VIETNAM ATOMIC ENERGY INSTITUTE
DALAT NUCLEAR RESEARCH INSTITUTE
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STUDY OF PHONON DENSITY OF STATES IN GRAPHITE USING DENSITY FUNCTIONAL THEORY

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TABLE OF CONTENTS

ABSTRACT

Aim & Scope

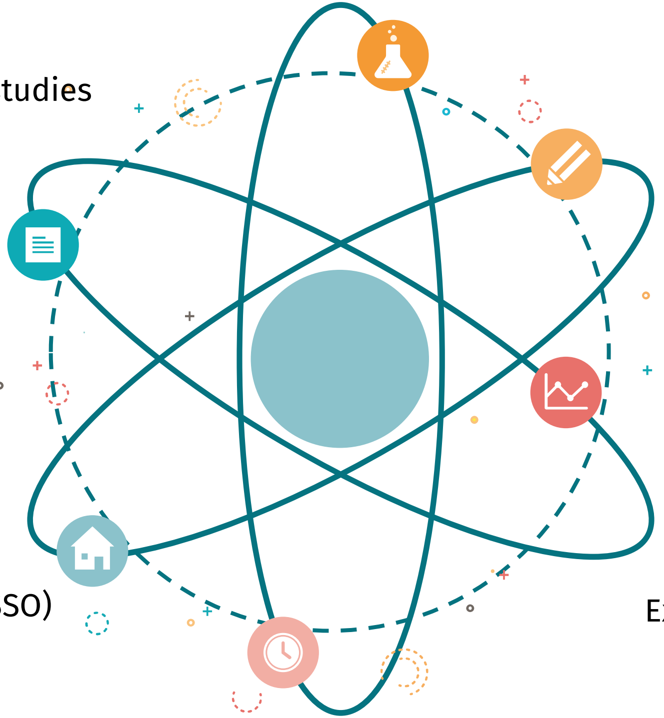
Compute graphite PhDOS for further studies on neutron interaction cross-sections

Research objects

Crystalline graphite (hexagonal), representative supercell and Brillouin zone sampling at 0 K.

Methods

First-principles DFT (Quantum ESPRESSO)
→ Phonon dispersion / PhDOS



Outputs

Phonon spectrum as a probability density function

Results

Physically consistent PhDOS for graphite; parameters suitable as initial data for TSL $S(\alpha,\beta)$.

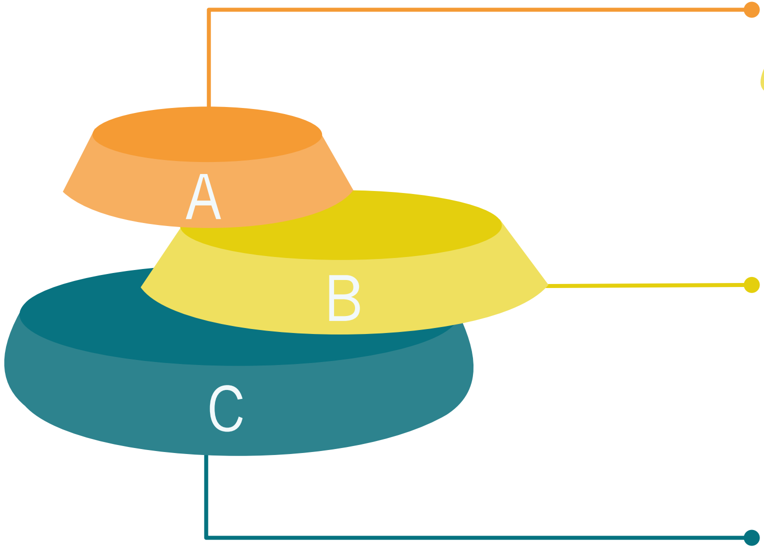
Impact

Expect improved thermal neutron cross-section accuracy

INTRODUCTION

Overview, objective, and scope of this work

FROM LATTICE DYNAMICS TO THERMAL NEUTRON SCATTERING IN GRAPHITE



Neutron Cross-sections

Accurate nuclear cross sections are foundational to reliable reactor modeling and safety assessment.

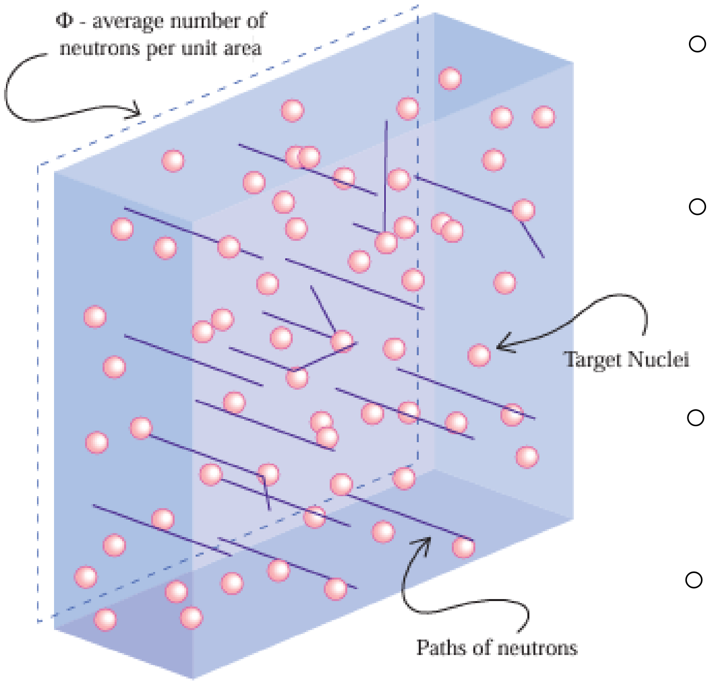
Thermal Scattering Law (TSL)

Thermal neutron interactions are governed by the thermal scattering law $S(\alpha,\beta)$, which depends on the phonon spectrum of the moderator.

Phonon Density Of State (PhDOS)

The PhDOS provides critical insights into vibrational dynamics and is a foundation for calculating the TSL

Why are cross sections important?



- Cross sections are extremely important to reactor design and control.
- They determine fuel usage, arrangement, moderator spacing, control rod design, and are vital for neutron filters and nuclear data analysis.
- Neutron cross-sections often require specialized instrumentation and sample preparation.
- Theoretical calculations using tabulated elemental or isotopic data are essential.

NEUTRON REACTION CROSS SECTIONS (THERMAL REGIME)

- In solids, thermal neutron scattering depends strongly on atomic structure and lattice dynamics (phonons).
- The total thermal cross section σ_{th} is composed of:

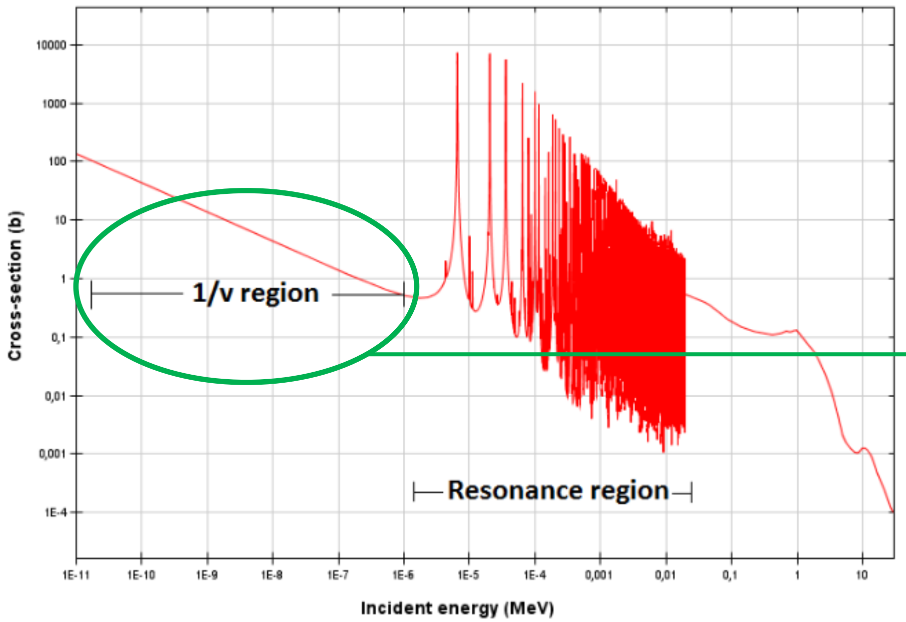
$$\sigma_{th} = \sigma_a + \sigma_{inel} + \sigma_{ela}$$

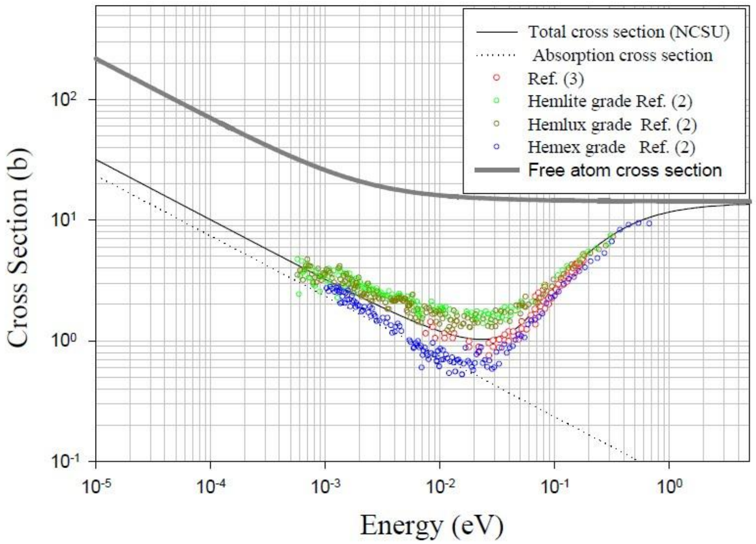
Where:

- σ_a is the absorption cross section - proportional to the neutron wavelength
- σ_{inel} is the inelastic-scattering cross section - depends on the crystal temperature (phonon distribution)
- σ_{ela} is elastic-scattering cross section (coherent Bragg + incoherent) - depends on wavelength, crystal orientation, and crystal perfection; coherent part vanishes below the first Bragg edge.

- In the thermal range, σ_{ela} and σ_{inel} are governed by the thermal scattering law $S(\alpha,\beta)$

Incident neutron data / ENDF/B-VII.1 / U238 / MT=102 : (z,γ) / Cross section





- Above ~1 eV, total cross section of many crystals is only a few barns.
- Below ~0.1 eV (Bragg mostly forbidden, λ ~ interatomic spacing), the single-crystal effective cross section is strongly reduced.

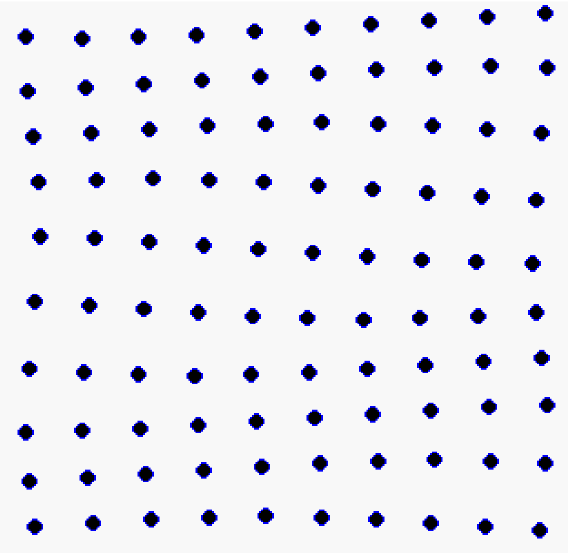
A. I. Hawari, I. I. Al-Qasir, K. K. Mishra, PHYSOR-2006, A146 (2006)

ATOMIC VIBRATIONS IN SOLIDS: PHONONS

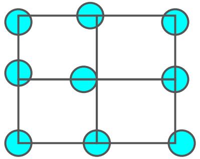
- Each type of crystal is characterized by a specific pattern of lattice vibrations known as its phonon spectrum.

- The phonon spectrum largely determines the important properties of solids, such as heat capacity, thermal conductivity, and thermal expansion coefficient,...

- In a solid, atoms vibrate collectively around their equilibrium positions. These quantized lattice vibrations are called phonons - the quantum mechanical analog of normal modes of vibration.



- Thermal Diffuse Scattering (TDS)



➢ The Thermal Scattering Law (TSL) serves as the theoretical framework for understanding Thermal Diffuse Scattering (TDS).

THERMAL SCATTERING LAW

- The double-differential inelastic scattering cross section can be written as

$$\frac{\partial^2 \sigma}{\partial \Omega dE'} = \frac{\sigma_b}{4\pi k_B T} \sqrt{\frac{E'}{E}} S(\alpha, \beta)$$

Thermal scattering law

Where:

- *E* and *E'* are the incident and outgoing neutron energies
- σ_b is the bound scattering cross section (material-dependent), with $\sigma_b = \sigma_{coh} + \sigma_{inc}$.

- The thermal scattering law $S(\alpha,\beta)$ is defined as:

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

Here:

- α: dimensionless momentum transfer
- β: dimensionless energy transfer.

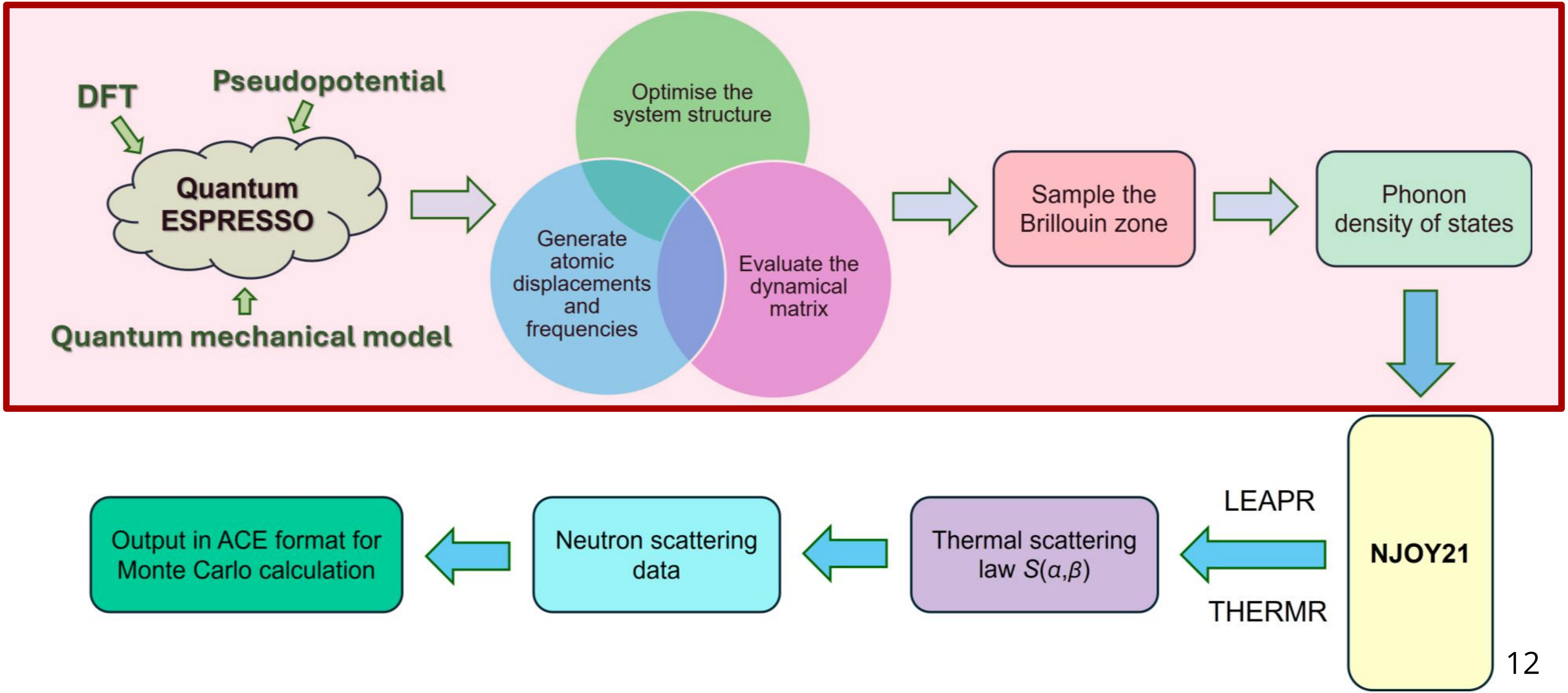
➢ In the thermal range, the **elastic (coherent)** and **inelastic** scattering components and thus the **effective cross sections** are governed by $S(\alpha,\beta)$ which encodes the lattice dynamics (phonons).

Phonon Density of States (PhDOS) - Foundation for $S(\alpha,\beta)$

The Phonon Density of States (PhDOS) describes how vibrational modes are distributed with frequency in a solid. It is a fundamental quantity linking atomic vibrations (phonons) to macroscopic thermodynamic and scattering properties, including the calculation of the thermal scattering law for neutron transport simulations.

First-principles calculations were performed using the Quantum ESPRESSO suite to compute the phonon dispersion and phonon density of states (PhDOS).

Workflow linking first-principles phonon calculations with NJOY processing for $S(\alpha,\beta)$ generation

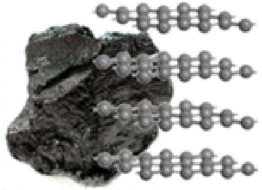


RESEARCH SUBJECTS AND METHODOLOGY

Research objects, data, and methods

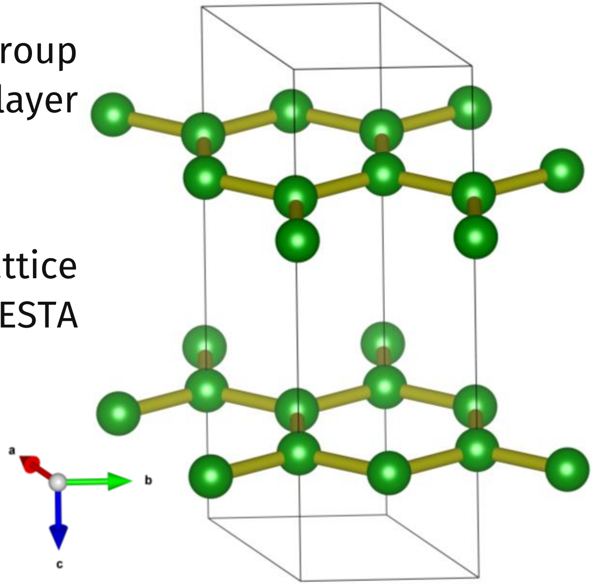
GRAPHITE

- Graphite is an important reactor material, used as a moderator and reflector due to its low neutron absorption, strong scattering, and high thermal stability.



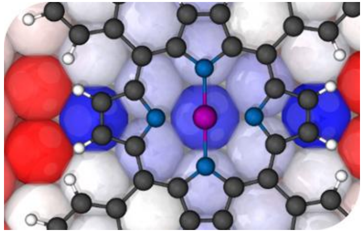
- Graphite crystallizes in a hexagonal structure (space group $P6_3/mmc$) with highly anisotropic bonding and weak interlayer van der Waals interactions.

- The crystal structure with cell formula C_4, including lattice parameters ($a \approx 2.46$ Å, $c \approx 6.70$ Å), was visualized using the VESTA software to verify atomic arrangement and symmetry.



QUANTUM ESPRESSO

QUANTUM ESPRESSO is an integrated suite of computer codes for electronic-structure calculations and materials modeling, based on density-functional theory, plane waves, and pseudopotentials (norm-conserving, ultrasoft, and projector-augmented wave)



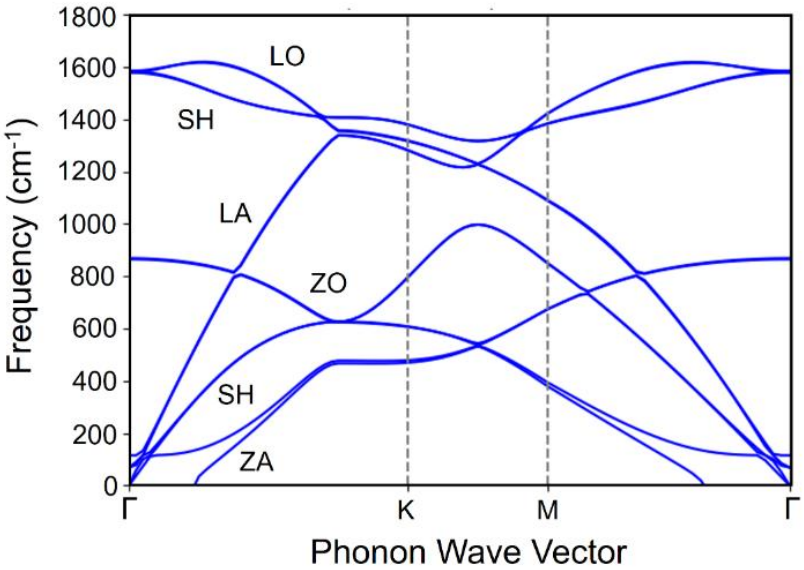
- *Ab initio* lattice dynamics calculations are computational methods that use first-principles quantum mechanics to study the atomic vibrations in a crystal.
- By calculating the interatomic forces based on the electronic structure (using techniques like Density Functional Theory), these methods can determine phonon frequencies

In general, the first step in establishing the phonon frequency spectrum for crystalline materials is to set up a dynamical matrix based on the crystal structure.

RESULTS AND DISCUSSION

Key results with interpretation

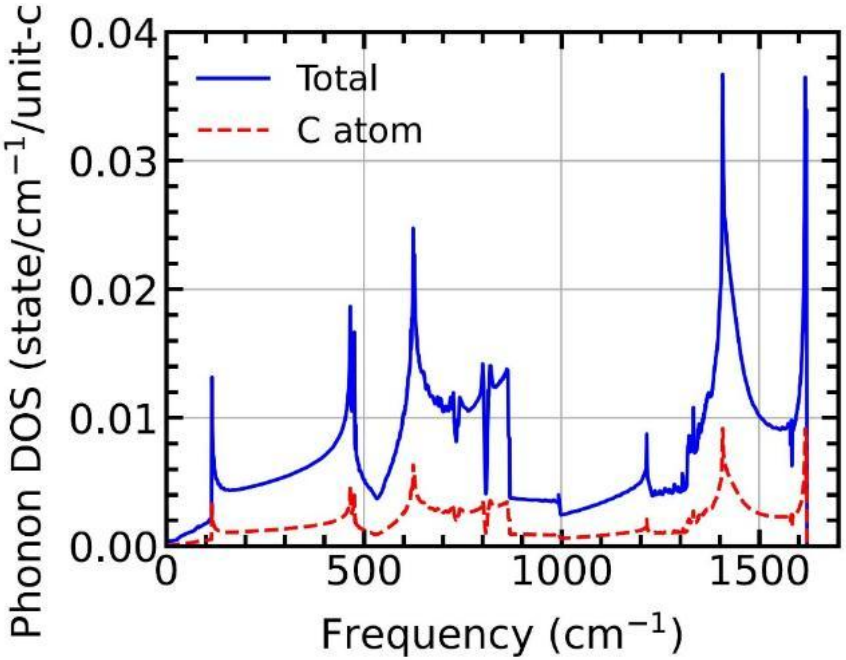
PHONON DISPERSION DIAGRAM



The phonon dispersion diagram shows key vibrational features. The high-energy longitudinal optical (LO) branch displays over-bending between Γ and M points, reflecting phonon-phonon interactions. The shear horizontal (SH) branch demonstrates theoretical inconsistencies, as the expected crossing with the zone optical (ZO) branch between the Γ and M points is not consistently observed across all ab initio calculations. These discrepancies suggest limitations in current theoretical models and the need for further refinement. Additionally, the out-of-plane acoustic (ZA) branch shows quadratic dispersion near Γ, indicative of weak interlayer bonding in graphite.

➢ These dispersion relations provide the foundation for analyzing the vibrational spectrum of graphite. Correspondingly, the PhDOS characterizes the distribution of these vibrational modes over energy.

PHONON DENSITY OF STATES



The PhDOS characterizes graphite vibrations. The total PhDOS (solid blue) is reported per primitive unit cell, and the partial density of states (PDOS, dashed red) is the site-averaged contribution per carbon atom. With this normalization, summing the per-atom PDOS over the four atoms in the cell gives the total. Low-frequency peaks arise from interlayer shear and breathing motions, whereas high-frequency peaks correspond to in-plane optical modes near 1580 cm^{-1}.

- These features underpin thermal transport and mechanical response in graphite

CONCLUSIONS

Key findings and future work

CONCLUSION

- This study employs the open-source Quantum ESPRESSO package to investigate the PhDOS and phonon dispersion of crystalline graphite using the package's density functional theory (DFT) option.
- The results provide detailed vibrational properties of graphite, highlighting its complex phonon behavior.
- The calculated phonon dispersion emphasizes the anisotropic vibrational characteristics of the material, while the PhDOS reveals dominant in-plane vibrational contributions.
- These results provide partial information for analyses of their influence on thermal neutron inelastic scattering in graphite.
- They provide preparatory data for constructing the thermal scattering law $S(\alpha,\beta)$ and may also be used as input to NJOY processing to generate ACE-formatted data libraries for Monte Carlo neutron transport simulations.

Thanks you for your attention!

Do you have any questions?

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