derived using the first Born approximation of scattering theory and assumes a highly localized nuclear potential known as the Fermi pseudopotential (e.g., see Ref. Squires, 1976). The outcome of this formulation is the following expression for the double differential thermal scattering cross section

$$\frac{d^2\sigma}{d\Omega dE} = \frac{1}{4\pi k_B T}\sqrt{\frac{E'}{E}}\left[\sigma_{coh}S(\alpha,\beta) + \sigma_{inc}S_s(\alpha,\beta)\right] \quad (1)$$

$$S(\alpha,\beta) = S_s(\alpha,\beta) + S_d(\alpha,\beta), \quad (2)$$

where, $S_s(\alpha,\beta)$ is the thermal scattering law, $S_d(\alpha,\beta)$ is the self scattering law, $S_d(\alpha,\beta)$ is the distinct scattering law, α and β are dimensionless momentum and energy exchange variables, σ_{coh} is the coherent bound atom cross section, σ_{inc} is the incoherent bound atom cross section, E is the incoming neutron energy, E' is the scattered neutron energy, Ω' is the scattering solid angel, k_B is Boltzmann's constant, and T is the temperature of the scattering medium. In this case, S_s is related to the fourier transform in space and time of the density correlation function for an atom at a given initial location in the atomic system with its location at time t. S_d is related to the Fourier transform in space and time of the density

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https://doi.org/10.1016/j.anucene.2019.106640
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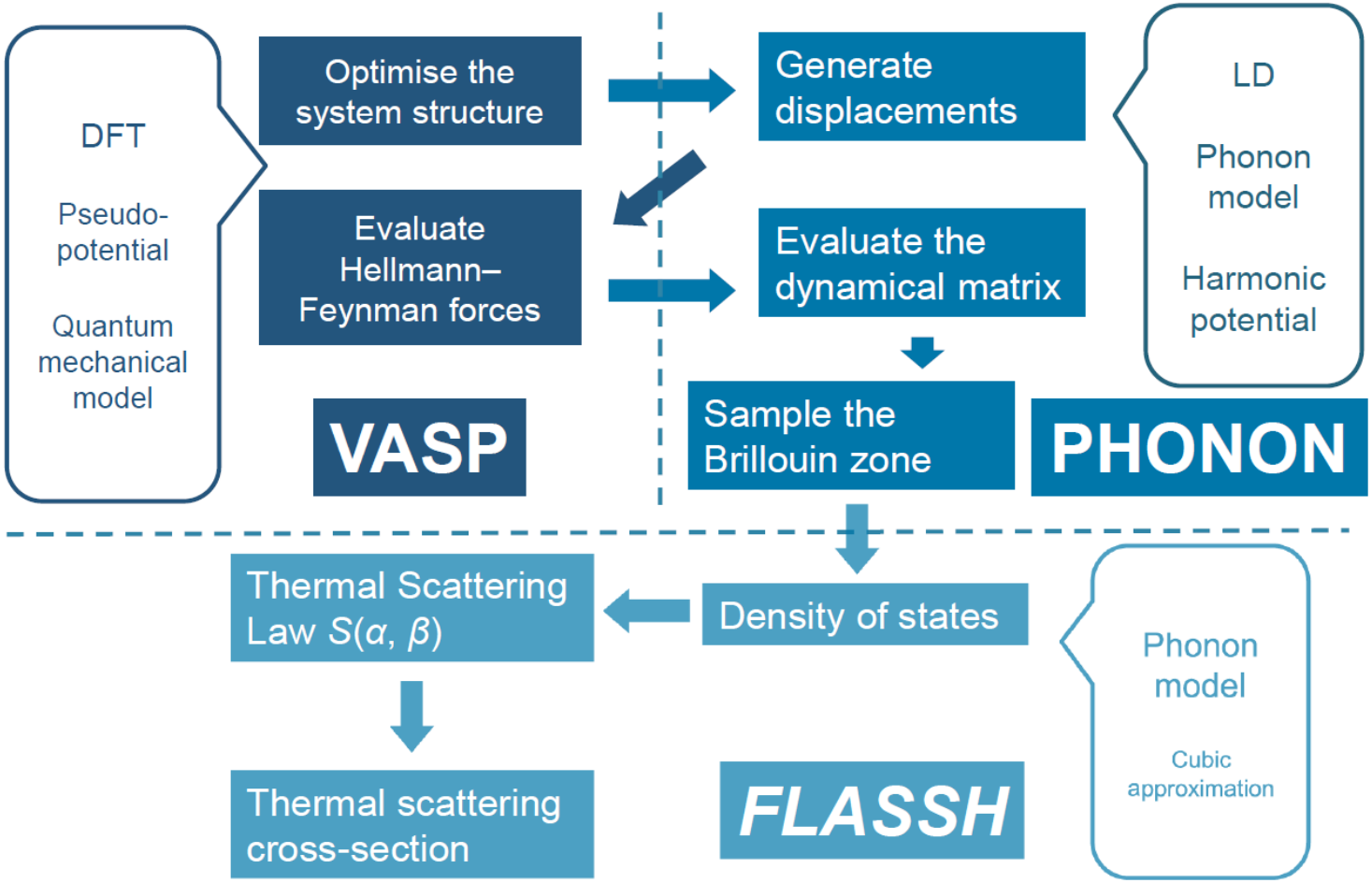
Analysis Approach

- Construct atomistic model of a material
- Verify ability of model to reproduce physical properties of the material (equilibrium conditions)
  - Density, thermal expansion, thermal conductivity,…
  - Ergodic behavior, correlations,…
- Generate input (DOS, …) for TSL calculations
- Calculate TSL and produce thermal scattering cross sections
  - Check consistency of results with computational assumptions/models
  - Compare to experimental data

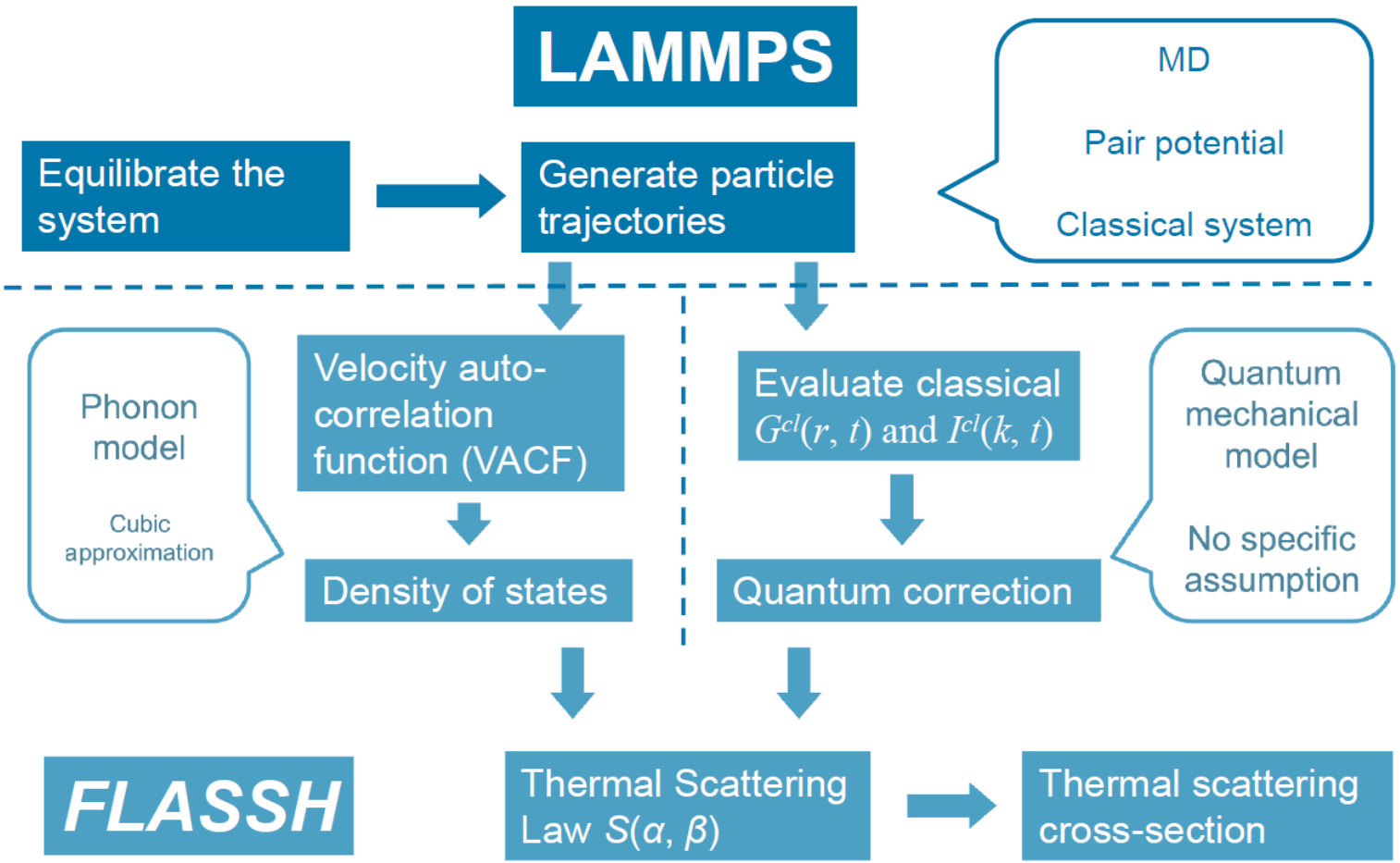
Thermal Scattering Law

DFT/LD



Thermal Scattering Law

MD/QM



ENDF/B-VIII TSL Evaluations

Material	ENDF Library Name	Evaluation Basis	Institution
Beryllium metal	tsl-Be-metal.endf	DFT/LD	NCSU
Beryllium oxide (beryllium)	tsl-BeinBeO.endf	DFT/LD	NCSU
Beryllium oxide (oxygen)	tsl-OinBeO.endf	DFT/LD	NCSU
Light water (hydrogen)	tsl-HinH2O.endf	MD	CAB
Light water ice (hydrogen)	tsl-HinIceIh.endf	DFT/LD	BAPL
Light water ice (oxygen)	tsl-OinIceIh.endf	DFT/LD	BAPL
Heavy water (deuterium)	tsl-DinD2O.endf	MD	CAB
Heavy water (oxygen)	tsl-OinD2O.endf	MD	CAB
Polymethyl Methacrylate (Lucite)	tsl-HinC5O2H8.endf	MD	NCSU
Polyethylene	tsl-HinCH2.endf	MD	NCSU
Crystalline graphite	tsl-graphite.endf	MD	NCSU
Reactor graphite (10% porosity)	tsl-reactor-graphite-10P.endf	MD	NCSU
Reactor graphite (30% porosity)	tsl-reactor-graphite-30P.endf	MD	NCSU
Silicon carbide (silicon)	tsl-CinSiC.endf	DFT/LD	NCSU
Silicon carbide (carbon)	tsl-SiinSiC.endf	DFT/LD	NCSU
Silicon dioxide (alpha phase)	tsl-SiO2-alpha.endf	DFT/LD	NCSU
Silicon dioxide (beta phase)	tsl-SiO2-beta.endf	DFT/LD	NCSU
Yttrium hydride (hydrogen)	tsl-HinYH2.endf	DFT/LD	BAPL
Yttrium hydride (yttrium)	tsl-YinYH2.endf	DFT/LD	BAPL
Uranium dioxide (oxygen)	tsl-OinUO2.endf	DFT/LD	NCSU
Uranium dioxide (uranium)	tsl-UinUO2.endf	DFT/LD	NCSU
Uranium nitride (nitrogen)	tsl-NinUN.endf	DFT/LD	NCSU
Uranium nitride (uranium)	tsl-UinUN.endf	DFT/LD	NCSU

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Light water (hydrogen)	tsl-HinH2O.endf	MD	CAB
Light water ice (hydrogen)	tsl-HinIceIh.endf	DFT/LD	BAPL
Light water ice (oxygen)	tsl-OinIceIh.endf	DFT/LD	BAPL
Heavy water (deuterium)	tsl-DinD2O.endf	MD	CAB
Heavy water (oxygen)	tsl-OinD2O.endf	MD	CAB
Polymethyl Methacrylate (Lucite)	tsl-HinC5O2H8.endf	MD	NCSU
Polyethylene	tsl-HinCH2.endf	MD	NCSU
Crystalline graphite	tsl-graphite.endf	MD	NCSU
Reactor graphite (10% porosity)	tsl-reactor-graphite-10P.endf	MD	NCSU
Reactor graphite (30% porosity)	tsl-reactor-graphite-30P.endf	MD	NCSU
Silicon carbide (silicon)	tsl-CinSiC.endf	DFT/LD	NCSU
Silicon carbide (carbon)	tsl-SiinSiC.endf	DFT/LD	NCSU
Silicon dioxide (alpha phase)	tsl-SiO2-alpha.endf	DFT/LD	NCSU
Silicon dioxide (beta phase)	tsl-SiO2-beta.endf	DFT/LD	NCSU
Yttrium hydride (hydrogen)	tsl-HinYH2.endf	DFT/LD	BAPL
Yttrium hydride (yttrium)	tsl-YinYH2.endf	DFT/LD	BAPL
Uranium dioxide (oxygen)	tsl-OinUO2.endf	DFT/LD	NCSU
Uranium dioxide (uranium)	tsl-UinUO2.endf	DFT/LD	NCSU
Uranium nitride (nitrogen)	tsl-NinUN.endf	DFT/LD	NCSU
Uranium nitride (uranium)	tsl-UinUN.endf	DFT/LD	NCSU

Thermal Scattering Law Analysis

2000 - Present

PHYSOR 2004 -The Physics of Fuel Cycles and Advanced Nuclear Systems: Global Developments
Chicago, Illinois, April 25-29, 2004, on CD-ROM, American Nuclear Society, Lagrange Park, IL (2004)

Ab Initio Generation of Thermal Neutron Scattering Cross Sections

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Department of Nuclear Engineering, P.O. Box 7909, North Carolina State University, Raleigh, NC 27695-7909

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***KEYWORDS:* neutron, thermal neutron, slow neutron, ab initio, VASP, graphite, beryllium, moderator, phonon frequency distribution, thermal neutron scattering cross section, nuclear reactor**

1. Introduction

Due to advances in computational power, the possibility now exists to perform detailed quantum mechanical ab initio (i.e., first principle) simulations of atomic systems. These simulations are currently used in fields such as physics, chemistry, and materials science to characterize and predict the behavior of new and exotic materials [1]. Using this approach, it is possible to establish the equilibrium atomic positions of a given material and predict the various properties of the material starting from such basic information as the coordinates of the atoms. Consequently, ab initio simulations seek to gain insight into the bonding forces in the material, which are usually variations of the Coulomb force that result in the formation of ionic, covalent, molecular, and van der Waals bonds.

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In the past, the thermal neutron scattering cross sections were derived from structure dynamics models that were fitted to experimental data in order to quantify the forces between the atoms and calculate the required excitation density of states [2]. However, by using the ab initio approach, the ability now exists to treat much larger systems of atoms, and arrive at more accurate and complete dynamical models from

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ScienceDirect
Nuclear Data Sheets 118 (2014) 172–175
Nuclear Data Sheets
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Modern Techniques for Inelastic Thermal Neutron Scattering Analysis

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A predictive approach based on ab initio quantum mechanics and/or classical molecular dynamics simulations has been formulated to calculate the scattering law, $S(\vec{\kappa},\omega)$, and the thermal neutron scattering cross sections of materials. In principle, these atomistic methods make it possible to generate the inelastic thermal neutron scattering cross sections of any material and to accurately reflect the physical conditions of the medium (i.e, temperature, pressure, etc.). In addition, the generated cross sections are free from assumptions such as the incoherent approximation of scattering theory and, in the case of solids, crystalline perfection. As a result, new and improved thermal neutron scattering data libraries have been generated for a variety of materials. Among those are materials used for reactor moderators and reflectors such as reactor-grade graphite and beryllium (including the coherent inelastic scattering component), silicon carbide, cold neutron media such as solid methane, and neutron beam filters such as sapphire and bismuth. Consequently, it is anticipated that the above approach will play a major role in providing the nuclear science and engineering community with its needs of thermal neutron scattering data especially when considering new materials where experimental information may be scarce or nonexistent.

I. INTRODUCTION

Low energy or "thermal" neutrons are characterized by energies that are on the order of the excitation (vibration, rotation etc.) energy in the medium in which they interact. Furthermore, their de Broglie wavelength is on the order of the separation distance in solids. Consequently, such neutrons are highly sensitized to the atomic binding details of the system that surrounds them including its structure and dynamics. In fact, the structural and dynamic properties of the atomic system are sampled through scattering interactions between the system's atoms and molecules and the neutrons. The scattering of low energy neutrons in an atomic system is generally described using thermal neutron scattering cross sections. Traditionally, the cross sections are quantified based on Born scattering theory combined with Fermi's Golden rule and the assumption of an extremely short range (delta function) nuclear potential [1]. The outcome of this approach, is an expression for the double differential scattering cross section given by

$$\frac{d^2\sigma}{d\Omega dE} = \frac{1}{4\pi}\frac{k'}{k}\left(\sigma_{coh}S\left(\vec{\kappa},\omega\right) + \sigma_{inc}S_s\left(\vec{\kappa},\omega\right)\right), \quad (1)$$

where $S(\vec{\kappa},\omega)$ is known as the scattering law, $\vec{\kappa}$ is the scattering vector, ω is the frequency, k and k' represent the magnitude of the wave vector of the incident and scattered neutron respectively, σ_{coh} is the bound atom coherent scattering cross section, and σ_{inc} is the bound atom incoherent scattering cross section. In general, S is composed of two terms as follows

$$S\left(\vec{\kappa},\omega\right) = S_s\left(\vec{\kappa},\omega\right) + S_d\left(\vec{\kappa},\omega\right), \quad (2)$$

where S_s is known as the self-scattering law, which accounts for non-interference (incoherent) effects, while S_d is the distinct scattering law and accounts for interference (coherent) effects. Examination of Eq. 1 shows that the thermal neutron scattering cross section depends on two factors: first, the neutron-nucleus interaction as represented by the bound atom cross sections, and second, a factor that represents the dynamics of the scattering system (i.e., the collection of atoms) as represented by the scattering law.

Frequently, the calculations of the thermal scattering cross section invoke the incoherent approximation where S_d is set equal to zero in Eq. 2. Based on this assumption, Eq. 1 is developed to give (e.g., see Ref. [2])

$$\frac{d^2\sigma}{d\Omega dE} = \frac{1}{4\pi}\frac{k'}{k}\left[\sigma_{coh} + \sigma_{incoh}\right]S_s\left(\vec{\kappa},\omega\right). \quad (3)$$

However, for some important neutronic materials such as graphite and beryllium, this assumption can introduce

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http://dx.doi.org/10.1016/j.nds.2014.04.029
0090-3752/2014 Published by Elsevier B.V.

Annals of Nuclear Energy 135 (2020) 106840
Contents lists available at ScienceDirect
Annals of Nuclear Energy
journal homepage: www.elsevier.com/locate/anucene

On a measurement approach to support evaluation of thermal scattering law data

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ARTICLE INFO
Article history:
Received 31 January 2019
Received in revised form 13 July 2019
Accepted 17 July 2019

Keywords:
Neutron
Thermal scattering law
Cross section
Measurement
ENDF

ABSTRACT
Inelastic thermal neutron scattering in materials that act as neutron moderators, reflectors, and filters results in shaping the neutron spectrum at low energies. This phenomenon is described using differential scattering cross sections calculated from three components including the bound atom (i.e., nuclear) scattering cross section of the neutron, the ratio of the outgoing and incoming neutron energy, and the thermal scattering law (TSL) i.e., $S(\alpha,\beta)$, where α and β represent dimensionless momentum and energy exchange variables, respectively. To date, no TSL libraries are generated using measured data. However, valuable information may be derived from measurements and "targeted" experiments that can validate TSL data and the related inelastic scattering cross sections. As a demonstration, a suite of coordinated measurements and experiments is described that was designed and used to support the evaluation of the TSL for "nuclear" graphite. This experimental suite includes neutron powder diffraction (for structure analysis), positron annihilation (for nano porosity assessment), inelastic neutron scattering measurements using a chopper spectrometer, transmission experiment using neutrons with energy below the Bragg cutoff thereby accessing the total (inelastic) cross section, and a slowing-down-time experiment to observe the developing neutron spectrum in the material. This experimental suite was key to understanding the difference in TSL between "nuclear" and "ideal" graphite and for the inclusion of "nuclear" graphite in the ENDF/B-VIII.0 nuclear data library release.

1. Introduction

Over the past 15 years, a general methodology has been developed to generate thermal neutron scattering law (TSL) data, and calculate the inelastic thermal neutron scattering cross sections, for materials under various conditions and while relaxing many of the traditional approximations (e.g., the incoherent approximation) (Hawari, 2014, 2004). The methodology is based on using molecular dynamics (MD) and density functional theory (DFT) atomistic simulation methods to derive the fundamental input needed for TSL calculations, such as the atomic and molecular system's excitation density of states. Alternatively, the TSL may be directly accessed from the atomistic simulations and the resulting atomic correlations. In this case, corrections are needed to account for missing quantum effects such as detailed balance (Nelte, 2016). This methodology has resulted in the largest contribution (in the last 50 years) of TSL data to the recently released ENDF/B-VIII.0 nuclear data libraries (Brown, 2018).

In general, the formulation that would be the subject of the evaluation process originates from the fundamental equations for the double differential thermal scattering cross section. This formulation is derived using the first Born approximation of scattering theory and assumes a highly localized nuclear potential known as the Fermi pseudopotential [e.g., see Ref. Squires, 1978]. The outcome of this formulation is the following expression for the double differential thermal scattering cross section

$$\frac{d^2\sigma}{d\Omega dE} = \frac{1}{4\pi k_B T}\sqrt{\frac{\beta'}{\beta}}\sigma_{coh}S(\alpha,\beta) + \sigma_{inc}S_s(\alpha,\beta)] \quad (1)$$

$$S(\alpha,\beta) = S_s(\alpha,\beta) + S_d(\alpha,\beta), \quad (2)$$

where, $S(\alpha,\beta)$ is the thermal scattering law, $S_s(\alpha,\beta)$ is the self scattering law, $S_d(\alpha,\beta)$ is the distinct scattering law, α and β are dimensionless momentum and energy exchange variables, σ_{coh} is the coherent bound atom cross section, σ_{inc} is the incoherent bound atom cross section, E is the incoming neutron energy, E is the scattered neutron energy, Ω is the scattering solid angle, k_B is Boltzmann's constant, and T is the temperature of the scattering medium. In this case, S_s is related to the Fourier transform in space and time of the density

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https://doi.org/10.1016/j.anucene.2019.106840
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Available online at www.sciencedirect.com
ScienceDirect
Nuclear Data Sheets 148 (2018) 1–142
Nuclear Data Sheets
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ENDF/B-VIII.0: The 8th Major Release of the Nuclear Reaction Data Library with CIELO-project Cross Sections, New Standards and Thermal Scattering Data

D. A. Brown,[1] M. B. Chadwick,[2,+] R. Capote,[3] A. C. Kahler,[2] A. Trkov,[3] M. W. Herman,[1] A. A. Sonzogni,[1] Y. Danon,[4] A. D. Carlson,[5] M. Dunn,[6] D. L. Smith,[7] G. M. Hale,[2] G. Arbanas,[8] R. Arcilla,[1] C.R. Bates,[2] B. Beck,[9] B. Becker,[10] F. Brown,[2] R. J. Casperson,[9] J. Conlin,[2] D. E. Cullen,[3] M.-A. Descalle,[9] R. Firestone,[11] T. Gaines,[12] K. H. Guber,[8] A. I. Hawari,[13] J. Holmes,[14] T. D. Johnson,[1] T. Kawano,[2] B. C. Kiedrowski,[15] A. J. Koning,[3] S. Kopecky,[16] L. Leal,[17] J. P. Lestone,[2] C. Lubitz,[18] J. I. Márquez Damián,[19] C. M. Mattoon,[9] E. A. McCutchan,[1] S. Mughabghab,[1] P. Navratil,[20] D. Neudecker,[2] G. P. A. Nobre,[1] G. Noguere,[21] M. Paris,[2] M. T. Pigni,[8] A. J. Plompen,[16] B. Pritychenko,[1] V. G. Pronyaev,[22] D. Roubtsov,[23] D. Rochman,[24] P. Romano,[7] P. Schillebeeckx,[16] S. Simakov,[25] M. Sin,[26] I. Sirakov,[27] B. Sleaford,[9] V. Sobes,[8] E. S. Soukhovitskii,[28] I. Stetcu,[2] P. Talou,[2] I. Thompson,[9] S. van der Marck,[29] L. Weiser-Sherrill,[2] D. Wiarda,[8] M. White,[2] J. L. Wormald,[12] R. Q. Wright,[8] M. Zerkle,[14] G. Zerovnik,[16] and Y. Zhu[12]

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(Received 18 September 2017; revised received 21 November 2017; accepted 14 December 2017)

We describe the new ENDF/B-VIII.0 evaluated nuclear reaction data library. ENDF/B-VIII.0 fully incorporates the new IAEA standards, includes improved thermal neutron scattering data and uses new evaluated data from the CIELO project for neutron reactions on ^{1}H, ^{16}O, ^{56}Fe, ^{235,238}U, ^{239}U and ^{239,240}Pu described in companion papers in the present issue of *Nuclear Data Sheets*. The evaluations benefit from recent experimental data obtained in the U.S. and Europe, and improvements in theory and simulation. Notable advances include updated evaluated data for light nuclei, structural materials, actinides, fission energy release, prompt fission neutron and γ-ray spectra, thermal neutron scattering data, and charged-particle reactions. Integral validation testing is shown for a wide range of criticality, reaction rate, and neutron transmission benchmarks. In general, integral validation performance of the library is improved relative to the previous ENDF/B-VII.1 library.

https://doi.org/10.1016/j.nds.2018.02.001
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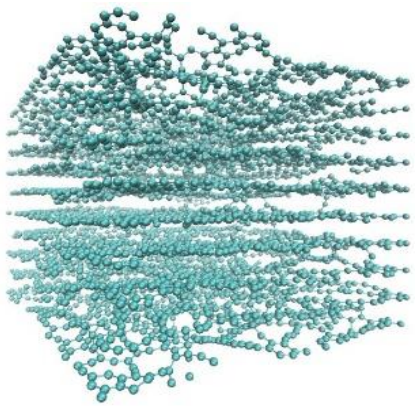
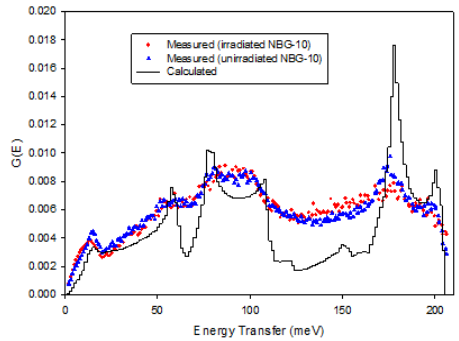
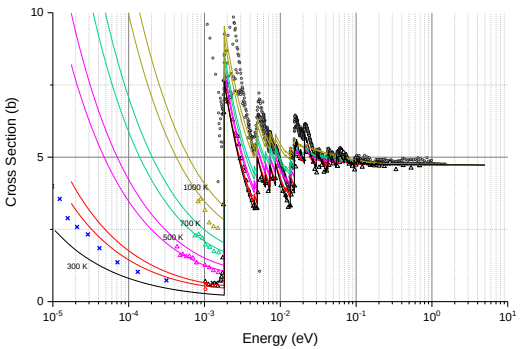
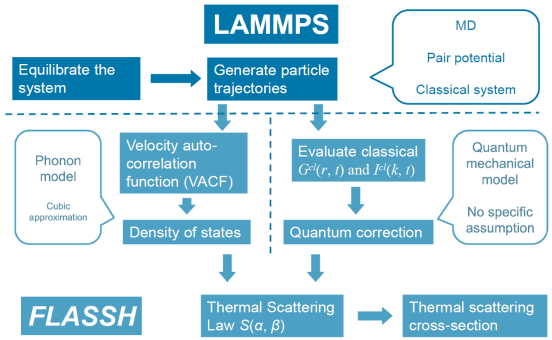
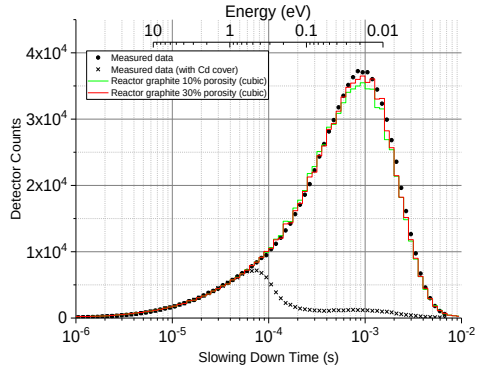
ENDF/B-VIII
Nuclear Graphite

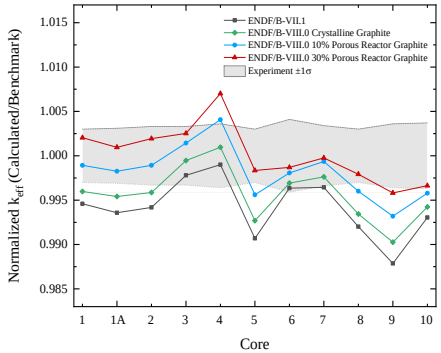










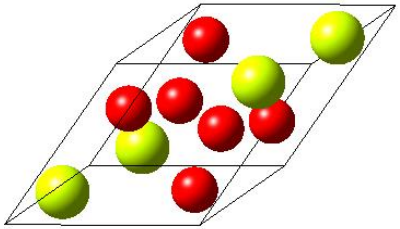


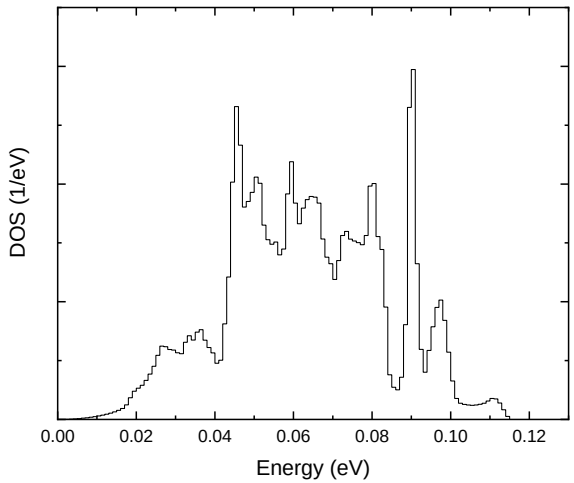
Evaluation

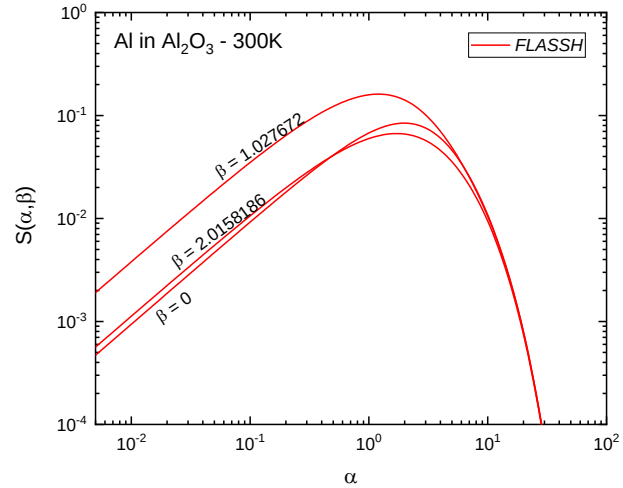
Measurements/Benchmarks

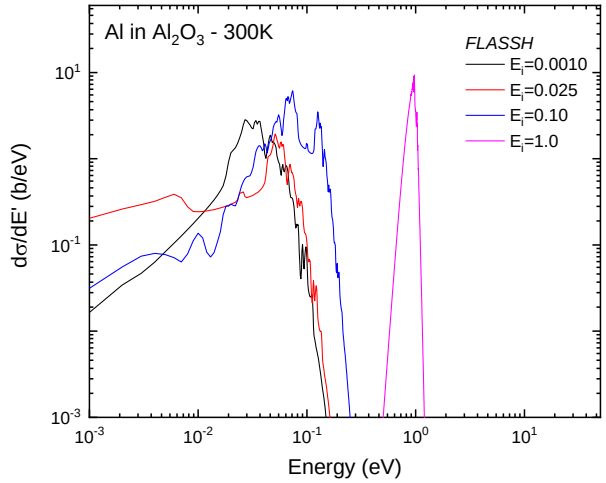
New to NNDC
Single Crystal Sapphire TSL Data

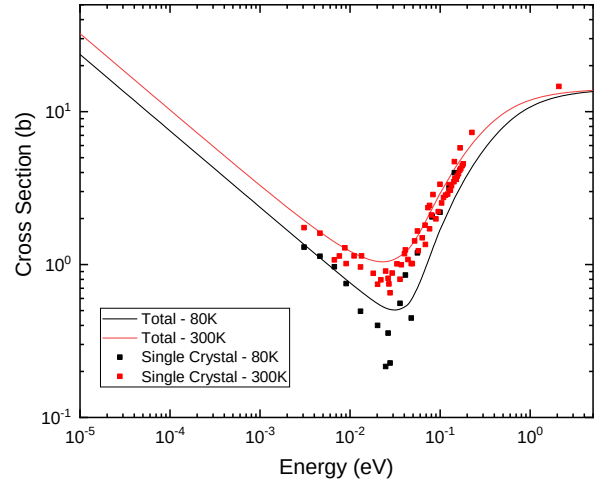
- *Ab initio* lattice dynamics
  - Predictive density of states (DOS)
  - Rhombohdral structure
  - 2x2x2 supercell
  - GGA-PBE





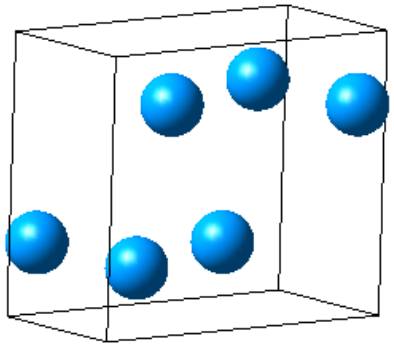




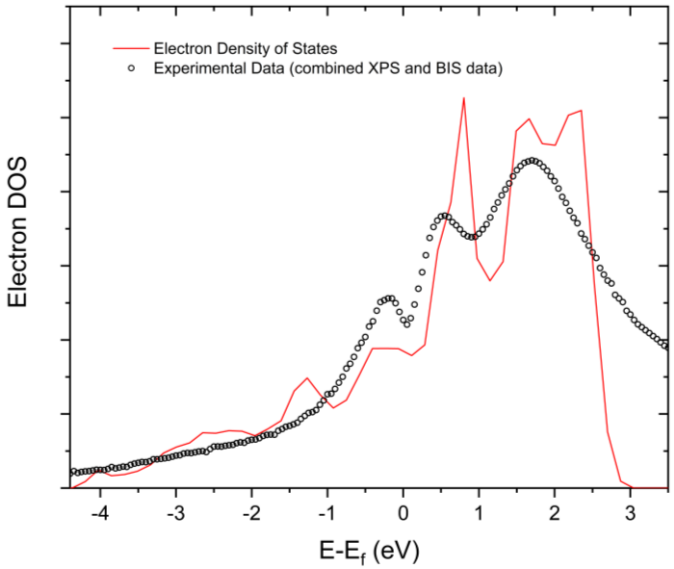


Uranium Metal

- α-Uranium Metal
  - Stable up to 668ºC
- *Ab initio* lattice dynamics
  - DFT – GGA-PBE plus an effective Coulomb term (+U) of 1eV for the *5f* electrons plus spin-orbit coupling



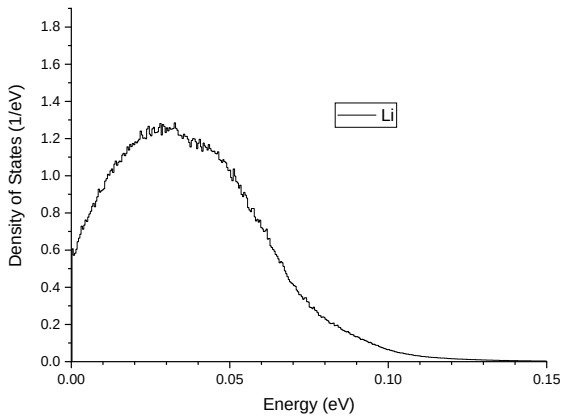
- Orthorhombic structure
- 550 eV plane wave cutoff
- 12x12x7 k-mesh



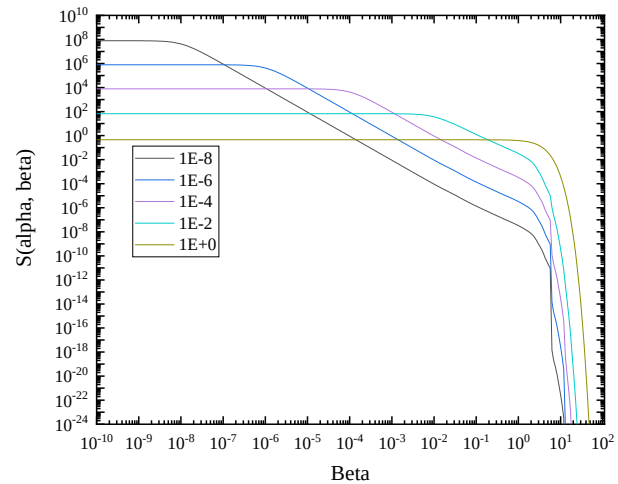
	Experiment (4.2 K)	Calculated	Diff. (%)
a (Å)	2.8444	2.8565	0.42
b (Å)	5.8689	5.8706	0.03
c (Å)	4.9316	4.9834	1.05

Completed FLiBe TSL Data

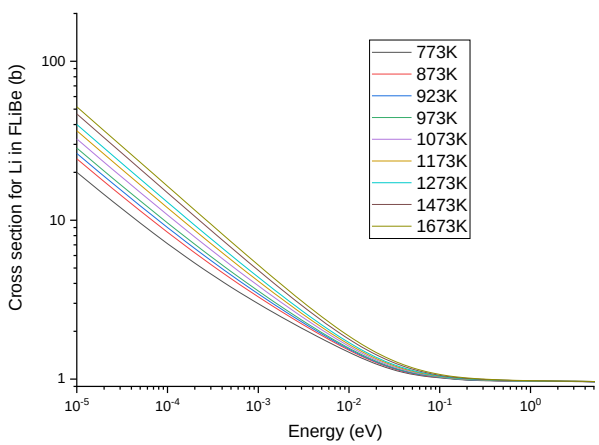
DOS using MD 773 K



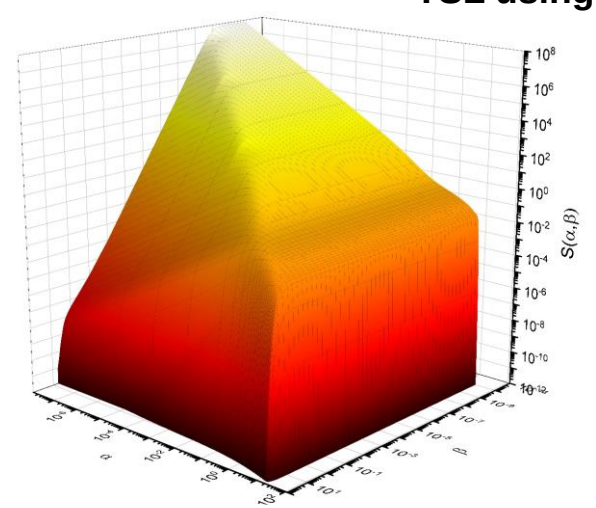
TSL using *FLASSH 773 K*



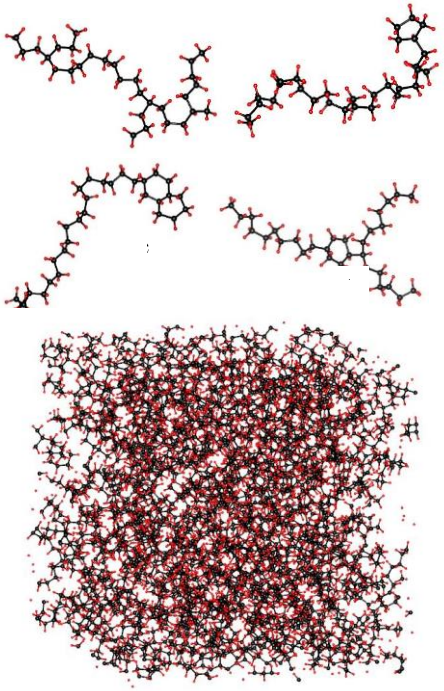
Cross section using *FLASSH*

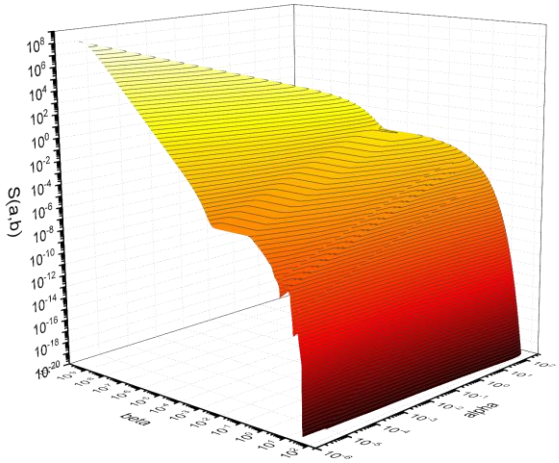


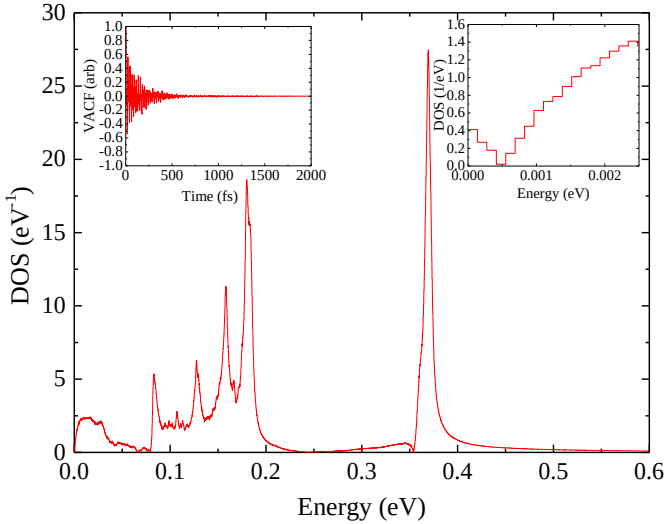
TSL using *FLASSH 773 K*



New to NNDC
Heavy Paraffinic Oil

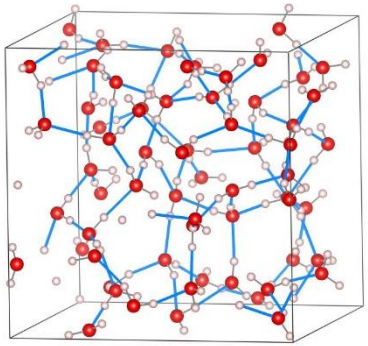




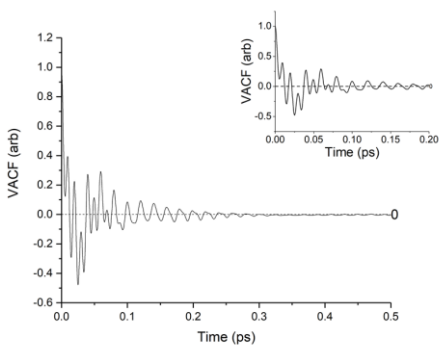


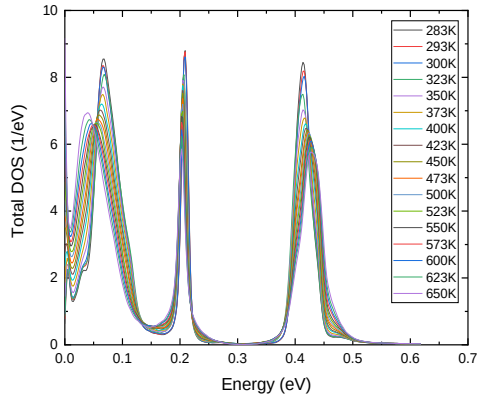
Light Water TSL Data

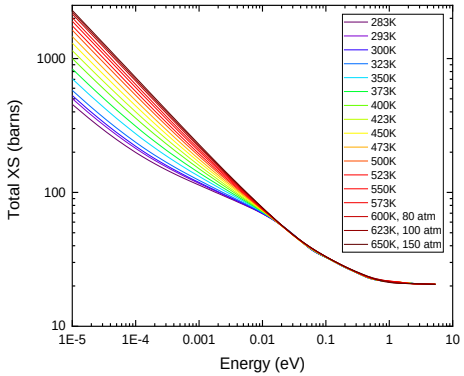
- Flexible TIP4P/2005 potential
- Molecular dynamics model
  - LAMMPS code
  - 1000 molecules



$$U^{tot} = U^{\text{inter}} + U^{\text{intra}} = \sum_{i \neq j} 4\varepsilon \left[\left(\frac{\sigma}{r_{O_i O_j}} \right)^{12} - \left(\frac{\sigma}{r_{O_i O_j}} \right)^{6} \right] + \sum_{i \neq j} \frac{q_i q_j}{r_{ij}} + D_r \left\{ 1 - \exp\left[-\beta (r_{OH} - r_{eq}) \right] \right\}^2 + \frac{1}{2} K_\theta (\theta - \theta_{eq})^2$$

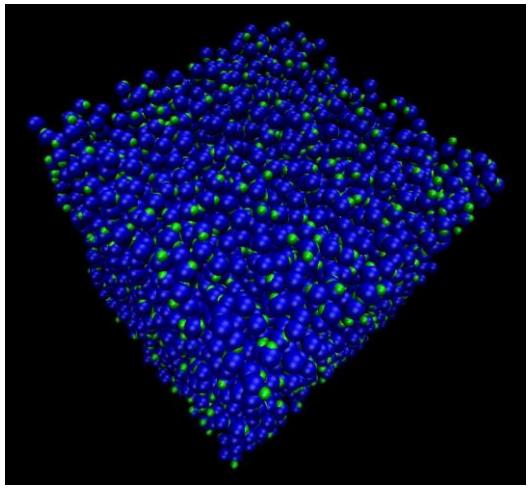




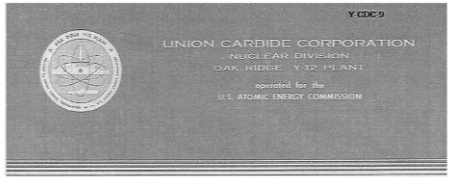


Hydrofluoric Acid MD

- Constructed potential function
- Molecular dynamics model
  - LAMMPS code
  - 4000 molecules

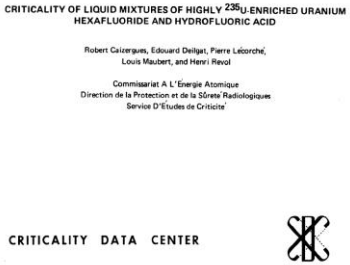


$$U^{total} = \sum_i \frac{1}{2} k_D \left|\boldsymbol{r}_{F,i} - \boldsymbol{d}_i\right|^2 + \frac{1}{2} k_{HF} (r_{HF} - d_{FH})^2 + \sum_{i<j} \sum_{s,\acute{s}} \frac{q_s q_{\acute{s}}}{\left|\boldsymbol{r}_{s,i} - \boldsymbol{r}_{\acute{s},i}\right|} + \sum_{i<j} 4\,\varepsilon_{FF} \left[\left(\frac{\sigma_{FF}}{\left|\boldsymbol{r}_{F,i} - \boldsymbol{r}_{F,j}\right|} \right)^{12} - \left(\frac{\sigma_{FF}}{\left|\boldsymbol{r}_{F,i} - \boldsymbol{r}_{F,j}\right|} \right)^{6} \right]$$



Y-CDC-9

UNION CARBIDE CORPORATION
NUCLEAR DIVISION
OAK RIDGE Y-12 PLANT
operated for the
U.S. ATOMIC ENERGY COMMISSION



CRITICALITY OF LIQUID MIXTURES OF HIGHLY ^{235}U-ENRICHED URANIUM HEXAFLUORIDE AND HYDROFLUORIC ACID

Robert Caizergues, Edouard Deilgat, Pierre Lécorché, Louis Maubert, and Henri Revol

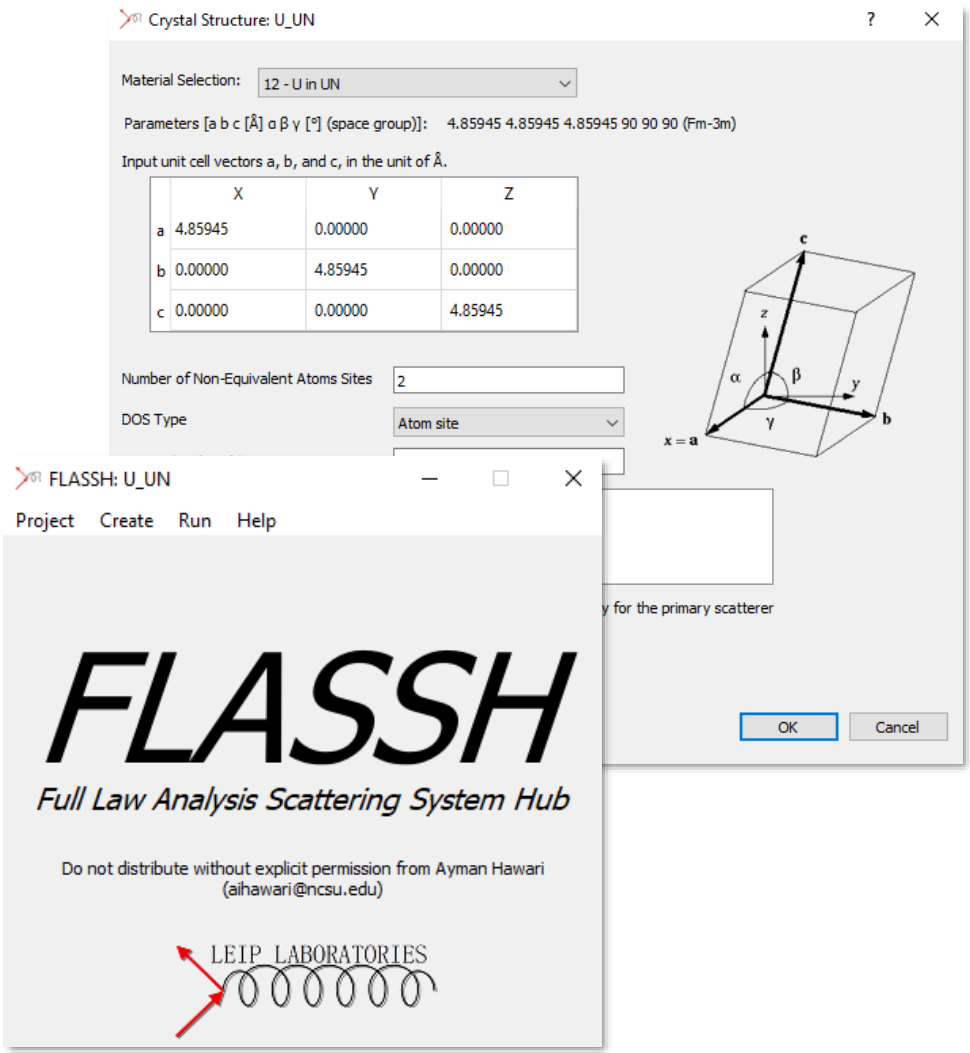
Commissariat A L'Energie Atomique
Direction de la Protection et de la Sûreté Radiologiques
Service D'Études de Criticité

CRITICALITY DATA CENTER

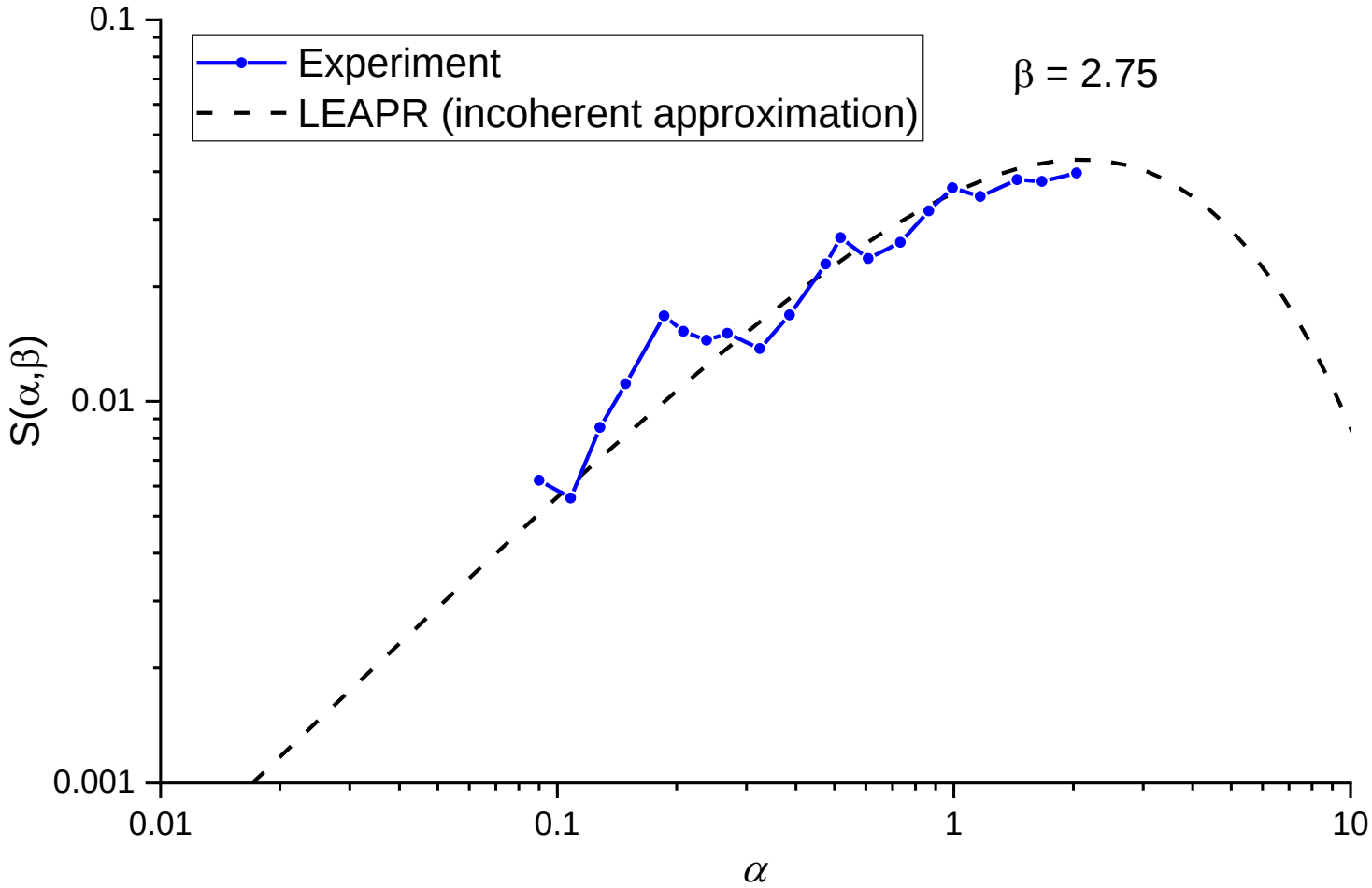
- Testing is underway using experimental data derived from "CRITICALITY OF LIQUID MIXTURES OF HIGHLY 235U-ENRICHED URANIUM HEXAFLUORIDE AND HYDROFLUORIDE ACID"

FLASSH Code

- *FLASSH* Code
  - Relaxed major approximations (incoherent, cubic, ect.)
  - Improved liquid physics
  - Improved Output Formatting
  - Warning Messages for the User
- *FLASSH* GUI
  - Error Checks
  - Crystal Structure Window/ Inputs
  - ENDF Header Formatting



NJOY
Be Scattering Law $S(\alpha, \beta)$



FLASSH

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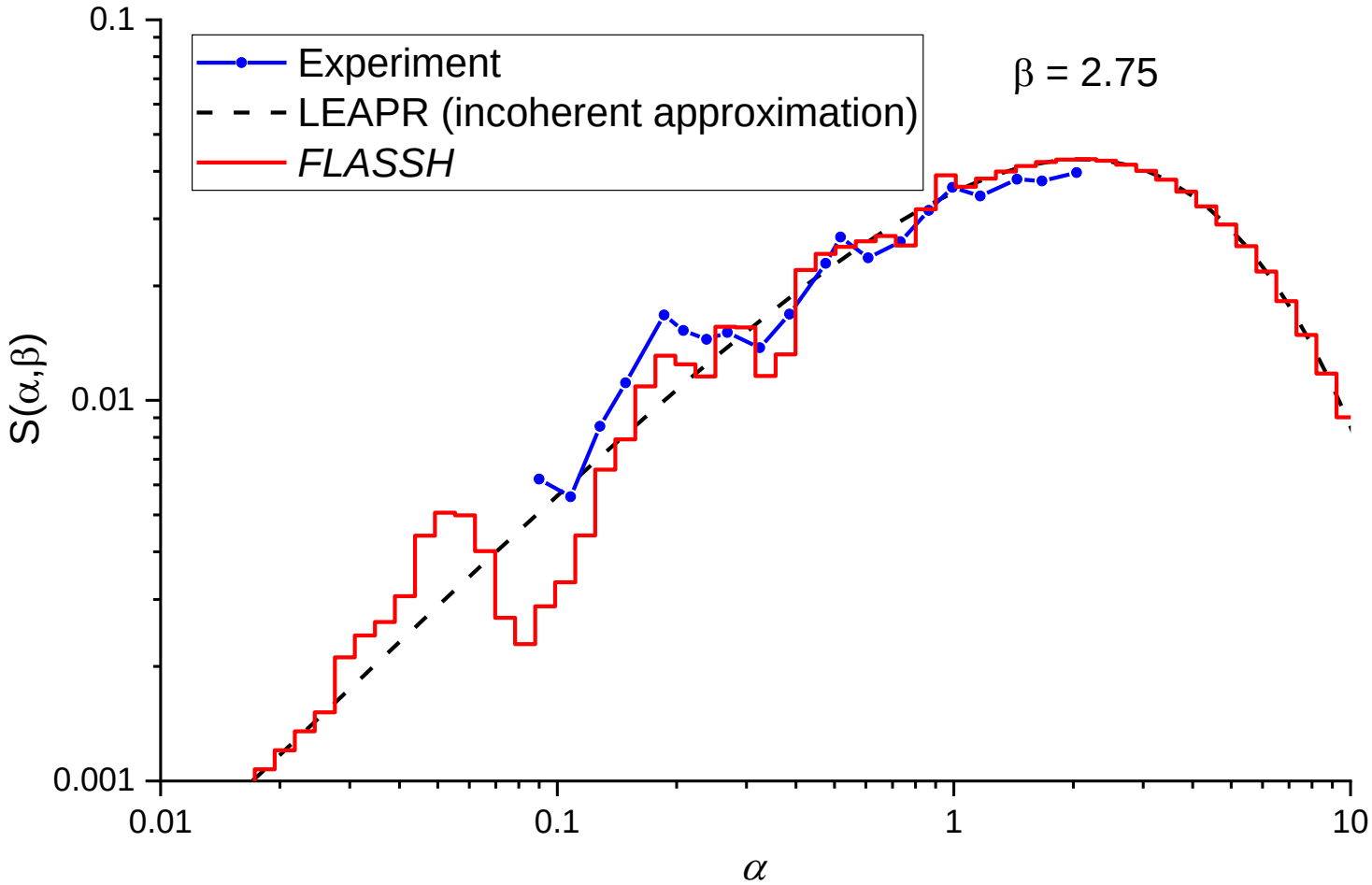
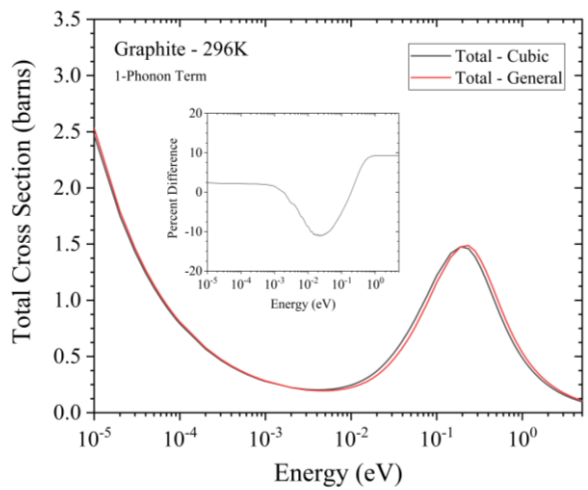


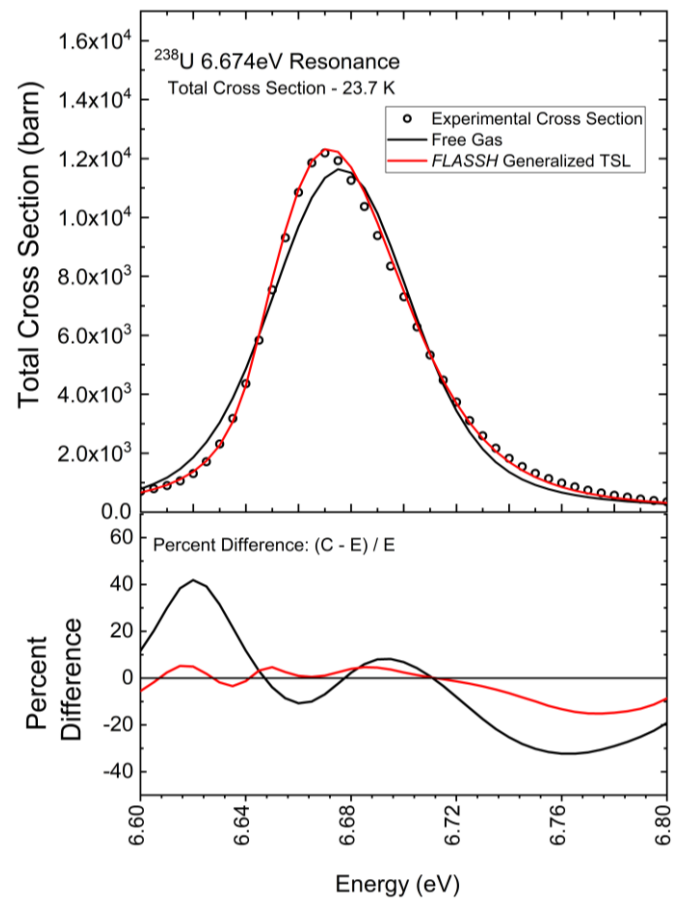
FLASSH – Generalized TSL & Doppler Treatment

- **Generalized TSL**
  - Exact Structure
  - Polarization vectors and frequencies directly input
  - Removes cubic and atom site approximation
- **Doppler Broadening**
  - *FLASSH* TSL to determine velocity distribution for broadening
  - Implements exact material structure
  - Consistent evaluation for both thermal scattering and Doppler broadening

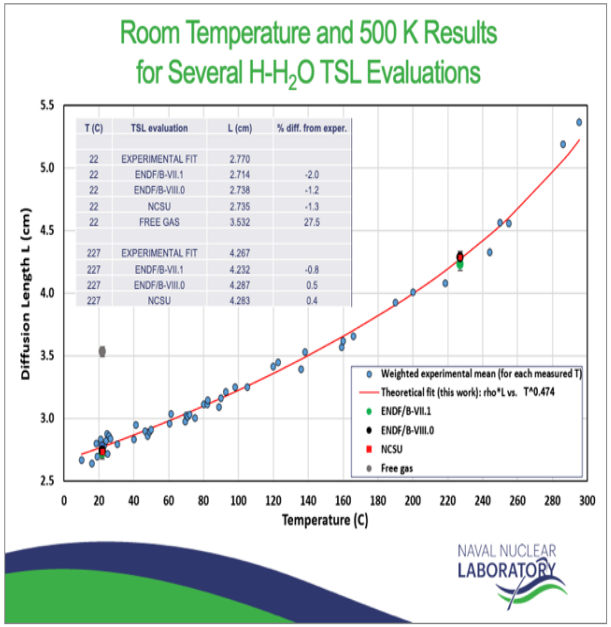
$$S_s^m(k,\omega) = \int e^{-i\omega t} I_s^m(k,t)\,dt$$

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

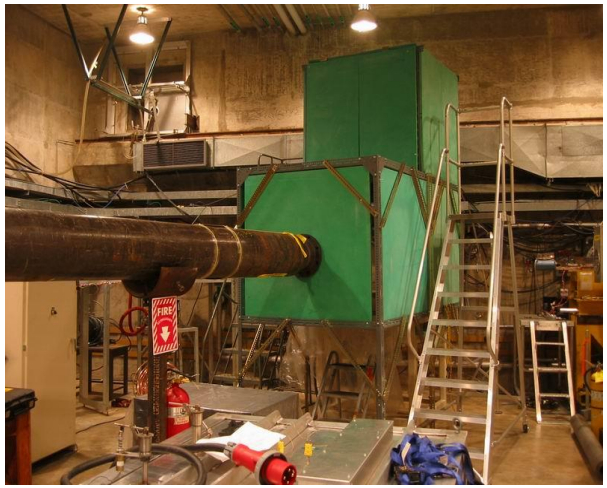


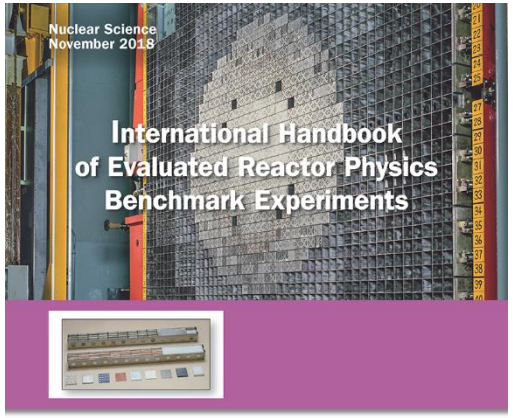


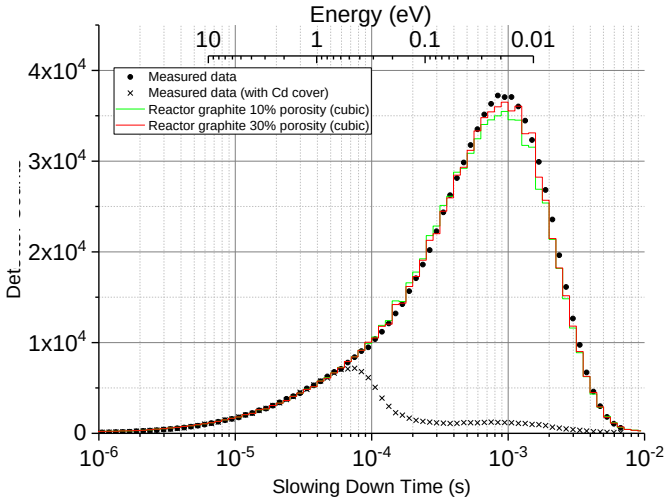
TSL Specific Benchmarks

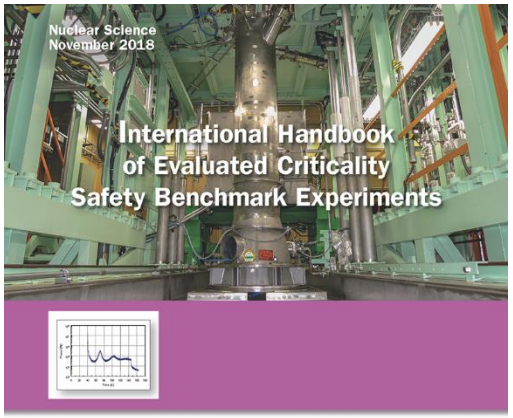


T (C)	TSL evaluation	L (cm)	% diff. from exper.
22	EXPERIMENTAL FIT	2.770	
22	ENDF/B-VII.1	2.714	-2.0
22	ENDF/B-VIII.0	2.738	-1.2
22	NCSU	2.735	-1.3
22	FREE GAS	3.532	27.5
227	EXPERIMENTAL FIT	4.267	
227	ENDF/B-VII.1	4.232	-0.8
227	ENDF/B-VIII.0	4.287	0.5
227	NCSU	4.283	0.4





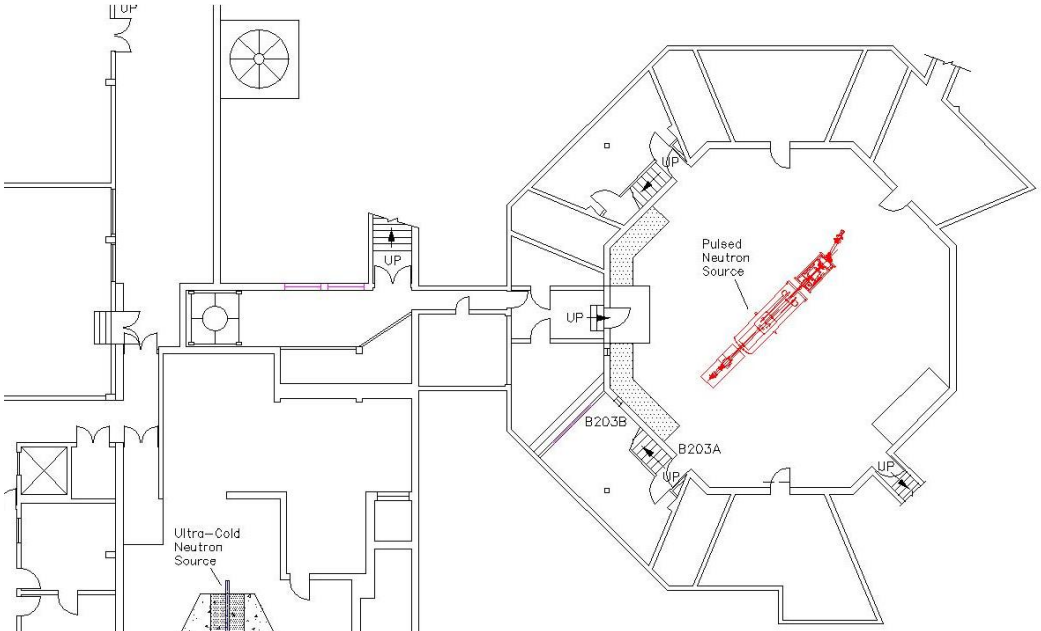




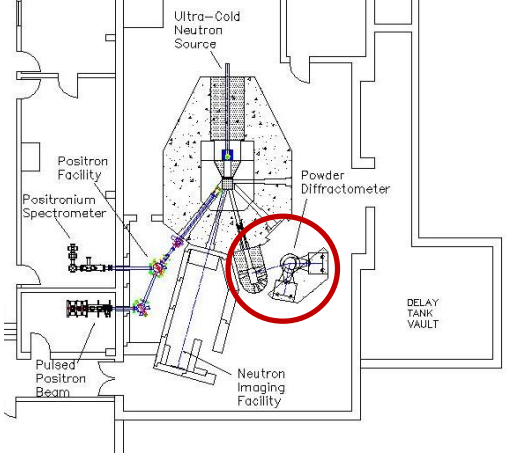
LEIP LABORATORIES

PULSTAR reactor

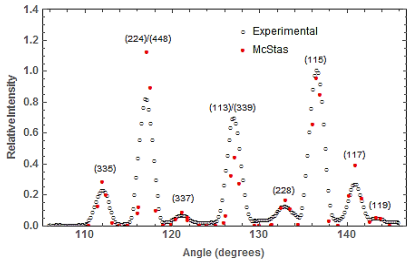
- **Current major facilities/capabilities**
  - **Neutron powder diffraction**
  - Neutron imaging
  - Intense positron beam
  - Ultracold neutron source (under testing)
  - Neutron activation analysis
  - In-pool irradiation testing facilities
- **Current projects**
  - PULSTAR power upgrade 1-2 MW (licensing stage)
  - Various instrument and facility upgrades
  - **Pulsed accelerator neutron source (under testing)**
  - Fuel loop for fission gas release studies



Old reactor bay



reactor bay



Cross Section Measurement Project

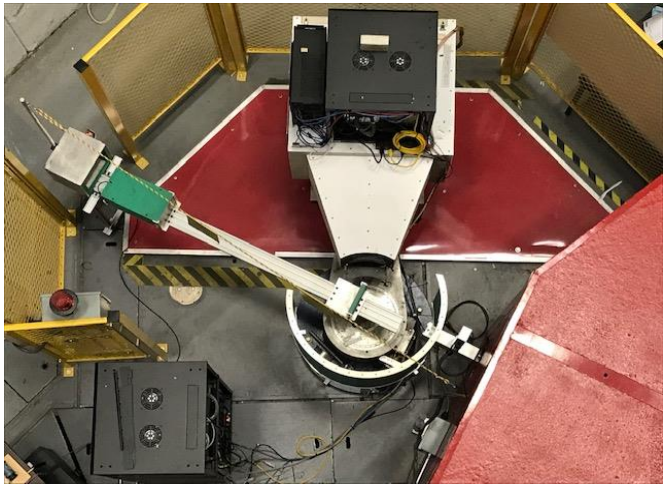
NPDF Facility Upgrades – Dual Purpose:

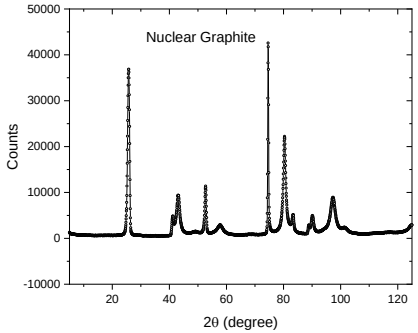
- Diffraction Measurements:

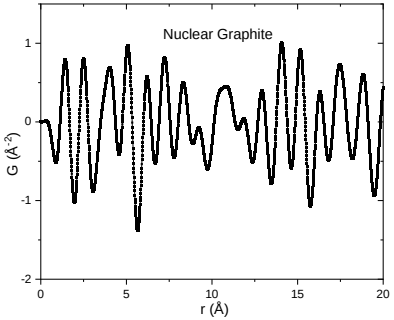
  15 New Position Encoding Modules (PEM) – improved diffraction measurement resolution $\Delta d/d$ of 2.9×10^{-3} for 3mm holder

- **Transmission Measurement Capabilities:**

  - Monochromator capable of providing beam wavelengths of 1.085 Å, 1.180 Å, 1.479 Å, and 1.762 Å
  - Transmission Detection Apparatus with collimator.

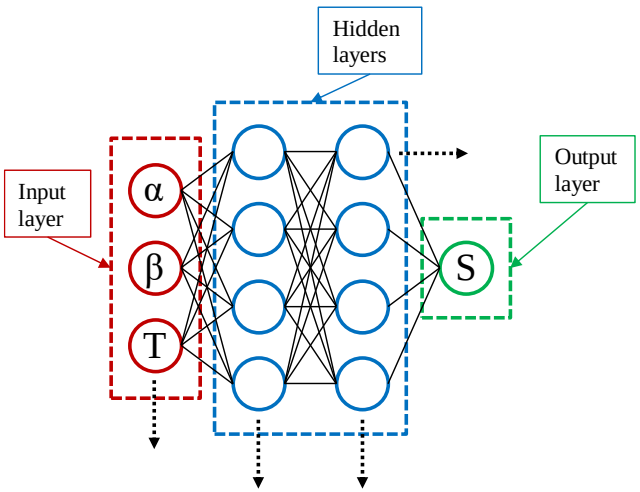


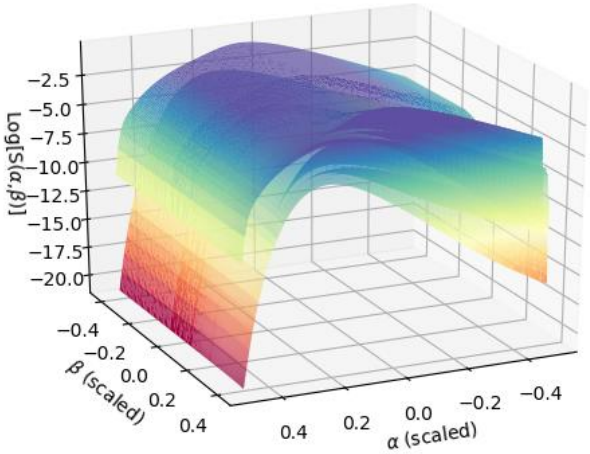




TSL NeTS

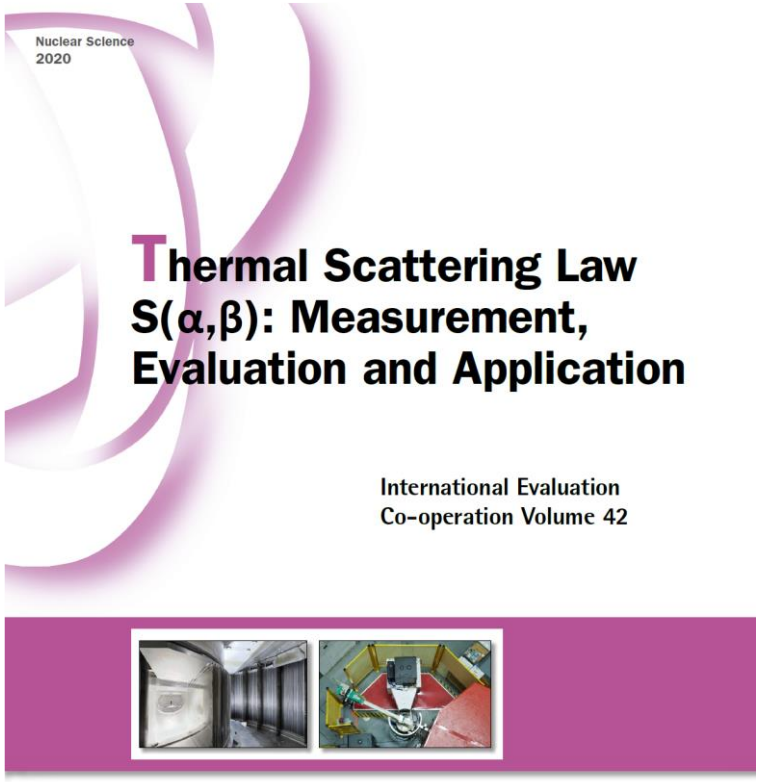
- New TSL paradigm
  - DL Neural Thermal Scattering (NeTS) modules
- See papers and presentations
  - ANS 2019 Winter Meeting, Washington, DC, USA
  - PHYSOR 2020 Meeting, Cambridge, UK





OECD/NEA WPEC Subgroup 48

- Kick-off in May 2020 during WPEC meeting
  - Motivate the TSL evaluation effort in support of various nuclear science and engineering applications
    - Advanced reactors (e.g., various molten salts)
    - Criticality safety (e.g., various U and Pu based fuels)
    - Neutron science (e.g., cryogenic moderators)
  - Review the development of advanced TSL evaluation methods and tools with consideration of modern simulation approaches
  - Address issues related to data validation, covariance generation, and data formats, …



OECD
BETTER POLICIES FOR BETTER LIVES

NEA
NUCLEAR ENERGY AGENCY

Summary

- TSL Activities continue including evaluations and methods development
- Evaluations are contributed to NNDC
  - Paraffinic oil and single crystal sapphire
  - Molten salt FLiBe will be submitted soon
  - Reevaluation of light water is ongoing
  - Hydrofluoric acid is initiated
  - Uranium metal is initiated
- *FLASSH* testing and several evaluations are underway
- Activities in data measurements and benchmark development are underway
- DL NeTS approach is under testing