Thermal Neutron Scattering in Graphite

by

Iyad I. Al-Qasir
Department of Physics
University of Jordan
Amman, Jordan

Supervisor

Dr. Ayman I. Hawari
Department of Nuclear Engineering
North Carolina State University
Raleigh, NC
Introduction

- Generation IV Very High Temperature Reactor (VHTR) are graphite moderated and gas cooled thermal spectrum reactors.

- The characteristics of the low energy (E < 1eV) neutron spectrum in these reactors will be dictated by the process of neutron slowing-down and thermalization in the graphite moderator.

- The ability to accurately predict this process in these reactors can have significant neutronic and safety implications.

- Currently used libraries (ENDF/B-VII) are a product of the 1960s and remain based on many physical approximations, these libraries show noticeable discrepancies with experimental data.
Graphite Inelastic Scattering Cross Section

[Graph showing cross section vs. energy with data points and curves for ENDF/B-VII (YK), Steyrel (1974), and Zhou (2006).]

T=300 K
Objectives

- To Review the currently used thermal neutron scattering laws of graphite as a function of temperature

- To Update models and models’ parameters by introducing
  - The new developments in solid-state physics (\textit{ab initio} approach)
  - The coherent part of the inelastic scattering

- To Develop and generate new sets of temperature dependent thermal neutron scattering laws
Outline

- Neutron Thermalization
- Lattice Dynamics
- Results
- Conclusions
Neutron Thermalization
The wavelength of thermal neutrons is comparable to the interatomic distances in solids and liquids.

The energy of thermal neutrons is of the same order of the excitations in condensed matter (e.g. phonons in crystalline materials).
Inelastic double differential scattering cross section

\[
\frac{d^2\sigma}{d\Omega \, dE'} = \frac{1}{4\pi} \frac{k'}{k} \left\{ \sigma_{coh} \sum_{P=1}^P S_d \left( \kappa, \omega \right) + \left( \sigma_{coh} + \sigma_{incoh} \right) \sum_{P=1}^P S_s \left( \kappa, \omega \right) \right\}
\]

Graphite → \( \sigma_{coh} = 5.50 \text{b} \)
\( \sigma_{incoh} \approx 0 \text{b} \)

Scattering Law \( S(\alpha, \beta) = k_B T e^{\beta/2} S(\kappa, \omega) \)

Momentum transfer \( \alpha = \frac{E' + E - 2\mu\sqrt{EE'}}{Ak_B T} \)

Energy Transfer \( \beta = \frac{E' - E}{k_B T} \)
Approximations

1- Incoherent approximation:

\[ S_d (\alpha, \beta) = 0 \]

\[ \Rightarrow \frac{d^2 \sigma}{dE' \, d\Omega} \approx \sigma_{coh} + \sigma_{incoh} \frac{E'}{E} e^{-\beta/2} \sum_{p=1} S_s (\alpha, \beta) \]

2- Harmonic interatomic forces
3- Monoatomic solid
4- The solid has one atom per unit cell
5- The unit cell has a cubic symmetry
6- The solid vibrational modes are described by a continuous spectrum, called the phonon frequency distribution \( \rho(\beta) \)
7- Gaussian Approximation

\[ \Rightarrow S_s (\alpha, \beta) = \frac{1}{2\pi} \int_{-\infty}^{\infty} e^{i\beta \tau} e^{-\gamma^2(\tau)} d\tau \]
In order to calculate the scattering law, all we need is the phonon frequency distribution $\rho(\beta)$.

The above formulation represents the basis of computer programs such as GASKET and LEAPR/NJOY.

Discrepancies between measurements and calculations (differential and integral)
Lattice Dynamics
Output:
- Dispersion relations: $\omega_j(\vec{q})$
- Polarization vectors: $\vec{e}_{dij}(\vec{q})$
- Phonon Distribution: $\rho(\omega)$

Phonon Frequency $\rho(\beta)$

Lattice dynamics
Force Constants Models

Early Work
- Force constant values obtained by fitting to thermodynamical experimental data (e.g., specific heat, compressibility, ...)

Graphite (1965)
Young-Koppel (ENDF/B-VII)

Later (Neutron Scattering)
- Force constant obtained by fitting to experimental dispersion curves along symmetry directions in the 1st Brillouin Zone

Graphite (1970)
Nicklow et al., (ORNL)

Recently
- (Ab Initio Calculations)
  Based on the Density Functional Theory (DFT) and the Pseudo-potential Approximation

Graphite (2007)
Al-Qasir-Hawari (NSCU)

DFT: Replace the many-electron problem by an exactly equivalent set of self-consistent one-electron equations
Results
Graphite

- Hexagonal structure
- Four atoms per unit cell
- $a=b=2.447\,\text{Å}$
- $c=6.639\,\text{Å}$
- 6x6x1 supercell used
Graphite Dispersion Relations

*Ab initio* vs. Experimental Data
Graphite Heat Capacity

\[ \text{Heat Capacity (J/mol deg)} \]

\[ \text{T(K)} \]

NCSU

DeSorbo, et al., (1953)
NCSU vs. Young-Koppel (YK)

Phonon Frequency distribution (1/eV)

Energy (eV)

- ZO'
- ZA
- ZO
- ZA' + TA
- TO
- LO
Inelastic Scattering Cross Section

\[ T = 300 \text{ K} \]
Scattering Law

\[ S(\alpha, \beta)/\alpha \]

\[ \begin{array}{c|c|c|c|c|c|c|c} \hline 10^{-3} & 10^{-2} & 10^{-1} & 10^0 & 10^1 \\ \hline \text{NCSU} & \text{ENDF/B-VII (YK)} & \text{NWS} & \text{Carvalho (1968)} \\ \hline \end{array} \]

\[ \beta = 0.2 \]

\[ T = 533 \text{K} \]
Cross Section (II)

Coherent One-Phonon
Coherent One-Phonon Contribution

Incoherent Approximation

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

Include Coherent 1ph contribution

$$\frac{d^2 \sigma}{d \Omega \, dE'} \approx \frac{\sigma_{coh}}{4\pi} \frac{k'}{k} \left\{ \sum_{P=2}^{P} S_s(\vec{k}, \omega) \right\}_{\text{Incoh Approx.}} + \left( S_s(\vec{k}, \omega) + S_d(\vec{k}, \omega) \right)_{\text{exact}}$$

Coherent One Phonon Scattering Law

$$1S = 1S_s + 1S_d$$

$$1S(\vec{k}, \omega) = \frac{(2\pi)^3}{2v} \frac{1}{M \mathcal{N}} \sum_{s} \sum_{\omega} \frac{1}{\epsilon_s} \left| \sum_{d} e^{i \vec{k} \cdot \vec{d}} e^{-W_d (\vec{k} \cdot \vec{e}_d)} \right|^2 \times$$

$$\left\{ (n_s + 1) \delta (\omega - \omega_s) \delta (\vec{k} - \vec{q} - \vec{\tau}) + \langle n_s \rangle \delta (\omega + \omega_s) \delta (\vec{k} + \vec{q} - \vec{\tau}) \right\}.$$
Inelastic Scattering Cross Section

Cross Section (b)

Energy (eV)

T=300 K

- NCSU(4ph) (2007)
- NCSU (2007)
- ENDF/B-VII (YK)
- Steyrel (1974)
- Zhou (2006)
Scattering Law

\[
S(\alpha, \beta) / \alpha = 10^{-3} \quad 10^{-2} \quad 10^{-1} \quad 10^0 \quad 10^1
\]

\[\beta = 0.2 \quad T = 533K\]
Scattering Law

\[
S(\alpha, \beta)/\alpha = \begin{cases} 
10^{-1} & \text{for } \alpha = 10^{-2} \\
10^{-2} & \text{for } \alpha = 10^{-1} \\
10^{-3} & \text{for } \alpha = 10^0 \\
10^{-4} & \text{for } \alpha = 10^1 
\end{cases}
\]

\(\beta = 0.2\)

\(T = 533K\)
Conclusions

- The *ab initio* direct approach was used to generate the dispersion relations, and phonon frequency distribution. Excellent agreements were observed between the dispersion relations, and heat capacity with experimental data.

- Some improvement was observed in graphite cross section using the *ab initio* phonon frequency distributions.

- Because graphite is a strong coherent scatterer, the scattering model was reformulated to account for the one phonon coherent scattering. Excellent agreement was found.
Reviewing and Generation of thermal neutron scattering cross sections for different crystalline moderators such as Be, BeO, ZrH$_2$, ZrH$_{1.75}$ and ZrH$_{1.6}$

Generation of thermal neutron scattering cross sections for new materials such as ThH$_2$, CaH$_2$, Bi, Al$_2$O$_3$

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