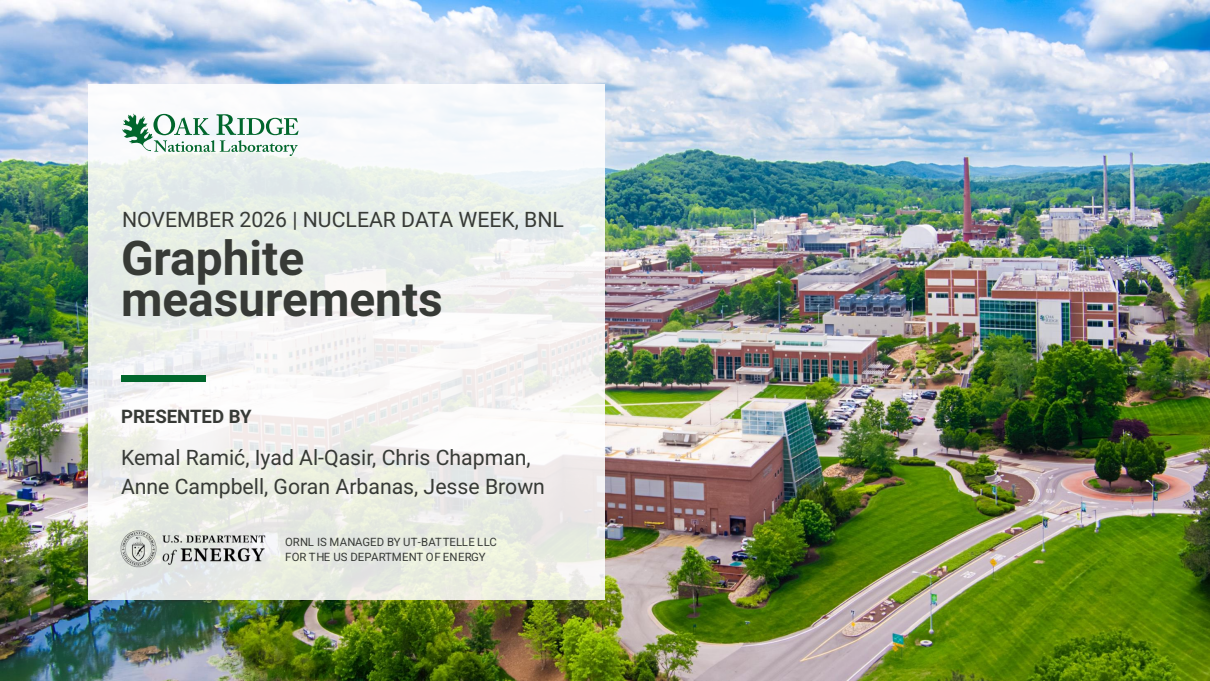


OAK RIDGE National Laboratory

NOVEMBER 2026 | NUCLEAR DATA WEEK, BNL

Graphite measurements

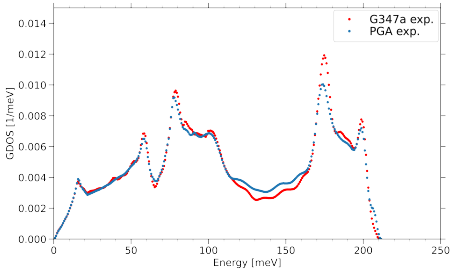
PRESENTED BY

Kemal Ramić, Iyad Al-Qasir, Chris Chapman, Anne Campbell, Goran Arbanas, Jesse Brown

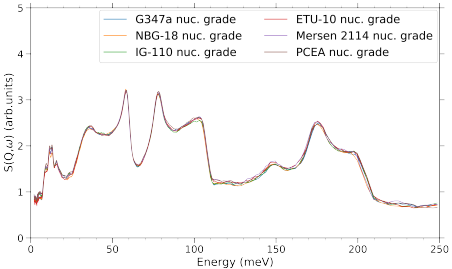
U.S. DEPARTMENT of ENERGY

ORNL IS MANAGED BY UT-BATTELLE LLC FOR THE US DEPARTMENT OF ENERGY

Inelastic neutron spectra are nearly identical across nuclear graphite grades



ARCS · SNS Phonon density of states, 2 graphite grades



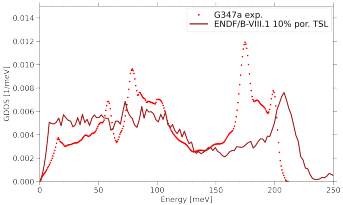
VISION · SNS S(Q, ω), 6 graphite grades, 5 K

Peak positions and intensities are nearly the same in every grade, although the grades differ in porosity. **Experimentally, porosity is not visible in the inelastic neutron spectrum.**

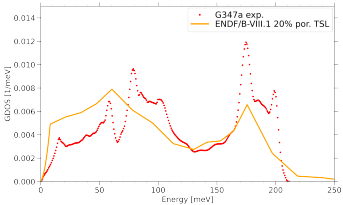
References

VISION: Ramić et al., *Carbon* (2025) · doi:10.1016/j.carbon.2025.120619
ARCS: Al-Qasir et al., *Carbon* 167 (2020) · doi:10.1016/j.carbon.2020.05.027
ARCS: Al-Qasir et al., *Ann. Nucl. Energy* 161 (2021) · doi:10.1016/j.anucene.2021.108437

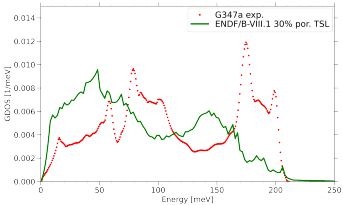
The 10, 20 and 30 % porosity TSLs use phonon spectra that no measurement shows



10 % porosity TSL



20 % porosity TSL

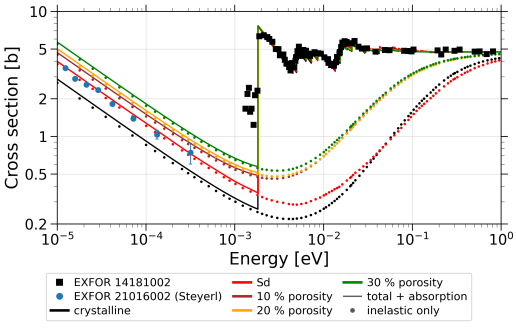


30 % porosity TSL

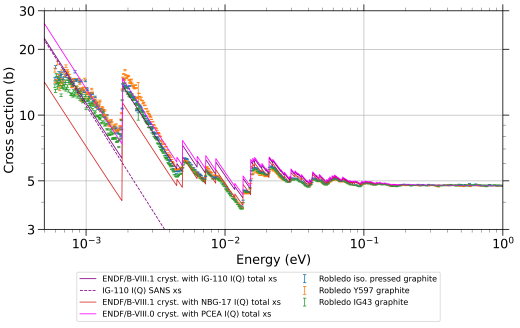
Is the difference from the measurement physics, or methodology?

It's the method! doi:10.1016/j.carbon.2025.120619

Pores show up as small-angle scattering, an elastic effect



Total cross section TSLs alone vs. measured



Crystalline TSL + measured SANS vs. three measured grades

Below 1.8 meV, every TSL falls below the measurement (left). The excess is **small-angle neutron scattering (SANS)** from the pores: adding a measured SANS curve to the crystalline TSL reproduces the measured cross sections (right) without a porosity TSL.

The distinction that matters

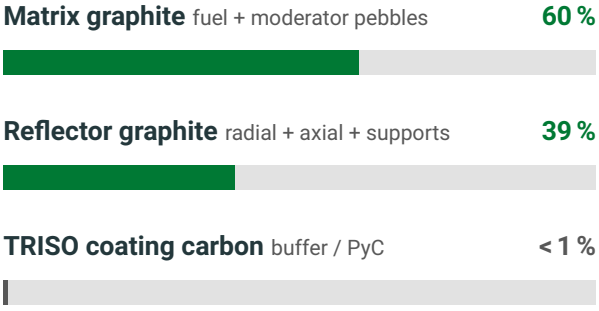
SANS is elastic: it changes the neutron's direction, not its energy, so it does not affect thermalization. It depends on pore size and distribution, not on a single porosity percentage.

Matching the porosity TSL to the real porosity never wins

Benchmark (porosity)	Crystalline	Sd	10 %	20 %	30 %	Best fit
Group A · graphite moderated & reflected						
HCT-016 (24–27 %)	**0.38**	0.39	0.77	1.00	1.24	Crystalline
LCT-060 (23–25 %)	**0.29**	0.31	0.35	0.43	0.73	Crystalline
Group B · TRISO / coated-particle						
ICT-010 / HTR-10 (22–30 %)	**3.36**	3.81	7.57	10.19	12.75	Crystalline
HTTR (22–23.5 %)	3.40	**3.35**	4.60	5.49	6.40	Sd
PROTEUS (22–23.5 %)	1.72	1.57	**0.65**	0.66	0.94	10 %
ICT-008 / ASTRA (25–27 %)	0.66	—	**0.43**	0.91	1.60	10 %

Reduced χ^2 of calculated vs. measured k_{eff} over each benchmark's cases (lower is better). Crystalline or Sd fits best in four of six benchmarks. Where a porosity TSL fits best, it is the **10 %** TSL, never one matching the 22–30 % porosity of the benchmark graphite.

Where k_{eff} is sensitive to graphite: matrix and reflector



PROTEUS core 1 · TSUNAMI-3D sensitivity (SCALE)

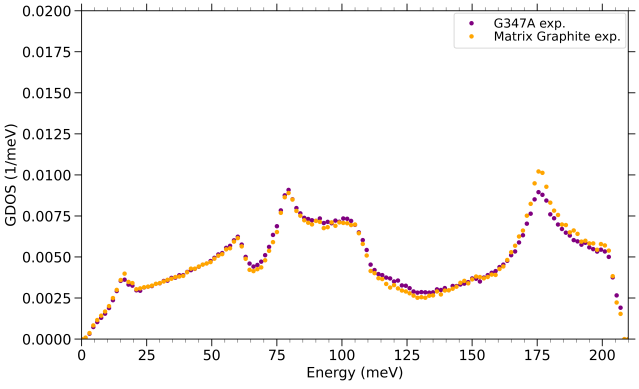
Share of the k_{eff} sensitivity to the carbon total cross section, summed over the regions of each type. TSUNAMI-3D, ENDF/B-VIII.0.

Why it matters

Nearly all of the k_{eff} sensitivity to graphite comes from graphite that would use the porosity TSL. The TRISO coating carbon contributes less than 1 %.

The TSL chosen for the matrix and reflector graphite therefore controls the graphite effect on k_{eff}. The next slide shows a measurement of the matrix graphite.

Matrix graphite has the same phonon spectrum as nuclear graphite



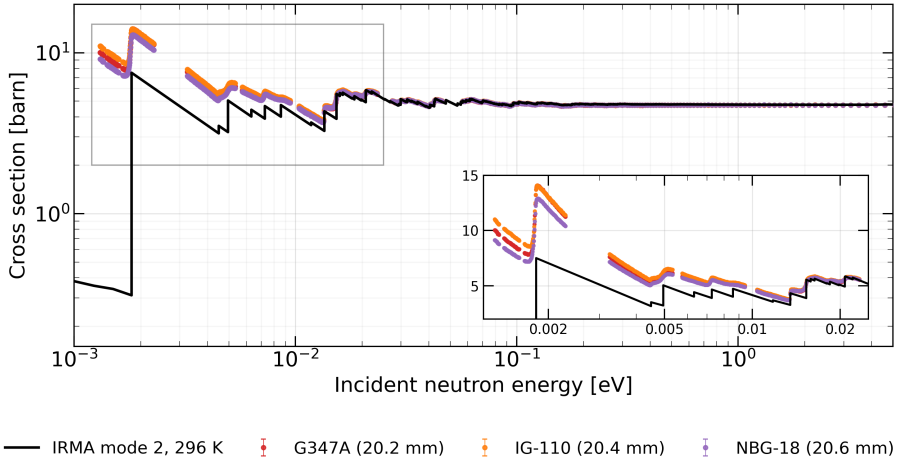
ARCS · SNS

Phonon DOS: fuel-matrix graphite vs. G347A nuclear graphite

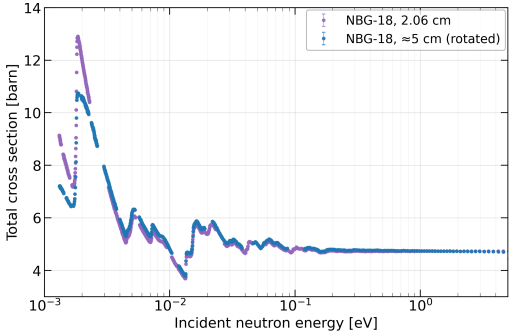
The graphite inside fuel and moderator pebbles is a different grade from reactor block graphite and is made for a different purpose.

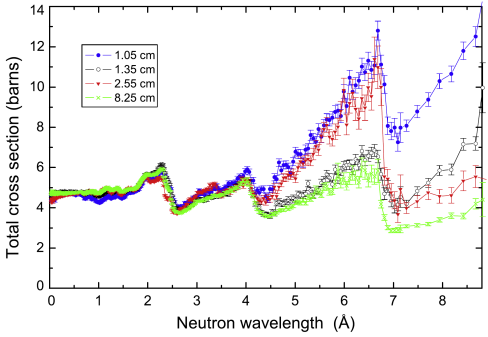
Measured on ARCS, its phonon spectrum is **nearly identical** to that of G347A nuclear graphite.

Total cross sections of three nuclear graphite grades at 20 °C



The cross section dependendance on sample thickness



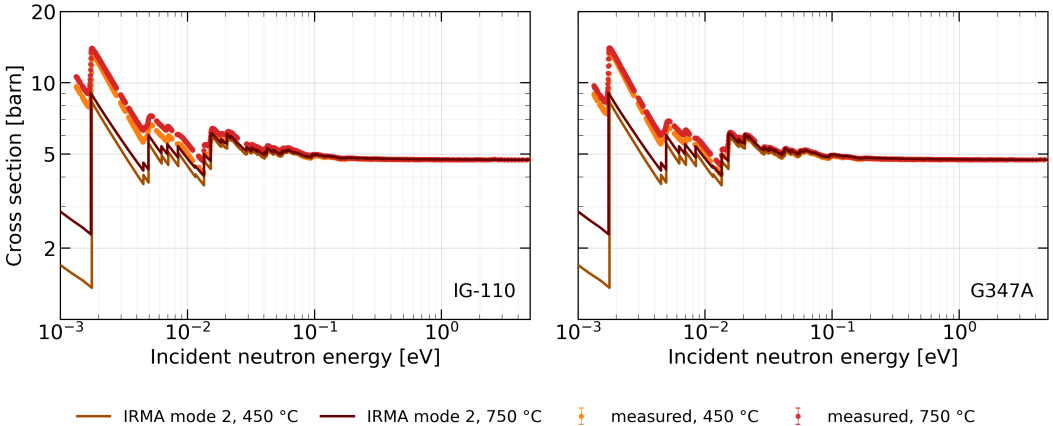


VENUS · SNS NBG-18, 20 °C, 2.06 cm and ≈5 cm thick **Petriw et al. (2010), Fig. 8** Needle-coke graphite, four thicknesses

In both measurements, the thicker sample gives a lower total cross section below the (002) Bragg edge (1.8 meV, 6.7 Å).

Petriw, Dawidowski, Santisteban, *J. Nucl. Mater.* 396 (2010) 181–188 · doi:10.1016/j.jnucmat.2009.11.003

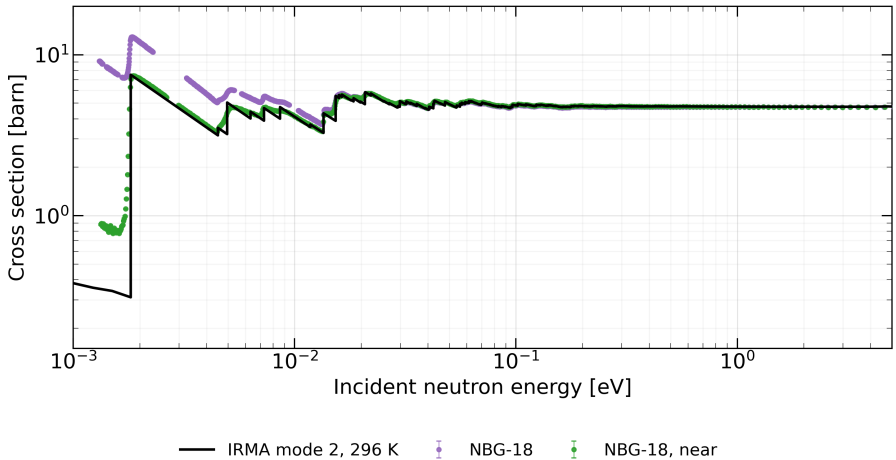
Total cross sections of IG-110 and G347A at 450 and 750 °C



VENUS · SNS

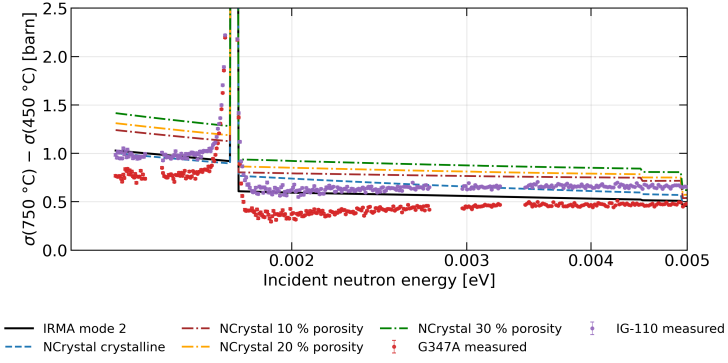
Total cross section, IG-110 and G347A, 450 and 750 °C

NBG-18 at 20 °C, sample 2 cm and 109 cm from the detector



Total cross section, NBG-18, 20 °C; sample 2 cm (near) or 109 cm from the detector

Porosity models **overpredict** the change from 450 to 750 °C



VENUS · SNS

Total cross section difference, G347A and IG-110; models with the measured lattice constants

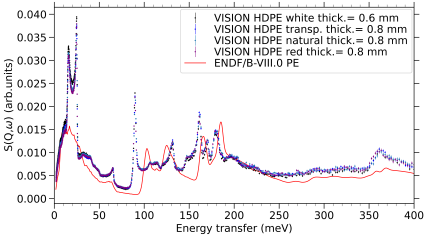
If SANS does not depend on temperature, it cancels in σ(750 °C) − σ(450 °C), leaving the temperature dependence of the thermal scattering model. **Porosity TSLs predict a larger difference than either grade shows; the crystalline models come closer.**

Porosity shows up as SANS, not in the phonon spectrum

- The measured phonon spectra (ARCS, VISION) are nearly the same in nuclear graphite grades of different porosity and in fuel-matrix graphite.
- The 10, 20 and 30 % porosity TSLs use phonon spectra that no measurement shows. In six criticality benchmarks, the porosity TSL that matches the benchmark porosity never fits the measured k_{eff} best.
- The low-energy excess in the measured total cross section is small-angle neutron scattering (SANS) from the pores; the crystalline TSL plus a measured SANS curve reproduces it.
- The measured low-energy cross section depends on the sample thickness and on the distance between the sample and the detector.
- With SANS cancelled in σ(750 °C) − σ(450 °C), the porosity models predict a larger temperature change than either measured grade; the crystalline models come closer.

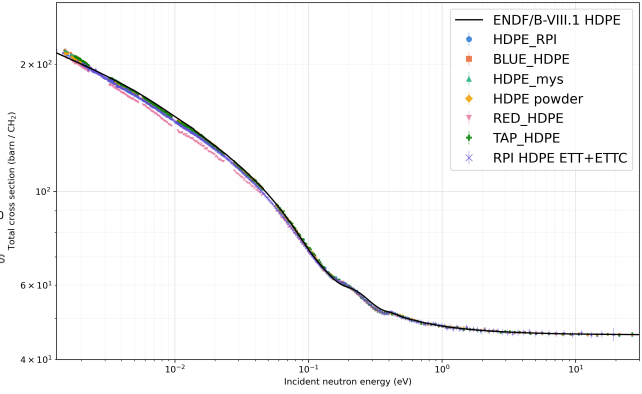
This work was performed under the framework of the DOE/US Nuclear Regulatory Commission collaboration for Criticality Safety support for the commercial-scale HALEU fuel cycles project (DNCSH). This research was supported by the Nuclear Criticality Safety Program, funded and managed by the National Nuclear Security Administration for the Department of Energy.

HDPE measurements



VISION · SNS

S(Q, ω), four HDPE samples



VENUS · SNS

Total cross section, HDPE samples

- Updated HDPE evaluation from ORNL is coming!

OAK RIDGE National Laboratory

November 5, 2026 | Nuclear Data Week| BNL, NY

Part I- Graphite Phonons at High Temperatures: X-Ray Diffraction, Inelastic Neutron Scattering, and Atomistic Simulations

Presented by

Iyad Al-Qasir

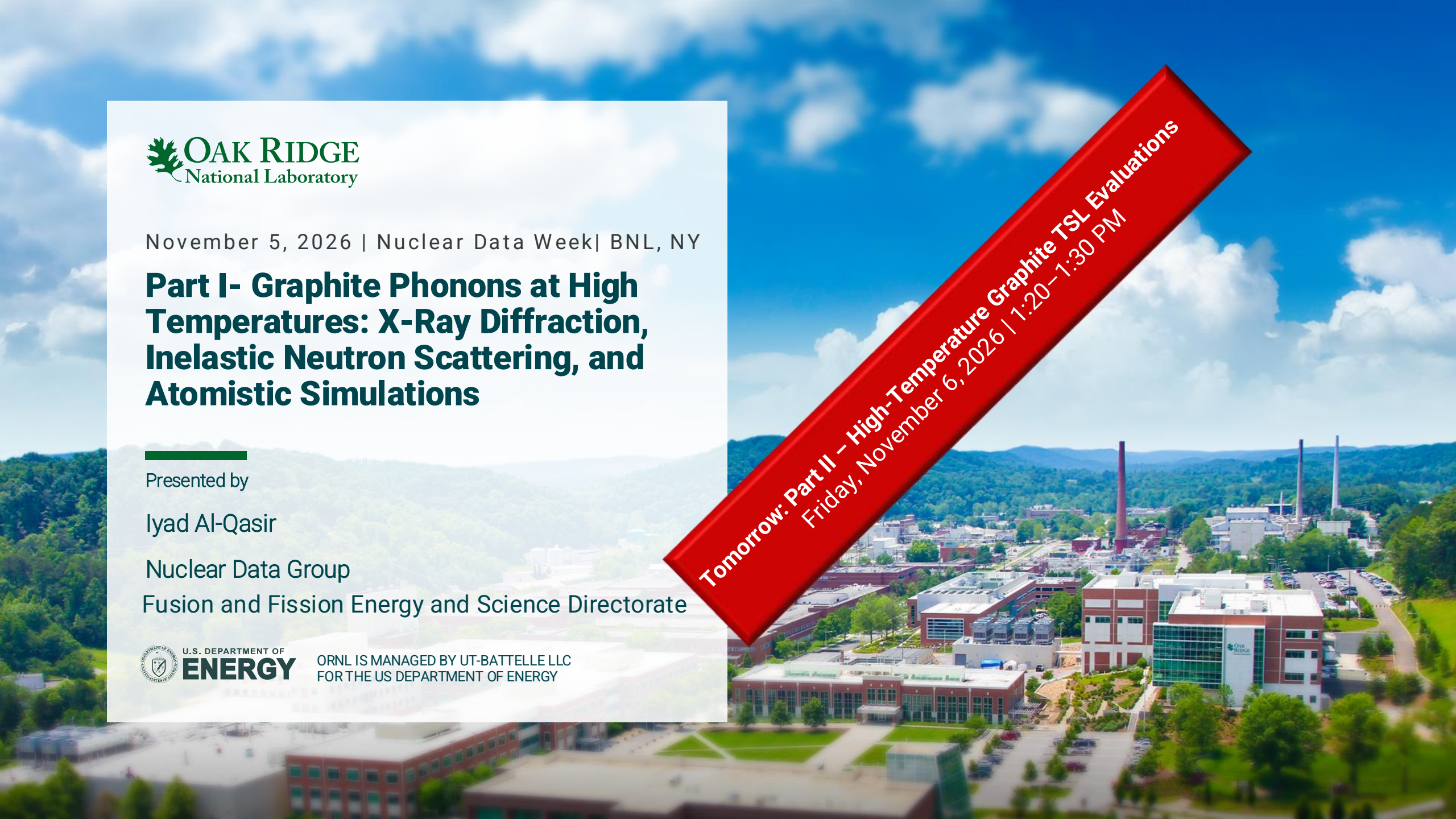
Nuclear Data Group

Fusion and Fission Energy and Science Directorate

U.S. DEPARTMENT OF ENERGY

ORNL IS MANAGED BY UT-BATTELLE LLC
FOR THE US DEPARTMENT OF ENERGY

Tomorrow: Part II – High-Temperature Graphite TSL Evaluations
Friday, November 6, 2026 | 1:20–1:30 PM



Team

Matthew Stone, Garrett Granroth, Anne Campbell, Yongqiang Cheng, Paul Kent, Ercan Cakmak, Alexander Kolesnikov, Michael Manley, Raphael Herman, Kemal Ramic, Luiz Leal, Chris W. Chapman, Douglas Abernathy, William Wieselquist

ORNL

Shinsuke Nakayama

ORNL

Nuclear Data Center- JAEA/Japan

Abdallah Qteish

Physics Department - Yarmouk University/Jordan

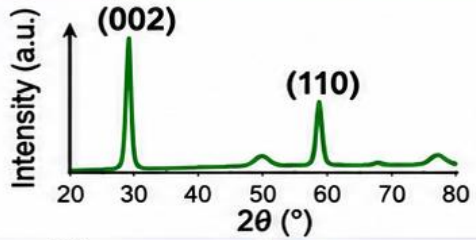
Riad Shaltaf

Physics Department – University of Jordan/Jordan

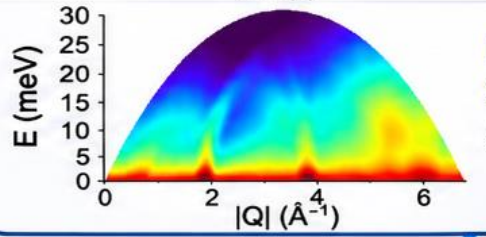
Building Confidence in Graphite TSL Evaluations

Goal: Reliable, experimentally validated graphite TSL evaluations

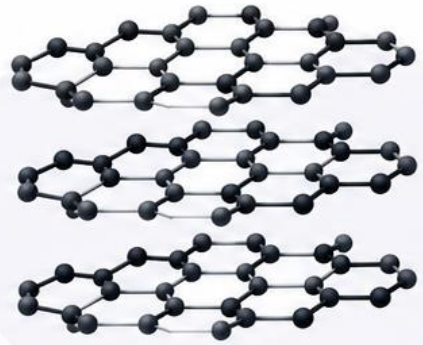
Experimental Measurements



X-ray Diffraction (XRD)
Thermal expansion and structural evolution.



Inelastic Neutron Scattering (INS)
S(Q, E)

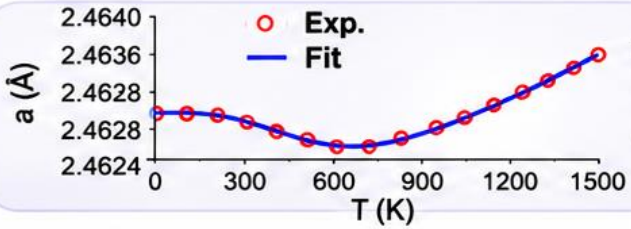


Graphite

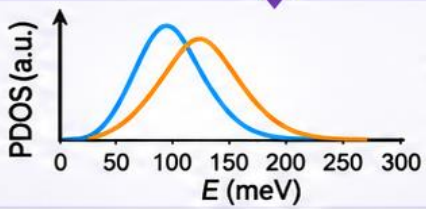
Atomistic Simulations

- Quasi-Harmonic Approximation (QHA)
- Stochastic Self-Consistent Harmonic Approximation (SSCHA)
- Machine-Learning Force-Field (MLFF) Simulations

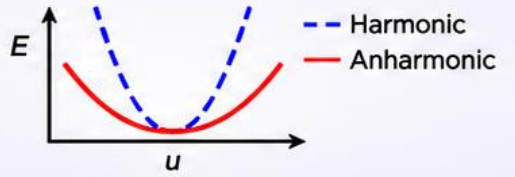
Cross-Validation



Validate thermal expansion



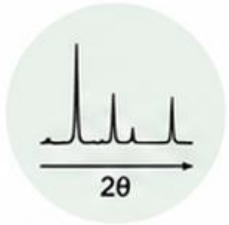
Validate PDOS



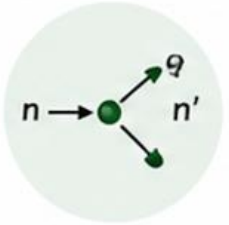
Quantify anharmonic effects

A robust, physically consistent, and experimentally supported foundation for trustworthy high-temperature graphite TSL evaluations

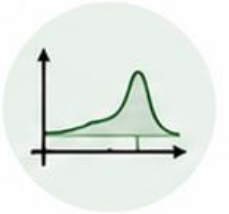
Outlines



1 X-ray Diffraction and Thermal Expansion



2 Inelastic Neutron Scattering Measurements



3 Calculated PDOSs

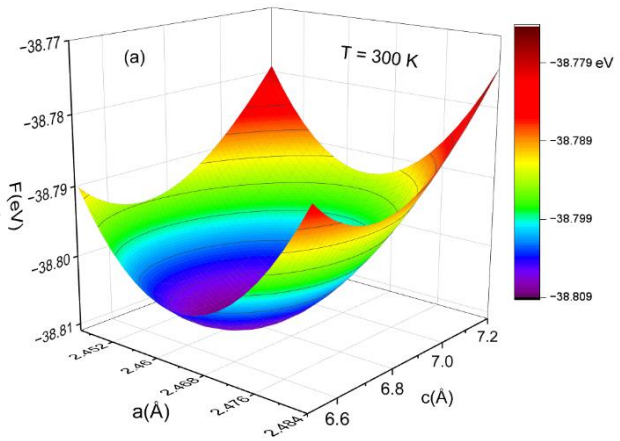


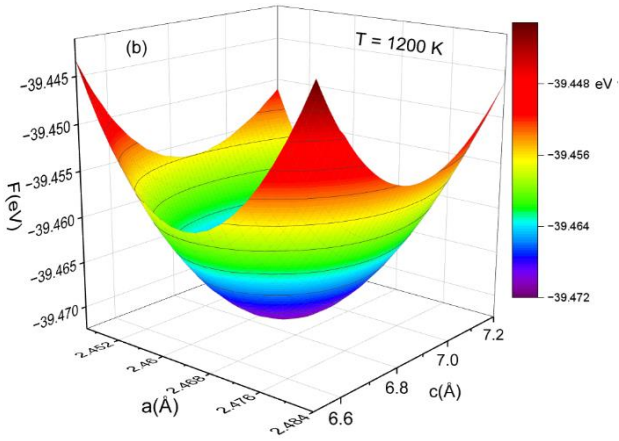
4 Conclusions

X-ray Diffraction
Thermal Expansion Measurements

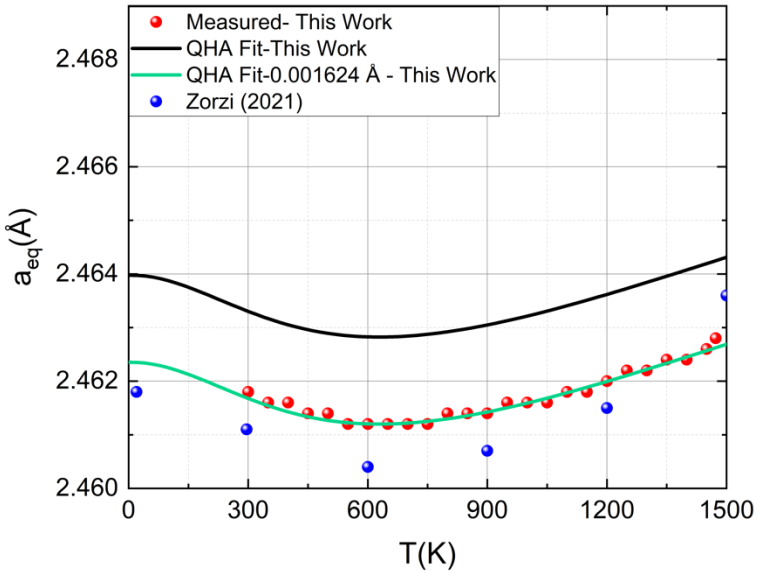
Graphite Thermal Expansion

- Highly anisotropic thermal expansion
- Negative a-axis expansion up to ~650 K
- Positive c-axis expansion ($\Delta c/c$ ~3.4 %)
- QHA reproduces the overall temperature dependence and the minimum in a(T)

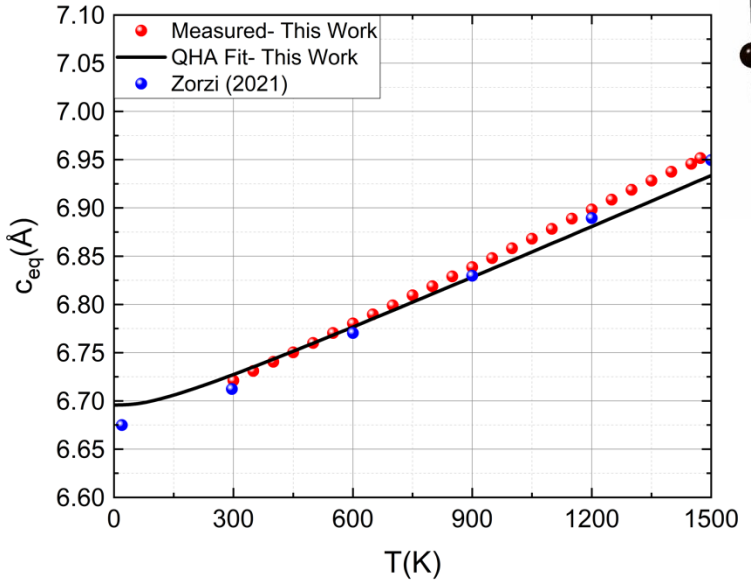


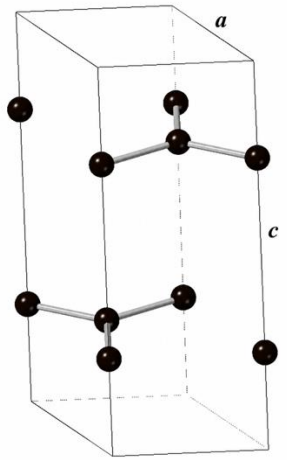


a-lattice constant



c-lattice constant





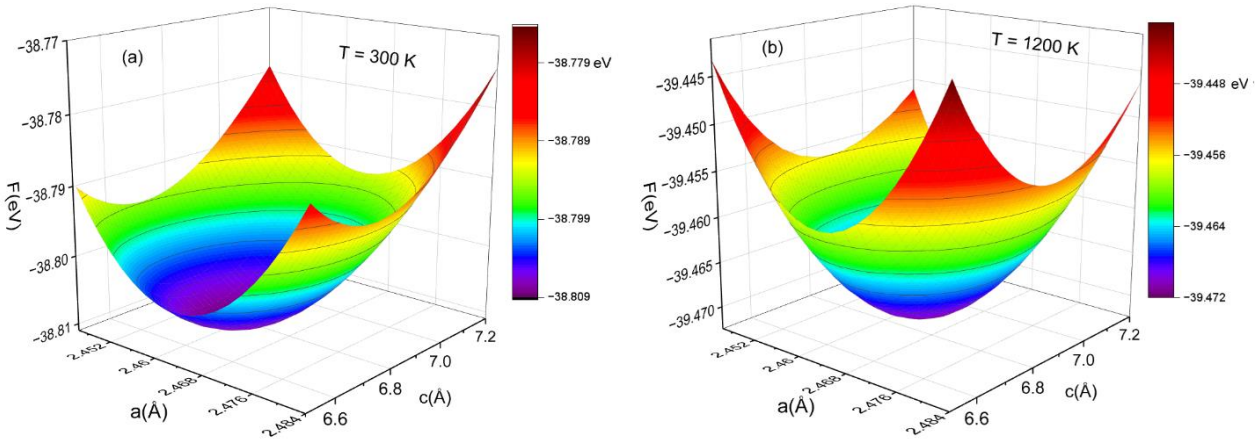
X-ray Diffraction
- ✓ 300–1500 K
- ✓ Cu Kα radiation
- ✓ (110) and (002) reflections

Quasi-Harmonic Approximation (QHA)
- ✓ DFT: r²SCAN + rVV10
- ✓ Helmholtz free-energy surface optimization

QHA captures the structural evolution of graphite, providing a reliable basis for the subsequent phonon calculations.

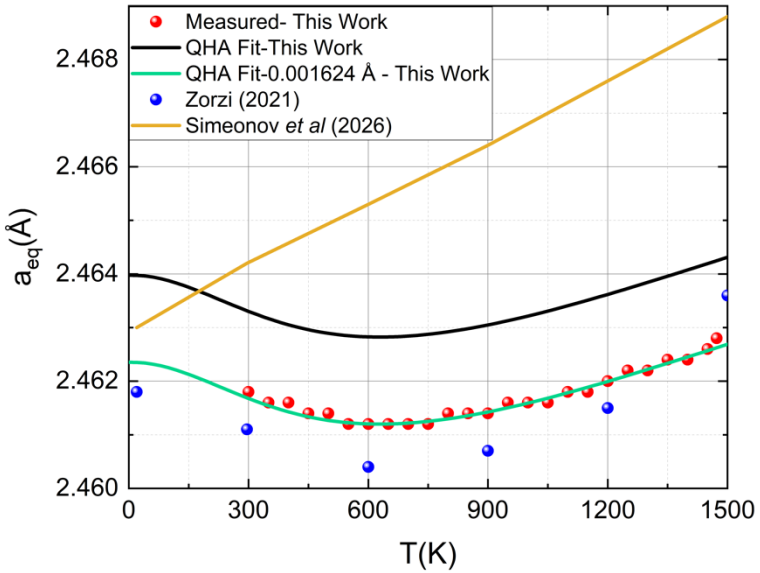
OAK RIDGE National Laboratory

Graphite Thermal Expansion

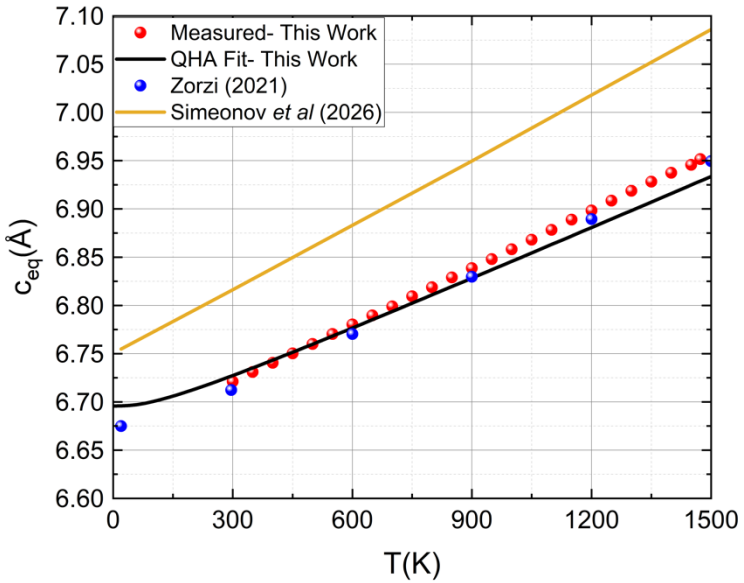


- Highly anisotropic thermal expansion
- Negative a-axis expansion up to ~700 K
- Positive c-axis expansion ($\Delta c/c$ ~3.4 %)
- QHA reproduces the overall temperature dependence and the minimum in a(T)

a-lattice constant



c-lattice constant



The choice of exchange–correlation (XC) functional and van der Waals (vdW) correction is critical, as different combinations can lead to unphysical continuous lattice expansion or unrealistic temperature dependence of the lattice parameters.

Zorzi & Perottoni, Computational Materials Science, **199**, 110719 (2021)
Simeonov, et al., ANS Transactions, 135 (2026)

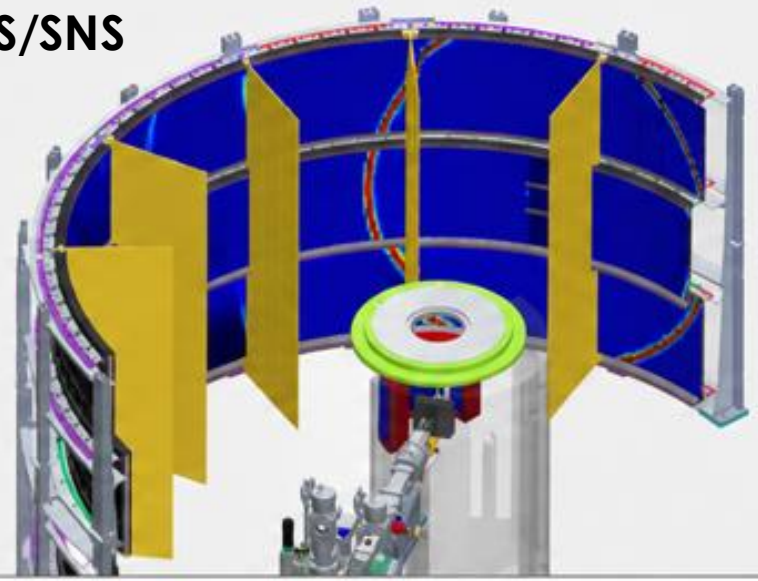
Inelastic Neutron Scattering Phonon Measurements

Inelastic Neutron Scattering Measurements

Experiment

- Ei = 30, 130, 215, 300 meV
- T = 300, 600, 900, 1200 K
- Graphite Grade: G347A
- Dimension: 3.96 cm × 5.00 cm × 0.400 cm
- Mass: 14.25 g

ARCS/SNS



Data Processing

Detector efficiency correction using vanadium standard → Subtract empty vanadium pouch background → Normalize by integrated proton current → Correct for multiple & multi-phonon scattering → Extract phonon density of states (multi-phonon code)

Temperature-Induced Crossover in the Graphite PDOS

Measured PDOS

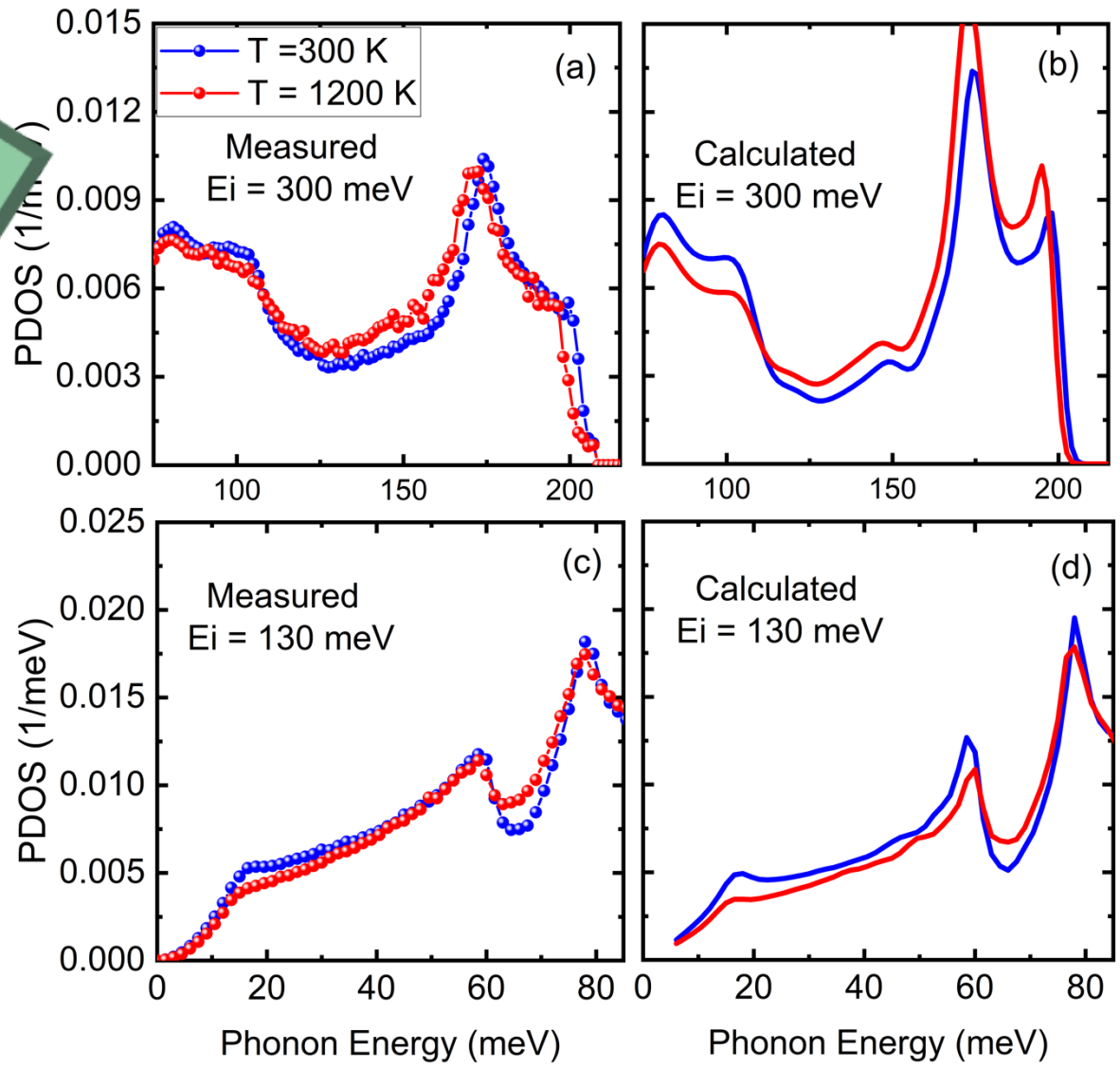
Temperature-dependent crossover near ~100 meV.

- E > 100 meV: PDOS (1200 K) > PDOS (300 K)
- E < 100 meV: PDOS (1200 K) < PDOS (300 K)

Calculated PDOS

Calculated S(Q,E)-derived PDOS reproduces the same crossover

Why validation is important?



Measured and calculated PDOSs exhibit the same crossover, confirming that the observed "flipping" is an intrinsic consequence of the temperature evolution of the graphite phonon spectrum.

Measured and Calculated PDOSs

1 Agreement with experiment

- Major phonon features reproduced
- Temperature evolution accurately captured

2 Temperature evolution

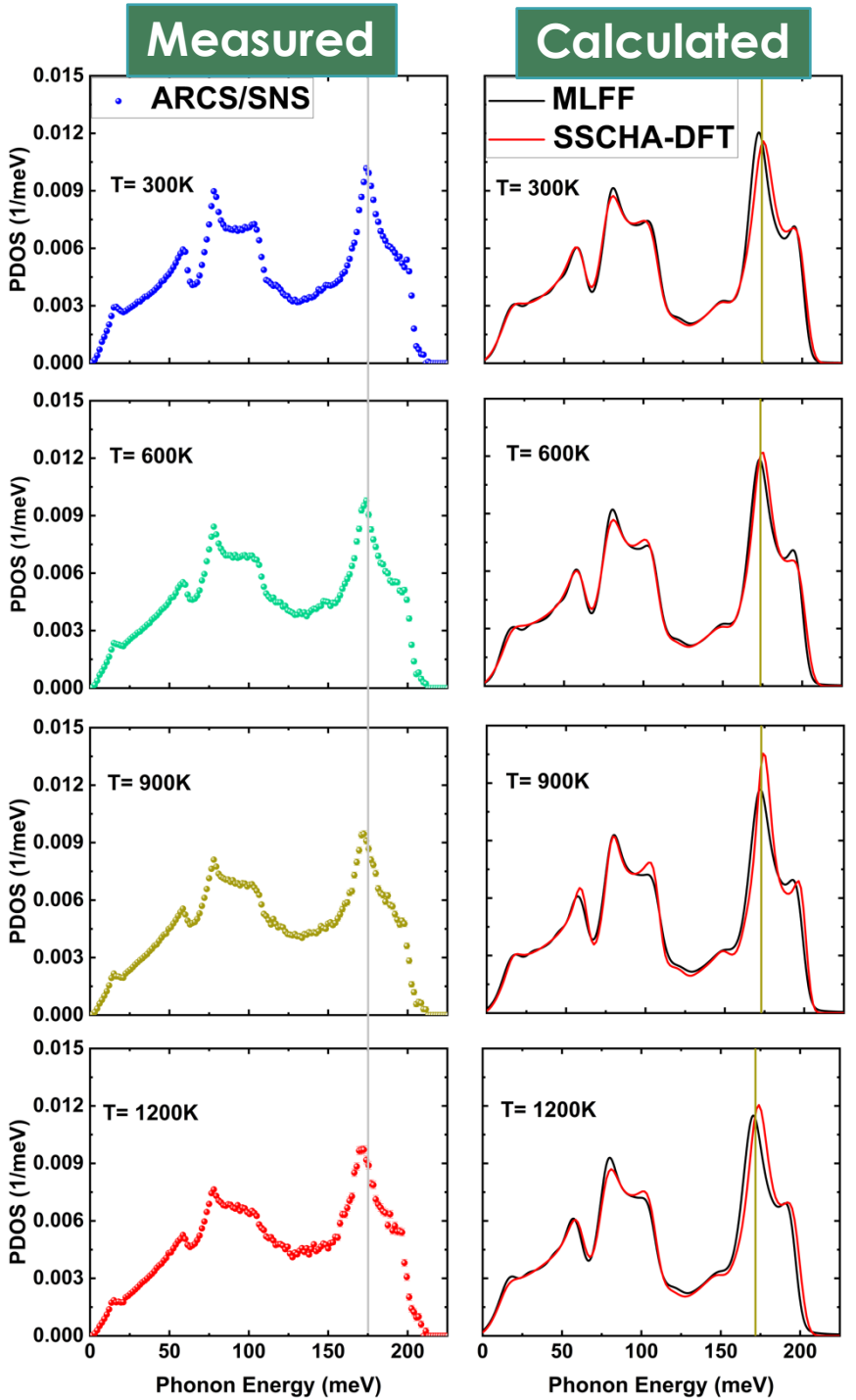
- Progressive softening of high-energy optical phonons
- Redistribution of spectral weight with increasing temperature

3 MLFF ≈ SSCHA-DFT

- Nearly identical PDOS over most of the phonon spectrum
- Excellent agreement at all temperatures

MLFF: Machine-Learning Force Field

SSCHA: Stochastic Self-Consistent Harmonic Approximation



OAK RIDGE National Laboratory

Determination of Phonon Peak Positions

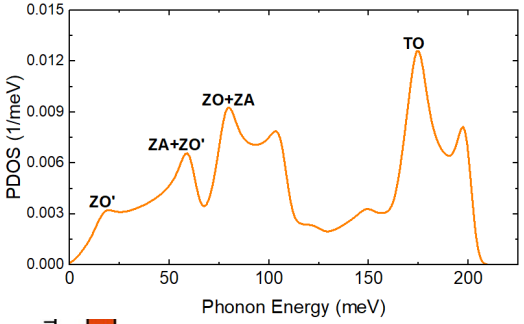
1. Directly from S(Q,E)
Phonon energies extracted directly from S(Q,E), not from the PDOS.

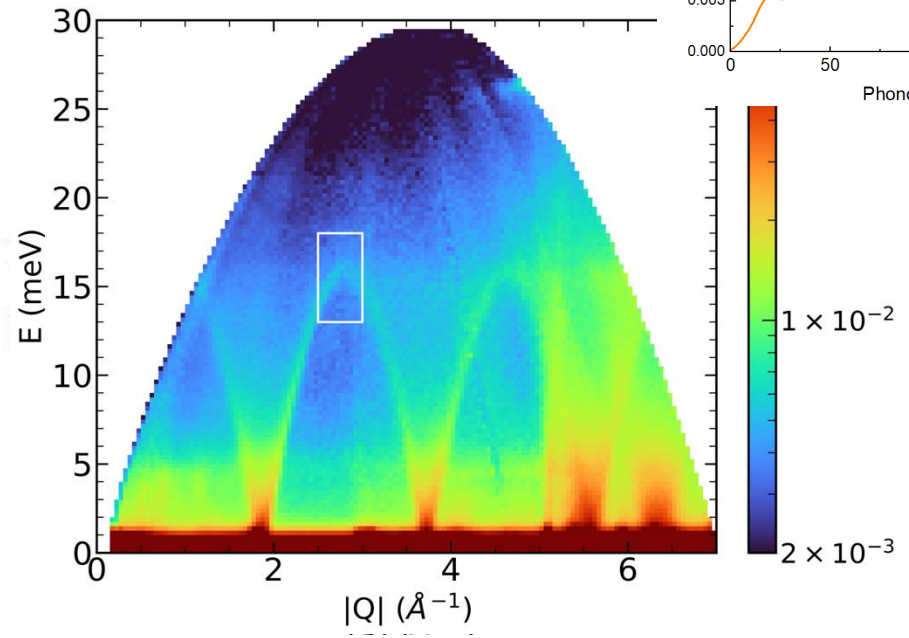
2. Consistent analysis
Same Q-integration ranges and fitting windows used at all temperatures.

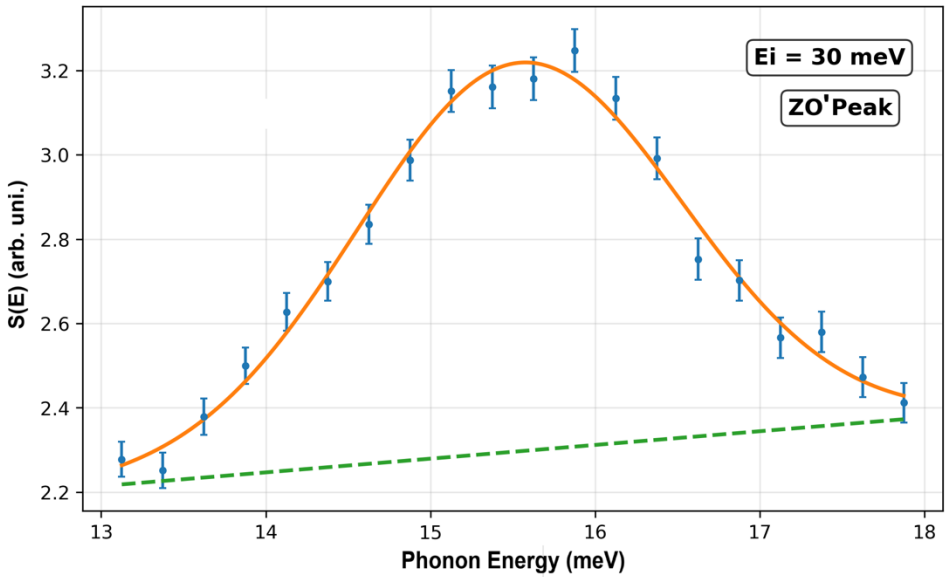
3. Peak fitting
Integrated spectra fitted with a Gaussian peak plus a linear background.

4. Reliable results
Direct fitting avoids uncertainties from PDOS smoothing and energy binning.

5. Quantify softening
Extracted peak positions used to quantify phonon softening.







OAK RIDGE National Laboratory

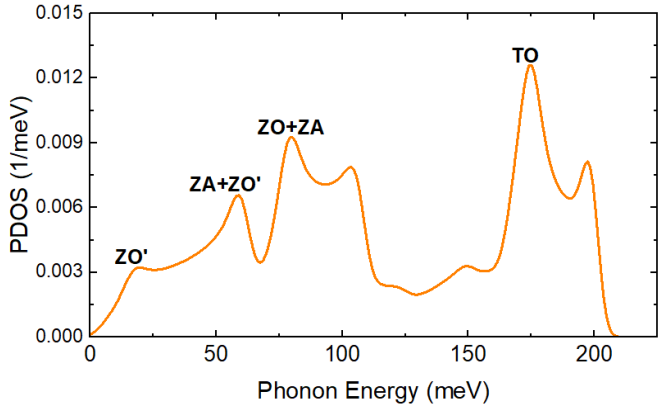
Phonon Softening

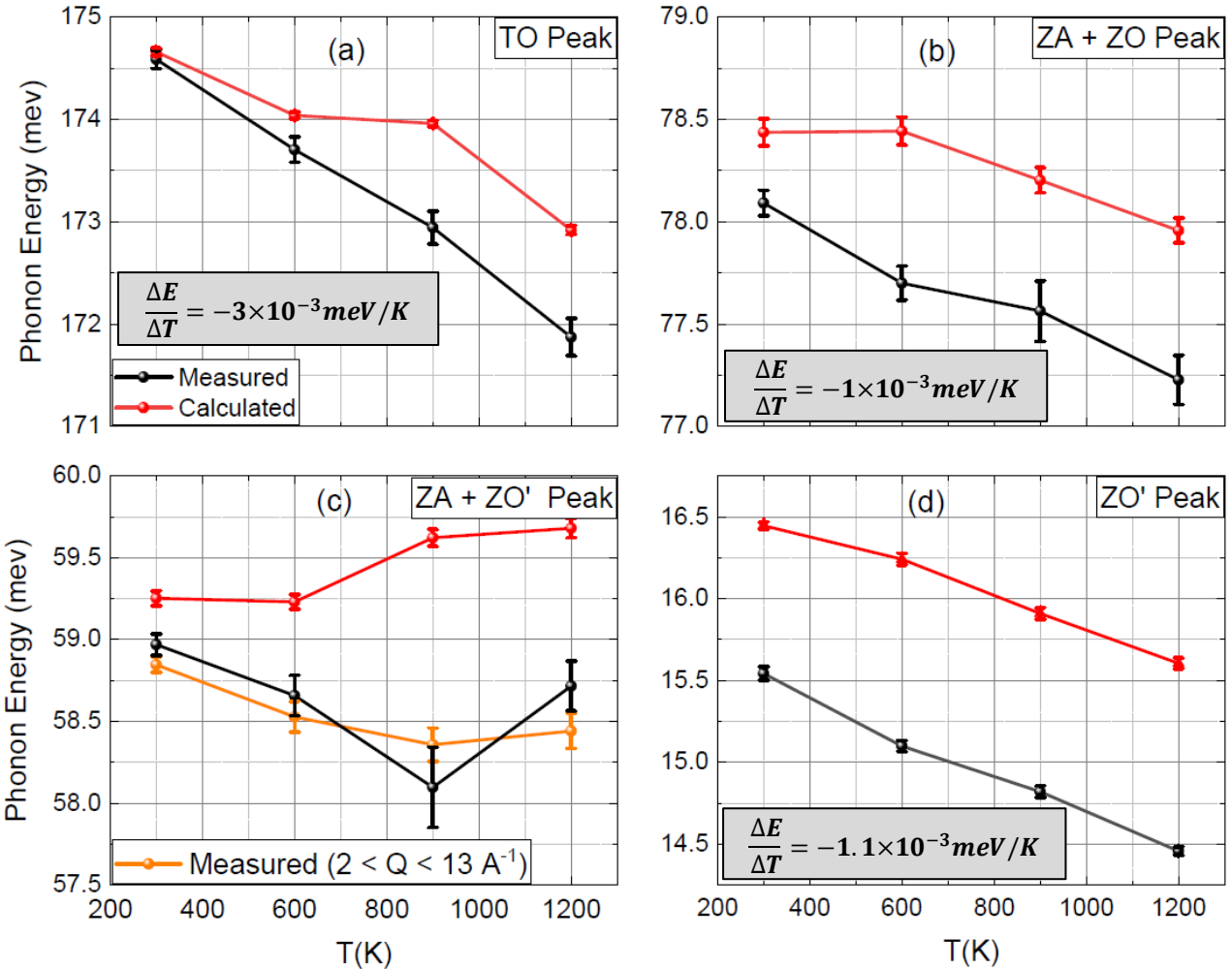
TO phonon shows the **largest** temperature-induced **softening** (~2.7 meV experimentally).

ZA+ZO and ZO' modes show **smaller shifts**, while ZA+ZO' mode is **nearly temperature independent.**

SSCHA-DFT reproduces the temperature dependence of all phonon modes but **systematically underestimates** the softening.

The agreement confirms that the **dominant anharmonic behavior** is captured by the atomistic model.





OAK RIDGE National Laboratory

Acknowledgment



Funding:

Supported by the **DOE/U.S. Nuclear Regulatory Commission (NRC)** collaboration for Criticality Safety in commercial-scale **HALEU fuel cycle** projects **(DNCSH)**



Computational Resources:

Simulations were performed using the **National Energy Research Scientific Computing Center (NERSC)**, a DOE Office of Science User Facility



Experimental Facilities:

Neutron scattering experiments were conducted at the **Spallation Neutron Source (SNS)** at Oak Ridge National Laboratory, (IPTS-34282).

Conclusions

01 **Combined experiments (XRD, INS) and simulations (DFT, QHA, SSCHA, MLFF)** provided a comprehensive understanding of graphite lattice dynamics from **300–1200 K.**

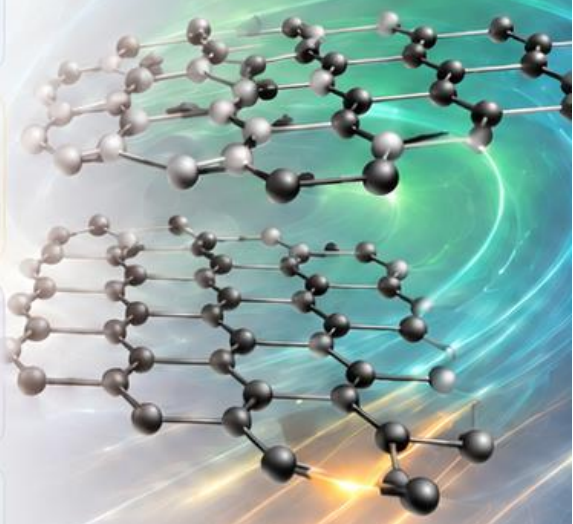
02 **High-temperature XRD** confirmed the **anisotropic thermal expansion** of graphite, which was well reproduced by **QHA** calculations.

03 **Negative in-plane thermal expansion** at low temperatures originates from **low-energy ZA phonons** with negative Grüneisen parameters; higher-energy phonons drive positive linear expansion at elevated temperatures.

04 **Temperature-dependent phonon softening is strongly mode dependent,** with **TO phonons** showing the largest softening, while **ZA+ZO'** remains nearly temperature independent.

05 **SSCHA and MLFF simulations** successfully reproduced the experimental lattice dynamics, validating **first-principles and machine-learning** approaches for predicting the high-temperature behavior of graphite.

06 Our validated phonon calculations are ready to generate temperature-dependent thermal scattering laws, **S(alpha,beta,T).**



Thank You

Graphite Phonons and the Origin of Anisotropic Thermal Expansion

1 Anisotropy

Strong in-plane covalent bonds and weak interlayer van der Waals interactions.

2 Phonon Energies

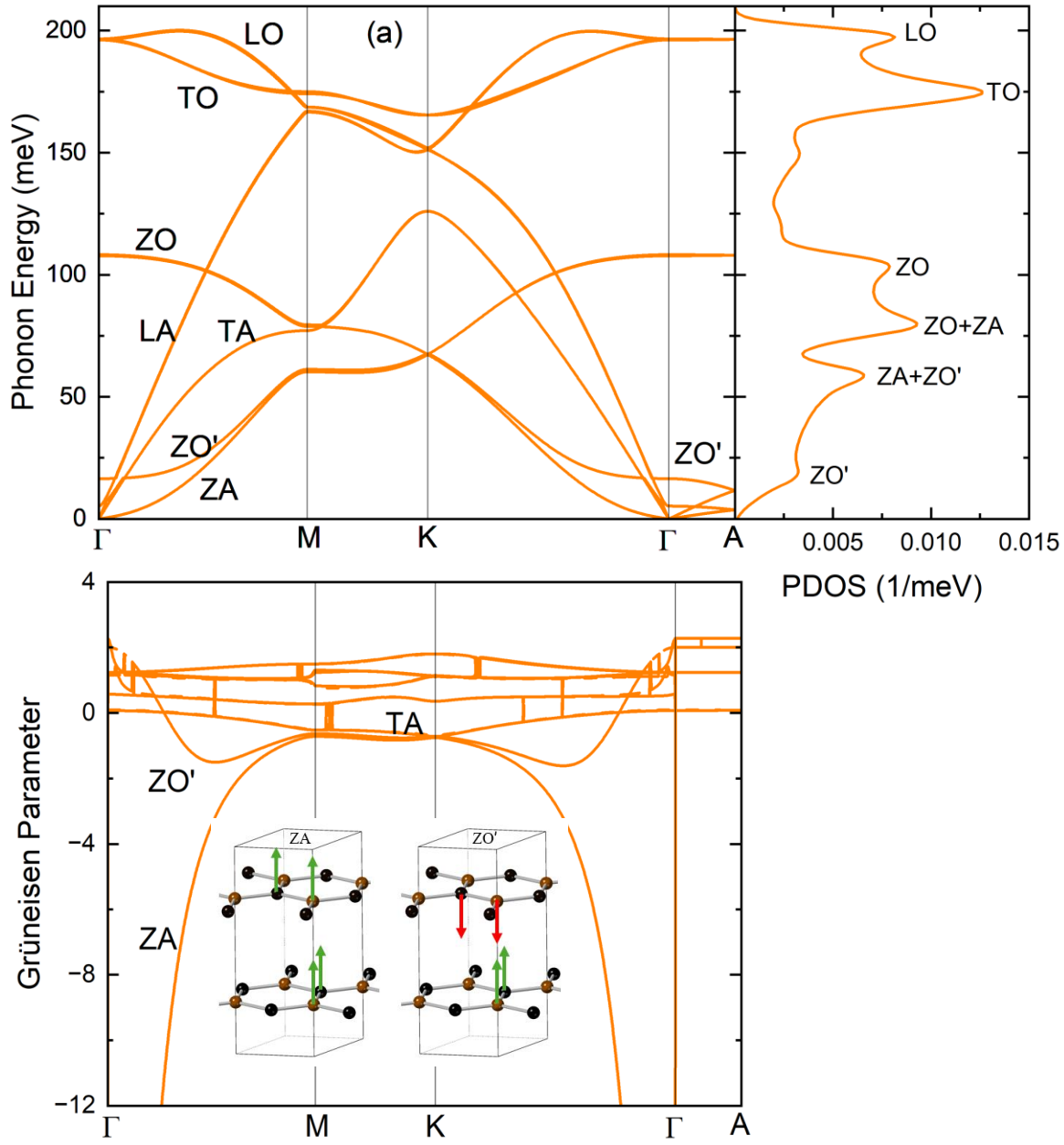
In-plane LO and TO optical modes reach ~200 meV, whereas interlayer ZO and ZO' modes are much softer.

3 PDOS Features

PDOS reflects the phonon dispersion, with pronounced peaks originating from flat branches.

4 Origin of NTE

ZA acoustic branch exhibits a large negative Grüneisen parameter, driving the negative in-plane thermal expansion.



OAK RIDGE National Laboratory