Thermal Neutron Scattering in Graphite

by

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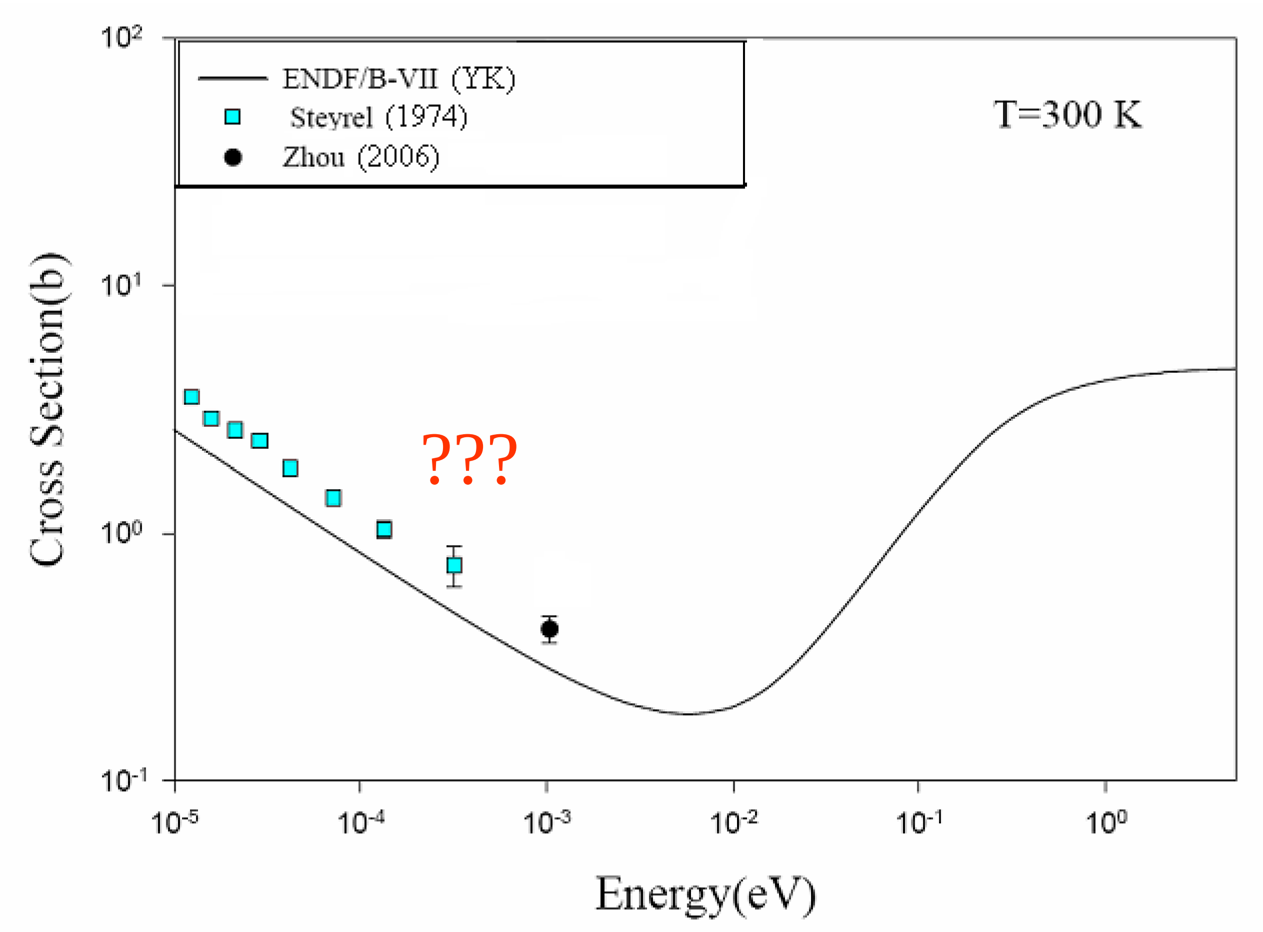
Introduction

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



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 (***ab initio*** approach)
  - The coherent part of the inelastic scattering

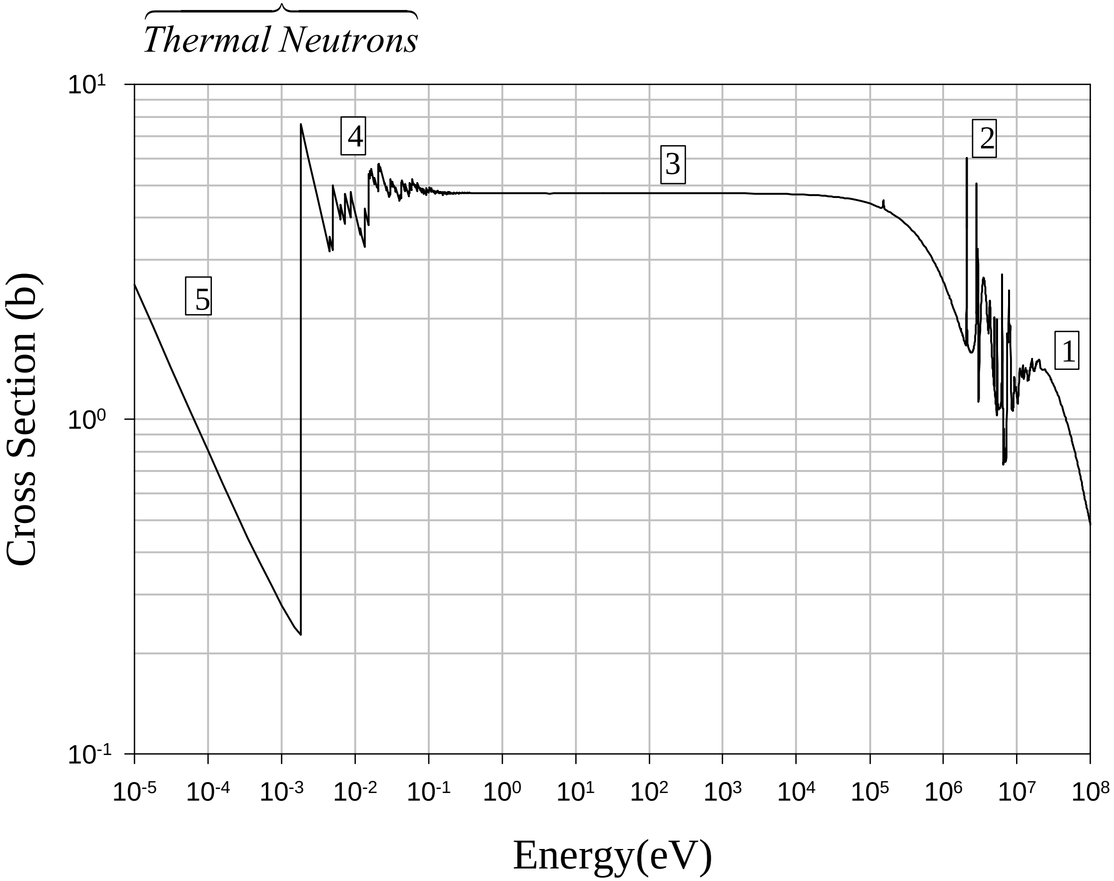
- To Develop and generate new sets of temperature dependent thermal neutron scattering laws

Outline

- Neutron Thermalization
- Lattice Dynamics
- Results
- Conclusions

Neutron Thermalization

Graphite Cross Section



- 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

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

Nuclear Part

Atomic (Dynamical) Part

Graphite → $\sigma_{coh} = 5.50\text{b}$

$\sigma_{incoh} \sim 0\text{b}$

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

Momentum transfer $\alpha = \dfrac{E' + E - 2\mu\sqrt{EE'}}{Ak_B T}$

Energy Transfer $\beta = \dfrac{E' - E}{k_B T}$

Approximations

1- Incoherent approximation:

$$S_d(\alpha,\beta)=0$$

$$\Rightarrow \quad \frac{d^2\sigma}{dE'\,d\Omega} \cong \frac{\sigma_{coh}+\sigma_{incoh}}{4\pi k_B T}\sqrt{\frac{E'}{E}}\,e^{-\beta/2}\sum_{P=1}^{P} S_s(\alpha,\beta)$$

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- Gaussain Approximation

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

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

- In order to calculate the scattering law, all we need is the phonon frequency distribution $\rho(\beta)$ (Lattice Dynamics)

- 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:

Disersion elations: $\omega_j(\vec{q})$

Polarization vectors: $\vec{e}_{dj}(\vec{q})$

Phonon Distribution: $\rho(\omega)$

Phonon Frequency
$\rho(\beta)$

Walter Khon:1998 Nobel Laureate in Chemistry for his development of the Density functional Theory

Lattice dynamics
Force Constants Models

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

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)

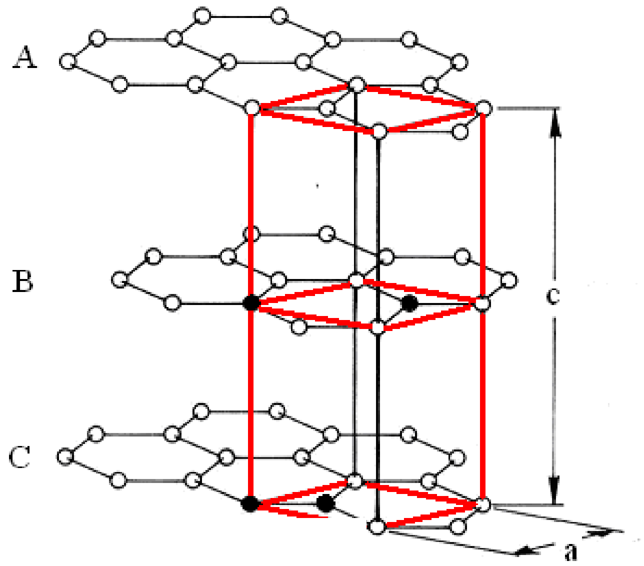
DFT: Replace the many-electron problem by an exactly equivalent set of self-consistent one-electron equations

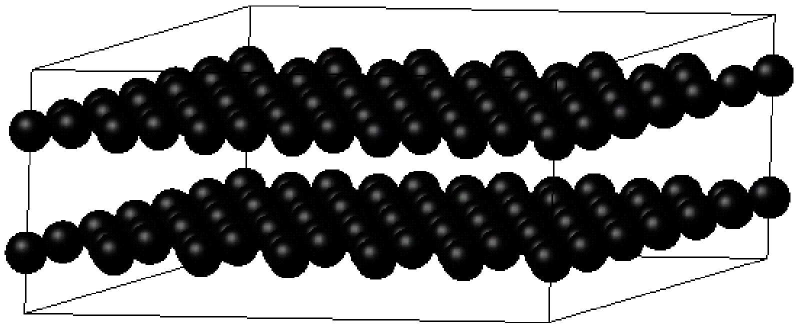
Results

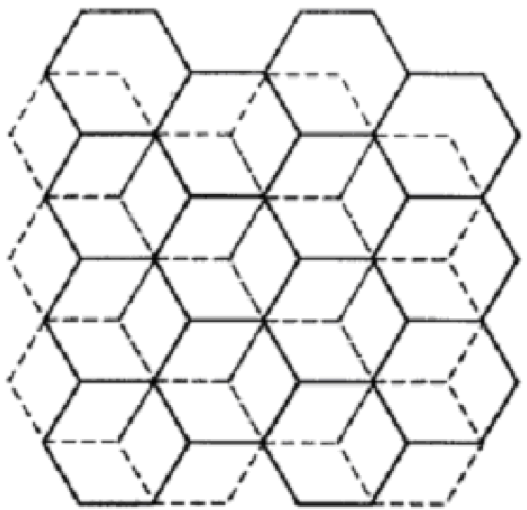
Graphite

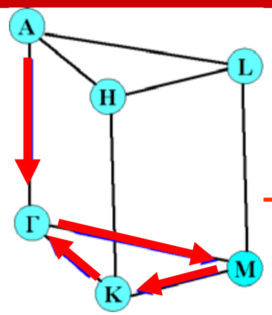
- Hexagonal structure
- Four atoms per unit cell
- a=b=2.447Å

  c= 6.639 Å
- 6x6x1 supercell used

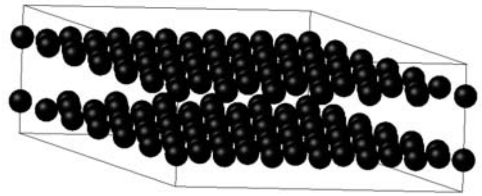


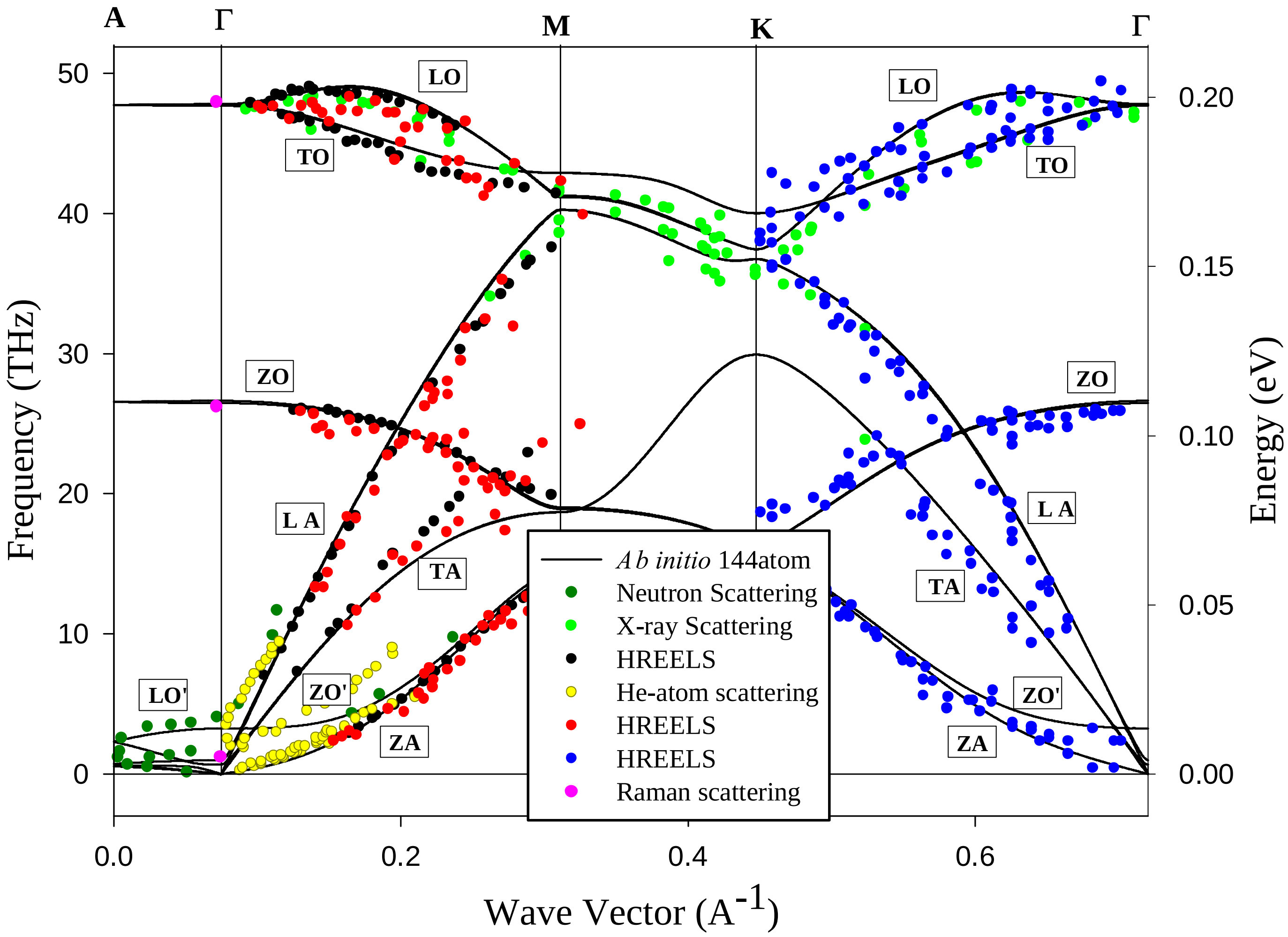




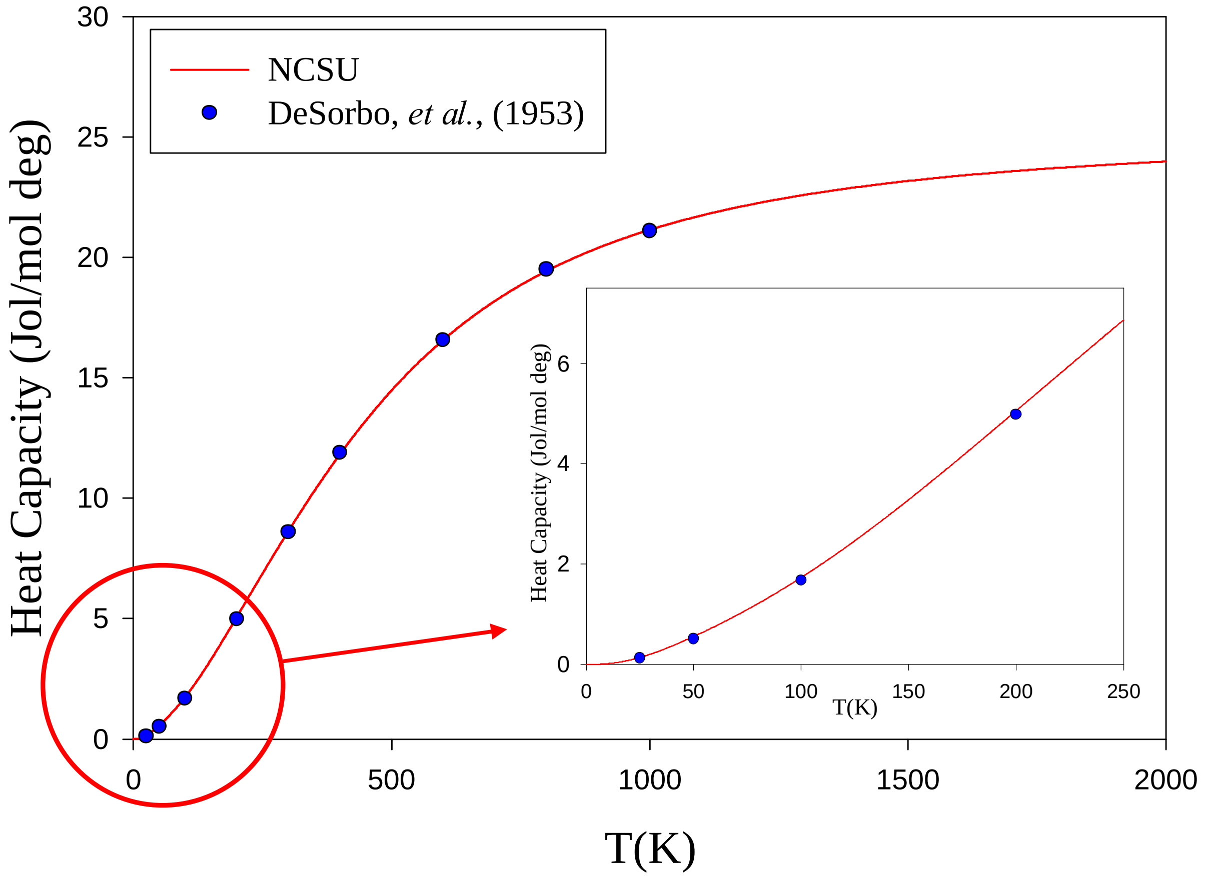


Graphite Dispersion Relations
Ab initio vs. Experimental Data

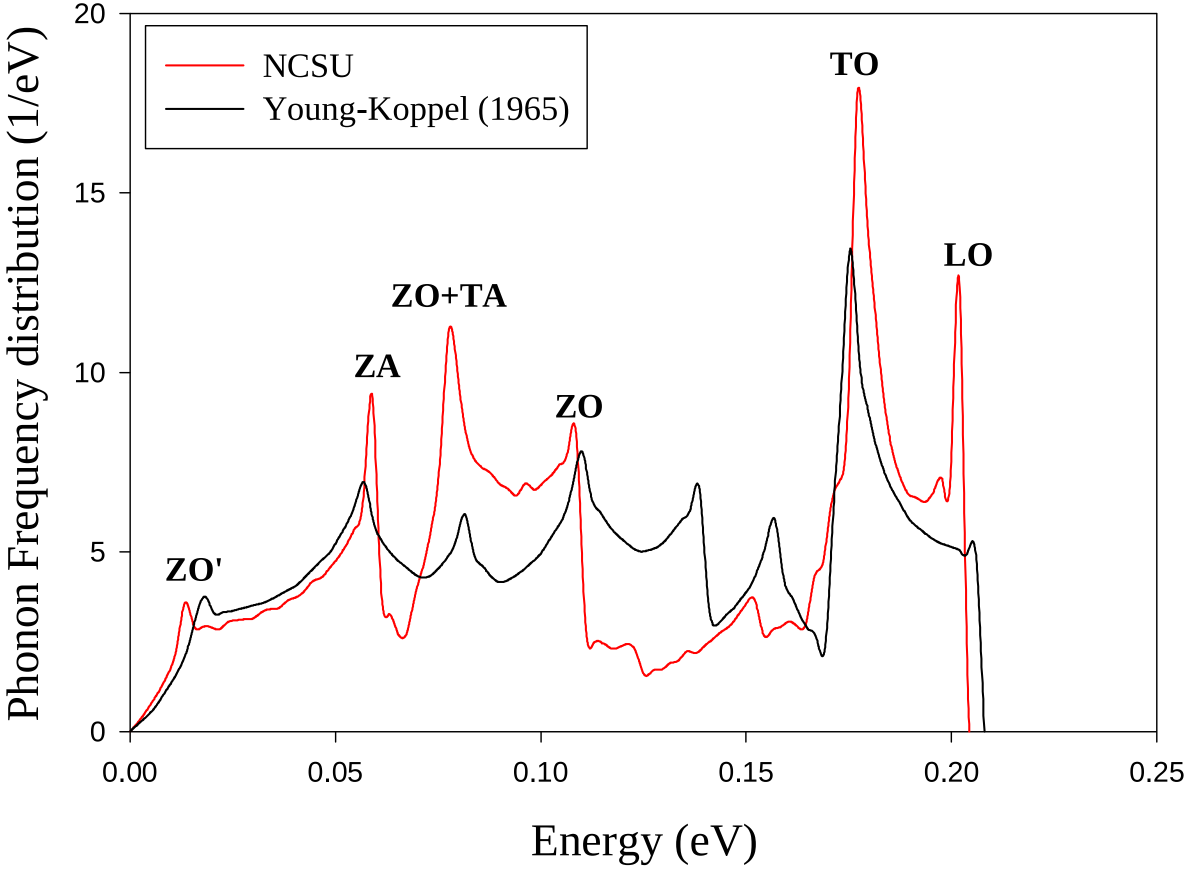




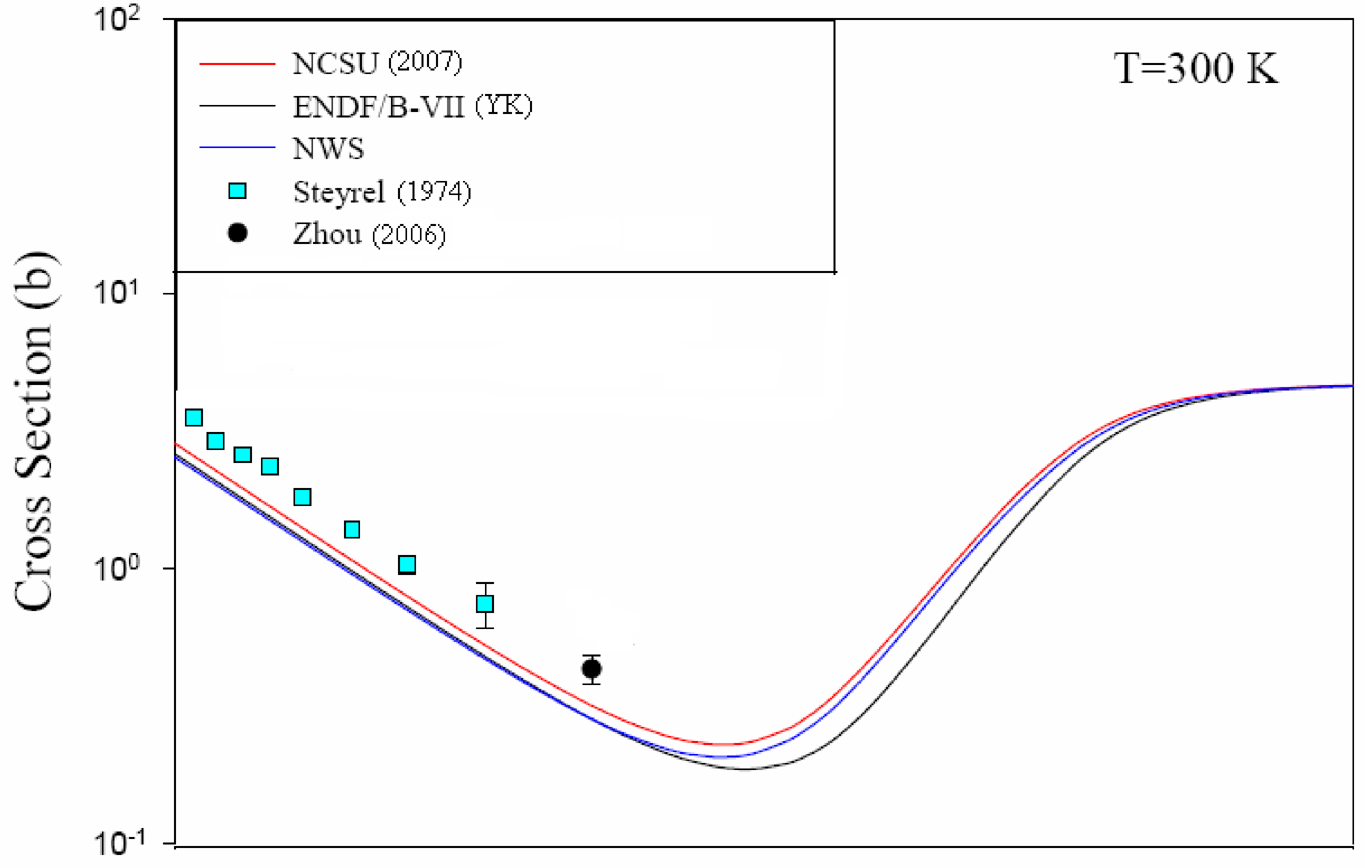
Graphite Heat Capacity



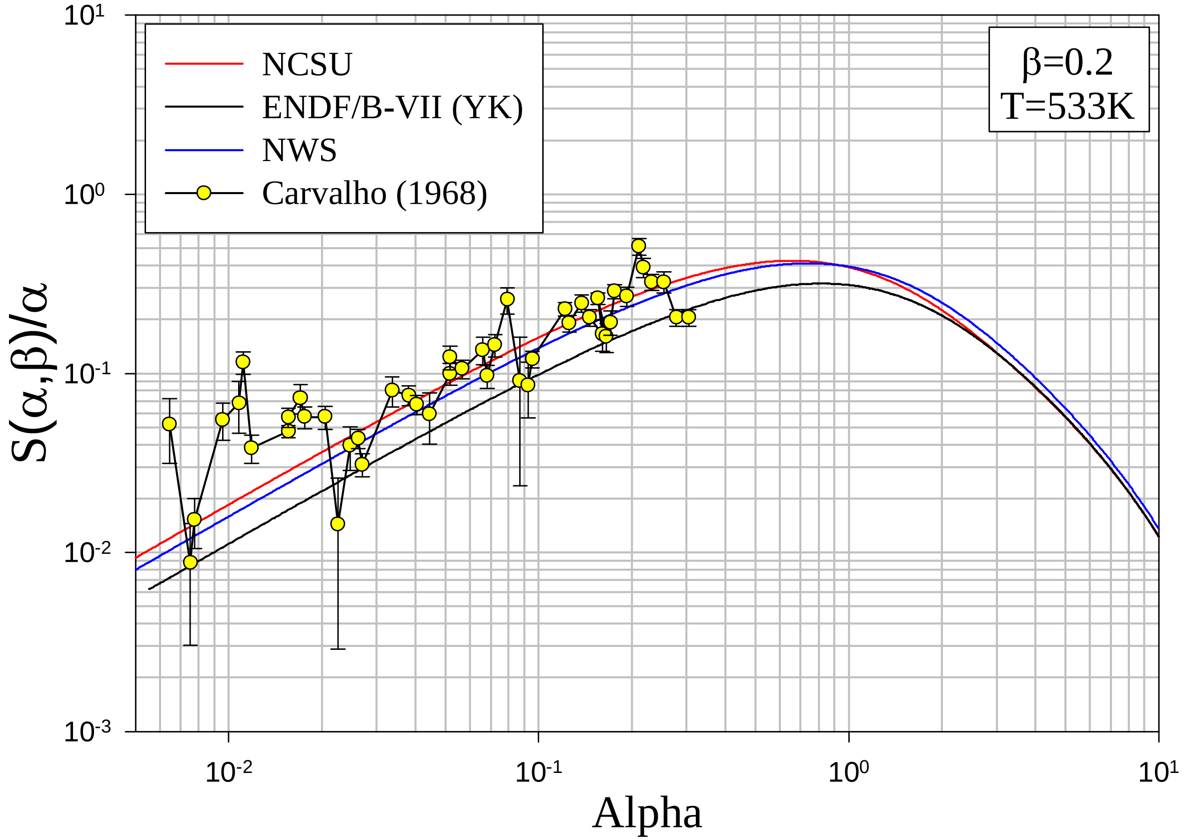
NCSU vs. Young-Koppel (YK)



Inelastic Scattering Cross Section



Scattering Law



Cross Section (II)

Coherent One-Phonon

Coherent One-Phonon Contribution

Incoherent Approximation

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

X

Include Coherent 1ph contribution

$$\frac{d^2\sigma}{d\Omega\, dE'} \cong \frac{\sigma_{coh}}{4\pi}\frac{k'}{k}\left\{\left(\sum_{P=2} {}^{P}S_s(\vec{\kappa},\omega)\right)_{Incoh\,Approx.} + \left({}^{1}S_s(\vec{\kappa},\omega) + {}^{1}S_d(\vec{\kappa},\omega)\right)_{exact}\right\}$$

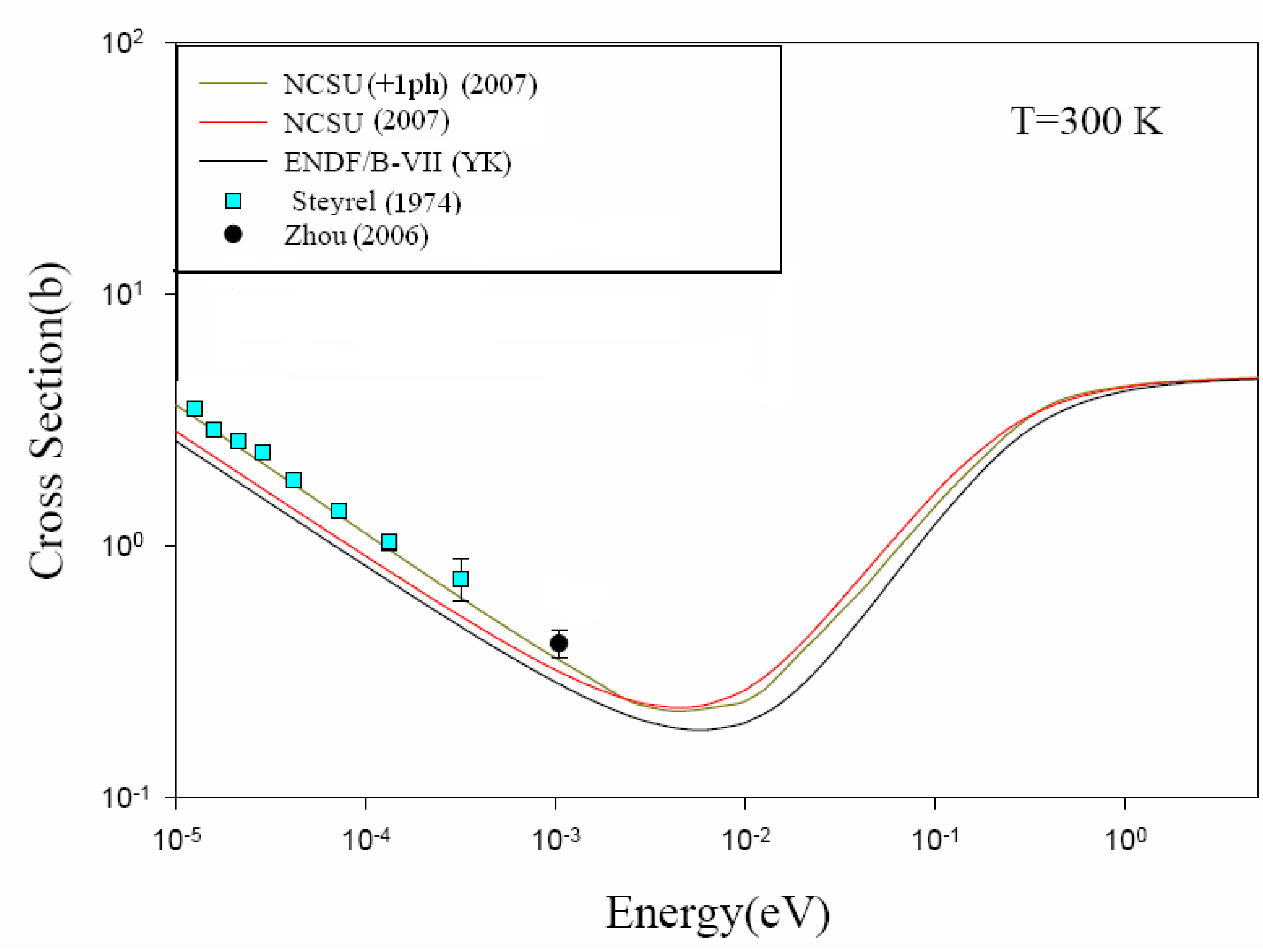
$${}^{1}S = {}^{1}S_s + {}^{1}S_d$$

Coherent One Phonon Scattering Law

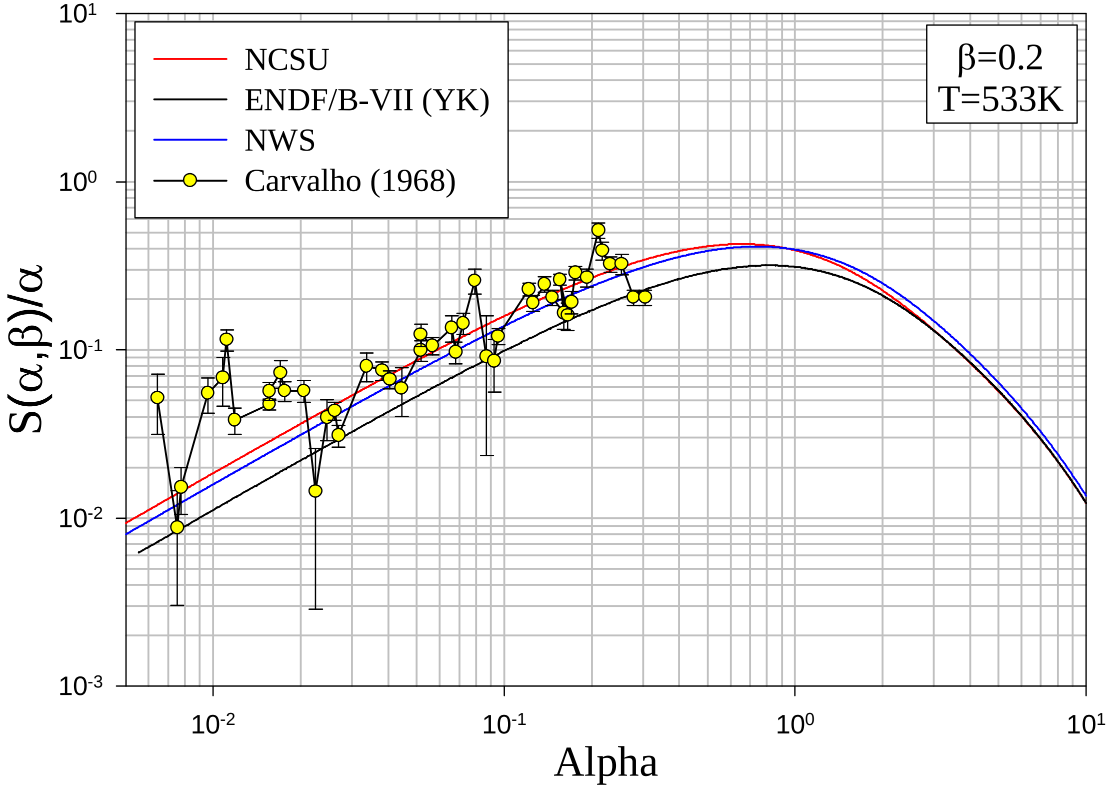
$${}^{1}S(\vec{\kappa},\omega) = \frac{(2\pi)^3}{2\,v}\frac{1}{M\mathcal{N}}\sum_s\sum_{\vec{\tau}}\frac{1}{\omega_s}\left|\sum_d e^{i\vec{\kappa}\cdot(\vec{d})} e^{-W_d}\left(\vec{\kappa}\cdot\vec{e}_{ds}\right)^2\right|^2 \times$$

$$\left\{\langle n_s+1\rangle \delta(\omega-\omega_s)\delta(\vec{\kappa}-\vec{q}-\vec{\tau}) + \langle n_s\rangle \delta(\omega+\omega_s)\delta(\vec{\kappa}+\vec{q}-\vec{\tau})\right\}.$$

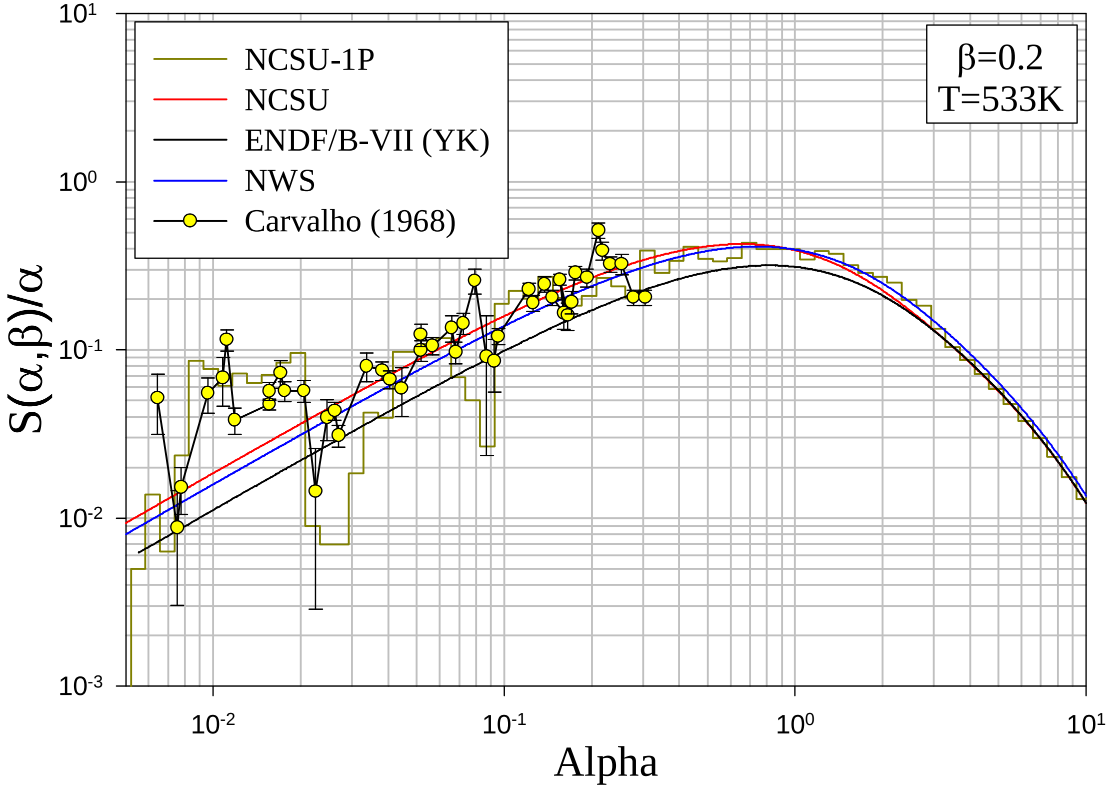
Inelastic Scattering Cross Section



Scattering Law



Scattering Law



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

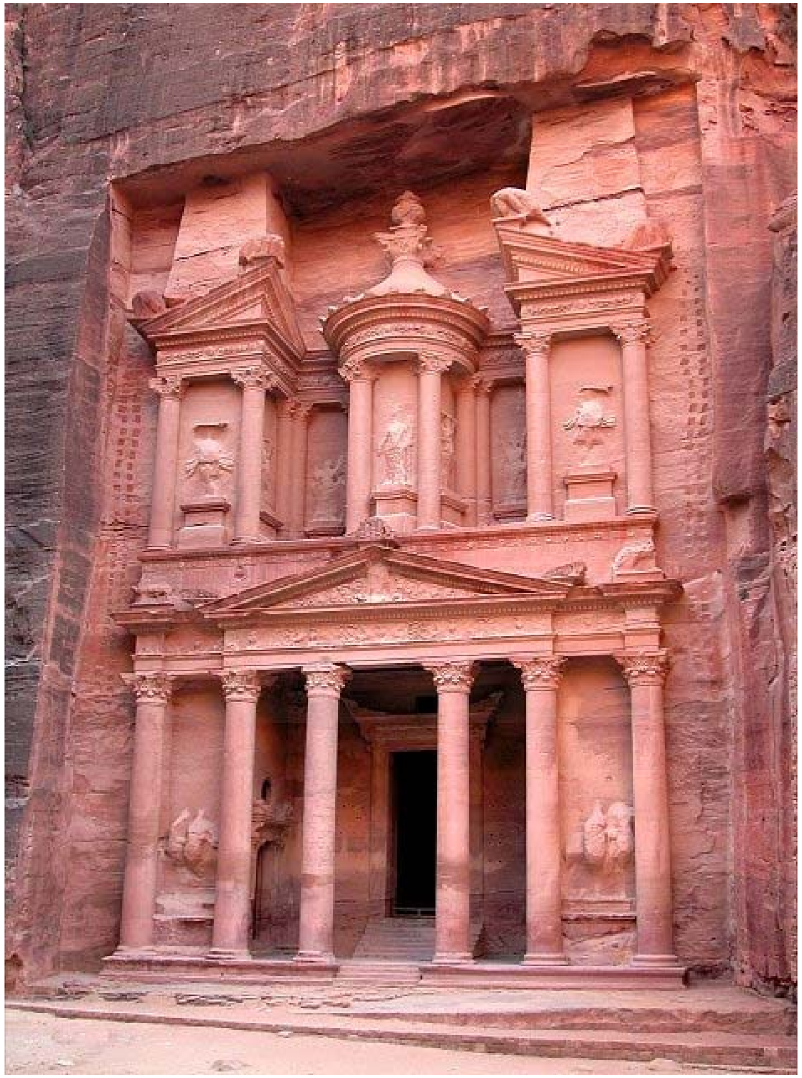
- Reviewing and Generation of thermal neutron scattering cross sections for different crystalline moderators such as Be, BeO, ZrH_2, $ZrH_{1.75}$ and $ZrH_{1.6}$

- Generation of thermal neutron scattering cross sections for new materials such as ThH_2, CaH_2, Bi, Al_2O_3

- Investigation of radiation effect on thermal neutron scattering in graphite "Impact of Simple Carbon Interstitial formations on Thermal Neutron

  Scattering in Graphite" Hawari, A. I., A. I. , **Al-Qasir, I. I,** and Ougouag, A. M, Nucl. Sci. Eng. 155, 449-462 (**2007**)

Welcome to Jordan



http://www.pbase.com/mansour_mouasher/