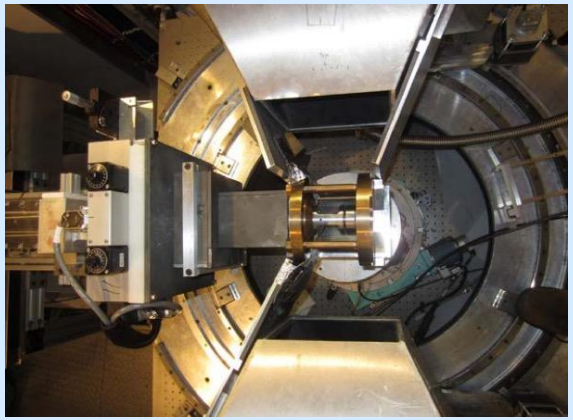
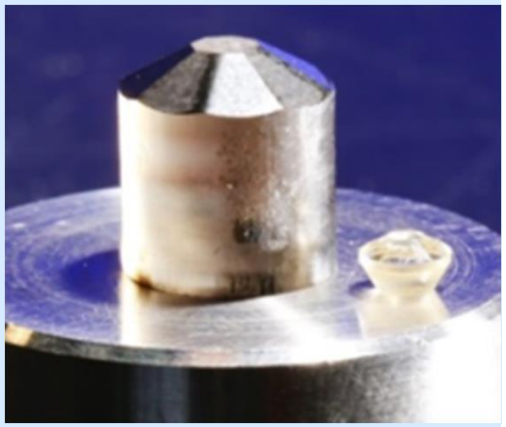
Neutron Scattering Developments and Progress in EFree

Chen Li

Geophysical Laboratory, Carnegie Institution at Washington

Midterm Review, February 5, 2016

U.S. DEPARTMENT OF ENERGY | Office of Science

EFree Energy Frontier Research Center

EFree Team at the SNS

Reinhard Boehler

High P-T Techniques Coordinator

Bianca Haberl

Weinberg Fellow
Oak Ridge National Laboratory
EFree Affiliate

Christopher Tulk

SNAP instrument scientist
Oak Ridge National Laboratory
EFree Affiliate

Malcolm Guthrie

European Spallation Source
EFree Affiliate

Chen Li

Neutron Scattering Coordinator

Technical Coordinators in EFree

Goal: *To facilitate the application of specialized experimental techniques to EFree Projects, and to enable advances in these technical areas to realize center goals.*

Current Technical Objectives – *Neutron Scattering Coordinator*:

Technique Development

- Further improve the high pressure neutron diffraction capacities at SNS
- Develop, test, and improve the large volume DAC for inelastic neutron scattering
- Further development of high pressure synthesis capabilities
- Expand high pressure capacity at other beamlines at SNS and other facilities to facilitate EFree objectives

Science Projects

- Support existing center-wide EFree projects using existing and new high-pressure neutron scattering techniques
- Proposal writing, experiment design, preparation, execution, and data analysis
- Conduct exploratory research in line with EFree objectives

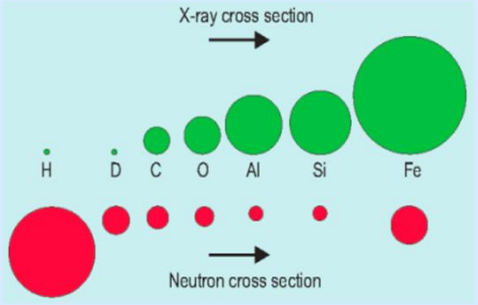
Neutron scattering advantages and its applications for materials under extreme environments

- Neutrons have distinct isotope dependent cross sections: very sensitive to light elements
- Neutrons have spins: direct magnetic properties measurements
- Accurate measurements of subtle structure distortions
- Energy-resolved area detectors: accurate inelastic measurements
- Excellent penetration: bulk properties and imaging

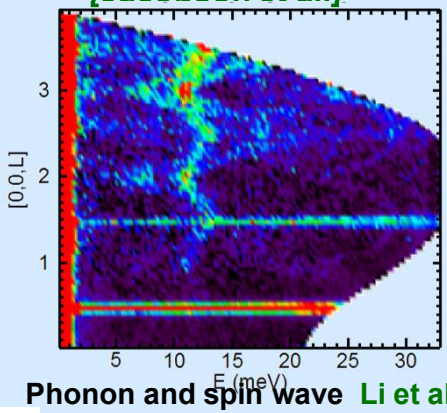
***Applications*:**

- Hydrogen bonding under extreme conditions
- Disordered systems at high pressure and temperature
- Structural and transport studies in functional materials such as thermoelectrics and ferroelectrics
- Magnetic structure and pressure-induced effects in non-conventional superconductivity

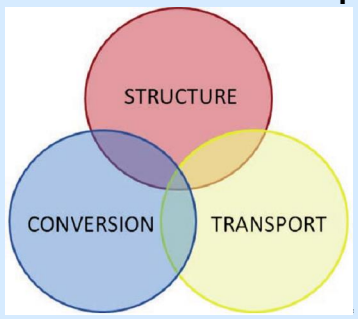
➢ ***Structural, dynamical, and transport properties***

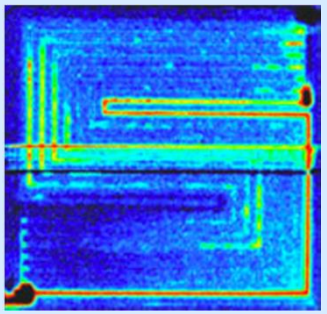


[Jacobson et al.]



Phonon and spin wave Li et al.





EESC Lab
UTK

Neutron imagining

US Neutron Scattering User Facilities

Spallation Neutron Source (and Center for Nanophase Materials Sciences)

High-Flux Isotope Reactor

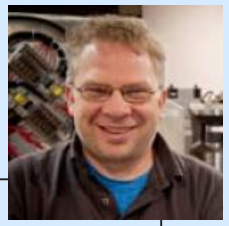
NIST Center for Neutron Research

Neutron scattering represented a major investment of EFree in its first five years EFree

- Custom cells for neutron diffraction reached megabar pressures:
  - *Pressure range extended by 4x*
  - *D_2O ice to 94 GPa on SNAP*

Reinhard Boehler

Malcolm Guthrie



Christopher Tulk

High Pressure Research, 2013
Vol. 33, No. 3, 546–554, http://dx.doi.org/10.1080/08957959.2013.823197

Taylor & Francis
Taylor & Francis Group

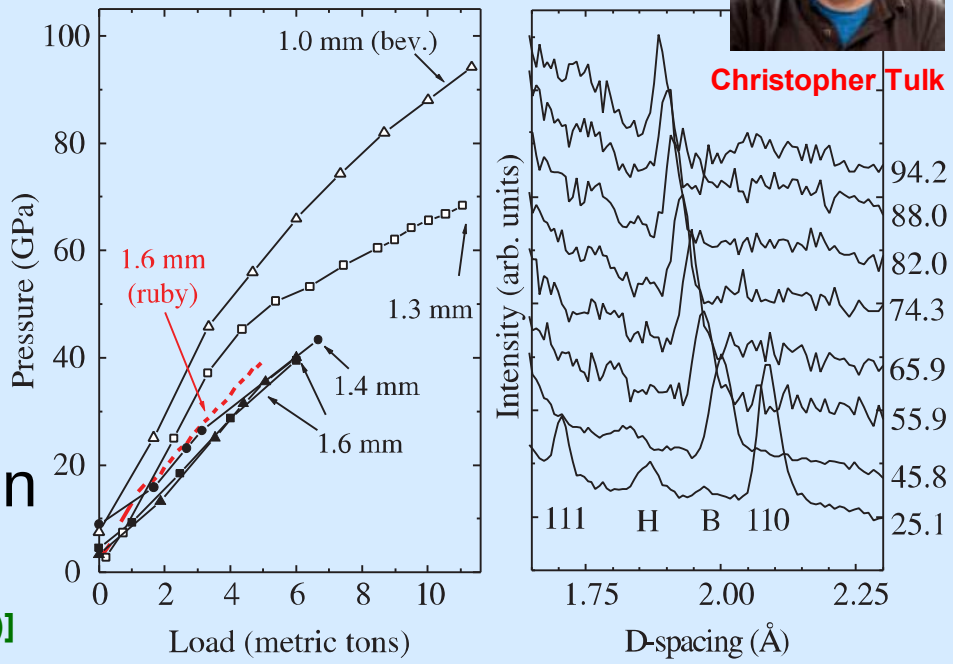
Large-volume diamond cells for neutron diffraction above 90 GPa

R. Boehler[a]*, M. Guthrie[a], J.J. Molaison[b], A.M. dos Santos[b], S. Sinogeikin[a,c], S. Machida[a], N. Pradhan[b] and C.A. Tulk[b]

[a]*Geophysical Laboratory, Carnegie Institution of Washington, Washington, DC 20015, USA;*
[b]*Neutron Sciences Directorate, Oak Ridge National Laboratory, Oak Ridge, TN 37831, USA;*
[c]*HPCAT, Advanced Photon Source, Argonne National Laboratory, Lemont, IL 60439, USA*

(*Received 5 June 2013; final version received 4 July 2013*)

Top ten of the Taylor & Francis Material's best 2013



- Discovery of interstitial proton ordering in dense ice

[M. Guthrie et al., *Proc. Nat. Acad. Sci.* 110, 10552 (2013)]

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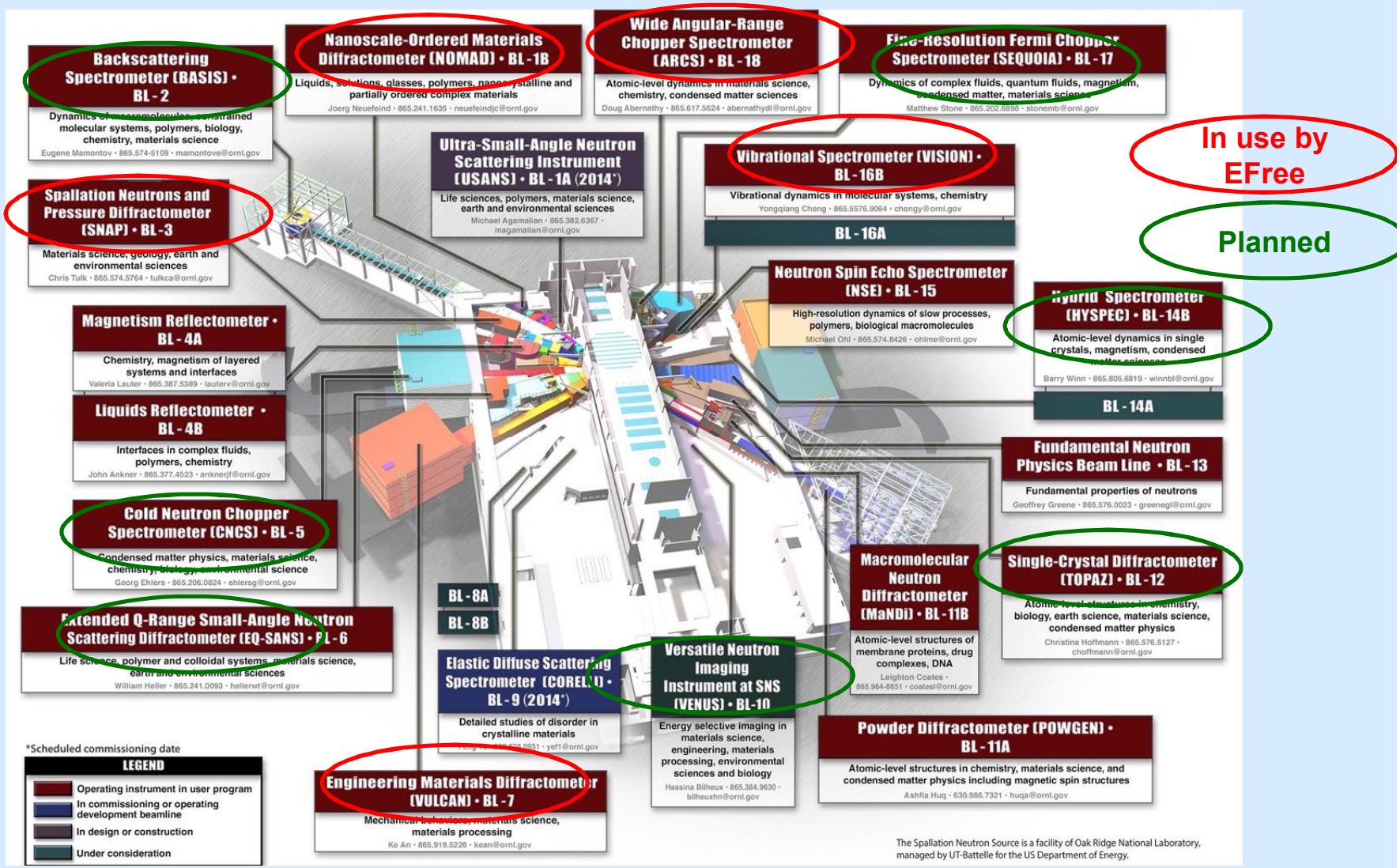
Spallation Neutron Source and EFree



Technique development for high-pressure neutron diffraction is a major goal of EFree

SNAP (Spallation Neutrons and Pressure)

- High-flux, medium resolution, focused neutron diffractometer for powder, single-crystal, and amorphous materials under pressure

Conventional high pressure cell

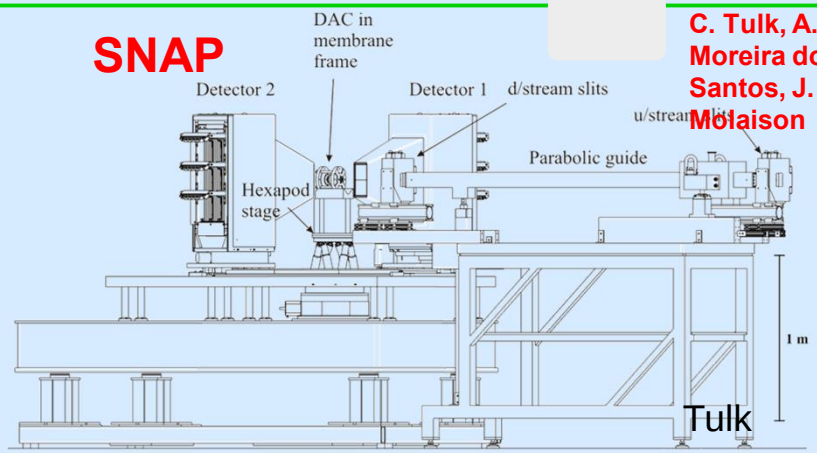
- PE cell sample volume 5.5 mm^3, up to 25 GPa
- Extremely valuable in sample synthesis

EFree large volume diamond anvil cell

- Sample volume 1.5 mm^3, up to ~100 GPa
- Significantly improved background, fast turn around
- In recent cycles, more than half of user proposals are based on these cells.

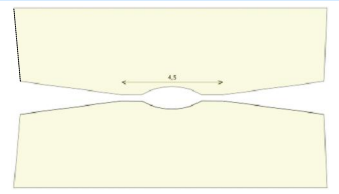
SNAP

C. Tulk, A. Moreira dos Santos, J. Molaison

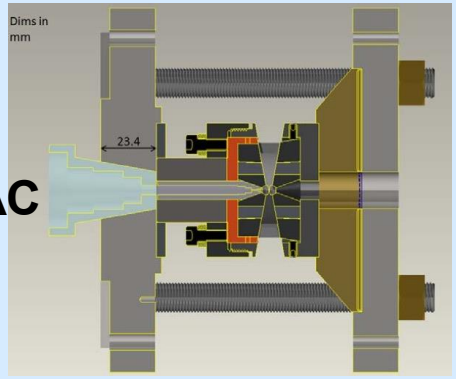


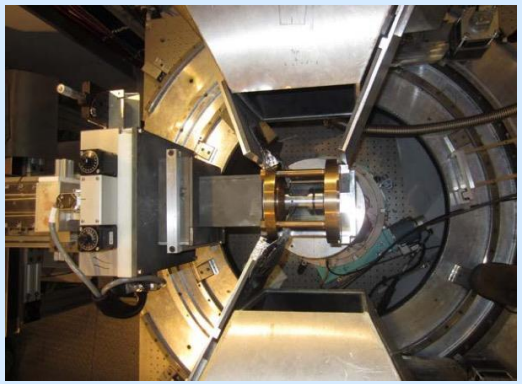
PE Press

Dimple Cell



DAC





[R. Boehler et al., *to be published*]

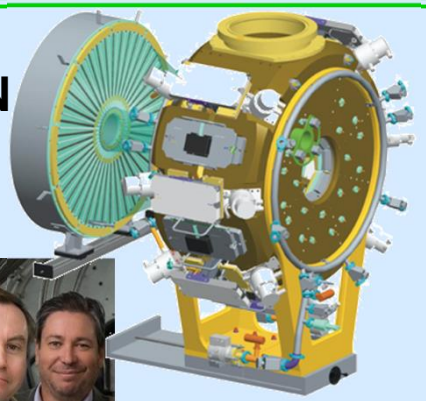
Technique development for high-pressure inelastic neutron scattering is a new thrust

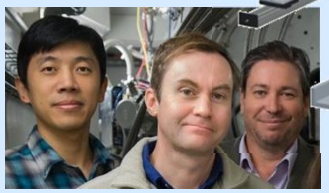
VISION: worldôs first high throughput indirect geometry spectrometer with high flux, large bandwidth (-2~1000 meV), and good resolution (1.5%).

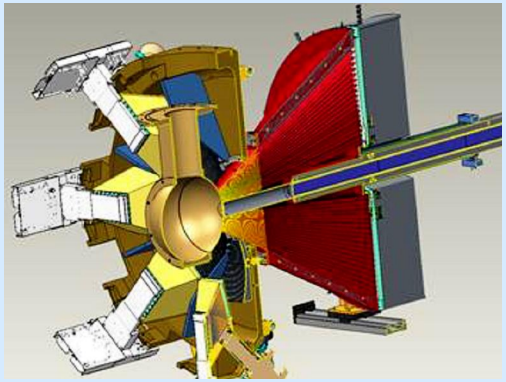
Extra large volume DAC designed for neutrons uses the largest single crystal diamonds for such applications. Shields and collimators are designed to minimize the background.

Several preliminary proof-of-principle experiments had been performed showing **excellent results** on a sample of 1.6 mm^3 in size. More are planned for additional characterization and improvements.

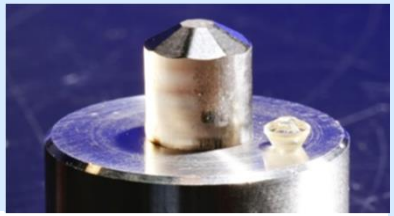
VISION

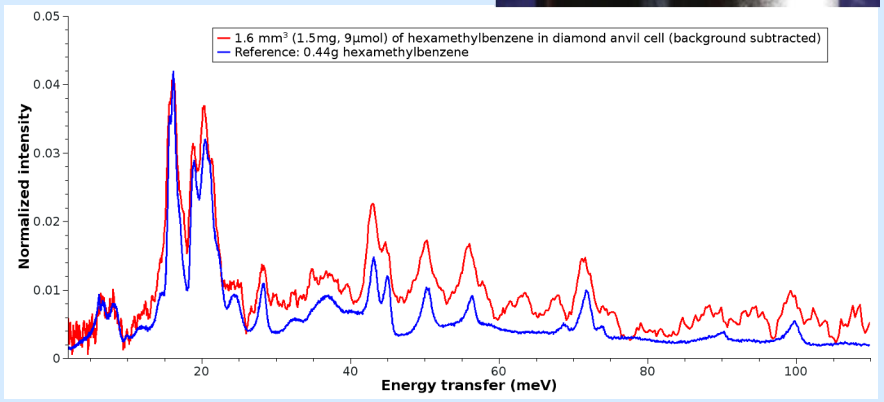






Y. Chen, L. L. Daemen, and A. J. Ramirez-Cuesta,
Extra large volume DAC designed for VISION using 11-ct diamonds





[R. Boehler, C. Li, B. Haberl, L. Daemen, Y. Cheng, A. Ramirez-Cuesta, *in progress*]

Other EFree Technique Development at SNS

Conventional piston-cylinder (clamp) cell

- Up to 3 GPa (limited by cell material)
- Large sample volume up to 10 cm

***Cells pre-stressed with CuBe** wire will be able to*

- Achieve much high pressure (up to 10 GPa)
- Or, achieve significantly less background

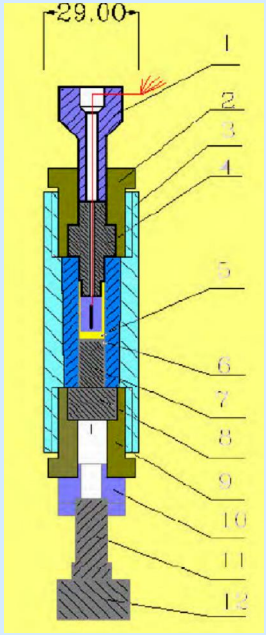
Applications will be direct geometry spectrometer such as CNCS, ARCS, SEQUOIA, HYSPEC and future instruments at STS

*Using **DAC for NOMAD** (Nanoscale-Ordered Materials) diffractometer*

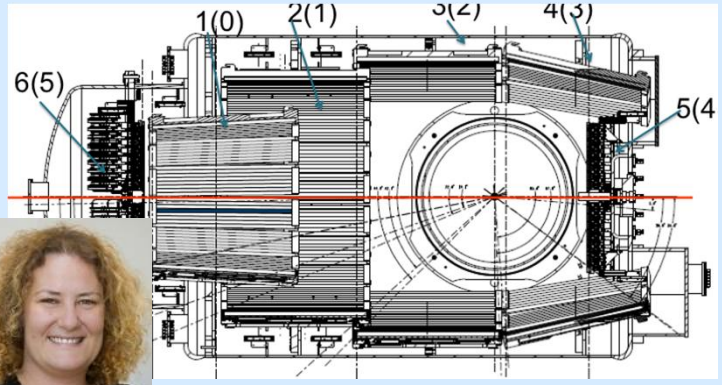
- Large bandwidth for structure determination of local order in crystalline and amorphous materials.
- Increase the current pressure limit of amorphous materials by a factor of 5 to 50 GPa
- Polymers, nanostructures: long range orders

Reinhard Boehler

Clamp cell



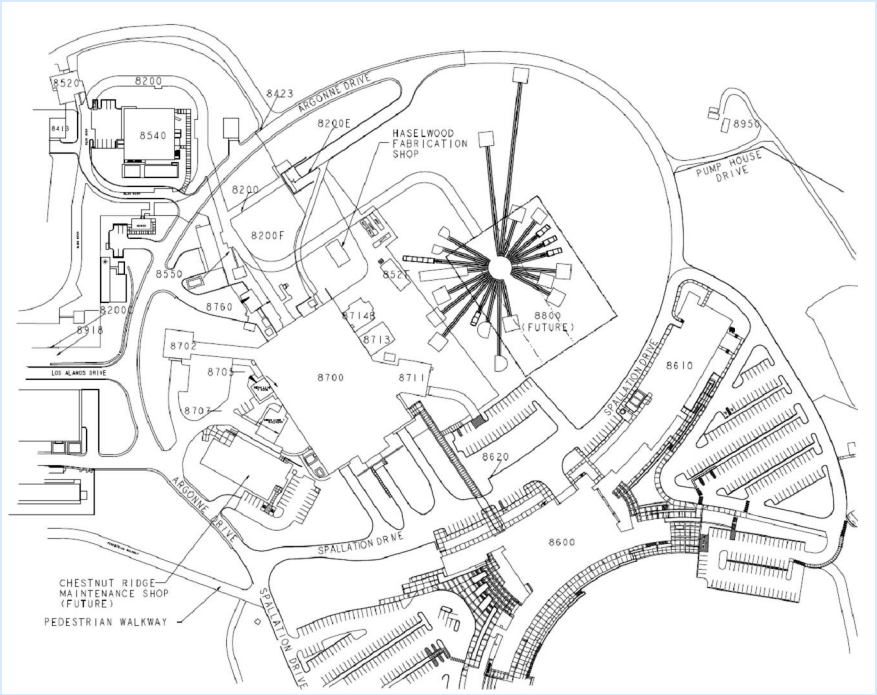
NOMAD

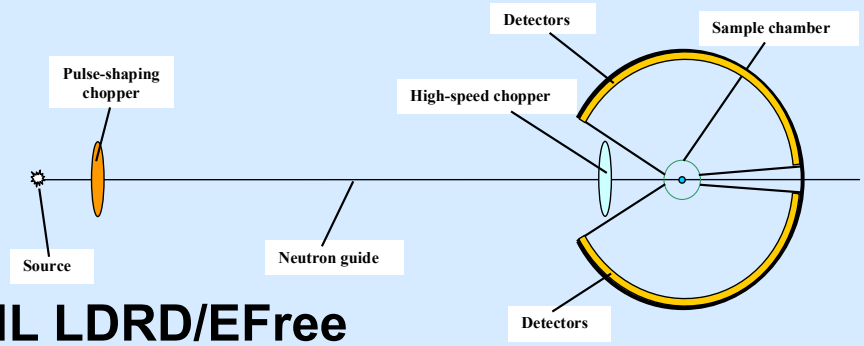


Bianca Haberl

EFree technique development is supporting the Second Target Station project at the SNS

- **Second Target Station (STS):**
  - Fourth generation neutron facility optimized for highest cold neutron peak brightness
- **ORNL LDRD**
  - ñ*ORNL is investing directly in challenge experiments designed to demonstrate the capabilities that will be routinely available at STS for addressing the science of tomorrow…establish infrastructure leading to inelastic measurements at extreme pressures of 40 GPa and above…*"
- **Chopper Spectrometer for Small Samples (CHESS)**:
  - Materials under most extreme conditions
  - Goal: measurements to >100 GPa
- **Boehler now jointly supported by ORNL LDRD/EFree**





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Structure and Vibrations of Carbon Nanothreads

Nanophase Carbons Project

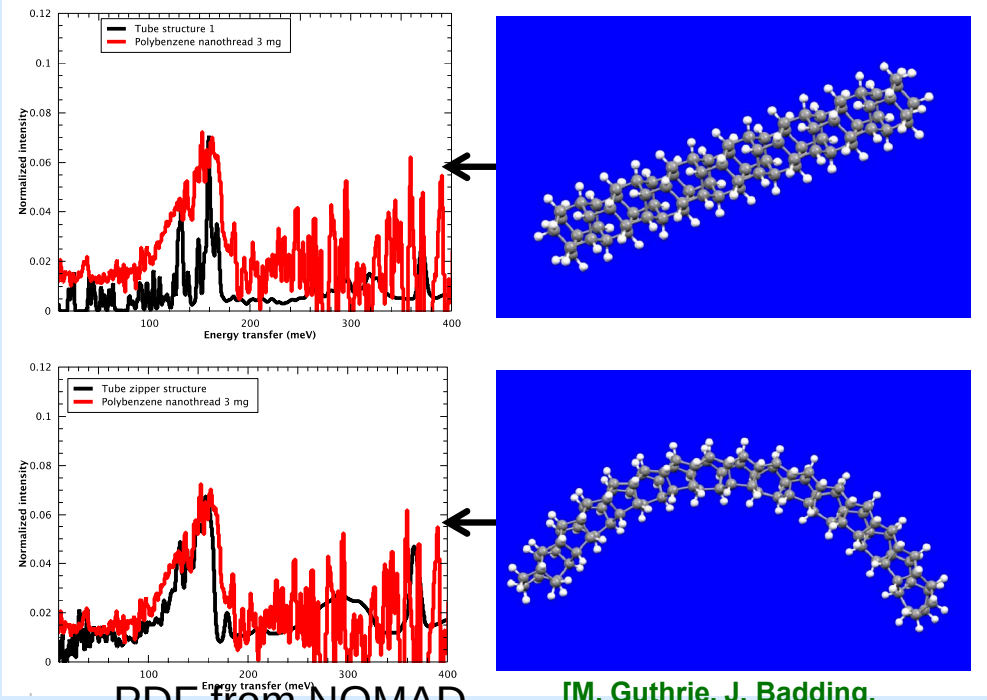
- **VISION inelastic measurements** provided the full vibrational density of states of on 3 mg of nanothreads!
- Provide another way for constraining atomic structure (paired with DFT)
- More experiments are planned on larger samples (VISION beamtime awarded)

- **NOMAD PDF measurements** provide more direct structural information.
- More experiments planned (NOMAD beamtime awarded)

- In-situ experiment performed at **PLANET** at J-PARC by Guthrie et al.

- PE cells at **SNAP** are used for sample synthesis by students

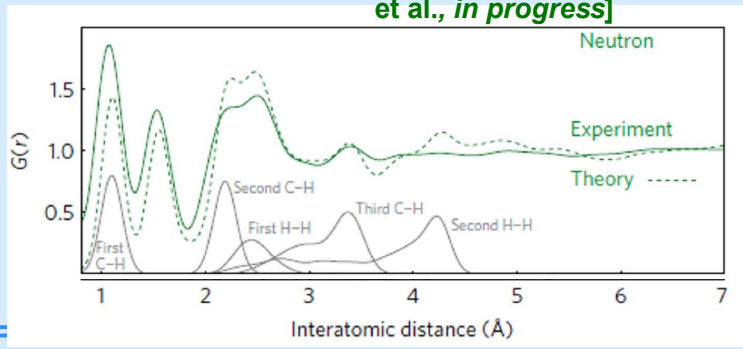
VISION spectra vs DFT simulation



[M. Guthrie, J. Badding, et al., *in progress*]

PDF from NOMAD

Xiang Li



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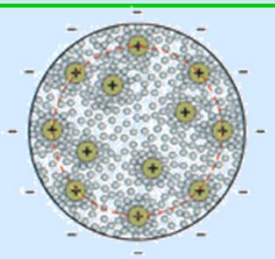
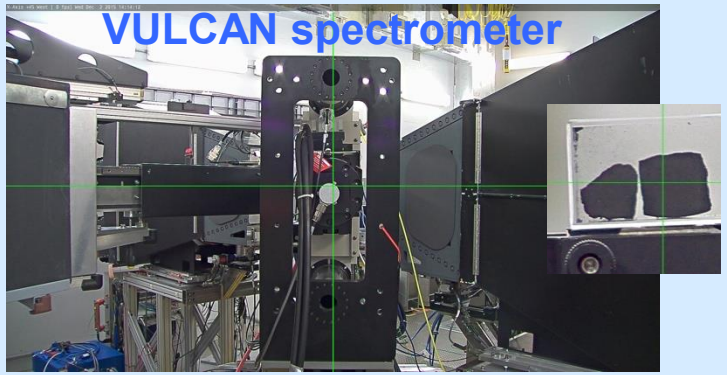
Electrolyte Infiltration in Porous Carbon

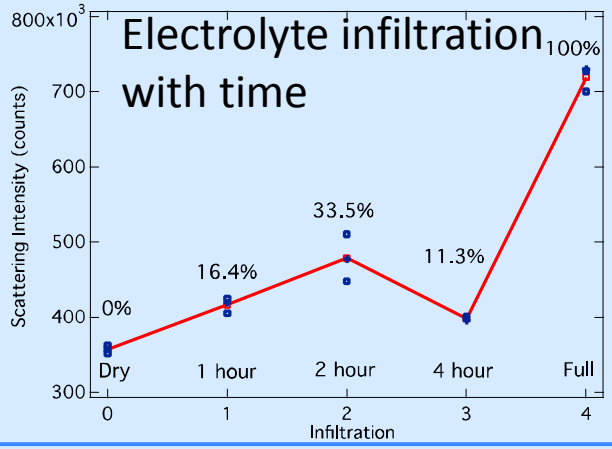
Porous Materials Project

- The goal is to study of the infiltration efficiency and the diffusion kinetics of electrolyte ions into porous electrode (used in super capacitors)
- Preliminary tests on **VULCAN** diffractometer proved the idea and showed promising results
- Follow-up experiments will be performed in-situ for more consistent and accurate results (beamline is secured through beamline development time)
- Ultimate goal is performing such measurement in working super capacitors using neutron imaging
- Small angle neutron scattering (**SANS**) will be able to provide with us the information on the spatial distribution of the electrolyte in porous electrodes

Cong Liu Yiqun Liu







[K. Landskron, C. Liu, Y. Liu, C. An, and C. Li, *in progress*]

Vibrational Properties of New Si Allotropes

Solar Materials Project

- Phonon DOS of silicon allotropes measured with 40 and 75 meV incident energies on ARCS.
- Diamond-structure silicon: 121 mg; BC8 silicon: 10 mg

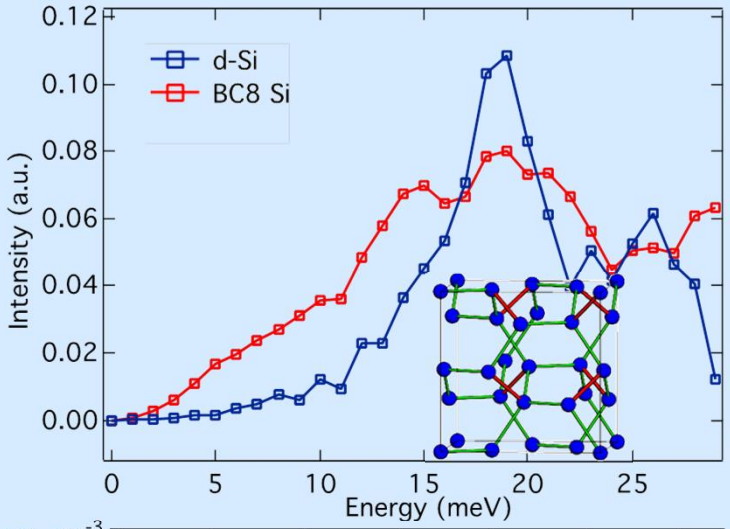
Small sample size works for direct geometry TOF spectrometers

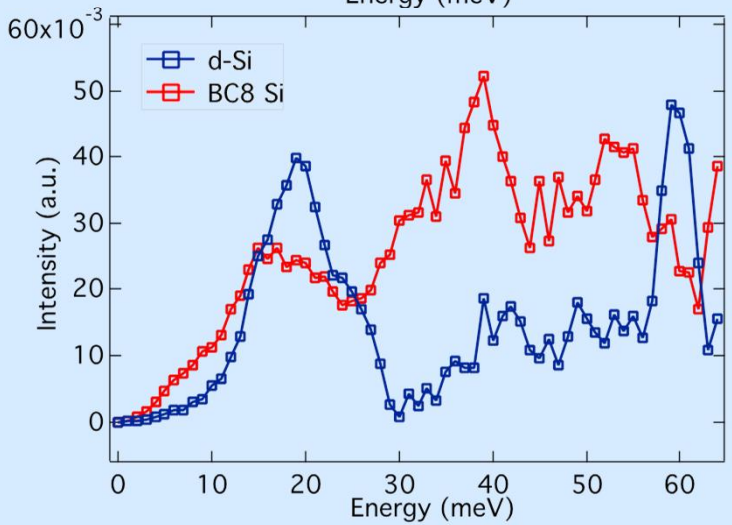
- Follow-up experiment planned on ARCS BC8 and Si_{24} (beamtime approved)
- Plan for VISION experiment is discussion

Timothy Strobel

Haidong Zhang

[T. Strobel, H. Zhang, D. Abernathy, and C. Li, *in progress*]





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Energy Frontier Research Center

Hydrogen Diffusion In Energy Materials

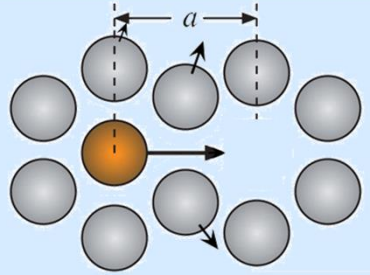
Ion Transport Project

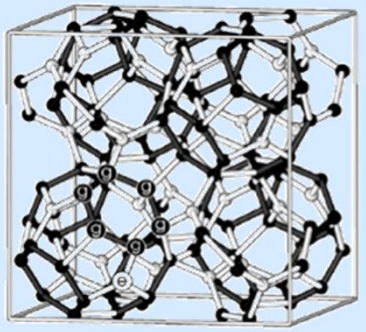
- Hydrogen doped YFe_2 is chosen for quasi-elastic neutron scattering (**QENS**) for its conveniently low activation energies at low temperatures (activation energies of 42 and 10 meV in the ranges of 295-390 K and 140-240 K)
- Possible change from an adiabatic to a non-adiabatic mechanism at low temperatures.
- Experiment for **DCS** at NIST Center for Neutron Research last week using gas pressure cell
- Future experiment plans for measurements on **backscattering spectrometer** at SNS (BASIS)

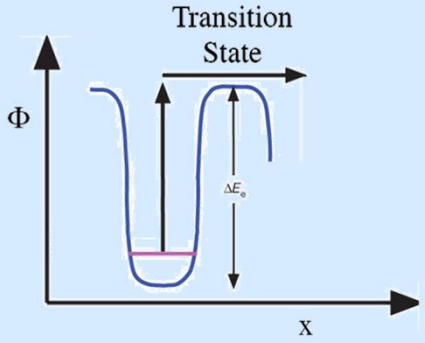
Brent Fultz

Hillary Smith

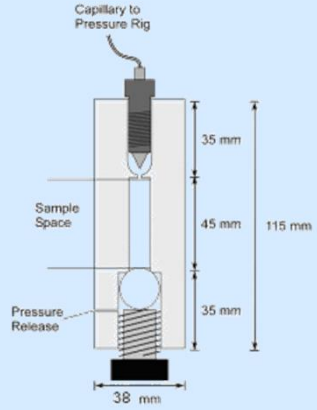
Max Murialdo







Spatial arrangement of hydrogen interstitial sites in cubic C-15 type AB_2



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High Pressure Structures of Novel Hydrides

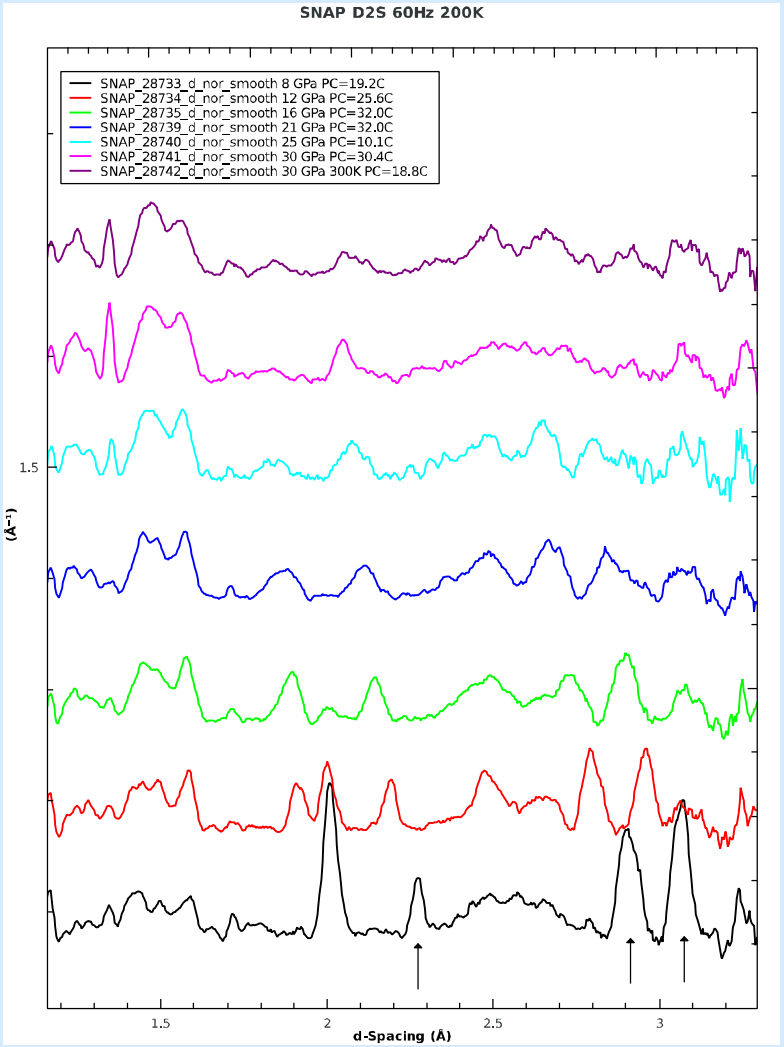
Electron Transport Project

- There are prediction and possible observation of high temperature superconductivity in simple hydrides, whose structures are still poorly understood
- Fully characterizing the structural properties of the lower pressure precursor phases of these materials using neutron scattering will help to determine the structure and dynamics of the H(D) sublattice of these materials
- Neutron diffraction experiment of D_2S on **SNAP** up to 30 GPa at 200 K was successful. Interesting behavior was observed
- More experiments on D_2S and other hydrides are planned (beam time granted)

[M. Somayazulu, *et al., in progress*]

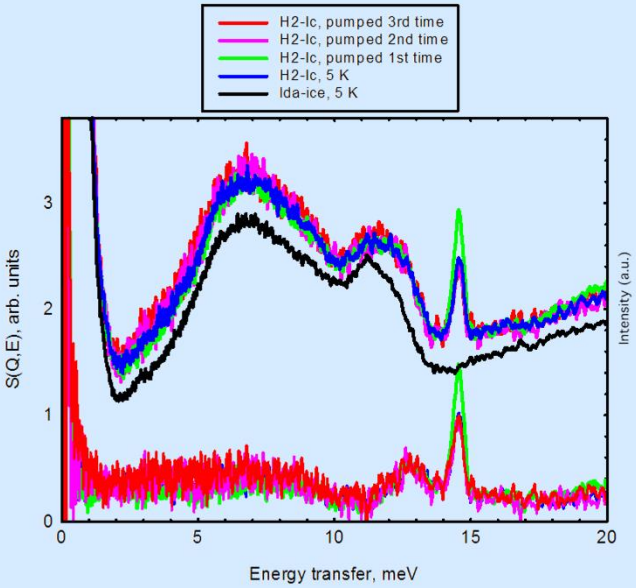
Maddury Somayazulu

Muhtar Ahart

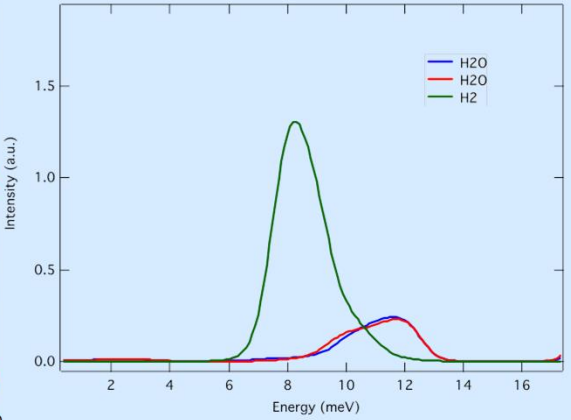


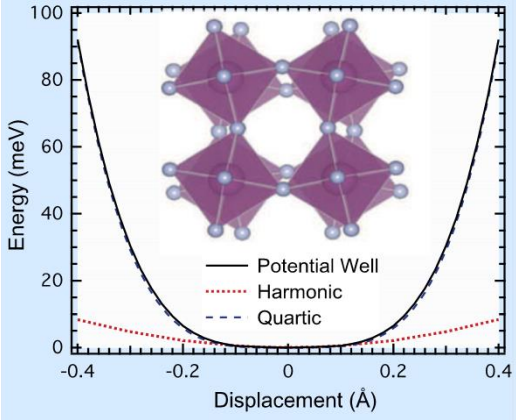
Selected Exploratory Projects

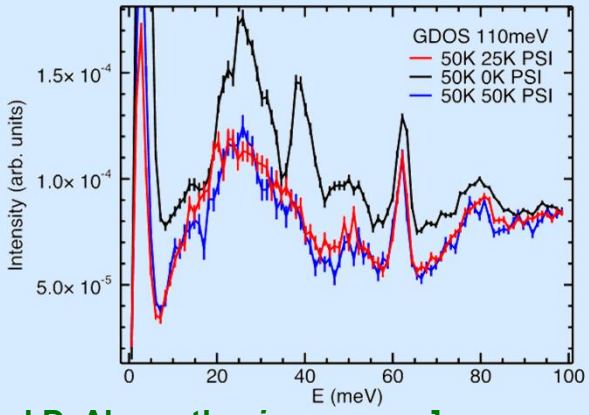
- INS spectra measured at VISION at 5 K for initial Ida-ice, and **H_2** in ice Ic
- The shape and intensity of the peak at 12.8 meV is very similar for all H_2-ice Ic samples, and relates to rattling vibration of H_2 in the voids of structure according to simulation



[C. Li, C. Tulk, and A. Kolesnikov, *in progress*]







[C. Li, B. Fultz, and D. Abernathy, *in progress*]

- **Negative thermal expansion** ScF_3 has strong anharmonic phonons
- TOF inelastic neutron scattering measurements performed on ARCS in helium gas cell at 0, 25, 50 kPSI and 50, 150, 300 K
- Strong dependence on both pressure and temperatures

Other EFree-related energy research is taking place at SNS

Bianca Haberl

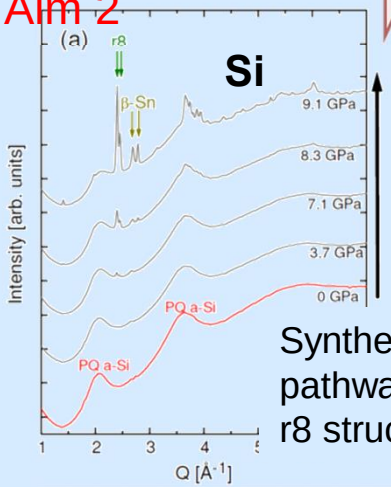
Specific Aim 1:
Understand the behavior of amorphous Group IV elements C, Si and Ge under extremes using neutron scattering.

Specific Aim 2:
Design and control synthesis pathways through tailored high pressure application and suitable precursor materials.

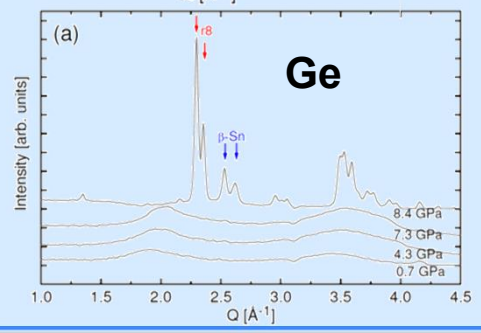
Specific Aim 3:
Transfer synthesis pathways to technologically more viable techniques such as indentation or also deposition.

Overarching Goal:
Enable controlled synthesis of new, functionally useful Group IV materials in a technologically useful manner.

Aim 2

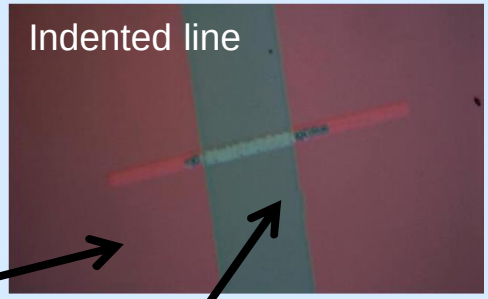


Synthesis pathway for r8 structure



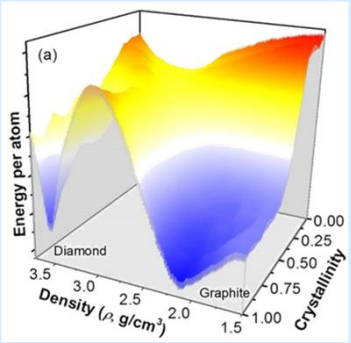
Aim 3

Nano-scaled device utilizing indentation



conductive c-Si insulating a-Si

Access to novel, inert and potentially supertough structures of carbon through in depth understanding and control over the free energy landscape



*This research is funded through an Alvin M. Weinberg Fellowship to U$ 116,343, a funding through the ORNL LDRD scheme under Project No. 7620.

In collaboration with the Solar Materials project

Goals Going Forward

Techniques

- Routine and reliable neutron diffraction up to 50 GPa with improved cell design, gasket materials, scattering geometry. Low background and reliable data reduction
- Inelastic neutron scattering up to 20 GPa using extra large volume diamond anvil cell

Science

- Structural and vibrational properties of materials with energy applications under high pressure
- Transport properties of energy materials, including *in situ* high pressure measurements

Conclusions

- **Neutron scattering is an invaluable tool for understanding structural and transport properties of energy materials**
- **The effort is crucial for realizing EFree project goals**
- **The EFree-SNS partnership has advanced neutron scattering and greatly benefited the US and international community**
- **Significant progress is being made to address the DOE-BES Grand Challenges through high pressure neutron scattering**

Thank you!

Outreach

- Neutron Day
- High Pressure Interest Group Seminar Series
- Wigner Seminar by Roald Hoffmann