Nuclear Graphite TSL Measurements

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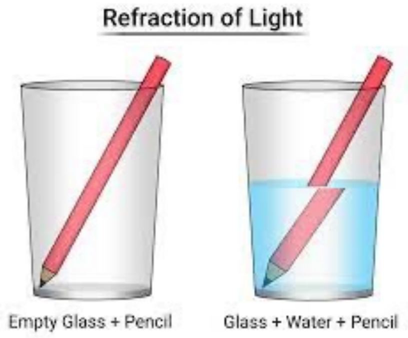
Outline

- PDOS Measurements (Inelastic Scattering)
- Diffraction Measurements (Elastic Scattering)
- Introduction to SANS

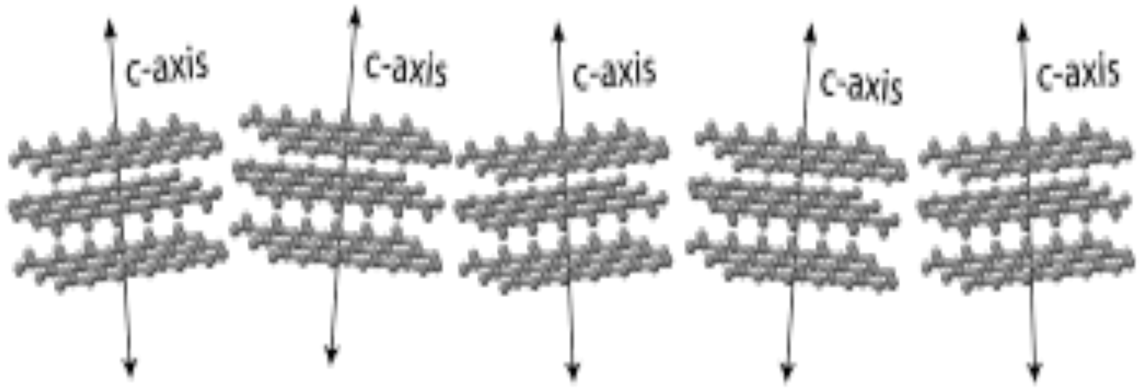
Scattering Type	Light	Thermal Neutron
Inelastic	Raman (photon-atom)	(phonon-atoms)
Elastic	Raleigh	Bragg



☼ There is an analogy between thermal neutrons interactions (neutron optics) and light photons interactions (light optics)

Crystalline Graphite

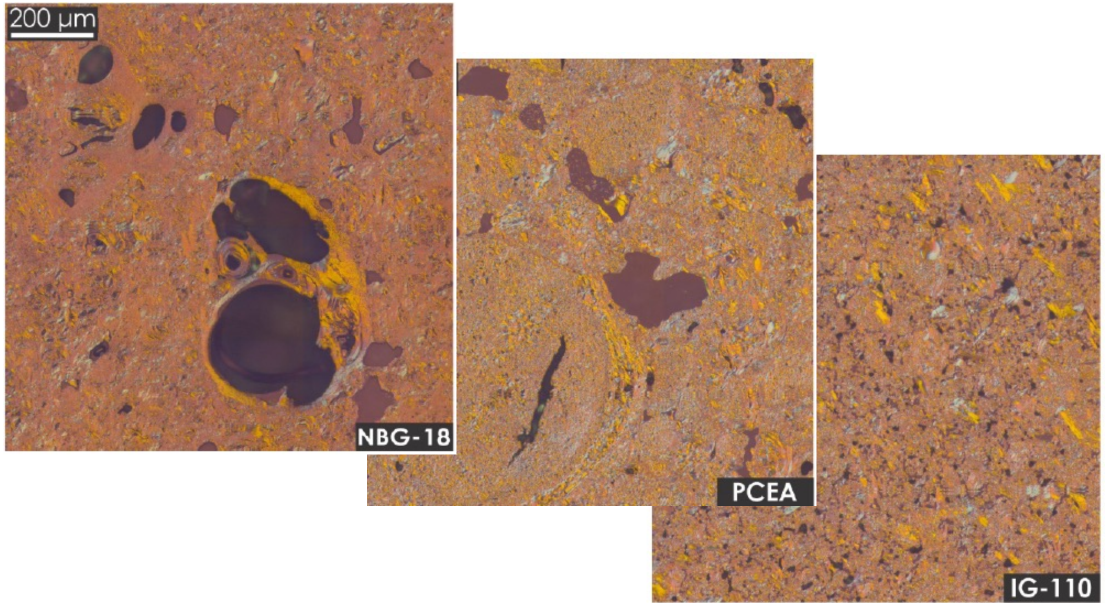
- **Density ~2.26 g/cm^3 (HOPG)**
- Huge number of small nearly perfect micro-crystallites that have the same c-axis but with different orientations in x-y plane
- Very close to the Ideal, prefect or theoretical graphite.

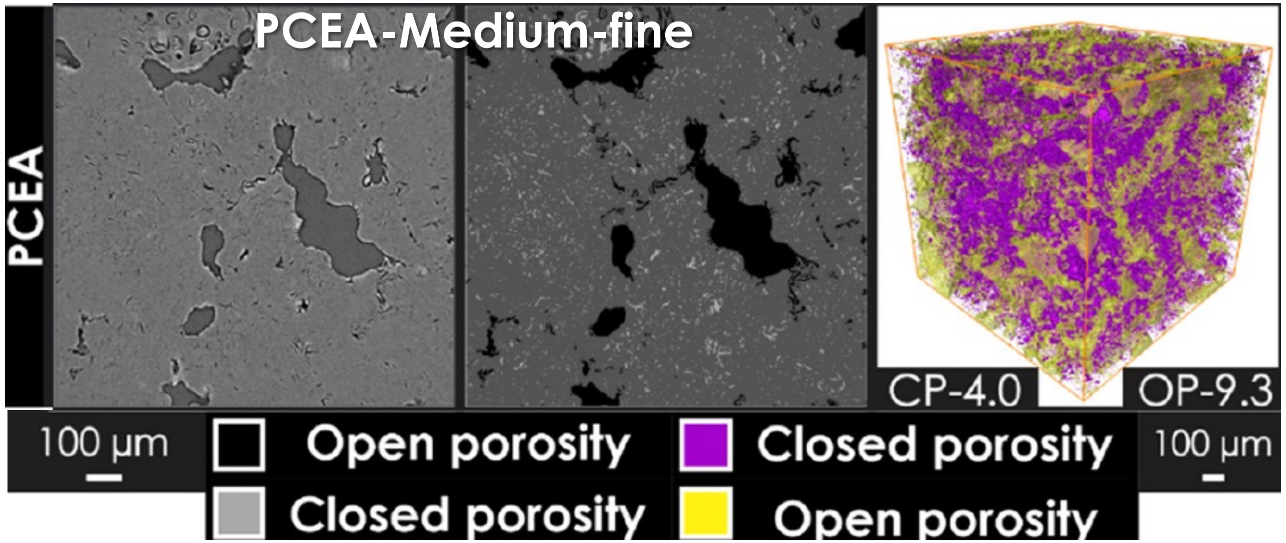


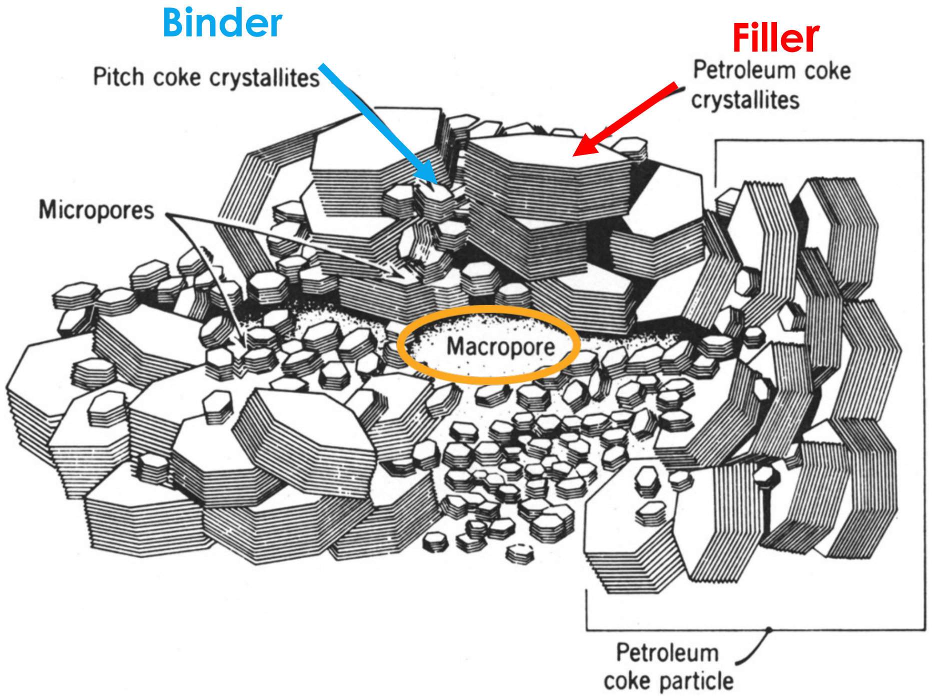
OAK RIDGE National Laboratory

Nuclear Graphite

- **Density**(1.5~1.90) g/cm^3
- Complex Microstructure- contains crystallite (filler and binder) & pores
- The size and shape of grains and pores vary from one graphite grade to another.
- Many nuclear grades are being developed, e.g., IG-430, PCEA, G347A, NBG-18, ZXF-5Q

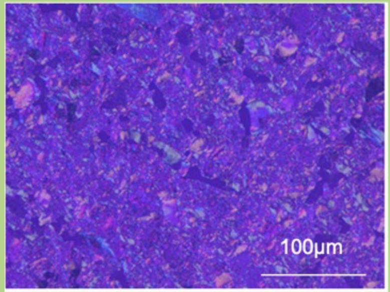
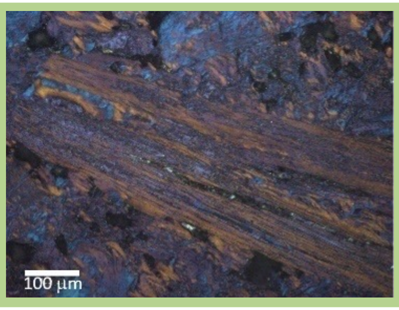




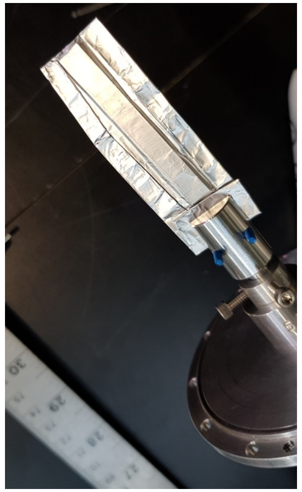


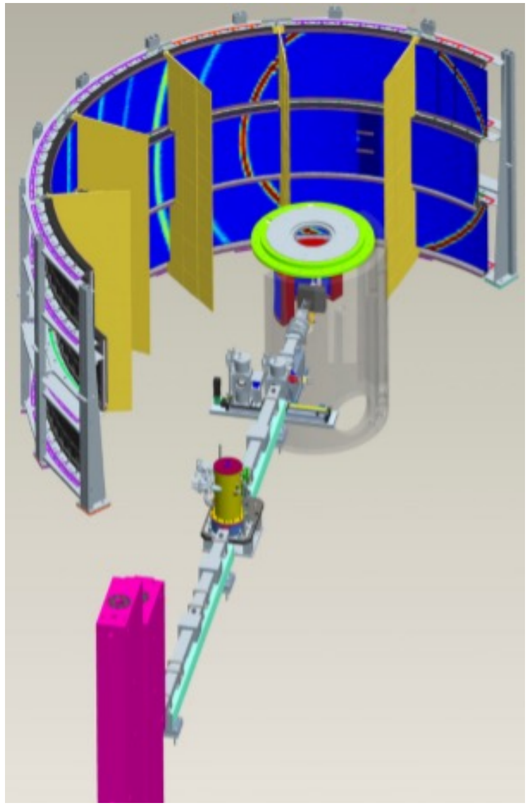
Contescu & Paul, ORNL/TM-2022/1839

Graphite Samples

Type	G347A	PGA
Processing	Isostatic Pressing	Extrusion
Grain Size (mm)	0.05	0.8
Isotropy	Near-isotropic	Non-isotropic
Density (g/cm^3)	1.85	1.70
Porosity (%)	18	25
Sample mass (g)	0.52	5.0
Source	Tokai Carbon	Uni. of Manchester
Polarized Optical Micrographs		

- *ARCS/SNS/ORNL*
- Ei = 30, 130, 300 meV
- Room Temperature

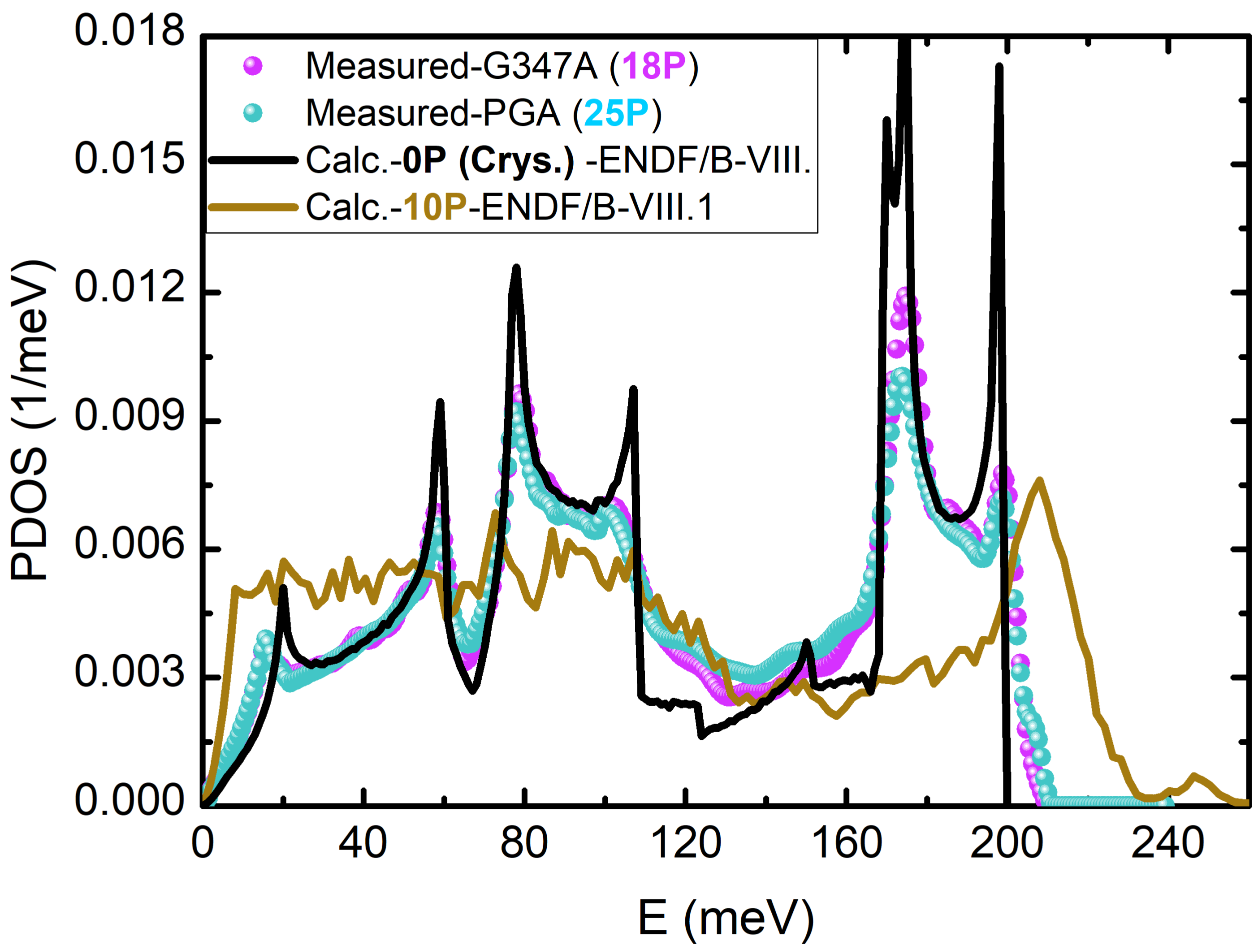


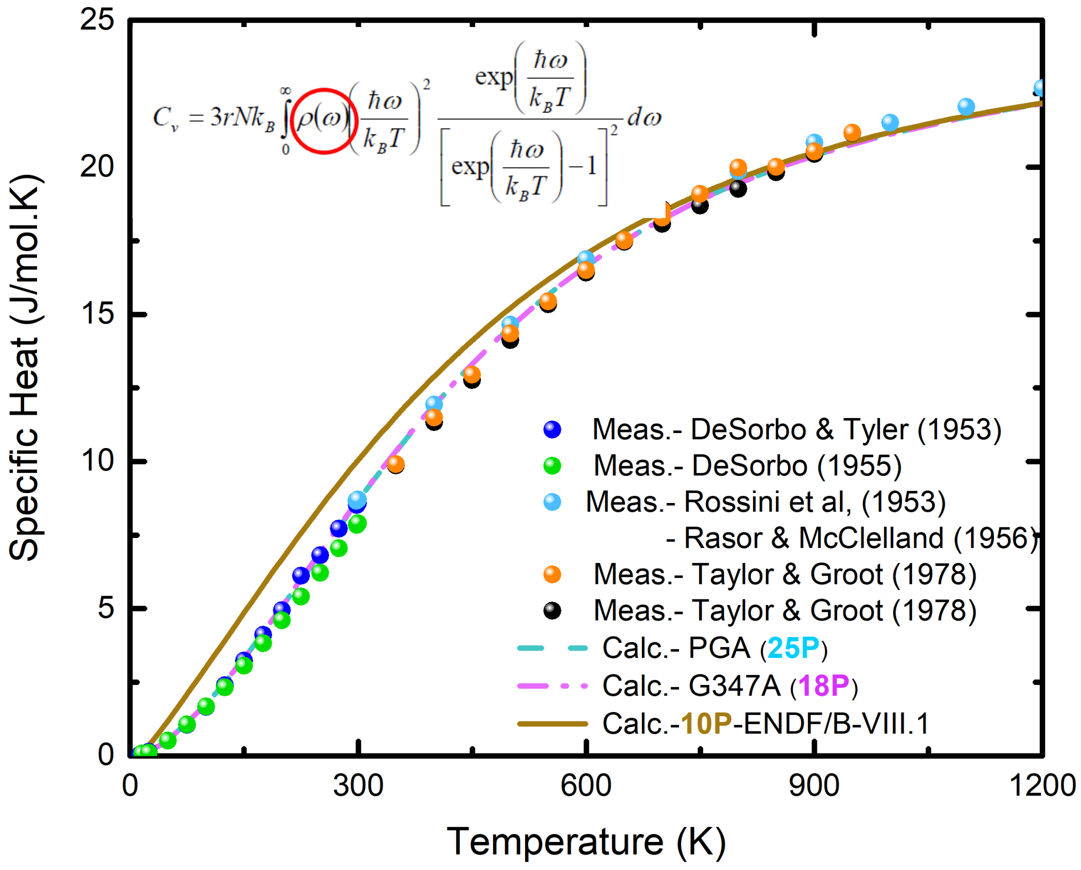


Inelastic Scattering

- Energy of thermal neutrons is of the same order of magnitude as that of excitations in a scattering medium (e.g., phonons in a solid).

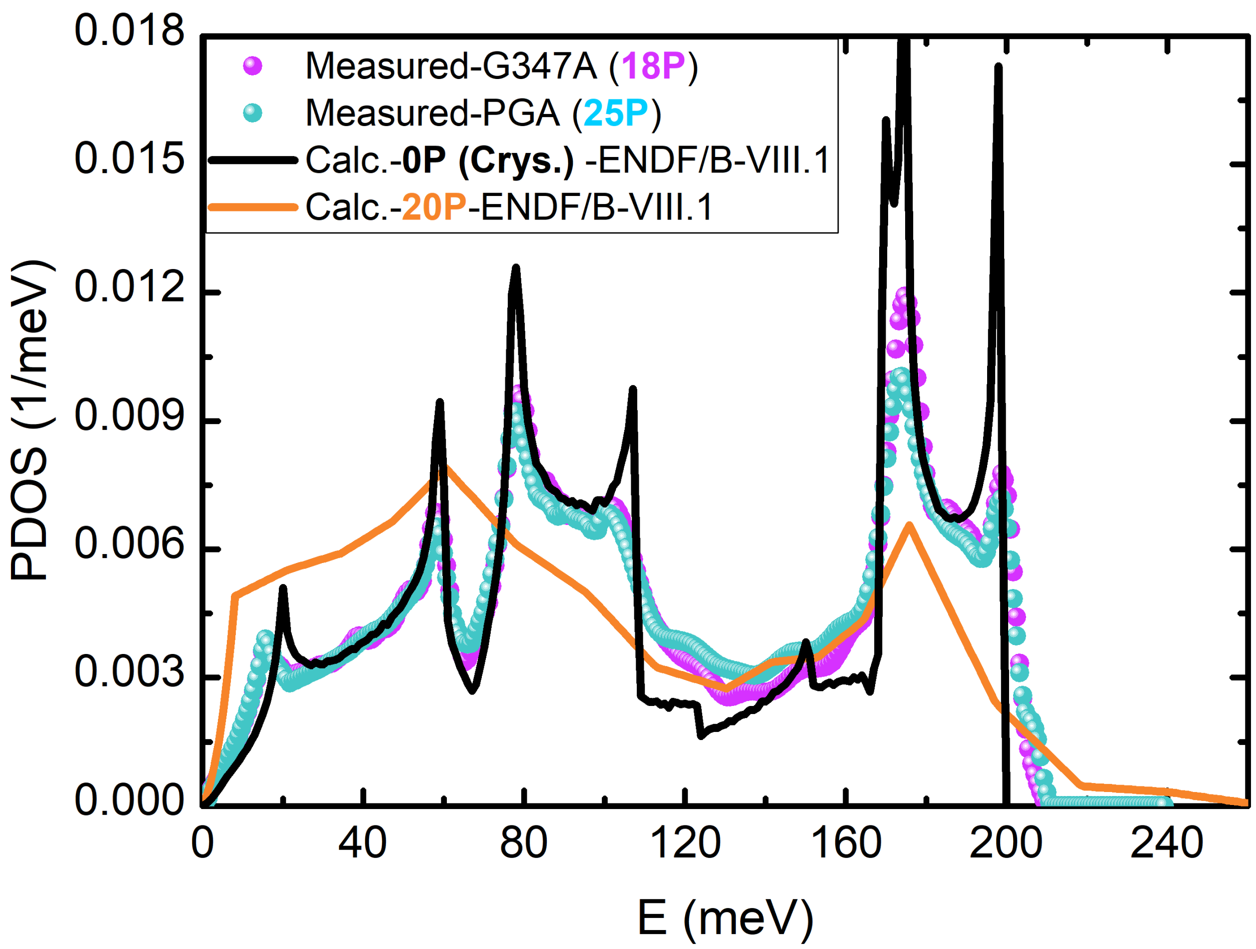
ARCS Measurements of Phonon Density of States (PDOSs)

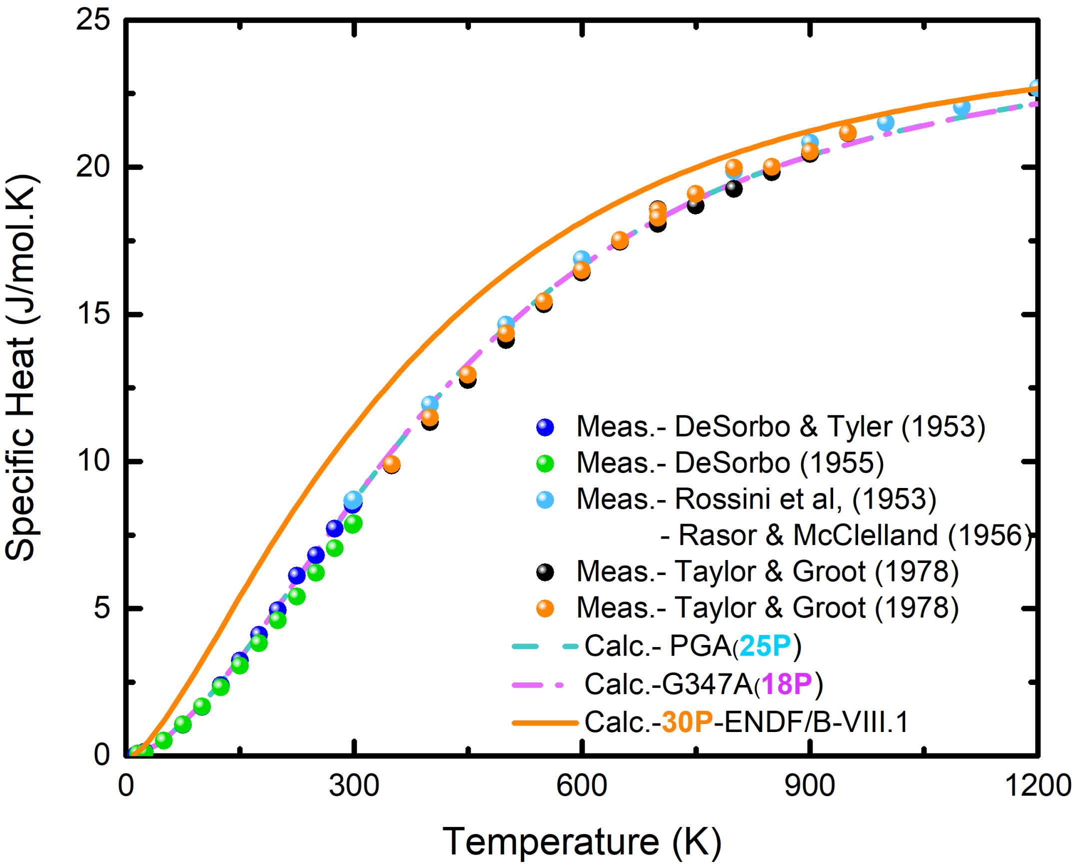




- ✓ **Specific heat as well as S(α , β) are both functions of the PDOS, $\rho(\omega)$**
- ✓ **Different graphite grades, show similar measured PDOS & specific heat**
- ✓ **10P-PDOS & Specific heat shows significant deviations from measured values**

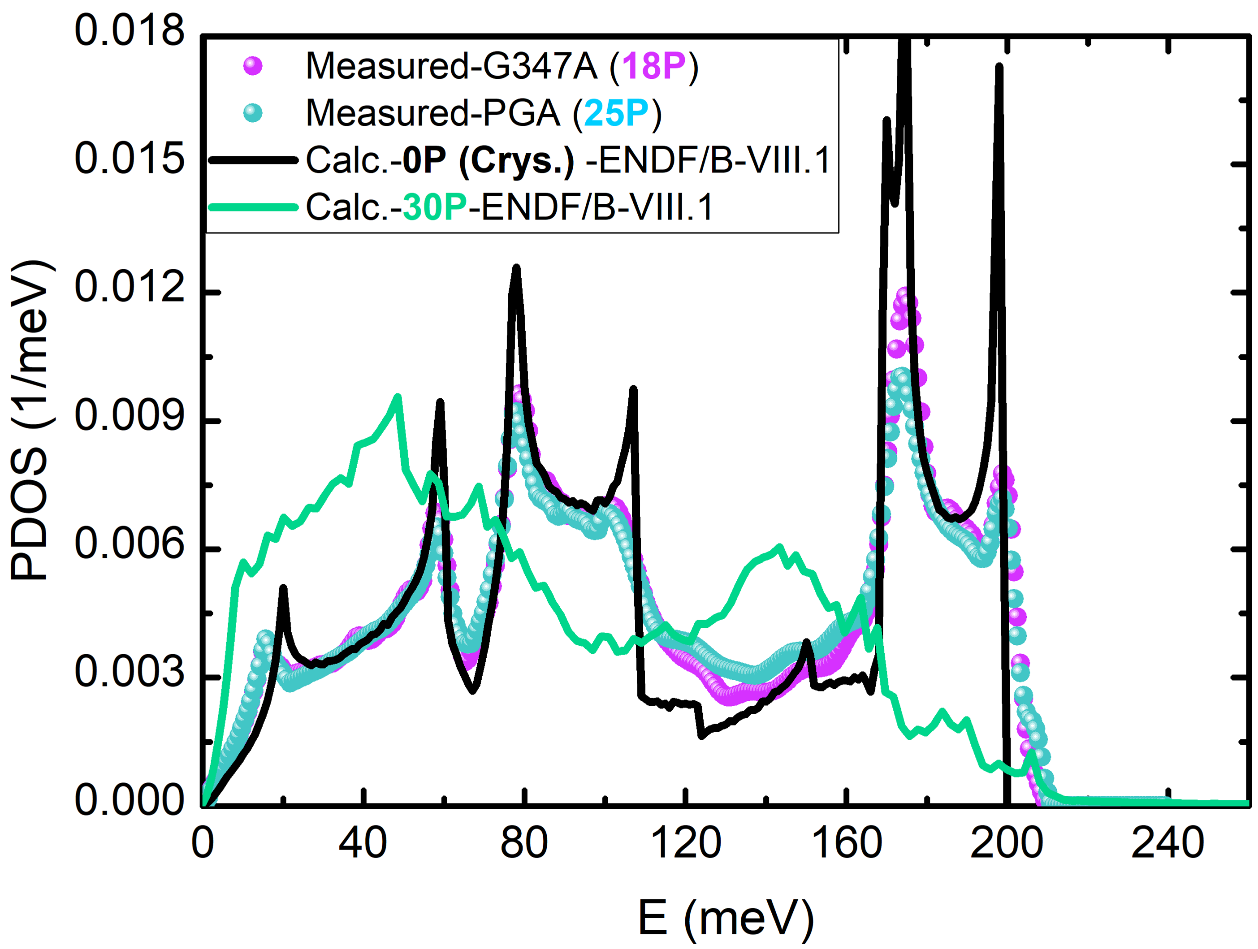
ARCS Measurements of Phonon Density of States (PDOSs)

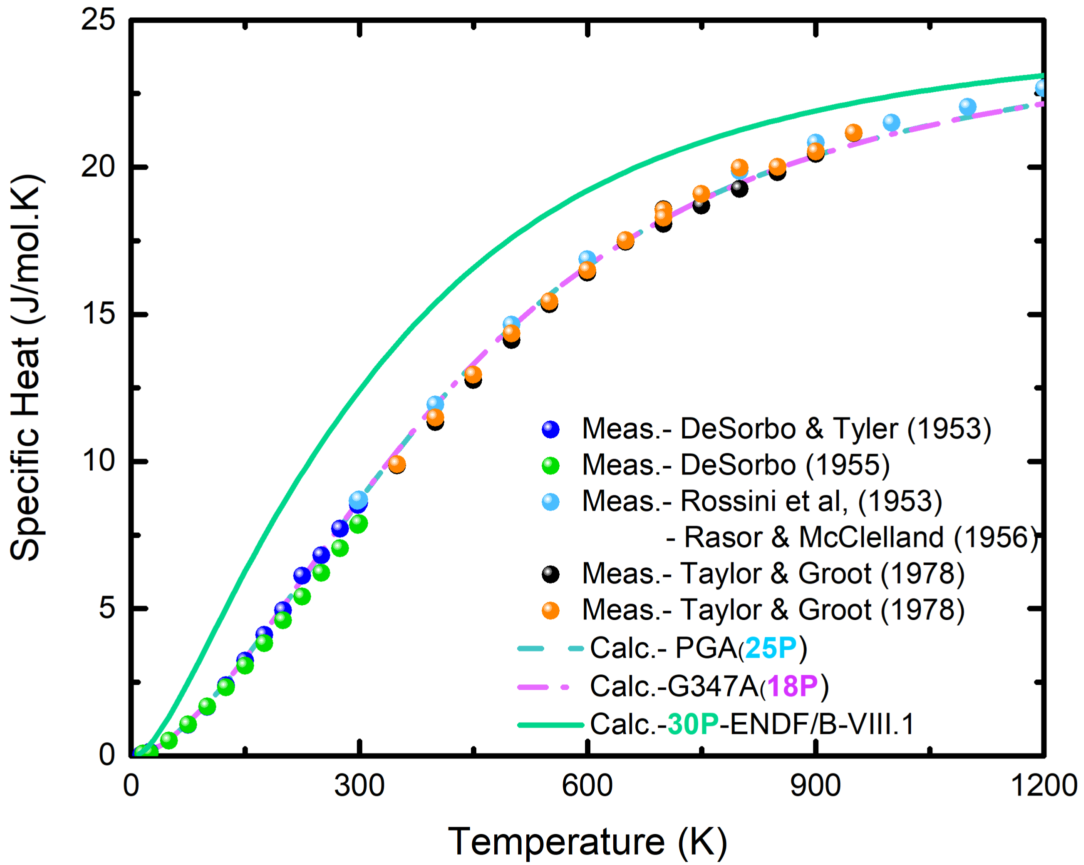




- ✓ **Specific heat as well as S(α , β) are both functions of the PDOS, $\rho(\omega)$**
- ✓ **Different graphite grades, show similar measured PDOS & specific heat**
- ✓ **20P-PDOS & Specific heat shows significant deviations from measured values**

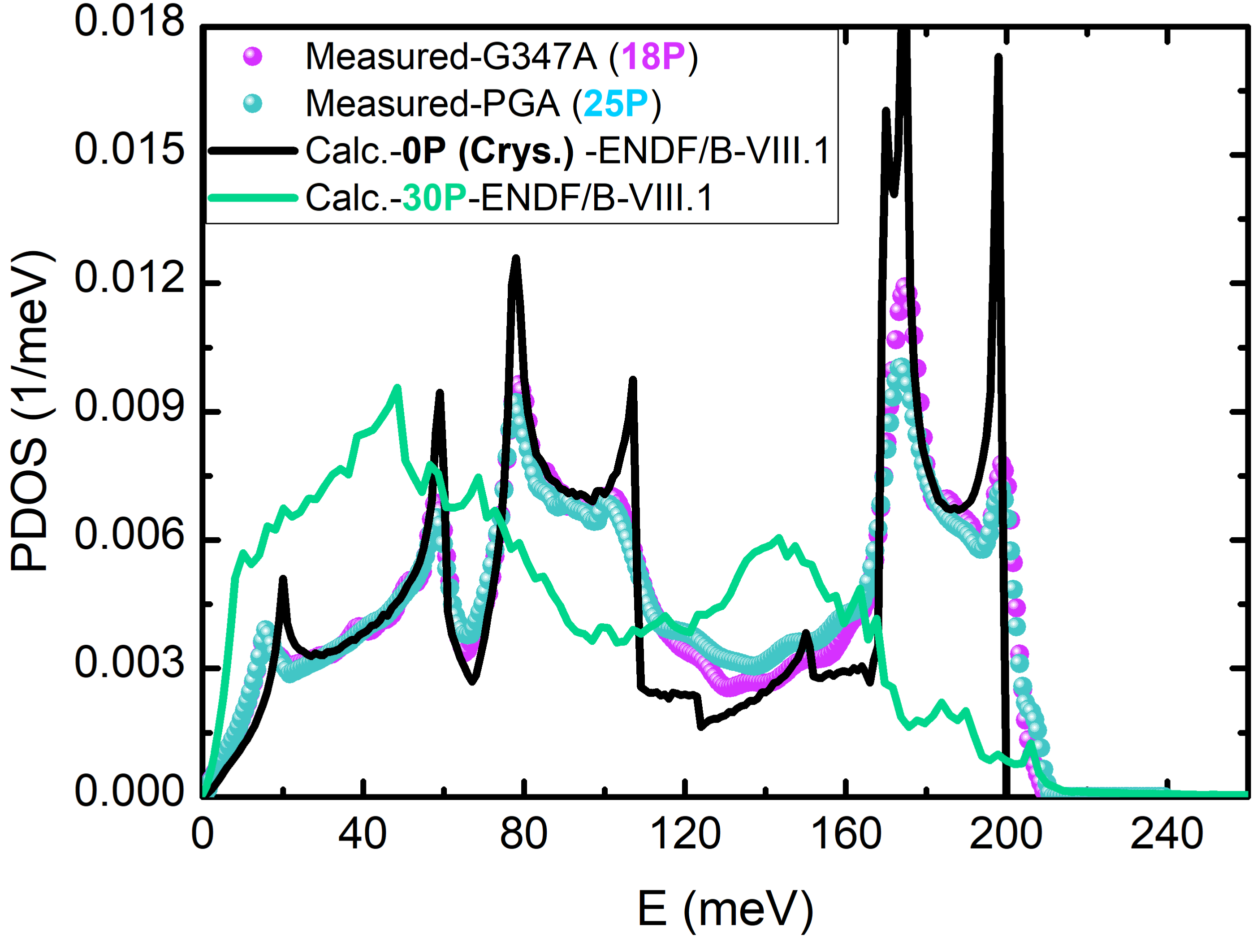
ARCS Measurements of Phonon Density of States (PDOSs)





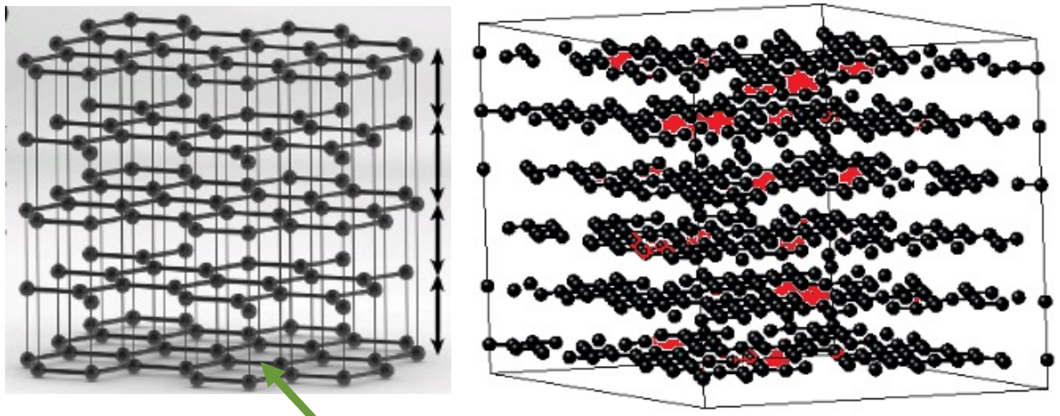
- ✓ **Specific heat as well as $S(\alpha, \beta)$ are both functions of the PDOS, $\rho(\omega)$**
- ✓ **Different graphite grades, show similar measured PDOS & specific heat**
- ✓ **30P-PDOS & Specific heat shows significant deviations from measured values**

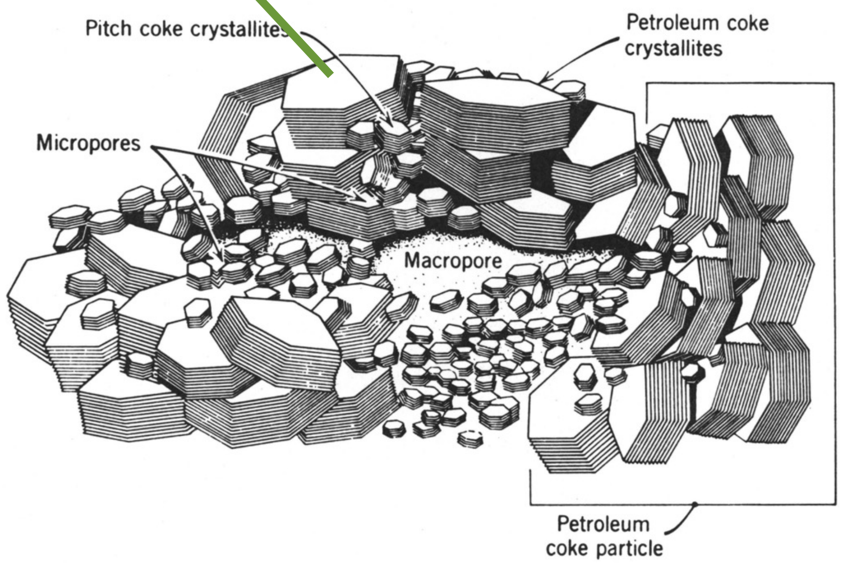
MD Porosity modeling??



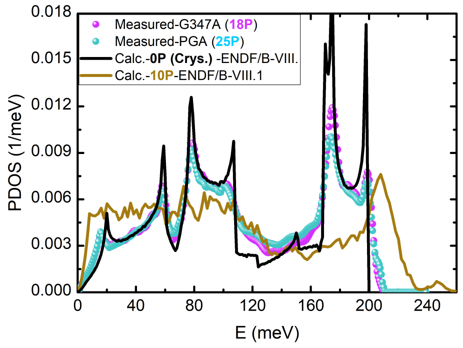
Perfect Graphite SC

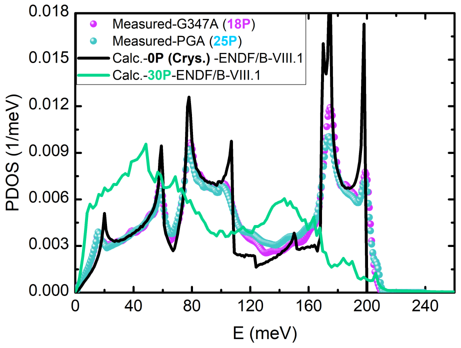
Defected graphite SC
Randomly removed atoms

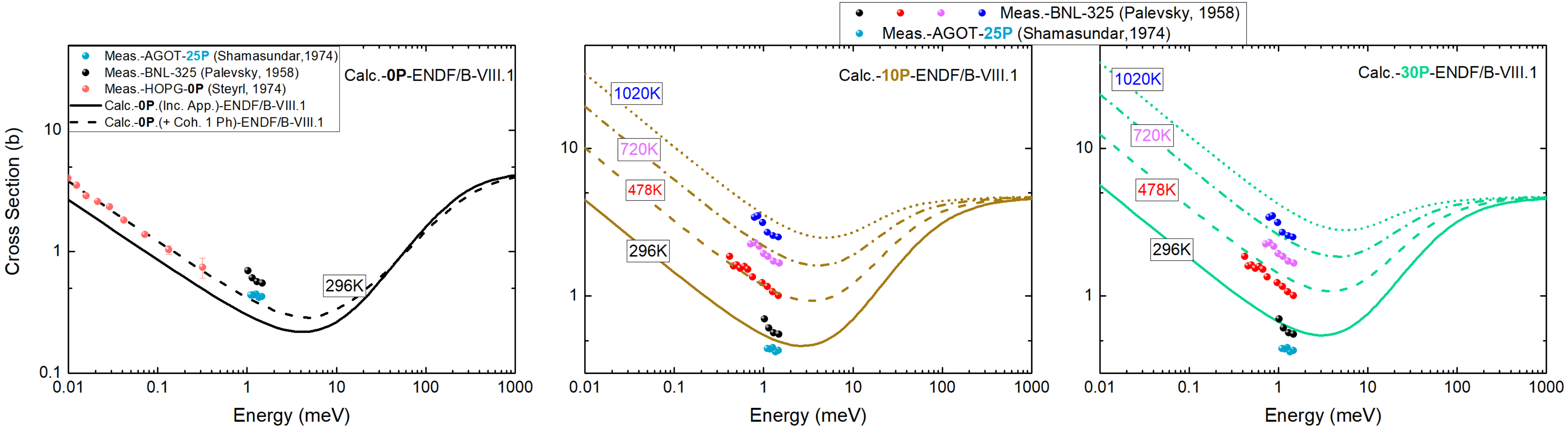




Inelastic Scattering XS







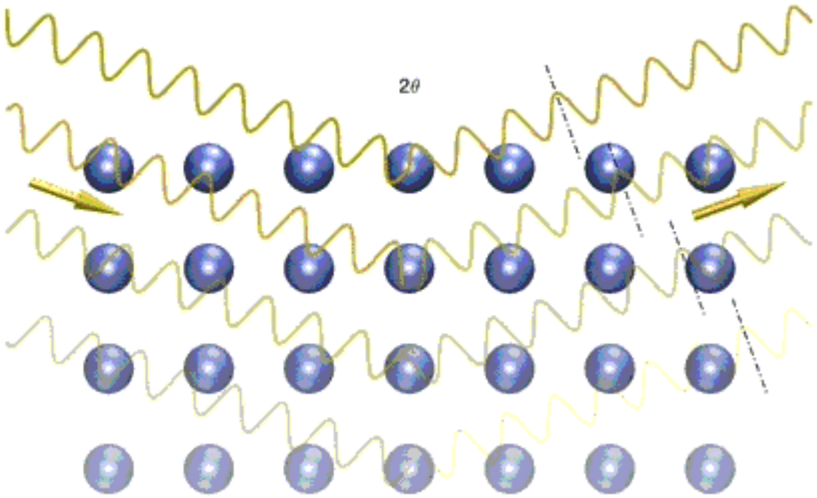
✓ As the temperature increases, both the 10P & 20P ENDF/B-VIII.1cross sections overestimate the measured cross sections of Palevsky.

✓ The higher the temperature, the higher overestimation. This is due to the phono excess in the low energy part of the PDOSs

Coherent Elastic Scattering (Bragg's Scattering)

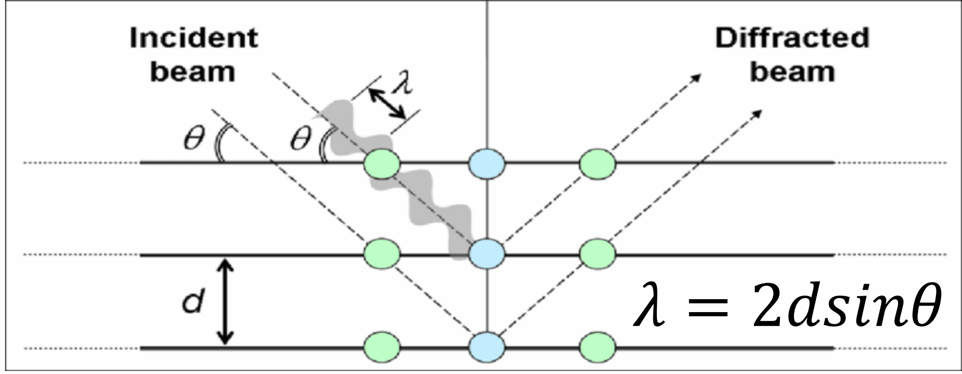
✓ Thermal neutrons have wavelengths (~Å) comparable to the separation distances of atoms in solids.

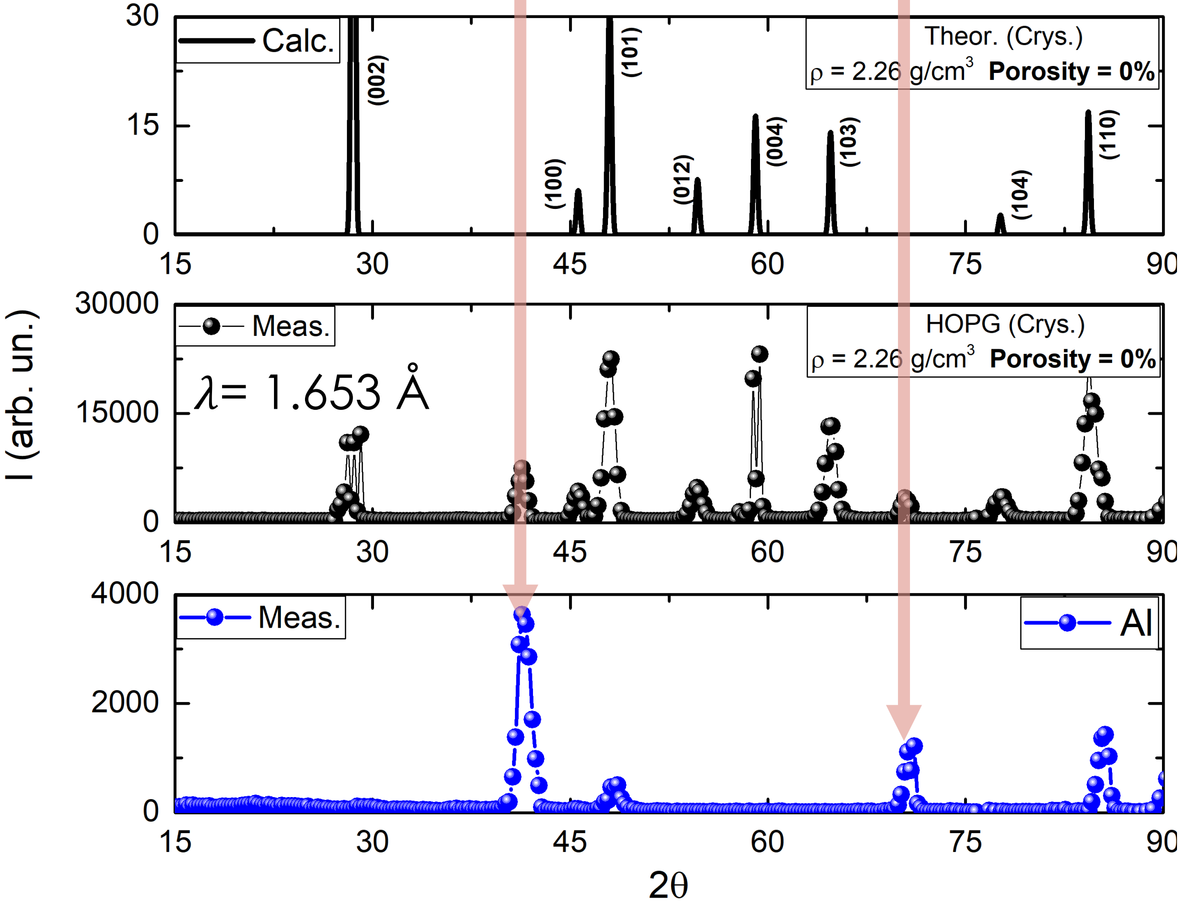
➔ As a consequence thermal neutrons are a useful tool in studying the structure of different scattering systems.

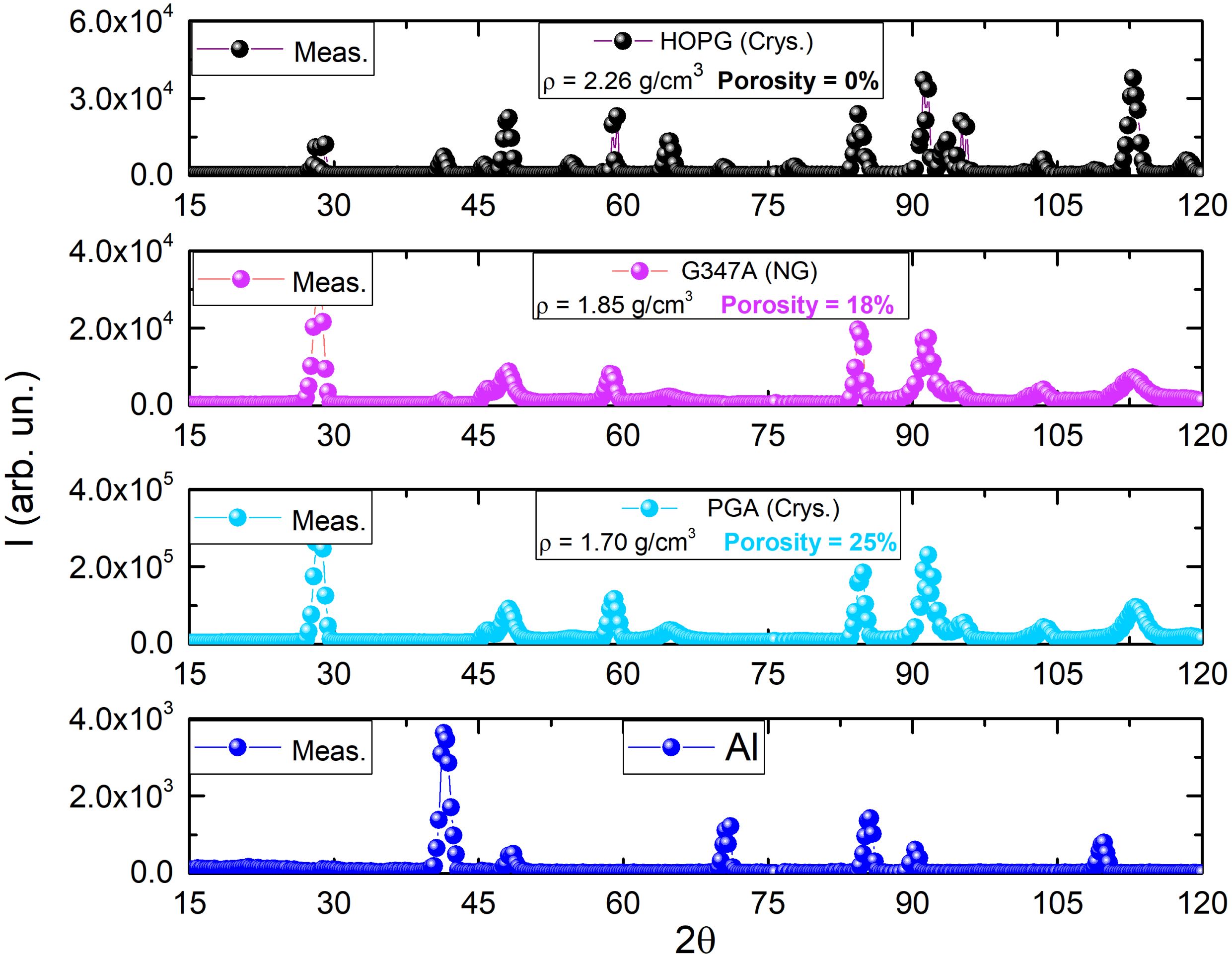


P. Willmott (Paul Scherrer Institute, Switzerland)

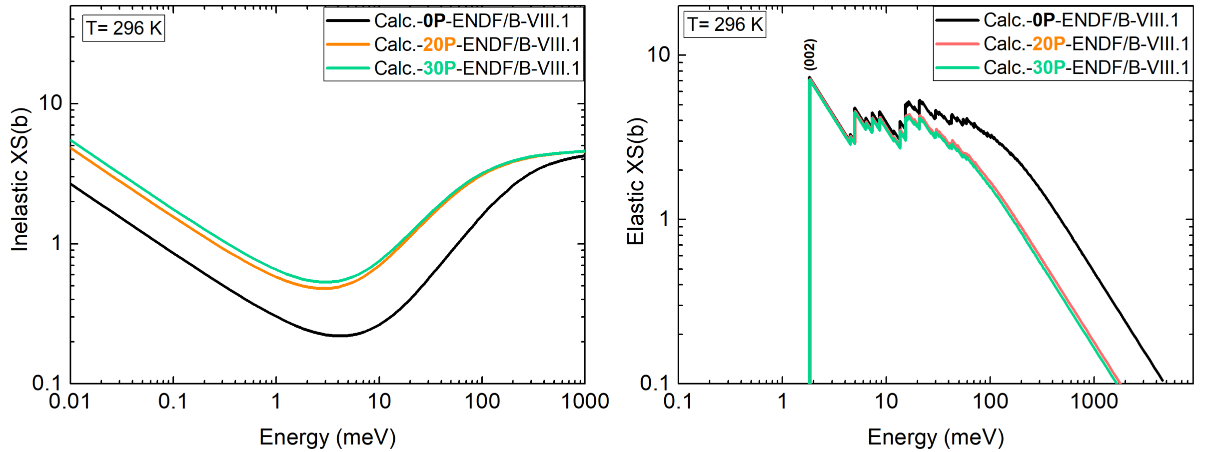
Coherent Elastic (Bragg') Scattering

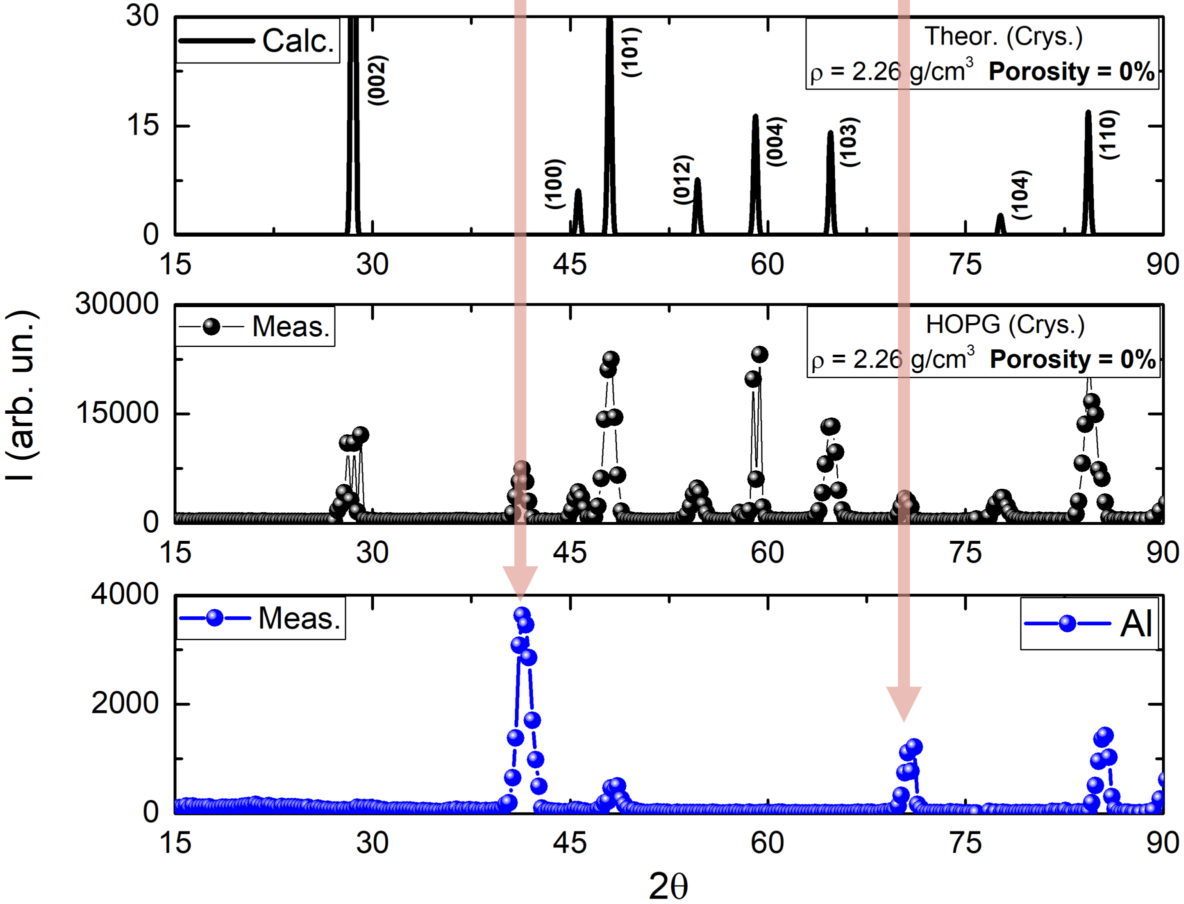


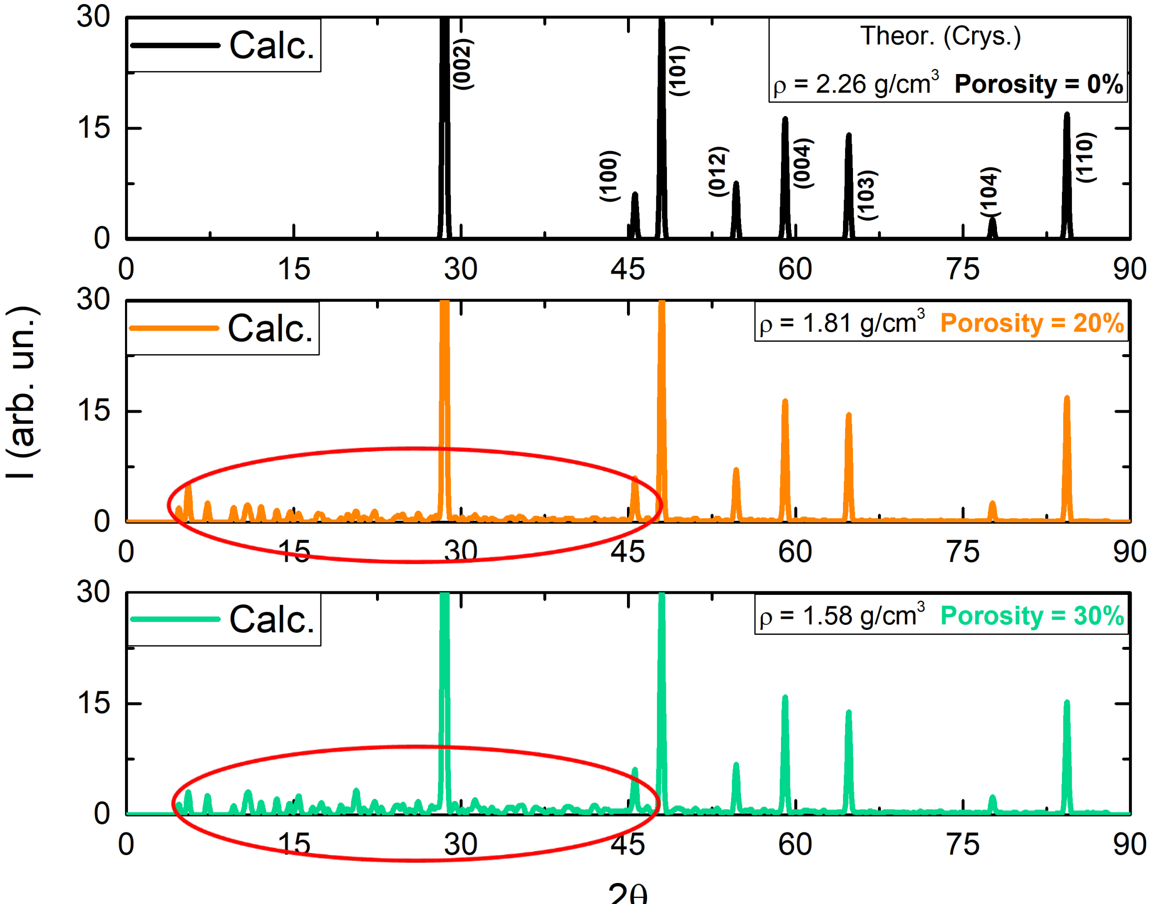




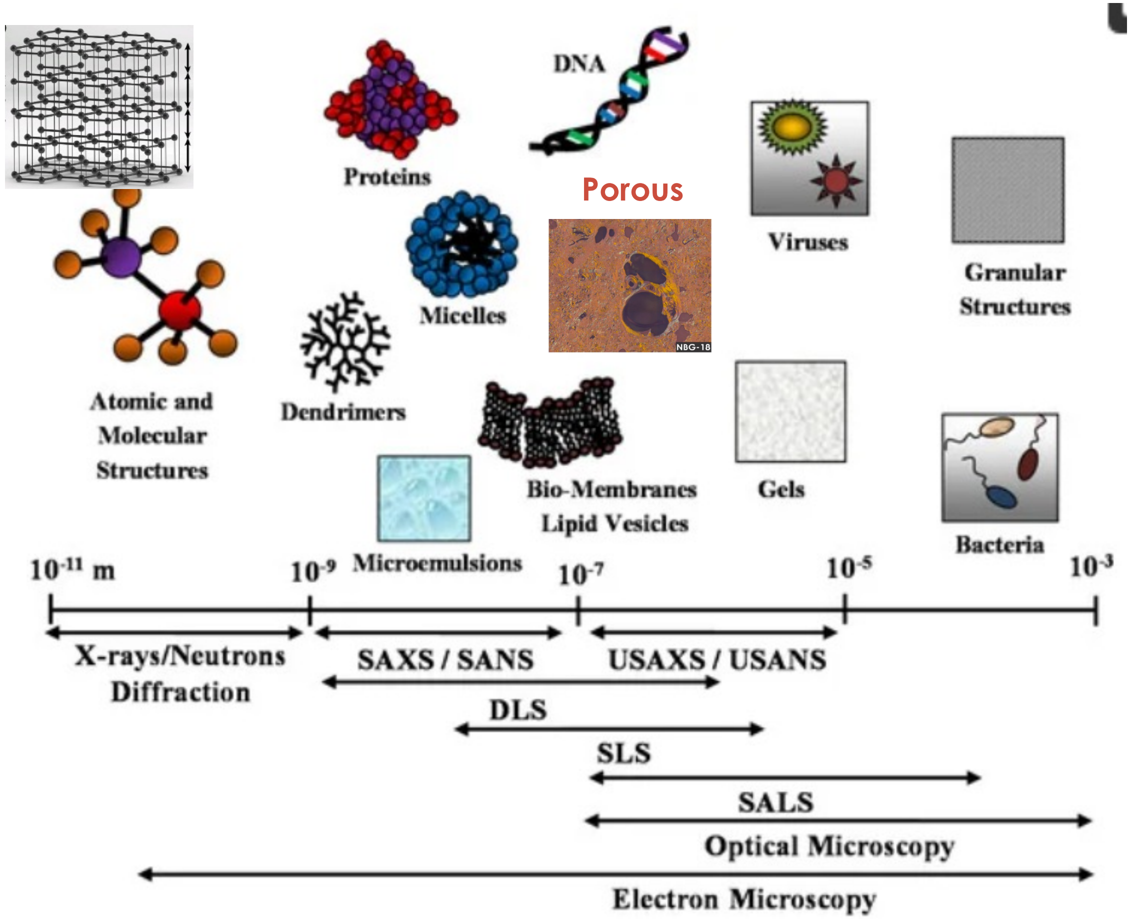
Coherent Elastic (Bragg's) Scattering







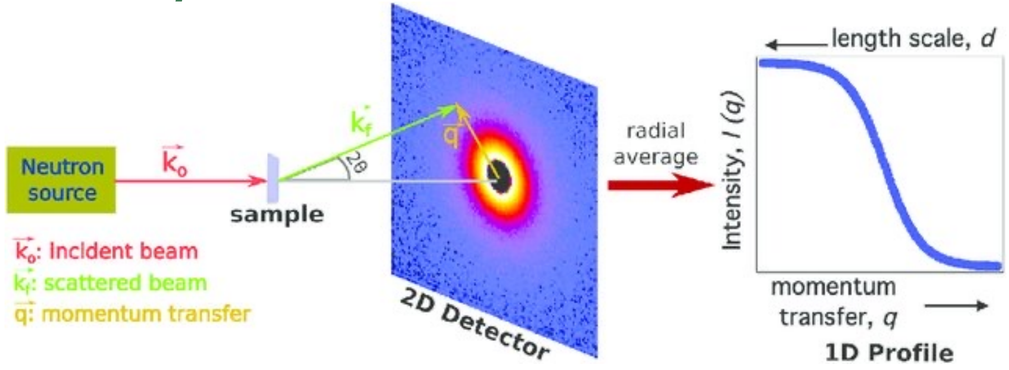
Small Angle Neutron Scattering (SANS)

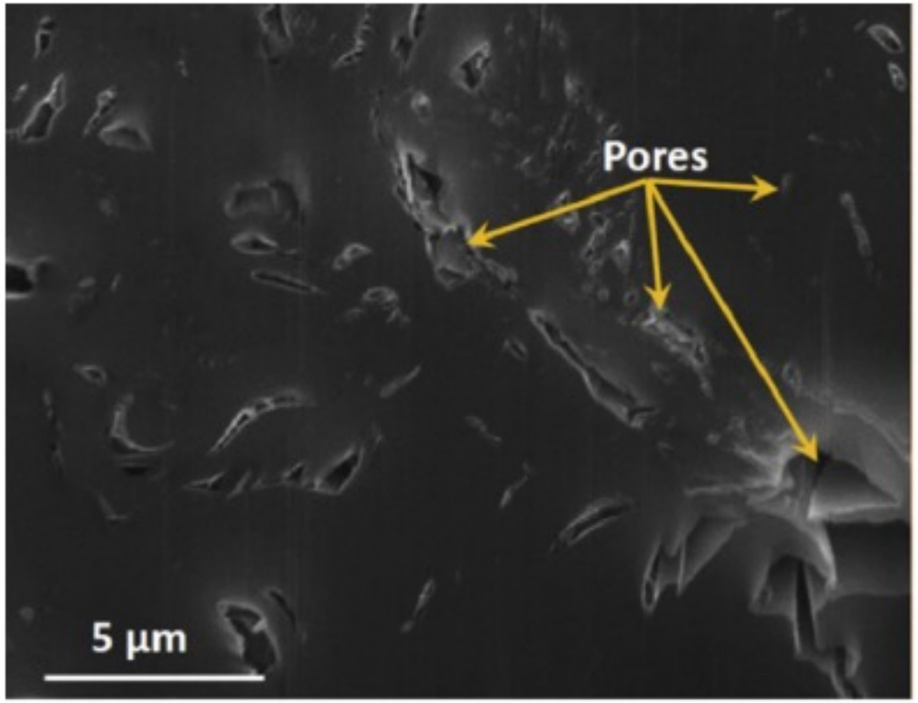


Lombardo et al., Molecules 25, 5624 (2020)

Small Angle Neutron Scattering (SANS)

- SANS is an elastic scattering phenomenon where neutrons **diverges** from their incident beam by a small scattering angle (generally, 2θ < 10 deg), as it penetrates through a sample.
- **SANS** arises from **the inhomogeneous microstructures** (voids, pores, cracks, interfaces, etc) on **mesoscopic** scale covering the length range typically between 10 and 1000 Å which translates into neutron scattering length density (SLD), ρ(r).
- In **nuclear graphite**, SANS occurs on cracks, voids, pores, etc., where the continuous distribution of carbon density is interrupted by defects.





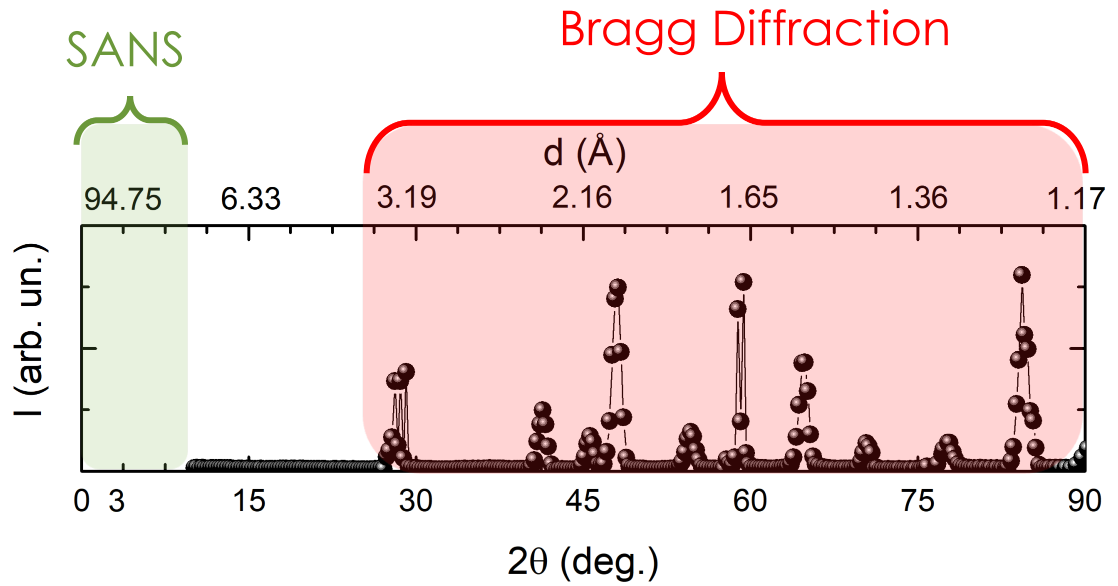
Liu et al., J. Nucl. Mater. ***493****, 246-54, (2017)*

Small Angle Neutron Scattering (SANS)

- The periodic arrangement of atoms in solids (**crystal structure**) can be observed by ~ Å neutrons or X-rays scattered over an angular range of 10 to 120 degrees (**Bragg's Scattering**).

- The **SANS** provides information on spatial inhomogeneities (voids, pores, cracks, interfaces, protein, etc.) with dimensions of 10 to 1000 Å. SANS probes structure on a scale, $d = \frac{\lambda}{2\theta}$

- There are no sharp Bragg peaks in SANS, and only an intensity profile smoothly varying with the scattering angle is obtained



For neutrons with:

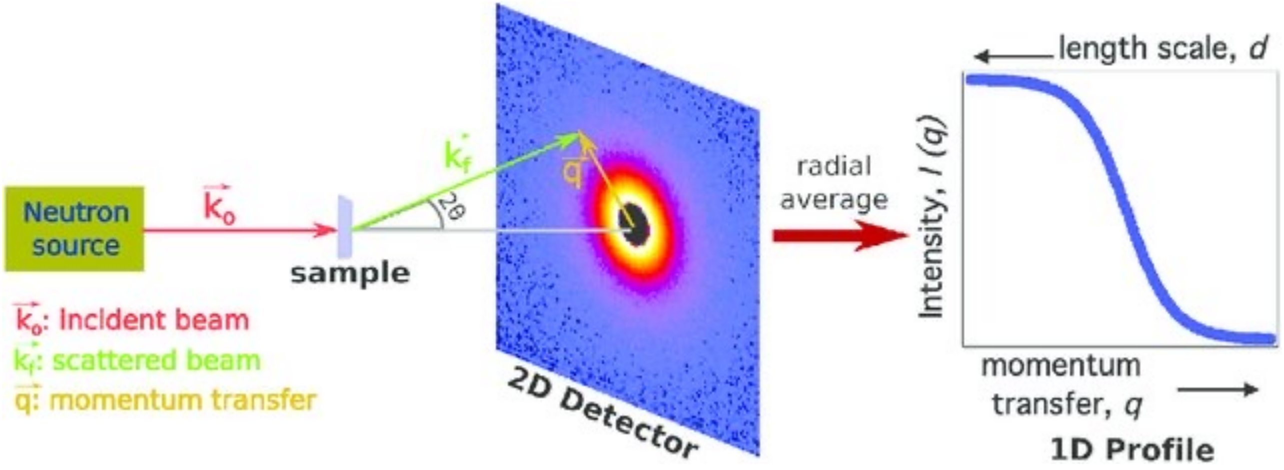
Energy range, $E \in [0.01, 100] meV$

Scatt. Ang range $2\theta \in [3, 6]$ deg

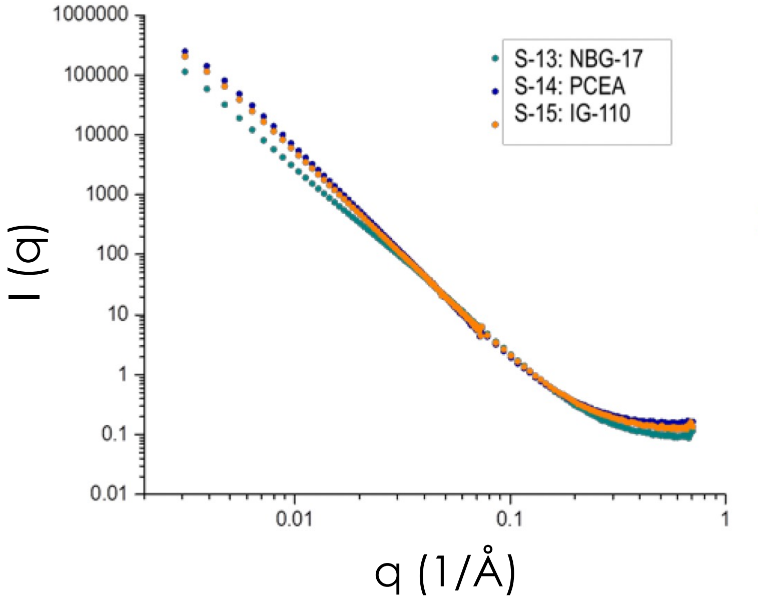
➔ Length Scale $d \in [4, 1250]$ Å

Small Angle Neutron Scattering (SANS)

- The scattered intensity I(q) is measured by a 2D detector. For samples with randomly oriented, contents (e.g., molecules, pores)
- The 2D intensity can be radially averaged to obtain a 1D profile of the scattered intensity I(q).
- For q < 0.2 Å,
  - Linear slope between -3 & -4
  - neutron scattering is caused by large voids, microcracks, and mesopores (30 Å - 1400 Å)

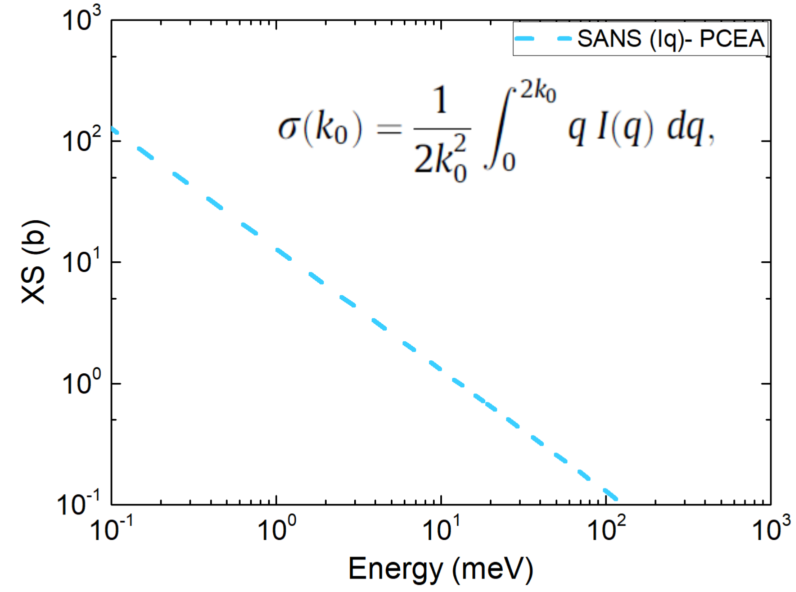


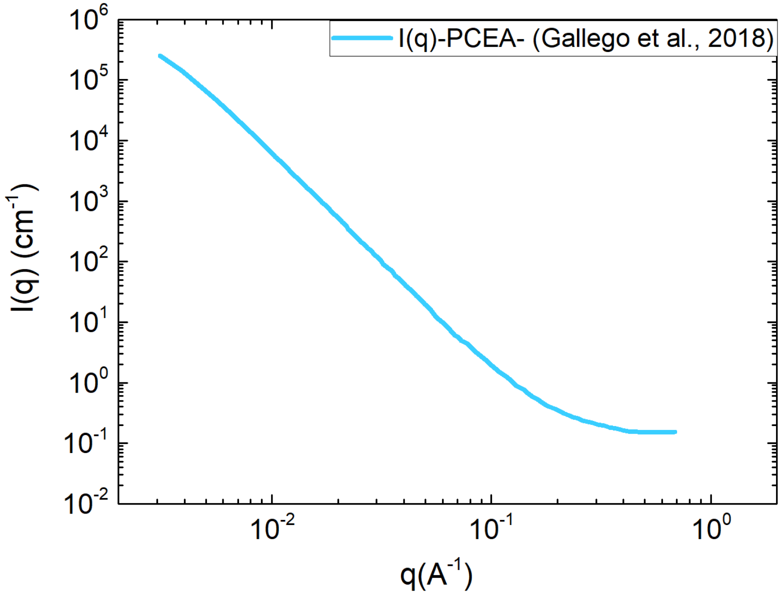
Castellanos et al, Comp. Struc. Biotech. J. **15**, 117 (2017)

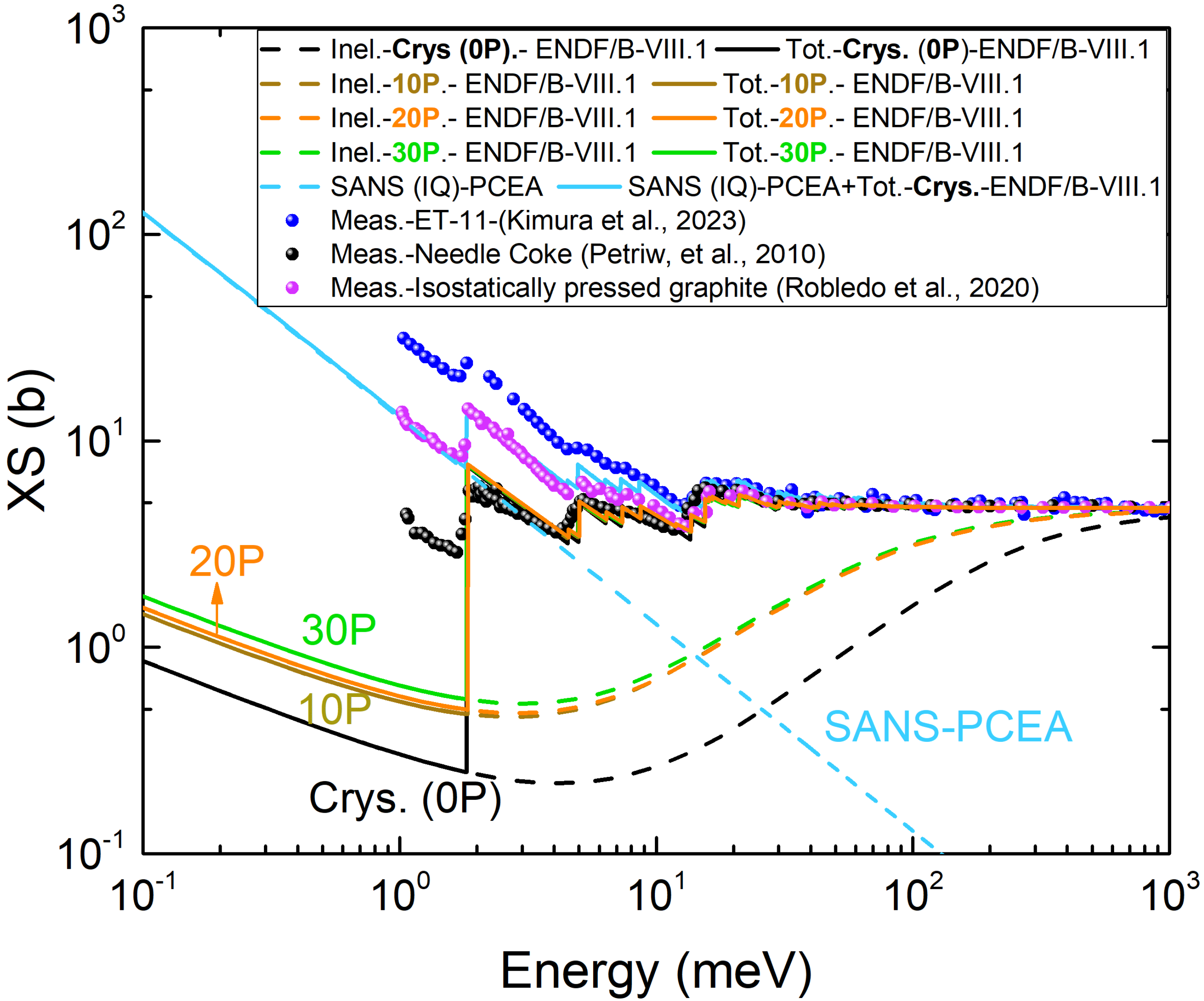


Gallego et al, ORNL/TM-2018/871

Small Angle Neutron Scattering (SANS)







Conclusion

- Excellent agreement is observed between the measured nuclear graphite PDOSs and the calculated PDOS of the crystalline (theoretical) graphite
- The PDOSs of porous graphite in ENDF/B-VIII.1 exhibit noticeable discrepancies when compared to the experimentally measured PDOSs of nuclear graphite. Furthermore, they fail to accurately predict the heat capacity and the inelastic scattering cross sections
- The porous graphite MD models lack the true description of nuclear graphite structure and introduce extra Braggs' peaks below the graphite Bragg edge
- Because of its porous structure, neutrons in in graphite do undergo SANS which results in significant increase in graphite total cross section
- *The high cross section of nuclear graphite cannot be attributed to neutron-phonon interaction,*
- Nuclear graphite is a complicated function of pore size, shape and distribution, Therefor, it essential to carry out both transmission and SANS measurements for a thorough understanding and accurate modeling of thermal neutron interactions.

Thank you