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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])

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

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

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inaccuracies in the calculated inelastic scattering cross section. Various methods can be used to remedy this effect by estimating S_d and introducing it into the expression for thermal neutron scattering cross sections. If the atoms are assumed to be bound by harmonic forces, an expansion can be performed to allow the decomposition of the coherent and incoherent double differential scattering cross section into elastic and inelastic components. In crystalline materials, this expansion is known as the phonon expansion. Assuming that the largest contribution to the scattering law is due to the creation or annihilation of a single phonon, the thermal neutron scattering cross section (including the S_d coherent inelastic contribution) can be written as

$$\frac{d^2\sigma}{d\Omega\, dE} \cong \frac{1}{4\pi}\frac{k'}{k}\Big\{((\sigma_{coh}+\sigma_{incoh})\left(\sum_{P=2}{}^{P}S_s\right) + \sigma_{coh}\left({}^{1}S_s + {}^{1}S_d\right)\Big\}, \quad (4)$$

where P represents the number of phonons created or annihilated in the inelastic scattering process (usually set large enough to ensure that high energy transfers are captured), and ${}^{1}S_d$ is the distinct scattering law for 1-phonon scattering [3].

II. METHODS FOR GENERATING THE SCATTERING LAW

As indicated above the scattering law, S, is an inherent property of the material in which the neutrons are interacting. Consequently, it can be estimated based on atomistic models of the material. For crystalline solids, the formulation for calculating the scattering law may be reduced to requiring the phonon density of states [e.g., see Ref. [2]]. In this case, a predictive (non-empirical) approach that is based on ab initio density functional theory (DFT) and/or classical molecular dynamics can be implemented.

A. Ab initio DFT Analysis

This approach treats the atomic system within the adiabatic Born-Oppenheimer approximation, which allows the electronic and nuclear components to be viewed as separable. The combination of the adiabatic assumption with DFT, and its principle assumption that the total energy of the electronic system can be set as a functional of the electronic density, allows the ground state energy of the system to be estimated. Once the ground state energy and its associated Hamiltonian are defined, then Hellmann-Feynman theorem can be used to calculate the inter-atomic forces in the system. Such modern methods that depend on ab initio DFT for the estimation of inter-atomic forces can then provide the basic input for executing classical dynamical matrix calculations of the phonon density of states and the associated S (e.g., see Ref. [4] and the references within).

The above represents a significant departure from past techniques that may have depended on simplistic atomistic models and fitting to experimental data to arrive at fundamental information such as the inter-atomic forces. Consequently, this approach expands substantially the ability to generate the thermal neutron scattering cross sections to many materials, (e.g., moderators, filters etc.) that may be of interest to nuclear reactor design and utilization.

B. Molecular Dynamics Analysis

More recently, the application of classical molecular dynamics (MD) techniques has been initiated to estimate the scattering law of materials [5]. In this approach, which also utilizes the Born-Oppenheimer approximation, the forces between atoms are calculated based on the choice and parameterization of appropriate potential functions (e.g., see Ref. [6]). The advantage of this approach over, e.g., the use of a dynamical matrix methodology, is the ability to relax the harmonic assumption and to treat imperfect materials that may contain symmetry breaking irregularities in the lattice structure. Furthermore, the MD approach is inherently suited for estimating the scattering law for materials that may be in the liquid or gaseous states. Information regarding the density of states and the scattering law can be extracted by calculating velocity and density autocorrelation functions, respectively, for the atoms in a system. However, as the calculations are classical in nature, elements of the thermal scattering phenomenon such as detailed balance and recoil effects are not typically captured. Therefore, quantum mechanical correction procedures may need to be introduced to account for such missing phenomena [7, 8]. The final outcome of this approach is a methodology that can treat the calculation of the scattering law of a given material in a general fashion including the estimation of the S_d component to relax the incoherent approximation.

III. THERMAL NEUTRON SCATTERING DATA

Based on the above discussion, DFT combined with the dynamical matrix approach and MD methods were used to generate the thermal neutron scattering cross section for neutron moderators/reflectors, neutron filters and cold neutron source materials. Important materials of interest are graphite, beryllium and silicon carbide. Graphite and beryllium (and to a lesser extent silicon carbide) are coherent scatterers where the incoherent approximation is known to introduce noticeable biases. Consequently, the calculations were performed by combining the fundamental input (i.e., the phonon density

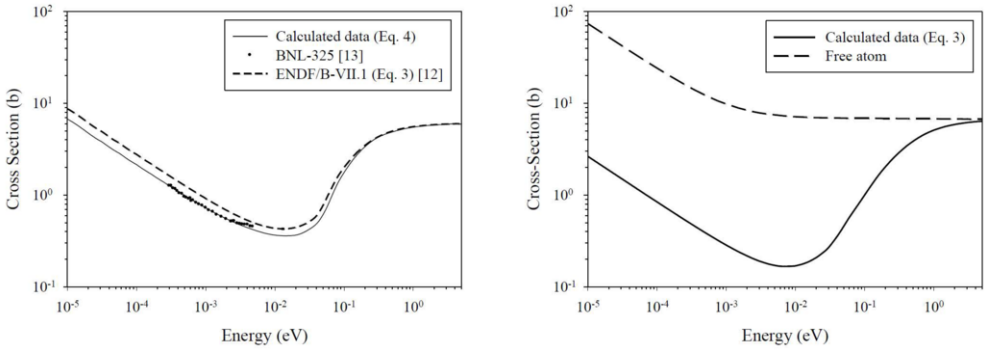

FIG. 1. The total inelastic scattering cross sections for beryllium (left) and silicon carbide (right) at 300 K.

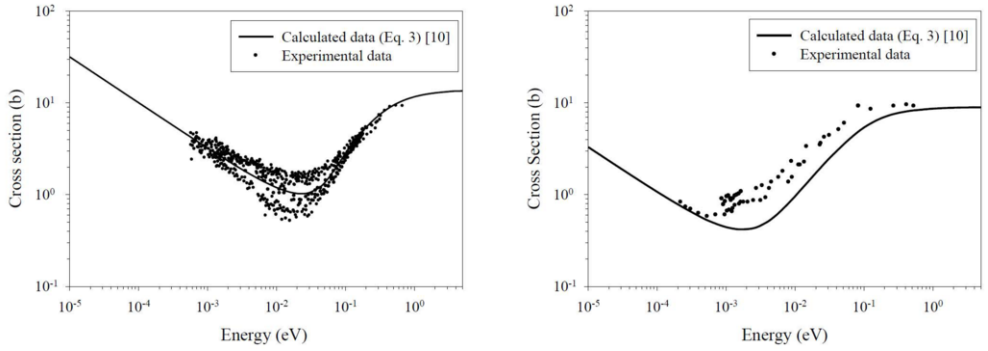

FIG. 2. The total inelastic scattering cross sections for sapphire (left) and bismuth (right) at 300 K [10].

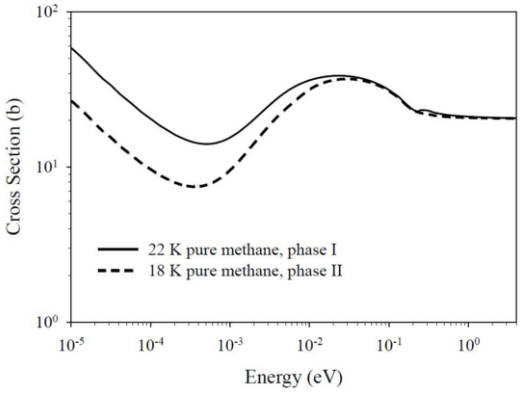

FIG. 3. The total inelastic scattering cross sections for solid methane [11].

of states) that was generated using DFT and dynamical matrix methods with Eq. 3 (i.e., the incoherent approximation) or Eq. 4 to account for the S_d component of the scattering law. A review of graphite data (including ideal and reactor-grade type materials) may be found in Ref. [9]. Figure 1 shows the data for beryllium (corected for the incoherent approximation) and silicon carbide, which gives the total inelastic scattering cross section for both materials. These represent new data sets that are not currently included in the ENDF/B-VII.1 compilation [12].

DFT and dynamical matrix techniques can be used to generate the thermal neutron scattering cross sections for neutron beam filters. In this case, two very popular filters are single crystal sapphire (used to remove fast neutrons) and single crystal bismuth (used to remove gamma-rays), which are usually implemented at room temperature. Figure 2 shows the total inelastic scattering cross section for these materials [10]. In both cases the calculations were performed within the limits of the incoherent approximation (i.e., Eq. 3). These represent new data sets that are not currently included in the ENDF/B-VII.1 compilation [12].

Another class of materials that is of interest is cryogenic moderators that serve as cold and ultracold neuron sources. As an example, MD techniques were used to study solid methane in its various phases to understand the impact of temperature dependent phase changes on its eventual performance as a cold neutron source [11]. In addition, the study demonstrated how dopants (e.g., krypton) may play a role in preserving the cold moderating properties even at temperatures below 22 K. Consequently, such MD simulations are helpful in understanding and interpreting experimental observations. Figure 3 shows the total inelastic scattering cross section for solid methane.

IV. CONCLUSIONS

Nowadays, various computational techniques have become available to generate the needed input for the calculation of the thermal neutron scattering cross sections. These techniques are of a general nature and, if needed, can quite easily overcome the handicaps of past methods (e.g., the incoherent approximation). In addition, the techniques take advantage of sophisticated atomistic simulations approaches based on DFT and MD that are predictive in nature, that can treat matter in all of its usual states (solid, liquid and gas), and that allow the treatment of irregular and imperfect materials. The results present data for various materials some of which are not currently available as part of the standard ENDF libraries [12]. Furthermore, methods are currently under development to allow the estimation of the data uncertainties and the generation of the associated covariance matrices for inclusion in the thermal neutron scattering data libraries [14].

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