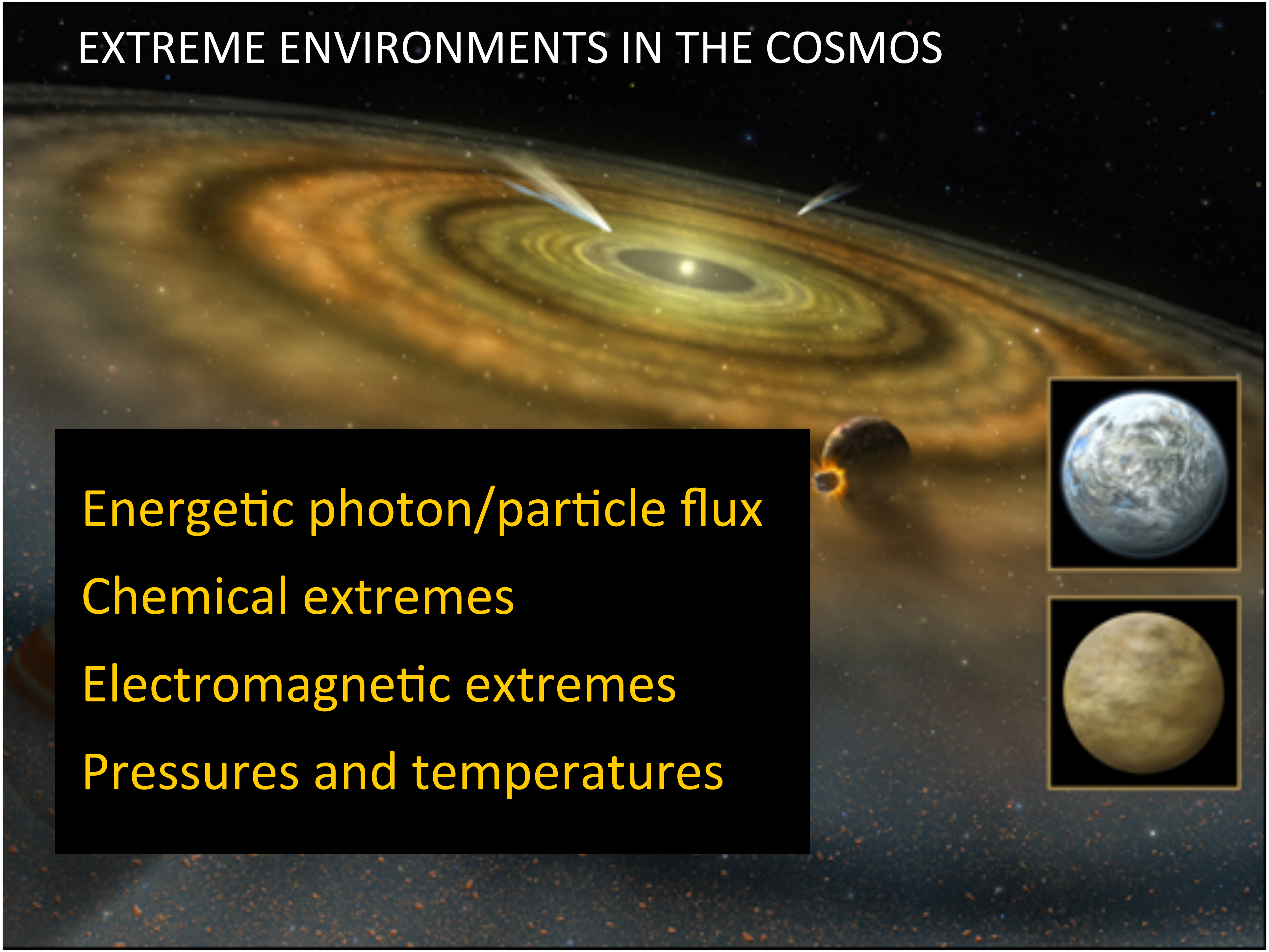
EFree Energy Frontier Research in Extreme Environments Center

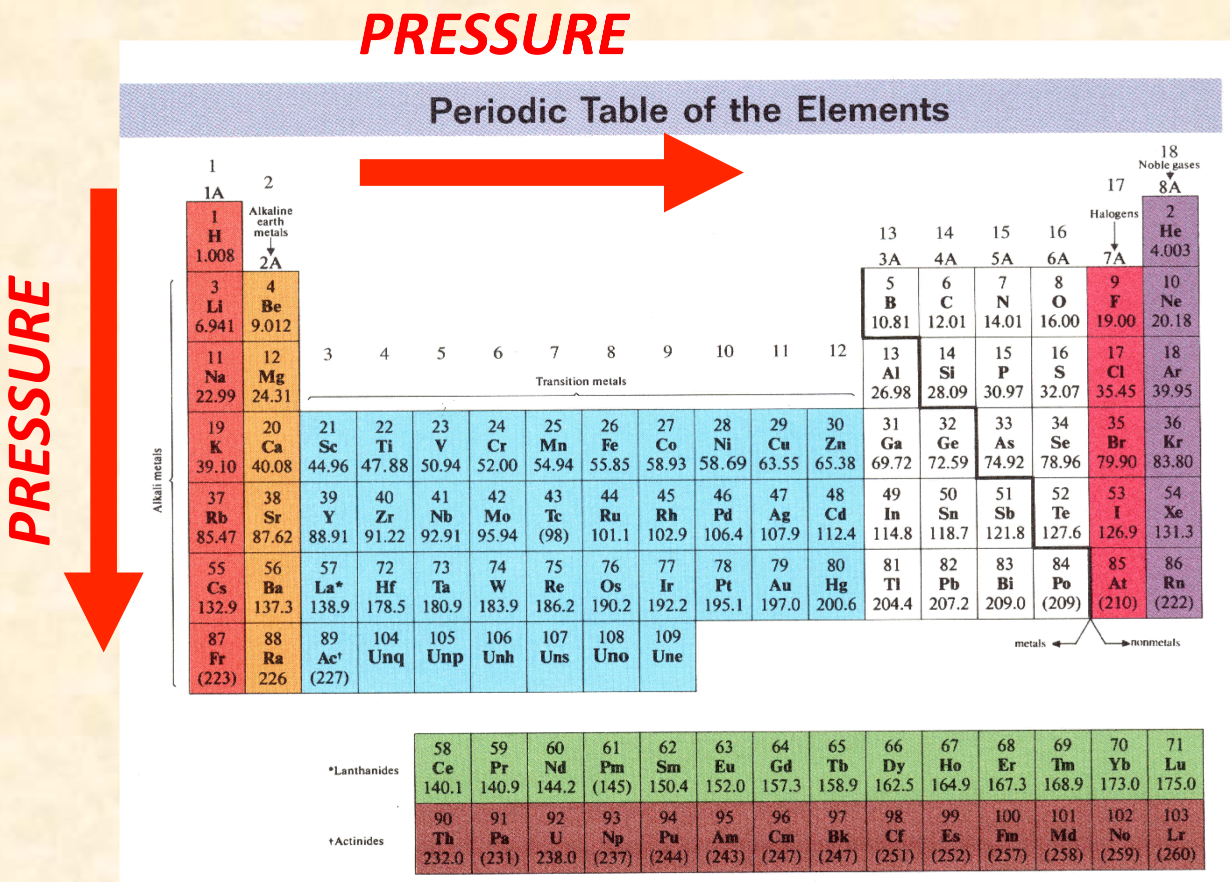
NEUTRON DAY

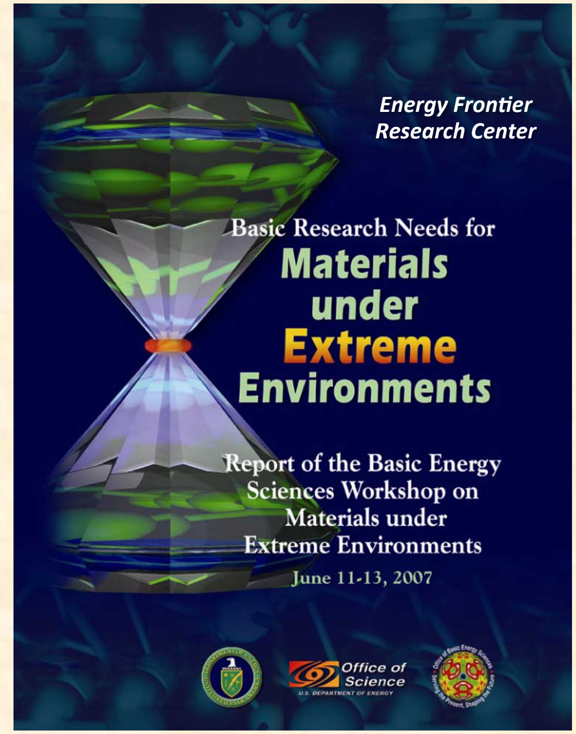
Dec. 10, 2015

Spallation Neutron Source

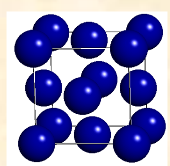
Oak Ridge National Laboratory

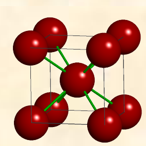

EXTREME ENVIRONMENTS IN THE COSMOS
Energetic photon/particle flux
Chemical extremes
Electromagnetic extremes
Pressures and temperatures

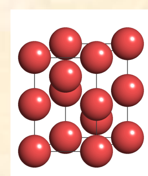




- Simple structures







Under Pressure

- Orbital hybridization (*e.g.*, *s* → *d*)
- Complex structures/electronic structure
- Valence-core hybrid. at more extremes

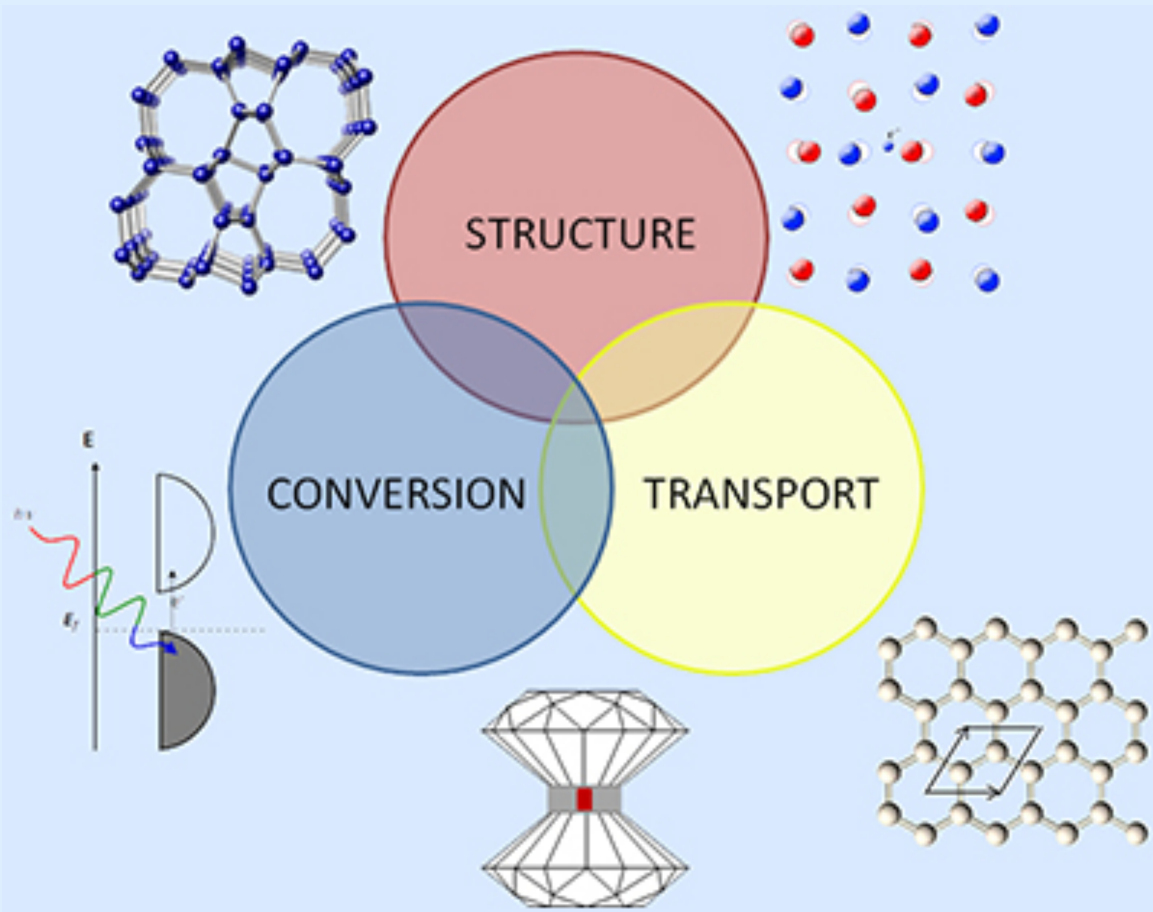
➢ Implications for making new energy materials

➢ Understanding behavior of energy materials in extreme conditions

EFree Energy Frontier Research in Extreme Environments Center

DOE Energy Frontier Research Center

Mission: To accelerate the discovery and synthesis of new energy materials using extreme conditions



Carnegie:

- **Director:** R. J. Hemley
- **Assoc. Director:** T. S. Strobel
- **Admin.** S. Gramsch, M. Phillips
- **Technical Coordinators:** R. Boehler, C. Li, M. Baldini, D. Kim

University Partners:

- **Penn State:** J. Badding, N. Alem, V. Crespi
- **Cornell:** R. Hoffmann, N. Ashcroft
- **Colorado School Mines:** C. Taylor
- **Caltech:** B. Fultz
- **Lehigh:** K. Landskron

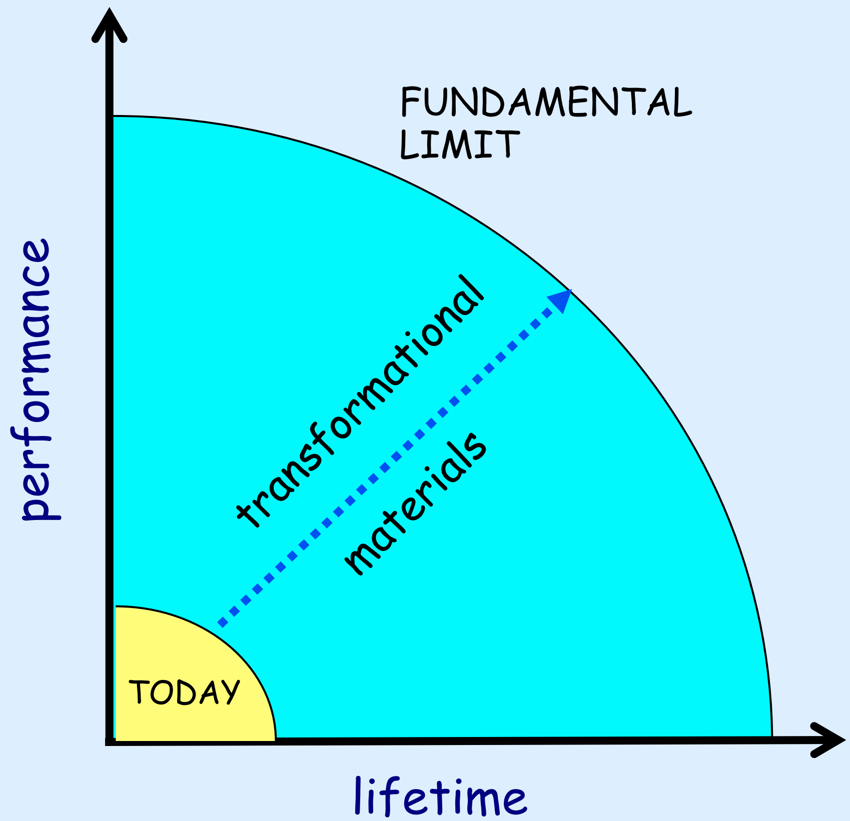
DOE Facilities

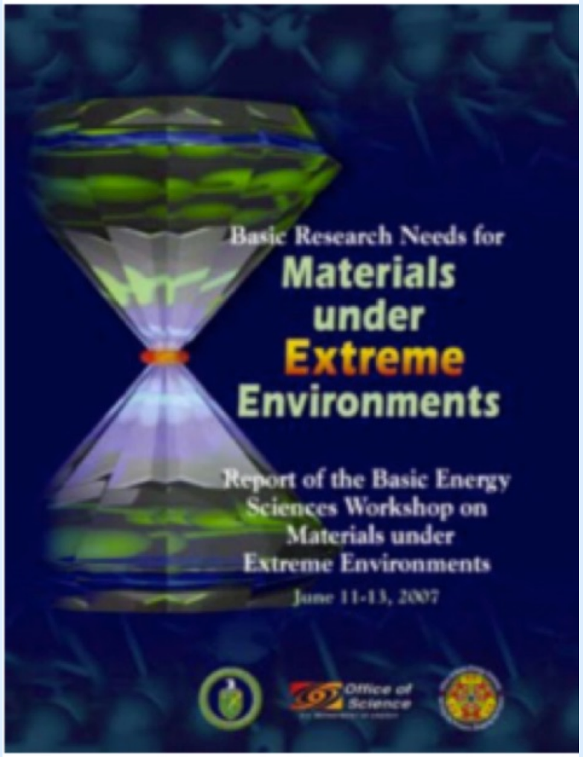
- ***APS, ANL (X-ray)***
- ***SNS, ORNL (Neutron)***
  C. Tulk, B. Haberl (*affiliates*)
- ***NSLS II, BNL (IR)***

U.S. DEPARTMENT OF ENERGY | Office of Science

EFree

The Energy Materials Challenge





Basic Research Needs Report (2007)

1. ***Characterize and understand existing materials in extreme environments***
2. ***Design, discover and synthesize new materials using extreme environments***

August
2002

High Pressure Beamline at the Spallation Neutron Source

OVERVIEW AND PREPROPOSAL

John B. Parise[1], Russell J. Hemley[2], Ho-kwang Mao[2], and Christopher A. Tulk[3]

[1] *Departments of Geosciences and Chemistry, State University of New York Stony Brook, NY 11794-2100*

[2] *Geophysical Laboratory, Carnegie Institution of Washington 5251 Broad Branch Road N. W., Washington, D. C. 20015-1305*

[3] *Research Staff Member, Experimental Facilities Division, Spallation Neutron Source, Oak Ridge National Laboratory, Oak Ridge*

Summary: This project will develop a high-pressure beamline facility at SNS that will revolutionize high-pressure neutron science. Capitalizing on myriad recent developments in studies of materials under pressure, the project requires three major elements to accomplish its mission: i.e., (A) optimization of the beamline components to fully utilize the high neutron flux of SNS, (B) development of state-of-the-art high-pressure devices for the beamline, and (C) advancing the pressure range beyond present limits (tens of GPa) and improvement of sample size by orders of magnitude, e.g., enabling neutron study of mm^3-size samples at megabar (>100 GPa) pressures. The roles of individual institutions and team members are as follows. J. B. Parise of SUNY Stony Brook will lead the development and construction of medium pressure devices, including the Paris-Edinburgh cell (PEC) which covers 12-20 GPa, large-volume gas cells reaching up to 3 GPa, and the ZAP press which provides additional loading for other devices. R. J. Hemley and H. K. Mao of Carnegie will lead the development of a new generation of diamond-anvil cell (DAC)-based devices that will allow mm^3 sample volumes above 100 GPa, and mossanite-anvil cell (MAC) device with 10 mm^3 sample volumes up to 50 GPa. C. A. Tulk of SNS will lead the development and construction of the beamline source, optics, and detector system to achieve at least one order of magnitude gain in flux and efficiency over all existing neutron sources. The 5-yr development will be carried out at ANL/ORNL. As such, the project will establish a much needed neutron facility in this country that will advance the frontier of high-pressure science in the US and set a new standard for the world community.

November 2002

SNAP - SPALLATION NEUTRONS AND PRESSURE

The High Pressure Instrument at the Spallation Neutron Source

John B. Parise[1], Russell J. Hemley[2], Ho-kwang Mao[2], and Christopher A. Tulk[3]

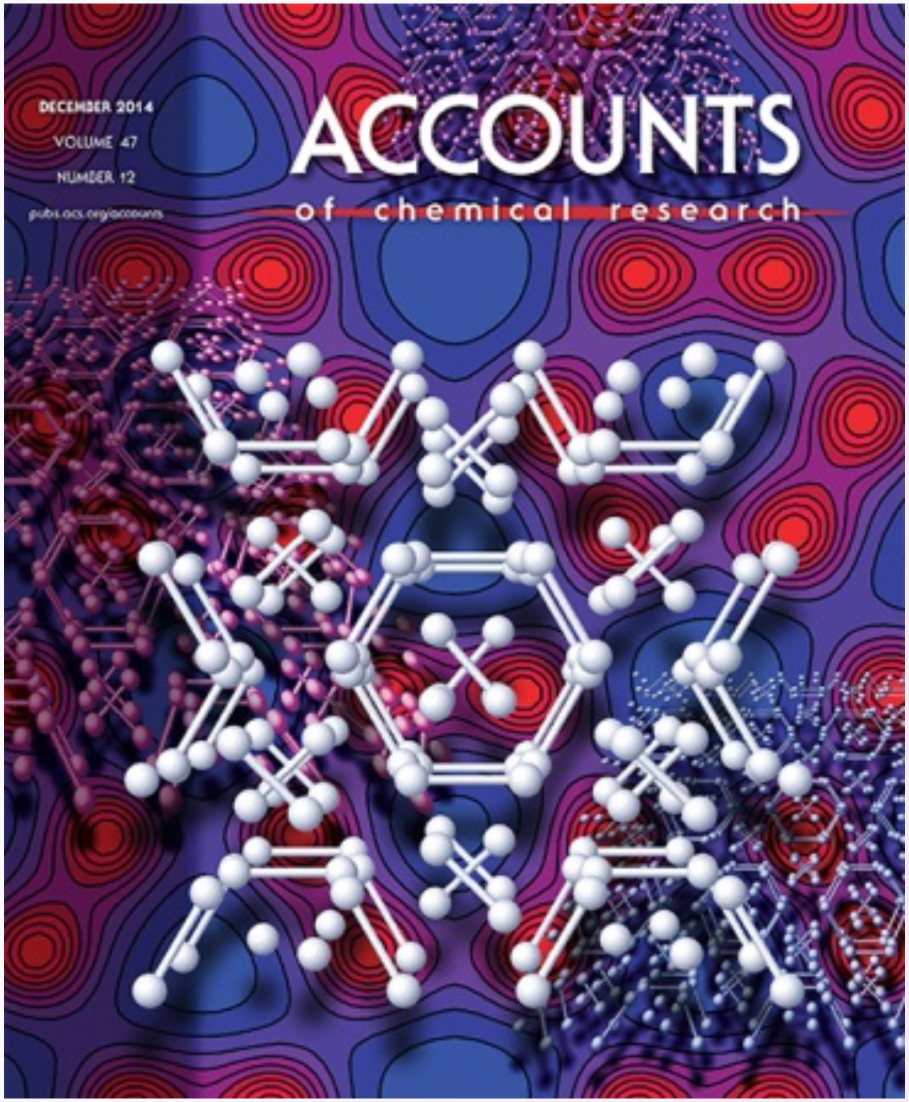
[1] *Departments of Geosciences and Chemistry, State University of New York Stony Brook, NY 11794-2100*

[2] *Geophysical Laboratory, Carnegie Institution of Washington, 5251 Broad Branch Road NW, Washington, DC 20015-1305*

[3] *Spallation Neutron Source and Chemical Sciences Division, Oak Ridge National Laboratory, Oak Ridge, TN 37831*

- Nature of dense hydrogen, including the metallic state
  From cryogenic to brown dwarf conditions
- Unraveling the mysteries of water and ice
  Structures and transformations from low to high pressures
- Behavior of novel clathrates
  Energy resources, hydrogen storage, and the solar nebula
- The complexity of simple molecular systems
  Fundamentals of chemical bonding to planetary interiors
- New chemistry in molecular mixtures
  Van der Waals compounds and hydrogen-bonded systems
- Rich structure and chemistry of metal hydrides
  New compounds under pressure and the Earth's core
- Structure and dynamics of amorphous materials
  Implications for glass technology and volcanism
- Iron and iron alloys
  Composition, elasticity, and state of the Earth's core
- Structures and stability hydrous minerals
  Oceans of water in the Earth's interior
- Minerals of the mantle and crust
  Transformations, defect structures, and equations of state
- Influence of pressure on magnetic ordering and collapse
  Direct observations of magnetic transformations
- Novel superconductivity at high pressure
  Structures, dynamics, and mechanisms
- Colossal magnetoresistance and strongly correlated systems
  Novel behavior from low to high pressures
- Structures of new ferroelectrics
  New materials and pressure tuning behavior
- Novel transformations in nanomaterials
  Nanotubes, fullerenes, and their derivatives
- Refractory materials and heavy elements
  Nuclear materials and stockpile stewardship
- Strength and rheology of materials
  Deformation mechanisms and texture development
- Organic systems and high-pressure biology
  From life at extreme conditions to food science

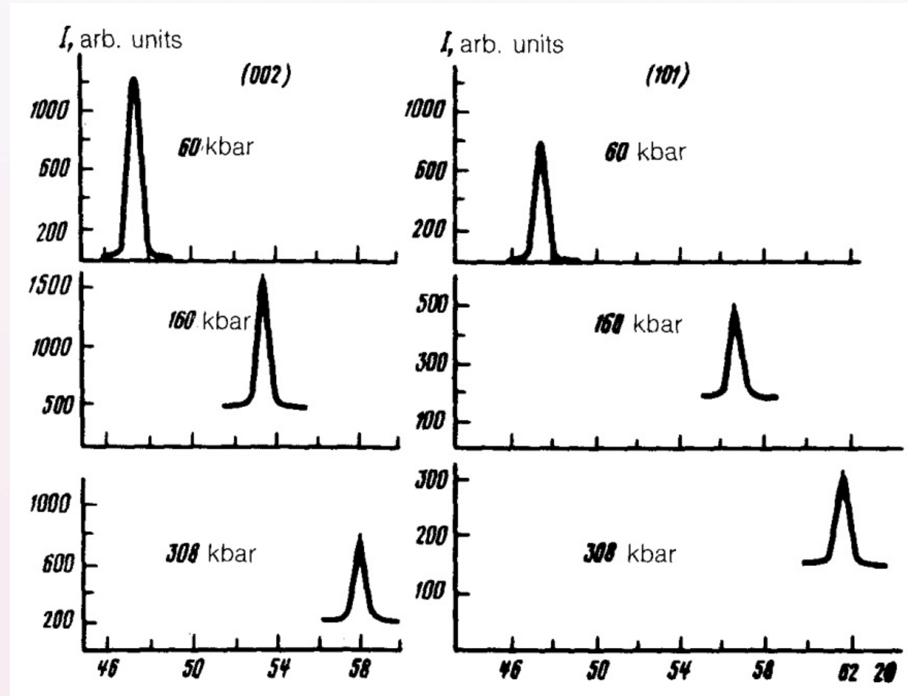
Hydrogen under pressure



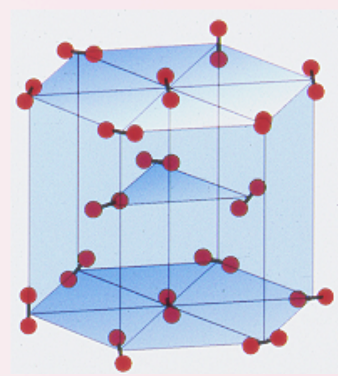
'Graphenic' hydrogen at 230 GPa (phase IV)

[Naumov & Hemley, *Accts. Chem. Res.* (2014)]

Neutron diffraction of D_2 to 30 GPa (phase I)

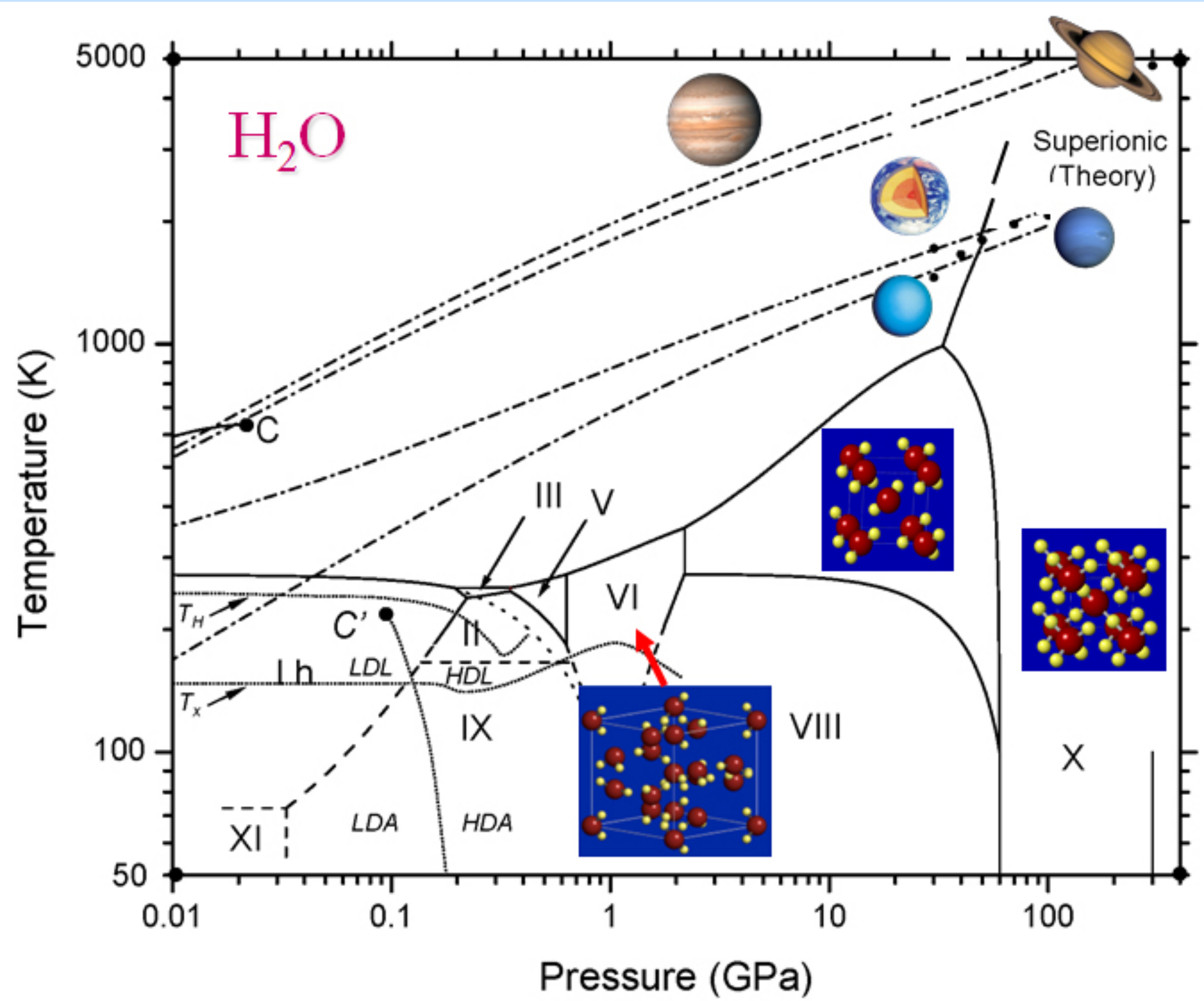


[Glazkov et al., *JETP Lett.* (1988)]



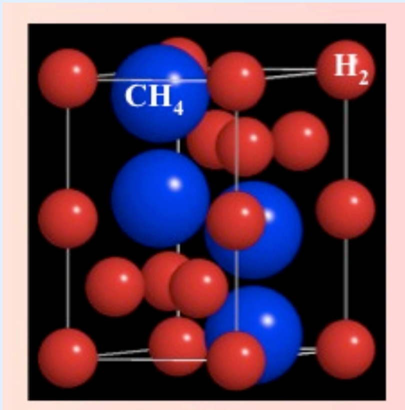
Rotational disordered hcp

High-pressure behavior of water:
new questions and surprises



- **~20 stable and metastable phases**
- **Novel transitions**
  - ***- non-molecular***
  - ***- amorphization***
  - ***- superionic***
  - ***- liquid/liquid trans***
- **High *P-T* fluid**
- **Electronic prop.**
- **New chemistry**
- **Breakdown of H_2O**
- **Supporting life at extreme *P-T***

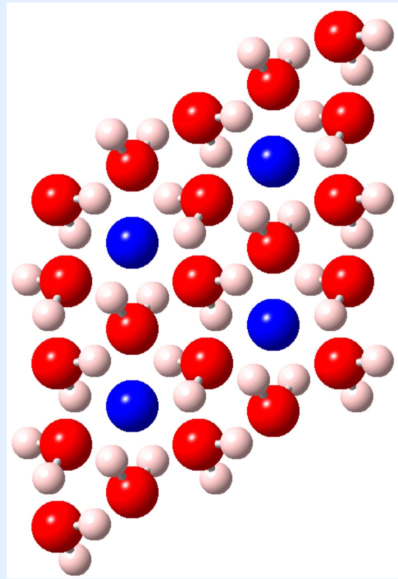
Novel Dense Molecular Compounds



$CH_4(H_2)_4$

33.4 wt% H_2

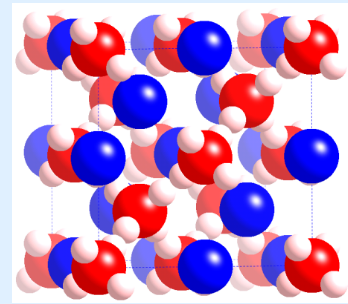
[Somayazulu *et al., Science* (1996);
W. Mao *et al. Chem. Phys. Lett.* (2005)]



$(H_2O)_2H_2$

α-quartz-type

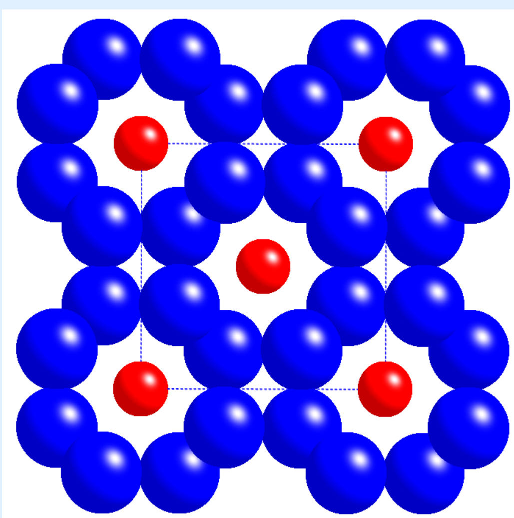
[Strobel et al.,
J. Phys. Chem.
(2011)]



H_2O-H_2

11.3 wt% H_2

[Vos et al., *Phys. Rev. Lett.* (1993)]



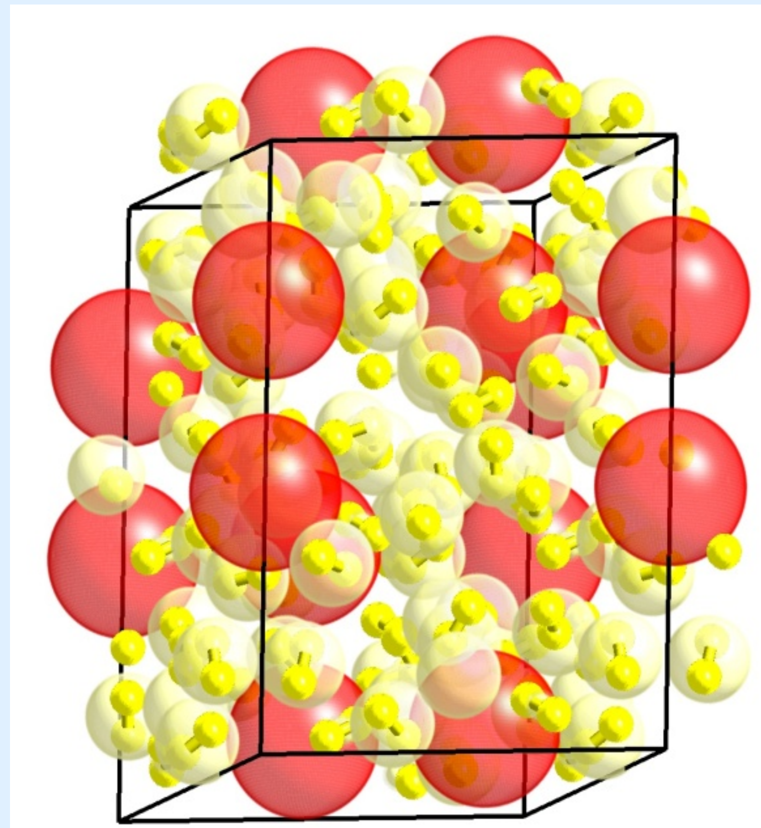
$(H_2S)_2H_2$

Al_2Cu type

[Strobel et al.,
Phys. Rev. Lett.
(2010)]

Higher pressure: Superconductor: T_c = 203 K

[Drozdov et al., *Nature* (2015)]



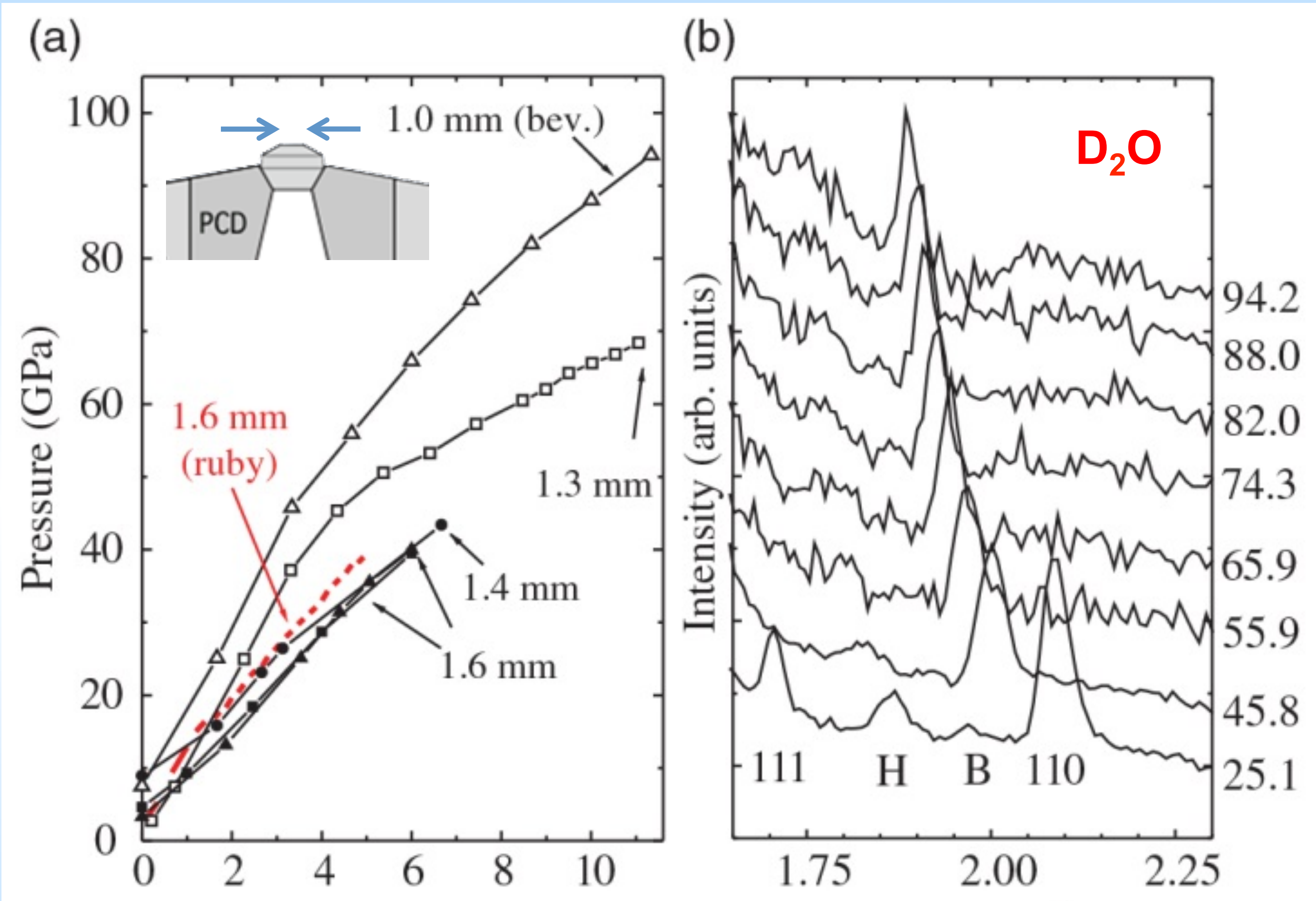
$Xe(H_2)_7$

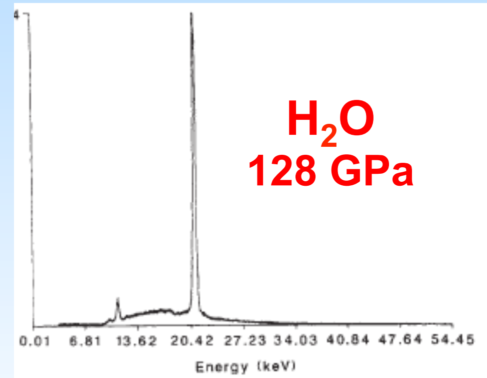
Insulating to >255 GPa

[Somayazulu et al., *Nature Chem.* (2009)]

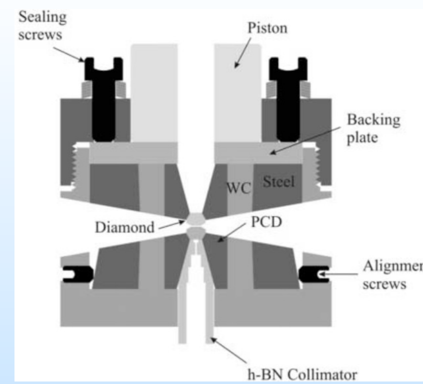
Neutron diffraction at megabar pressures

X-ray H_2O





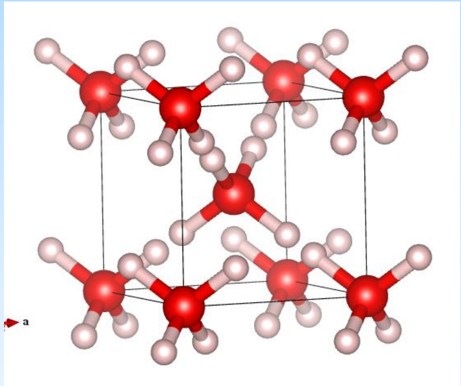
[Hemley et al., *Nature* (1987)]



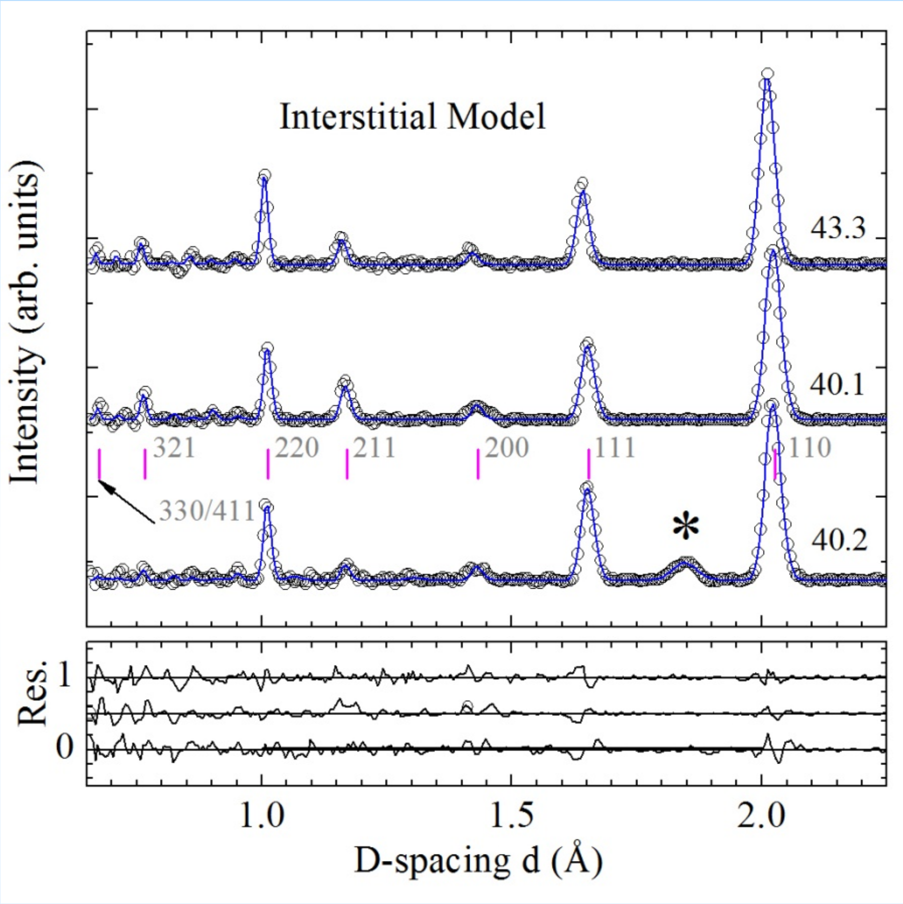
Increase sample volume by x100 at 100 GPa
(10^{-4} mm^3 to ~2 x 10^{-2} mm^3)

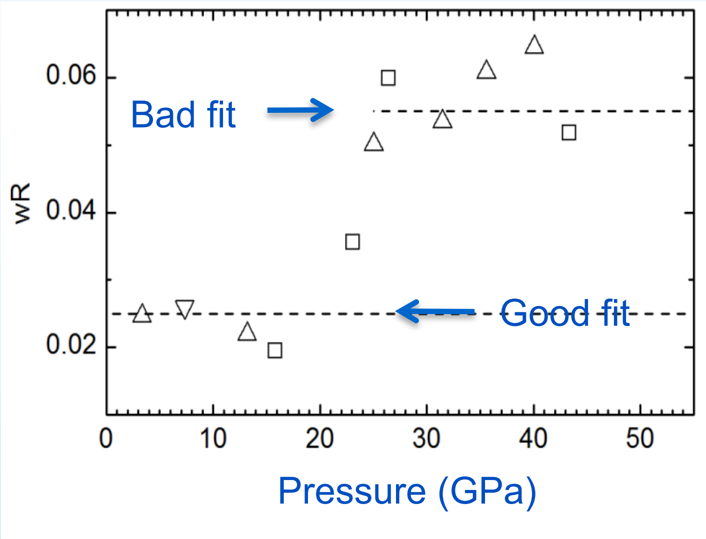
[Boehler et al., *High Pressure Res.* (2013)]

New insights into ‘proton centering in dense ice



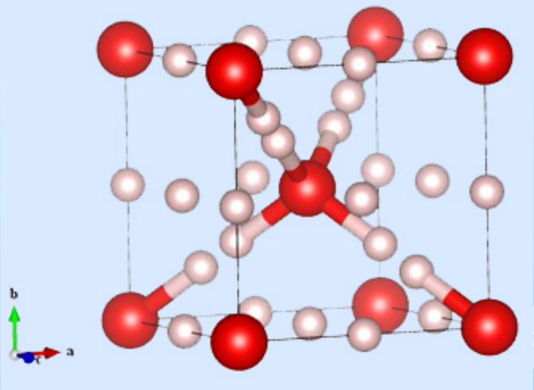
- Rietveld refinement
- Low pressure data agree well with previous work
- Abrupt reduction in quality of fit above 25





Observed intensities consistent with scattering density in the octahedral voids of O lattice

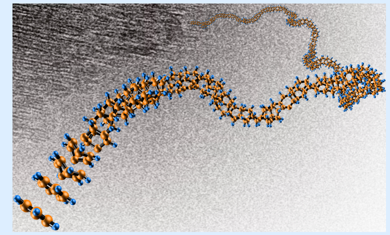
[Guthrie et al., *PNAS* (2013)]



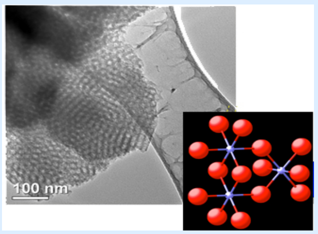
EFree currently supports five center-wide projects

1. New Nanophase Carbons

To stabilize and characterize new forms of carbon through tailored synthetic processes for the development of new structural materials *(Lead: Badding, Penn State)*.



Carbon Nanothreads



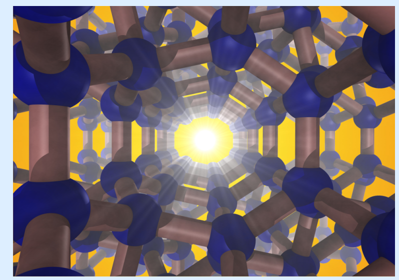
Mesoporous Stishovite

2. Next Generation Porous Materials

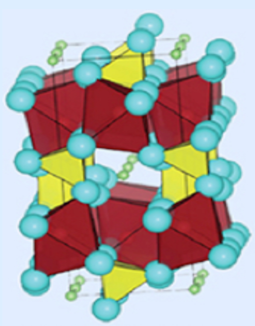
To obtain porous, mesoporous and mesostructured crystalline materials at high pressure for catalysis, gas storage, and other applications *(Lead: Landskron, Lehigh)*.

3. New Solar Energy Materials

To synthesize new materials that will have a transformative impact on solar light absorption, conversion efficiencies and manufacturing costs *(Lead: Strobel, Carnegie)*.



Open-Framework Si_{24}



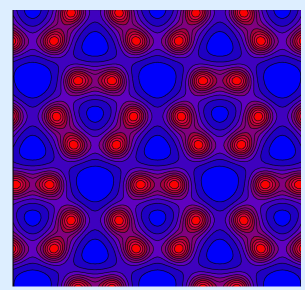
$Li_x FePO_4$

4. Ion Transport Processes

To characterize the structural features that underlie ion transport in battery materials *(Lead: Fultz, Caltech)*.

5. Electron Transport in Light Element Materials

To uncover novel electron transport in light element containing materials for enhanced electrical transport *(Lead: Hemley, Carnegie)*.



Graphenic Hydrogen

The Pressure is on at Oak Ridge

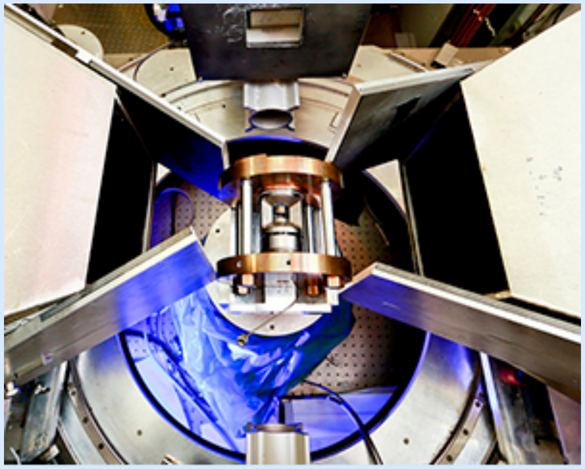


Figure 1. Diamond anvil cell within a press as prepared for an experiment at SNAP. Image credit: Jason Richards/ORNL.

Researchers at Oak Ridge National Laboratory's Spallation Neutron Source, including EFree Technical Coordinators **Reinhard Boehler** and **Chen Li**, and EFree Affiliated Scientists **Bianca Haberl** and **Chris Tulk**, have developed technology to use diamond anvil cells to squeeze materials to more than 100 GPa while studying them with neutrons. Their work has been featured on the ORNL website.

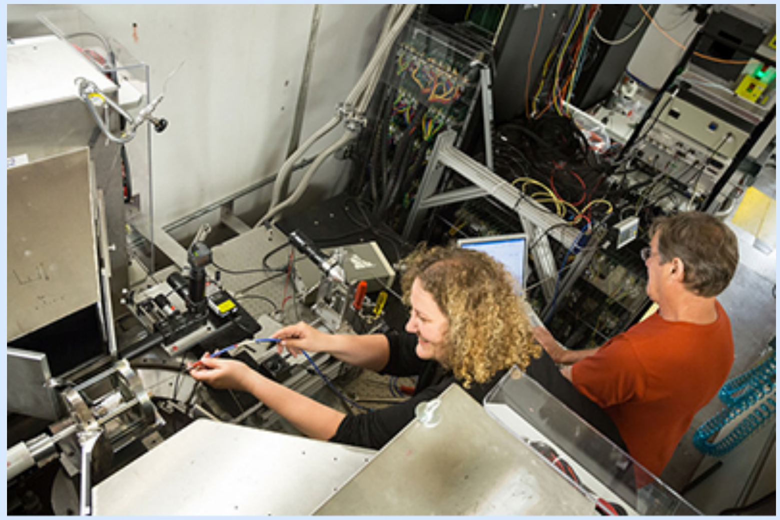


Figure 2. Bianca Haberl and Reinhard Boehler working at the SNAP Instrument, SNS beamline 3. Image credit: Geneviev Martin/ORNL.

EFree Neutron Day

December 10, 2015, Oak Ridge National Laboratory

Neutron Day will give personnel from across the Center an opportunity to share ideas and plan for the future application of neutron techniques to energy materials problems.

Goals of the Meeting

1. Update on capabilities?

- SNAP, VISION, NOMAD, ARCS, other instruments

2. Neutron scattering needs/opportunities for projects?

- 1. carbons, 2. porous, 3. solar, 4. ion transport, 5. low-Z

3. What new developments are needed?

- how can EFree help, resources required, leverage

4. New projects?

- brainstorm, feasibility studies, near- and long-term