CARNEGIE / DOE ALLIANCE CENTER:

A Center of Excellence for High Pressure Science and Technology

Russell J. Hemley

SSAP Symposium
February 17-18, 2016

OUTLINE

1. **Overview**

   MOTIVATION, CENTER STRUCTURE, PERSONNEL

2. **Training**

   EDUCATION, OUTREACH

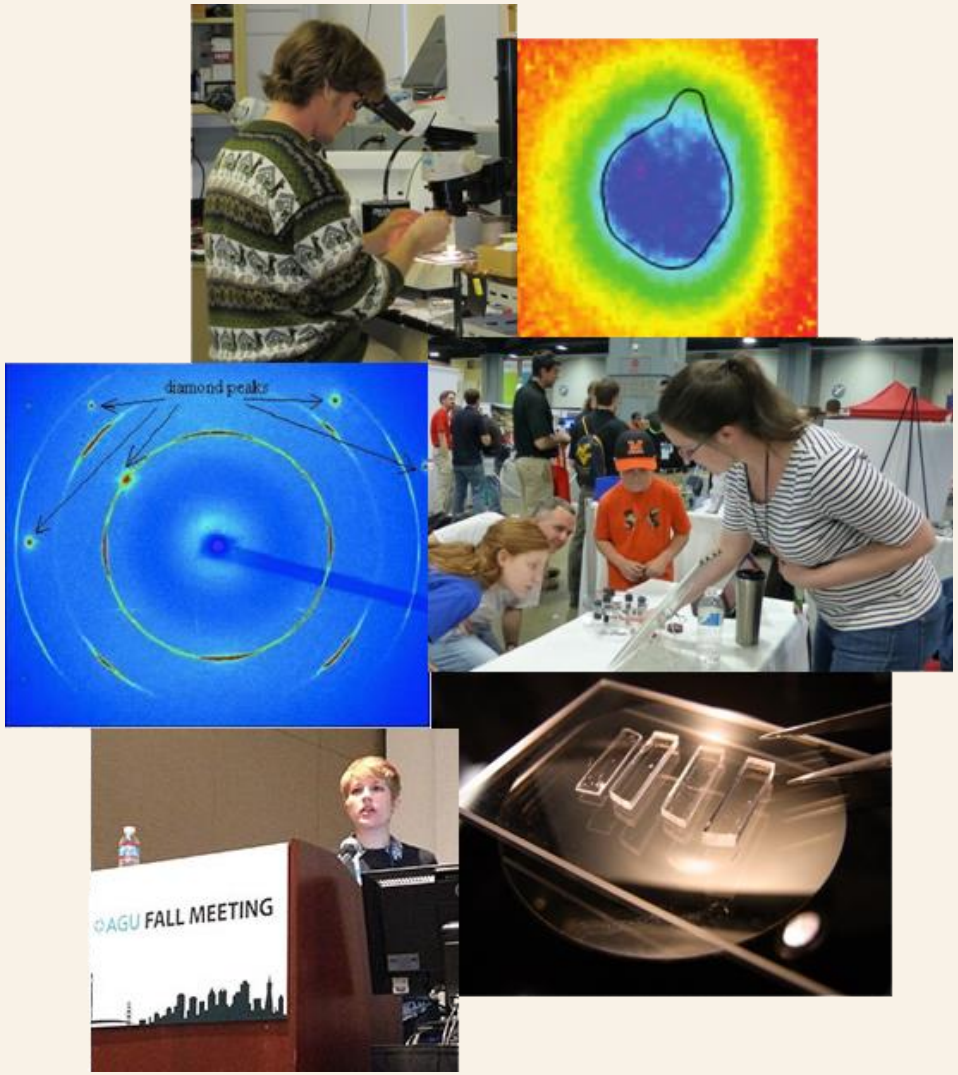
   RESEARCH TRAINING

3. **Selected Science**

   STUDENT AND NNSA LAB PROJECTS

4. **Outlook**

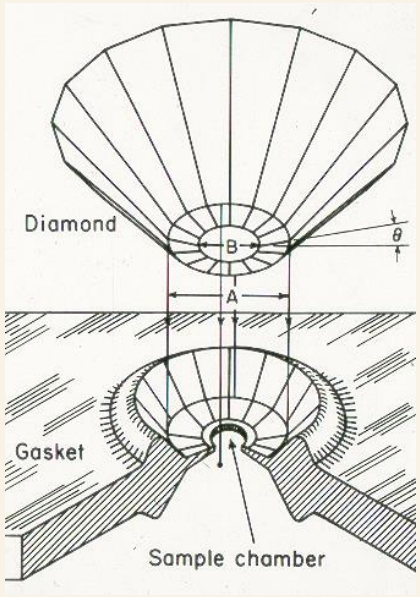
   OPPORTUNITIES

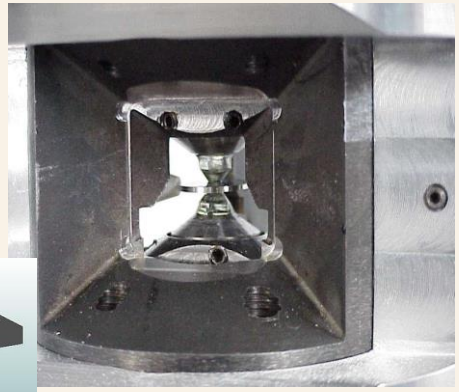


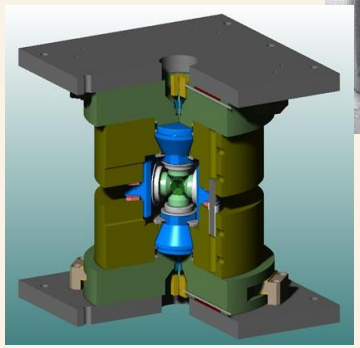
CDAC

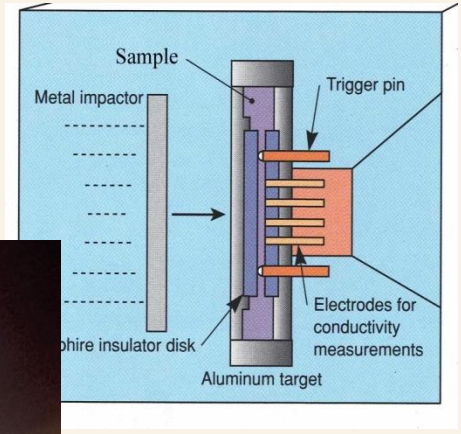
NNSA

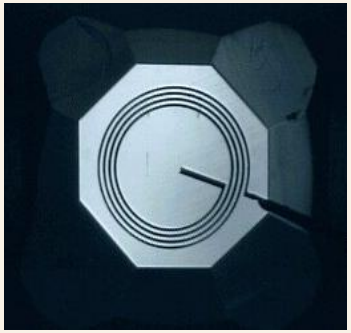
New tools have opened a new world on materials behavior under extreme *P-T* conditions

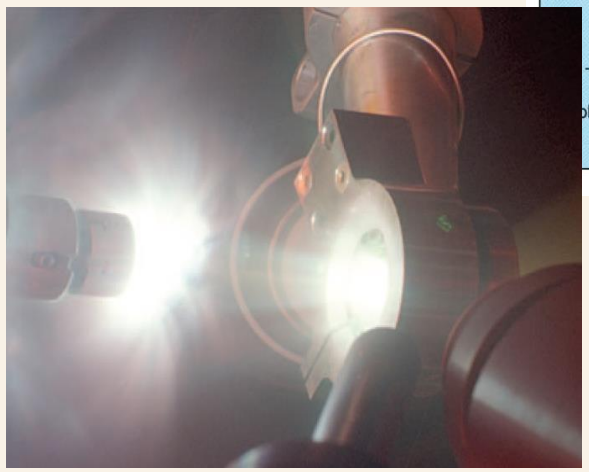










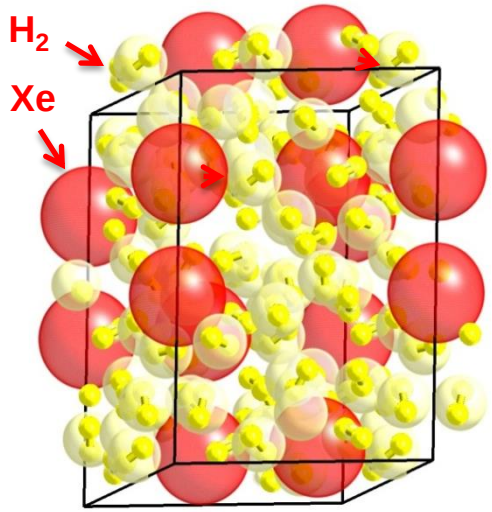


CDAC

NNSA

Novel High-Pressure Materials

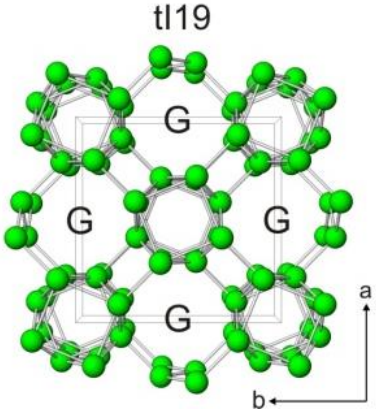
Novel Compounds



$Xe(H_2)_8$

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

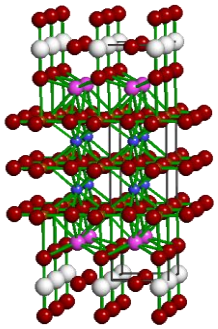
'INSULATING' METALS



Na >200 GPa

Ma et al., *Nature* 2009); Lazicki et al., *PNAS* (2009)]

HIGHEST T_c SUPERCONDUCTORS



$HgBa_2Ca_2Cu_3O_{8+d}$

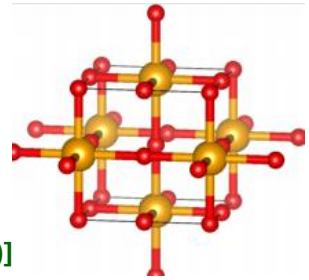
164 K at 30 GPa

[Gao *et al., Phys. Rev. B* (1994); Lokshin *et al. ibid.* (2002)]

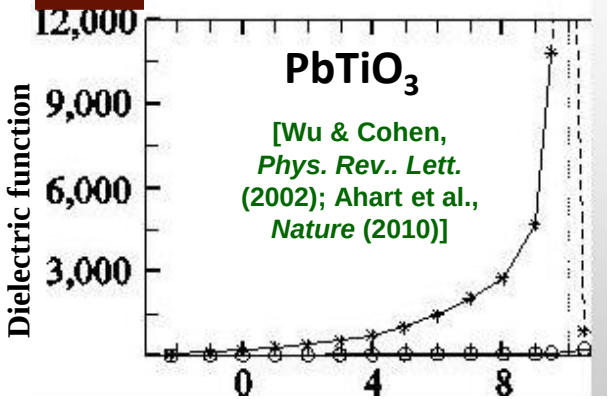
'H_3S'

203 K at 200 GPa

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

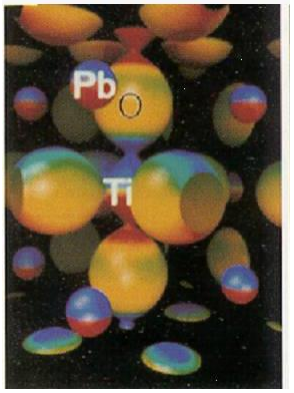


Colossal Ferroelectricity



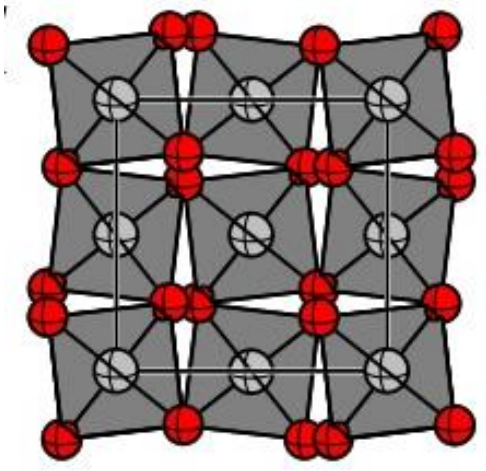
$PbTiO_3$

[Wu & Cohen, *Phys. Rev.. Lett.* (2002); Ahart et al., *Nature* (2010)]



'Polymeric' CO_2 (>30 GPa)

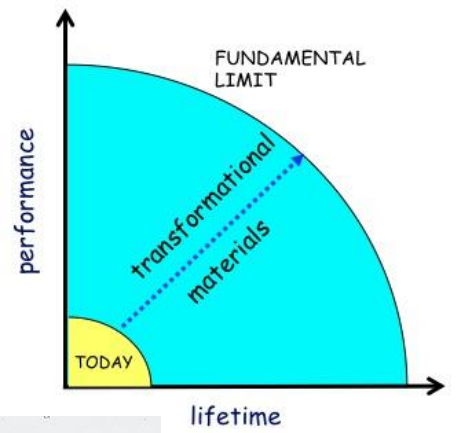
[Santoro et al. *PNAS (2012]];* Datchi et al., *Phys. Rev. Lett.* (2012)

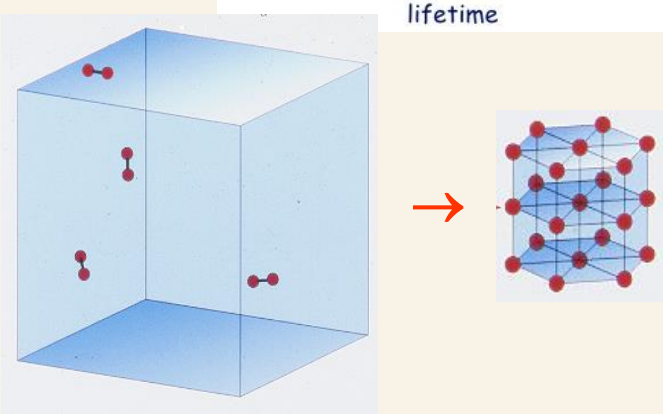


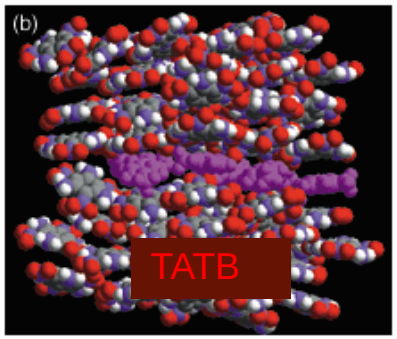
Center Goals

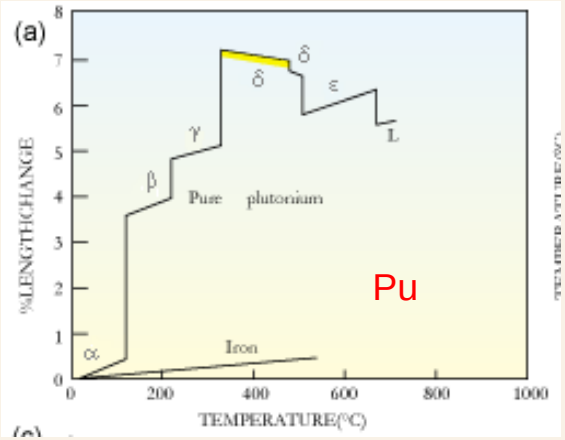
Mission

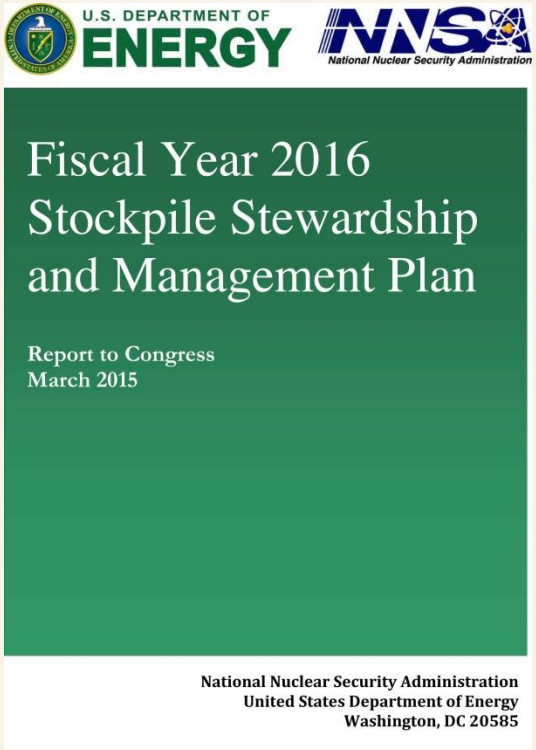
Develop techniques and training to examine the full complement of high *P-T* materials problems essential for stewardship science







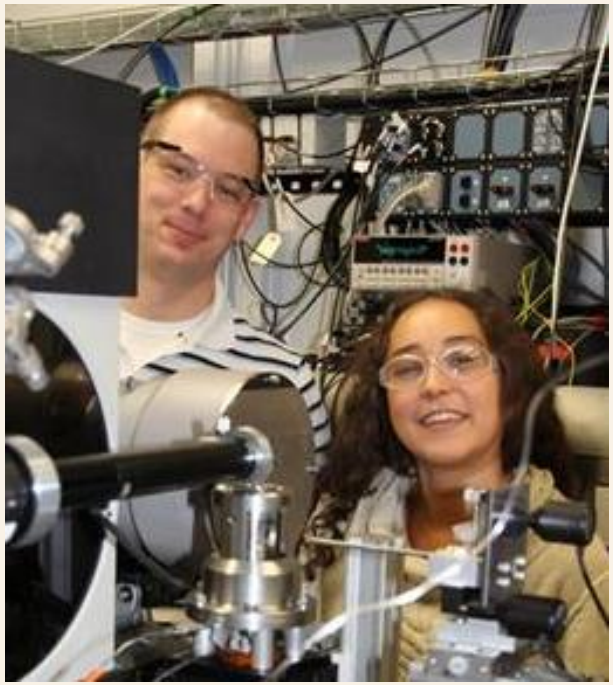


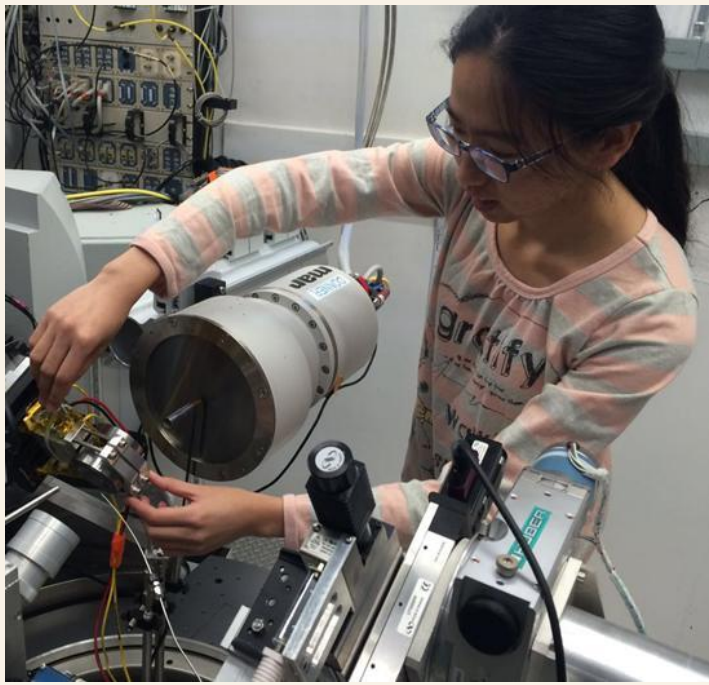


Center Goals

Mission

Develop techniques and training to examine the full complement of high *P-T* materials problems essential for stewardship science





CDAC

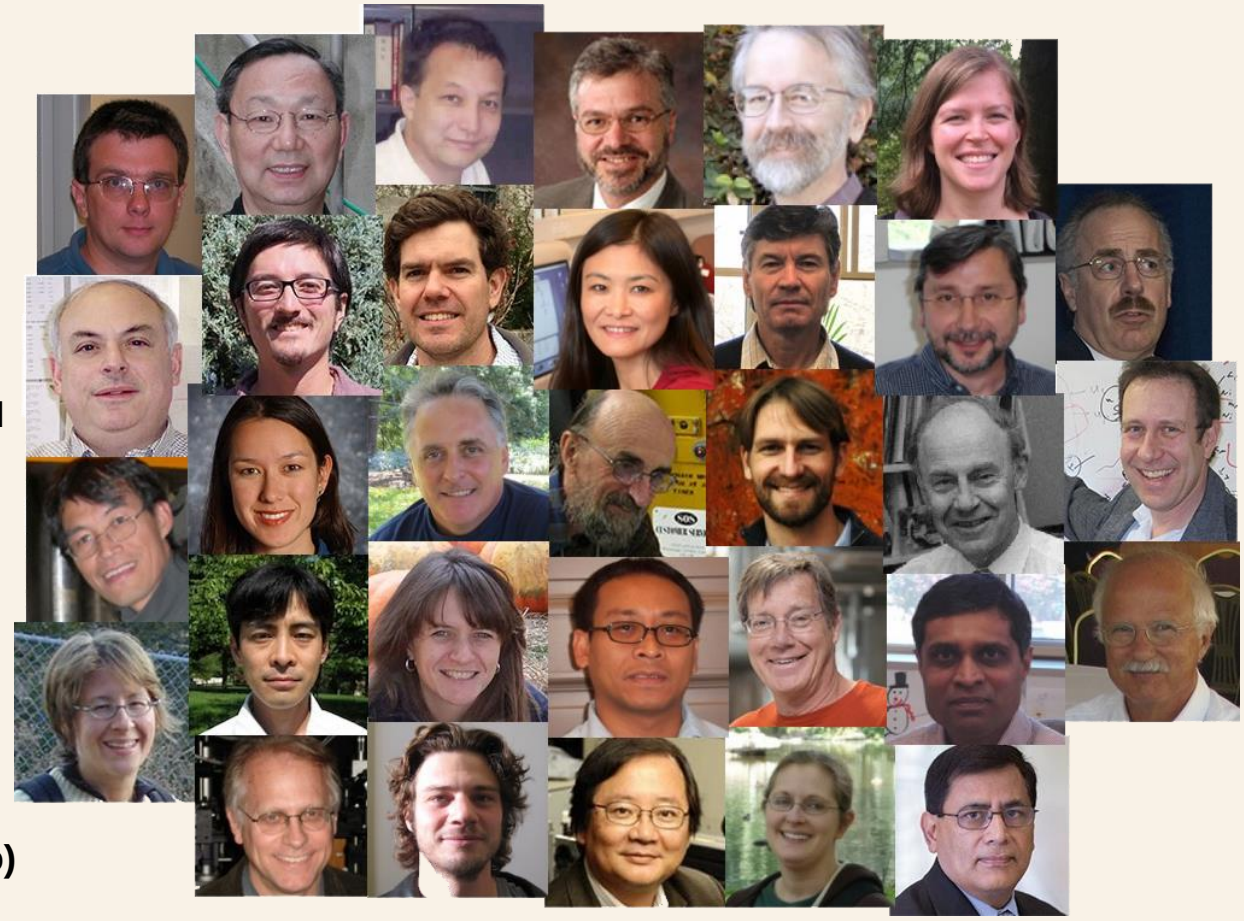
➢ ***Train the next generation***

NNSA

Components of the Center

Academic Partners

- CARNEGIE INST. (Hemley)
- CALTECH (Fultz)
- GEORGETOWN (Ichiye)
- NORTHWESTERN UNIV. (Jacobsen)
- UCLA (Kavner)
- UNIV. ALABAMA – BIRMINGHAM (Vohra)
- UNIV. at BUFFALO (Zurek)
- UNIV. CALIF. – BERKELEY (Wenk & Jeanloz)
- UNIV. HAWAI'I (Dera)
- UNIV. ILLINOIS (Dlott & Cahill)
- UNIV. UTAH (Miyagi)
- WASHINGTON UNIV. (Schilling)
- WASHINGTON STATE UNIV. (Yoo)
- YALE UNIV. (Lee)



Academic Collaborators

FACILITY USERS

NNSA Laboratory Partners

ALL HIGH *P-T* GROUPS AT LLNL, LANL, SNL;
STEERING/ADVISORY COMMITTEE MEMBERS

CDAC

NNSA

CDAC manages and coordinates activities at major facilities for high *P-T* research

- **Carnegie facilities**

  High P-T technology
  Spectroscopy labs
  Diffraction and microanalysis
  Computational resources
  CVD diamond/materials growth
  Sample preparation (beamlines
  e.g., gas loadings ~50/yr)

- **CDAC Headquarters**

Steve Gramsch
Coordinator/
Research Scientist

Morgan Phillips
Administrator

Maddury Somayazulu
Senior Lab Manager/
General high pressure

Muhtar Ahart
Ferroelectrics,
Polymeric Materials

Ivan Naumov
Theory and
Computation

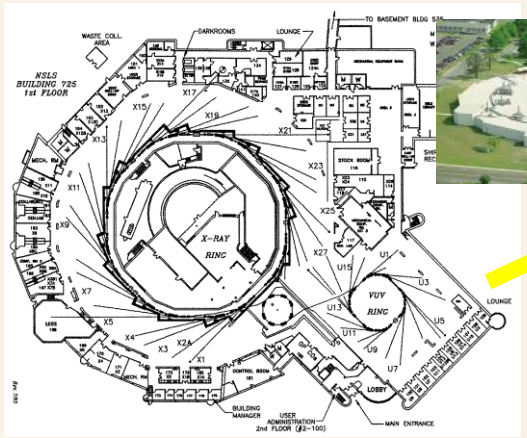
Chang-sheng Zha
Hydrogen/
Molecular Systems

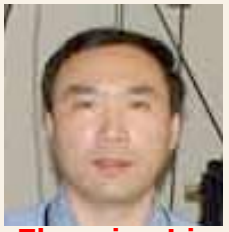
- **Manage facilities**
- **Student mentoring**
- **Visitor training**
- **Technique development**

Kadek Hemawan
CVD, Synthesis

CDAC manages and coordinates activities at major facilities for high *P-T* research

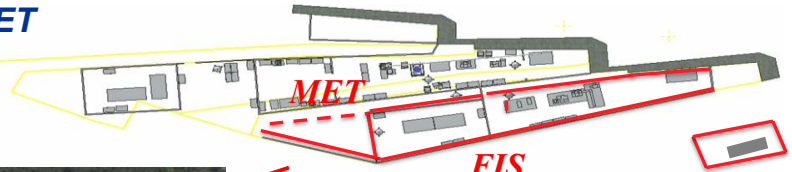
- High *P-T* synchrotron IR beamline at BNL (NSL-U2A)
- Major component of CDAC
- Academic and NNSA Lab users (*e.g.*, LANL and SNL)



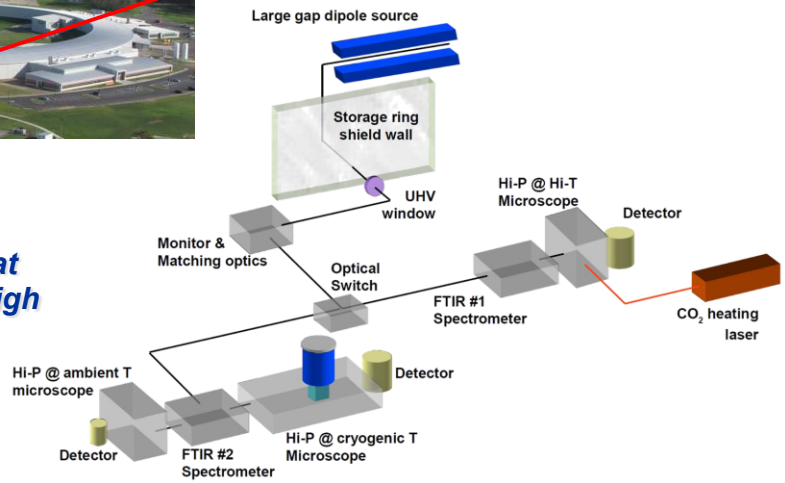


Zhenxian Liu

Floor layout of FIS/MET infrared beamlines at NSLS II



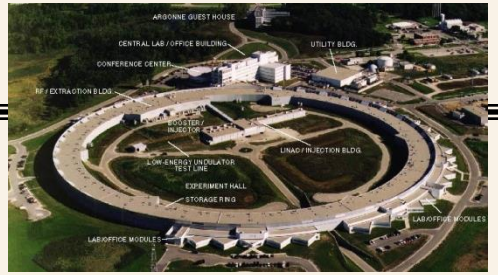
Large-gap IR Beamline at NSLS-II: Endstation for High P-T Devices



NEW NSLS-II IR BEAMLINE

- Frontier Infrared Spectroscopy (FIS) beamline to be built
- Improved perform. (stability, far-IR)
- New opportunity for NNSA Labs and SSAP

Dedicated high *P-T* facilities at the Advanced Photon Source



HPCAT
HIGH PRESSURE COLLABORATIVE ACCESS TEAM
at the Advanced Photon Source
GEOPHYSICAL LABORATORY, Carnegie Institution of Washington

- **HPCAT (Sector 16) launched in 1998**
- **Dedicated high-pressure facility**
  - Physics, chemistry, materials
  - Advanced techniques
  - Programmatic work (NNSA Labs)
- **>6100 person visits**
- **>950 peer reviewed publications**
- **Training and education**
  - More than 60% users are students and post-docs
- **Enhanced capabilities**
- **2012 Trilab (LLNL, LANL, SNL)**
- **Upgrades of APS and HPCAT**

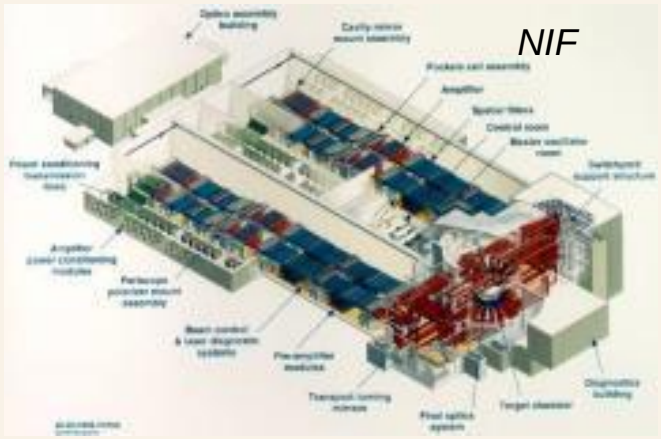
Guoyin Shen
HPCAT Director

- 9 hutches
- 4 independently operating stations
- support laboratories

DOE NNSA/SC Partnership

CDAC supports research activities at major DOE facilities

- **Technique development/support for NNSA Lab facilities**
- **CDAC co-leads two NIF Discovery Science Campaigns**



1. **Hydrogen 'PPT' Fluid Transition**
2. **Fe Melting 10-20 Mbar in 'habitable' exoplanets**

CDAC

CDAC HIGHLIGHTS 2015-2016:

Education, training and outreach

- **Supported 19 PhD students – 17 PhDs awarded**
- **52 total PhDs awarded with CDAC support**
- **4 early career scientists join DOE/NNSA labs/HQ**
  - Jeffrey Montgomery (UAB) & Suzanne Ali (UC Berkeley) to LLNL
  - Maneeshika Madduri (Stanford, Carnegie Intern) to SNL Staff
  - Caitlin Murphy (Carnegie) joined DOE HQ
  - Two postdocs offered positions (turned down)
- **10 undergraduate/high school interns (2 yrs)**
- **CDAC/HPCAT/Lab collaborations**
  - 680+ collaborators/coauthors from 170+ institutions
- **Presentations at major national meetings**
  - AGU – Fall 2015: 32 abstracts
  - APS – March 2015: 20 abstracts
- **Partner Awards**
  - Dana Dlott (Lippincott); Brent Fultz (Hume-Rothery); David Cahill (Touloukian); James Schilling (AIRAPT VP)

Jeffrey Montgomery (LLNL)

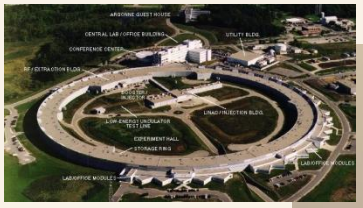
Maneeshika Madduri (SNL)

Caitlin Murphy (DOE HQ)

CDAC Annual Meeting / NNSA Review

December 8-9, 2015

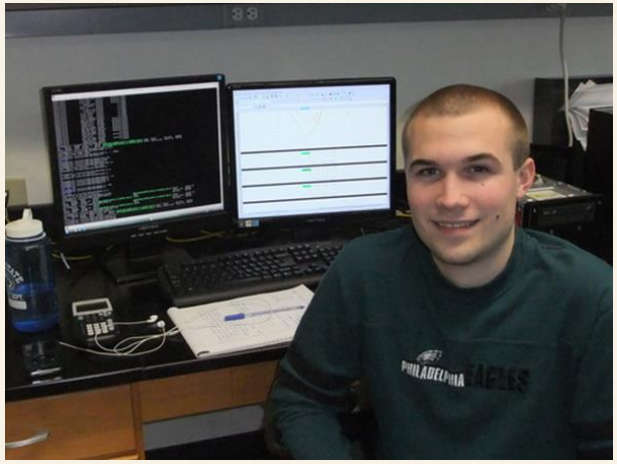
- 21 Student Posters
- 9 Academic Partners
- 2 National Lab Partners
- Program Overview
- HPCAT Overview



Educational Enrichment at NNSA Labs

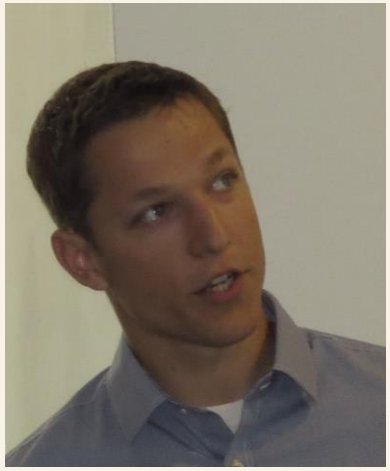
Andrew Shamp
University at Buffalo

LLNL
Quantum Simulations
Summer 2014
E. Schwegler, S. Hamel, T. Ogitsu
Theoretical Studies of the Primary Hugoniot of Boron Carbide in Extreme Conditions



John Lazarz
Northwestern University

LANL
Shock and Detonation Physics
Fall 2014 + September 2015 - Present
K. Ramos, C. Bolme
Measurement of Elasticity at Extreme Conditions



Eloisa Zepeda-Alarcón
University of California-Berkeley

LANL
Materials Science in Radiation and Dynamics Extremes
Summer 2014
R. Lebensohn, C. Tomé
Modeling Two-Phase Deformation in Polycrystalline Aggregates Relevant to the Lower Mantle

CDAC

NNSA

Summer Interns at Carnegie

Keenan Brownsberger
Whitworth University
Synthesis of Palladium Hydrides at High Pressure
APS March Meeting 2016

Reed Mershon
University of Chicago
The Role of Oxygen Fugacity in Elemental Fractionation Between Basaltic and Sulfidic Liquids
AGU December 2015

Anne Davis
California Institute of Technology
Phase Transitions in Silicon Quantum Dots for Solar Energy Conversion
MRS November 2015

CDAC

NNSA

A broad range of fundamental problems in high *P-T* science is being investigated

- ***STRUCTURES AND PHASE RELATIONS***
- ***EQUATIONS OF STATE***
- ***ELASTICITY, RHEOLOGY, STRENGTH***
- ***ELECTRON AND PHONON DYNAMICS***
- ***TRANSPORT PROPERTIES***
- ***EXTREME CONDITIONS CHEMISTRY***

DIVERSE MATERIALS

Molecules
Metals
Low-Z gases
High explosives
Polymers
Composites
…

A broad range of fundamental problems in high *P-T* science is being investigated

- ***STRUCTURES AND PHASE RELATIONS***
- ***EQUATIONS OF STATE***
- ***ELASTICITY, RHEOLOGY, STRENGTH***
- ***ELECTRON AND PHONON DYNAMICS***
- ***TRANSPORT PROPERTIES***
- ***EXTREME CONDITIONS CHEMISTRY***

DIVERSE MATERIALS

Molecules
Metals
Low-Z gases
High explosives
Polymers
Composites
…

2015-2016: 156 Publications (including in press)

- 24 Student papers (18 Student First Author Papers)

Since 2003: 1635+ Publications

(224+ Student Publications – 145+ Student First Author Papers)

- 95 Phys. Rev. Lett., 73 Nature, 25 Science, 75 PNAS

CDAC HIGHLIGHTS 2015-2016:

Student / Postdoc Presentations

17 posters at this meeting

Will Bassett (UIUC) - *32-Channel Emission Spectrometer for Studies of Energetic Materials*

Kierstin Daviau (Yale) - *Inside a Diamond Planet: Dissociation of SiC at High P-T*

Sakun Duwal (WSU) - *Isostructural Transition and Metallization in WS_2*

Yi Hu (Hawai'i) – *Five-Coordinated Silicon in Diopside at High Pressure by Single-Crystal X-ray Diffraction and First Principles Calculations.*

May-Ling Li (UIUC) – *Thermal Conductivity of Anisotropic Materials at High Pressure*

Feng Lin (Utah) – *Elastic Visco-Plastic Self-Consistent Analysis of Periclase Deformation*

Chris McGuire (UCLA) - *Fe_5Si_3 : High Pressure and High Temperature Equation of State up to 90 GPa from Diamond Anvil Cell Experiments*

Samuel Moore (UAB) - *Fabrication of Designer Diamond Anvils Using Maskless Lithography with Integrated Wireless Data Transmission*

Raul Palomares (Tennessee) - *Controlling the Stability of Octahedral GeO_2 Glass Using in situ Ion Irradiation at High Pressure*

Andrew Shamp (Buffalo) - *Theoretical Studies of the Primary Hugoniot and Solid State Properties of Boron Carbide in Extreme Conditions*

Will Shaw (UIUC) - *Shock Wave Energy Dissipation of Nanoporous Materials*

Hannah Shelton (Hawai'i) - *Evolution of Interatomic and Intermolecular Interactions of Melamine at Pressure*

Spencer Smith (UAB) - *High Pressure and High Temperature Structural Behavior of the Organic Crystal Paracetamol*

Jing Song (WUStL) - *Magnetic Ordering at Anomalously High Temperatures in Nd and Dy under Pressure*

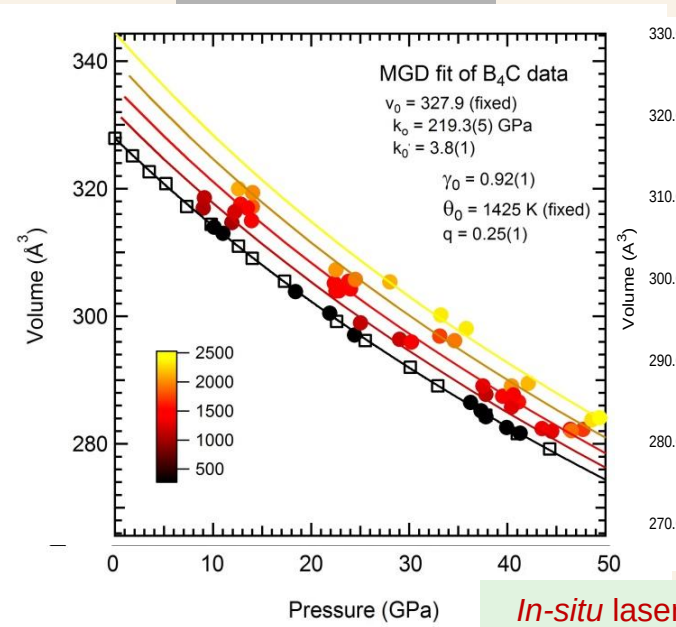
Josh Townsend (Northwestern) - *First-Principles Investigation of Hydrous Post-Perovskite*

Eloisa Zepeda-Alarcon (Berkeley) – *Modeling Two-Phase Deformation in Polycrystalline Aggregates Relevant to the Lower Mantle*

Jocelyn Rodgers (Carnegie) – *Proteins Under Extreme Pressures – A Computational Study*

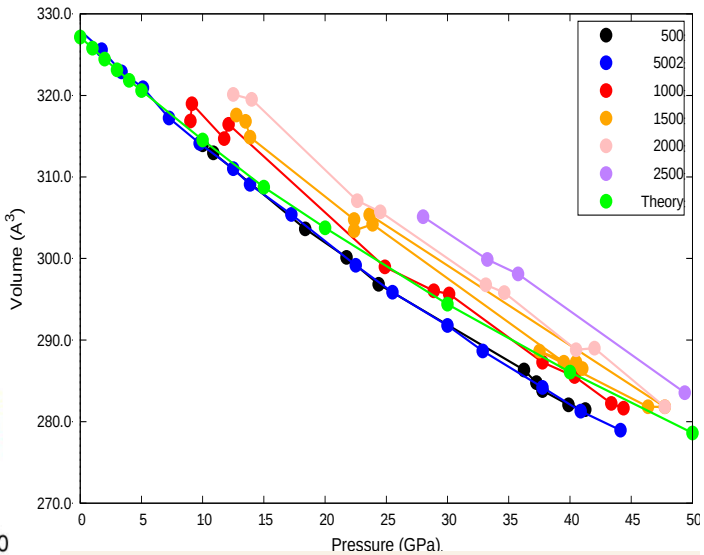
P-V-T EOS and strength measurements in B_4C and B_4C-Si mixtures

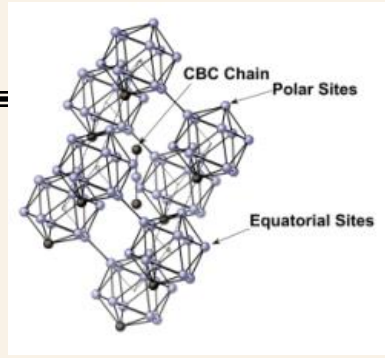
Experiment



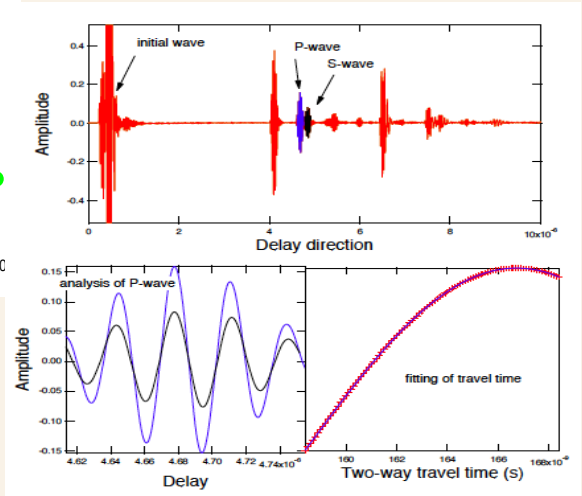
In-situ laser heating at HPCAT 16ID-B

Theory

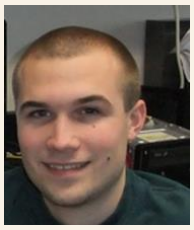




In-situ ultrasonic and x-ray diffraction at HPCAT 16BM-D



Ultrasonic measurements of B_4C sample at 3 GPa and 1073 K. Upper panel shows the whole spectrum. Lower panel shows the analyzed P-wave and that allow us to obtain accurate velocity


Andrew Shamp


Eva Zurek


M. Somayazulu


Muhtar Ahart

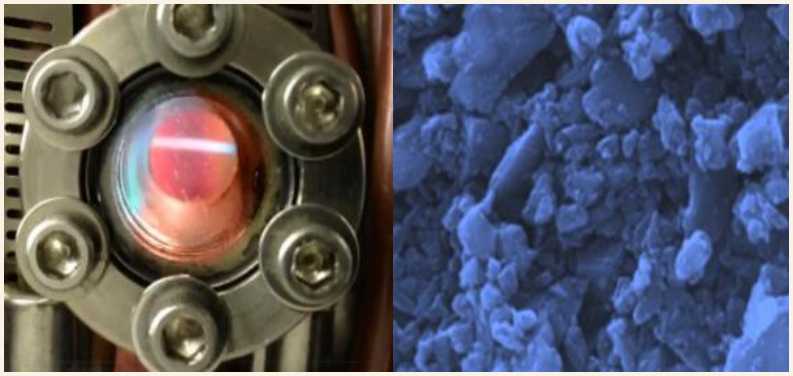
Buffalo-HPCAT-LLNL-Carnegie

New developments in CVD diamond

1. **Growing diamond at atmospheric pressure**
   - *New micro-wave plasma CVD methods*
   - *Increasing pressure enhance diamond growth rates*
2. **Metastable growth of other materials**
   - *Other materials in kinetically stabilized states such as Si*
3. **N-V centers in nanodiamonds**

Single-crystal diamond grown at <200 torr

[K. Hemawan *et al.*, *JVST A* 33, 061302 (2015)]



Polycrystalline diamond produced by atmospheric pressure CVD

[K. Hemawan *et al.*, *Appl. Phys. Lett.* 107, 181901 (2015)]

Derek Keefer (Penn State)

Todd Zapata (TAMU)

Huiyang Gou

Kadek Hemawan

PSU-TAMU-Carnegie

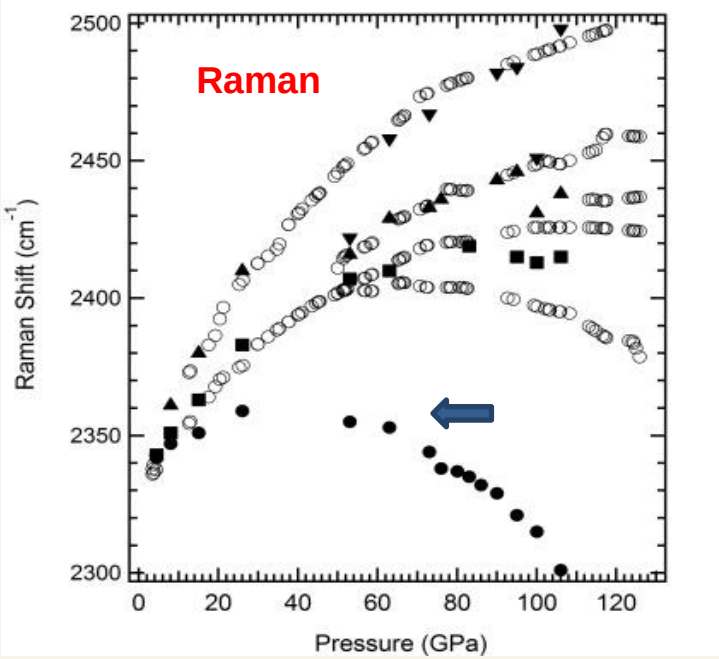
LANL collaboration: Novel behavior of molecular mixtures

- **Hydrazine-H_2 and hydrazine-N_2**
- **Xe-N_2**

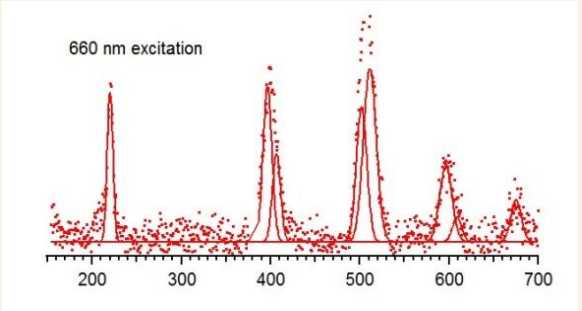
Hydrazine compression - observe phase transitions at 12 GPa and 35 GPa and at 300 K.

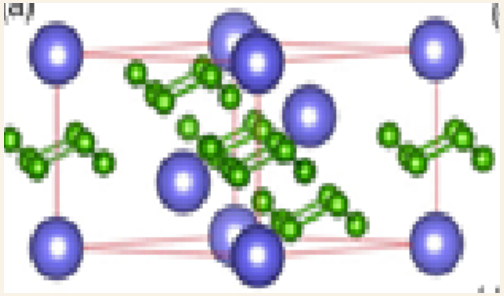
Observe a new phase at 8 GPa in hydrazine-H_2 characterized by a very large unit cell.

vdW compound $Xe(N_2)_4$ forms at 2.5 GPa and is stable to 120 GPa.



$Xe(N_2)_4$





[Ping et. al, arXiv]

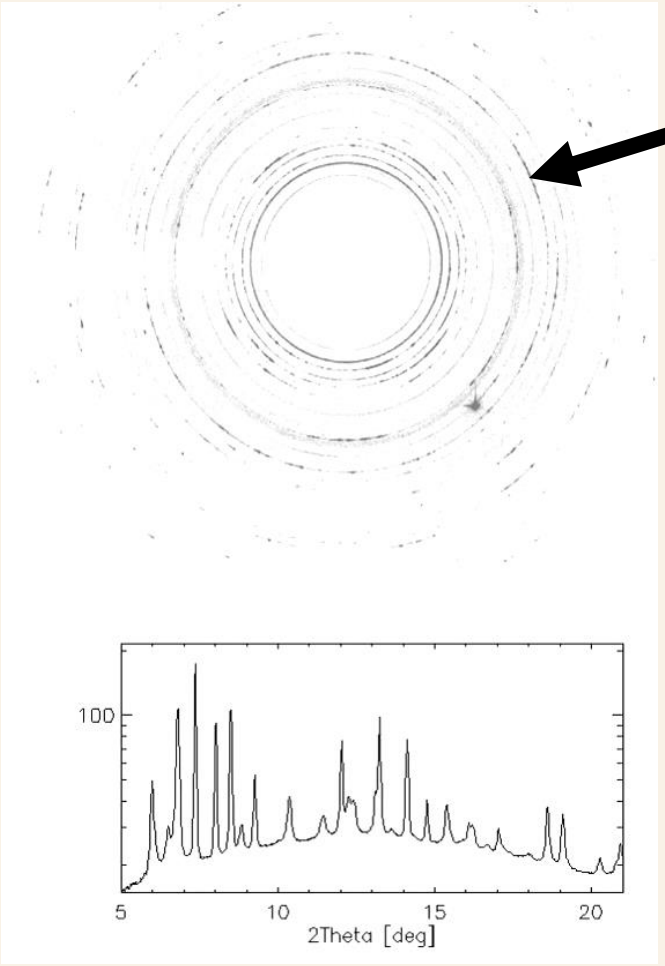
M. Somayazulu

Dana Dattlebaum

LANL-Carnegie

Raman and IR spectra obtained from laser heated sample at 120 GPa shows a mixture of N_2 and a new Xe-N_2 compound that displays 'ring-like' nitrogens. This is confirmed from XRD.

LANL: Time-resolved XRD measurements of Zr α-ω phase transformation

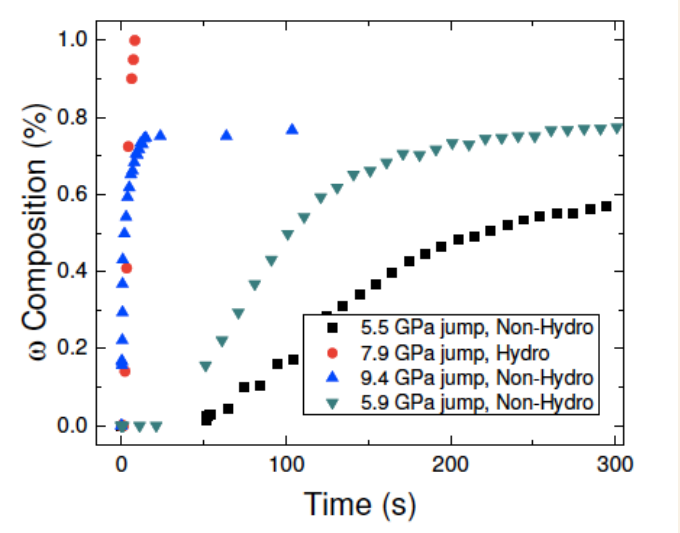


Controlled-Continuous Compression

- *Improved measurement of structural phase "boundary"*
- *Shift in phase boundary as a function of compression rate*
- *Increased efficiency in obtaining P-V data*

Pressure-Jump Experiments

- *Pressure increased in <0.1 s*
- *Time--resolved data collected at constant P*
- *Information on transition kinetics, metastable phases, etc.*



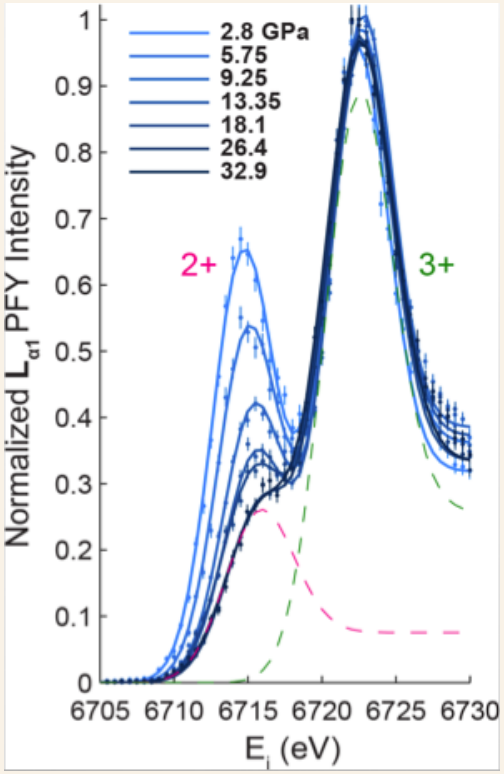
Results indicate a sluggish transition, and in some case full conversion to ω is not observed even after 10+ min

Nenad Velisavljevic

Northwestern-HPCAT-LANL

[M. Jacobsen and N. Velisavljevic, *J. Appl. Phys.* 118, 025902 (2015); N. Velisvljevic et al., *Mat. Res. Exp.* 1, 035044 (2014).]

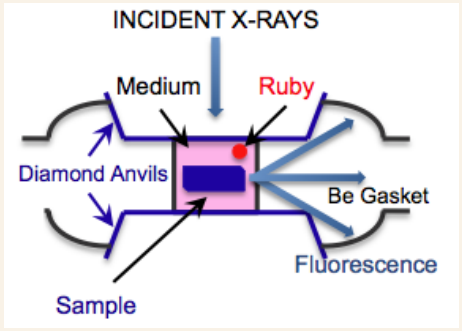
LLNL: Sm valence in SmB_6 under pressure

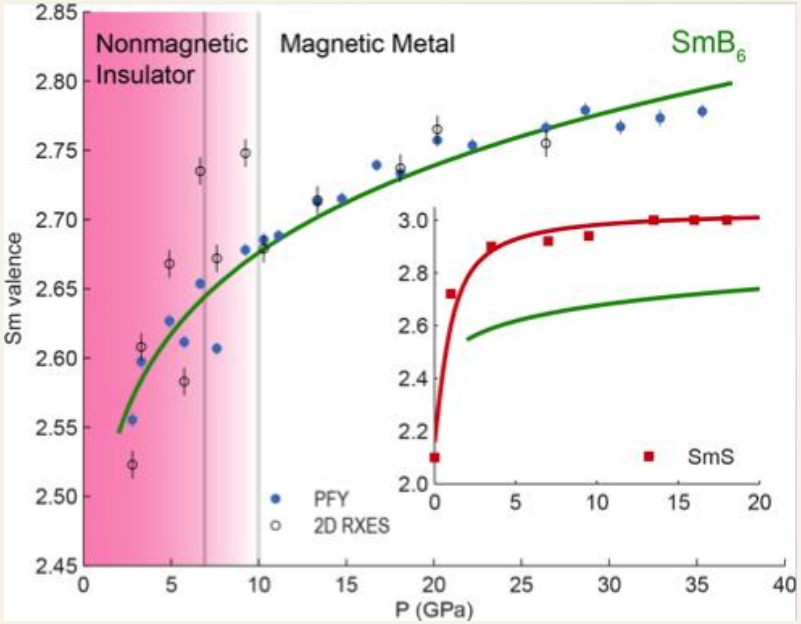


RXES and Lα PFY at HPCAT

- Incident energy scanned around L3 absorption
- Emitted energy scanned around Lα emission

Jason Jeffries

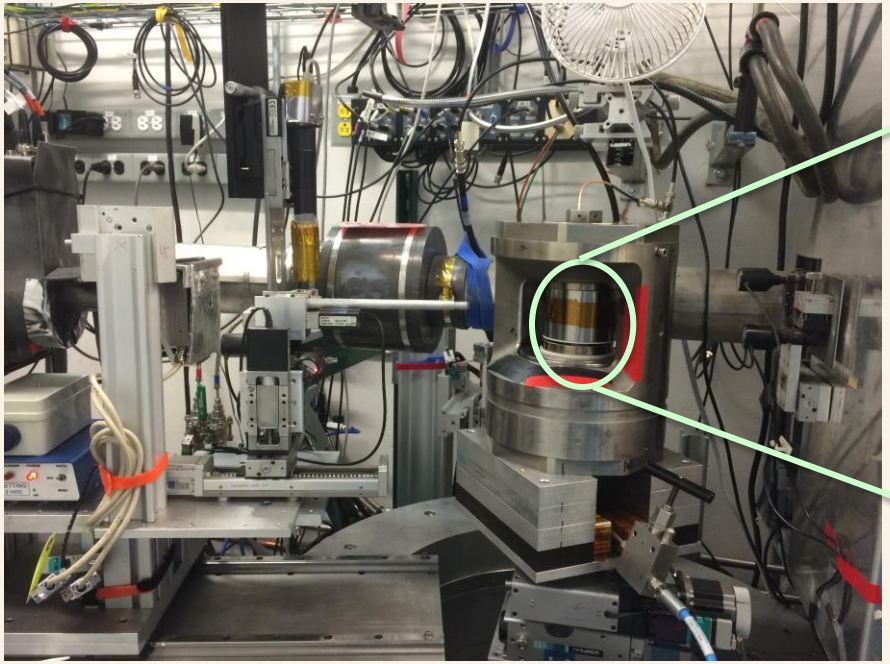


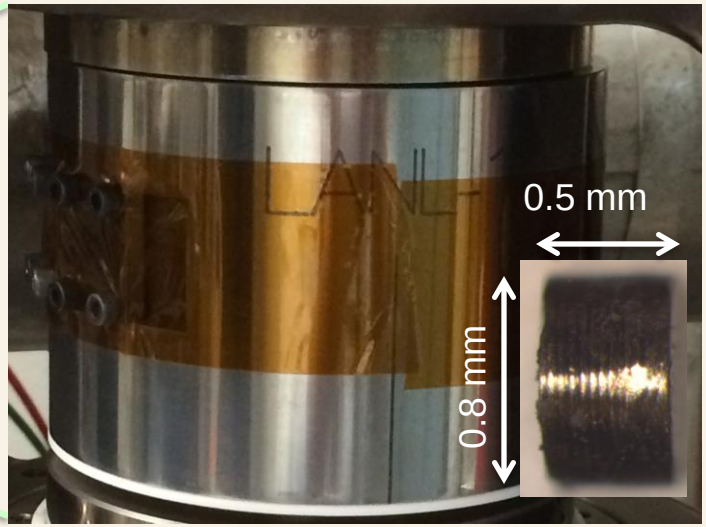


- Monotonic increase of valence with pressure
- PFY and RXES show comparable trends
- Contrasts with SmS (insulator-metal and valence change at ~5 GPa)
- Valence insensitive to gap closure and onset of magnetism near 10 GPa

LLNL-HPCAT

SNL-LANL: New device for multiple simultaneous measurements at high *P* and *T*

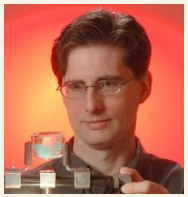




Approved Triple Containment Vessel

[M. Jacobsen and N. Velisavljevic, *Rev. Sci. Instrum.* 86, 113904 (2016)]

- Long-term collaboration with SNL and LANL
- First radioactive material experiments in ‘PE’ cell at APS
- Supports diffraction, radiography, ultrasonic, thermal, electrical measurements
- Thermally and electrically insulated for high P-T exps
- Feasibility studies on Zr, ongoing work on depleted uranium
- Beam time provided by CDAC, then supplemented by Tri-Lab

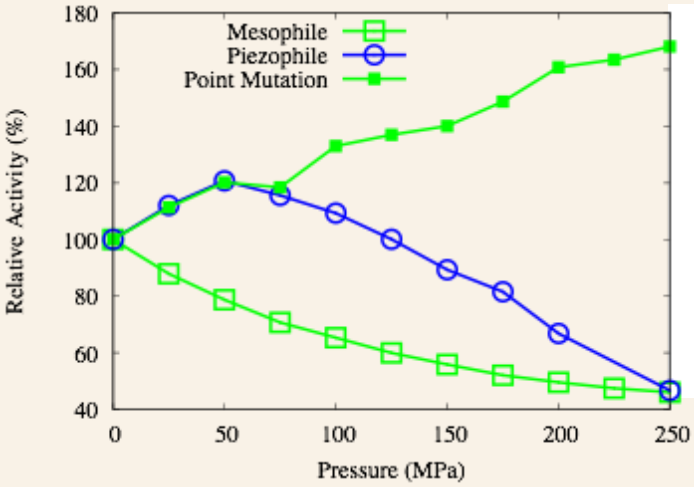



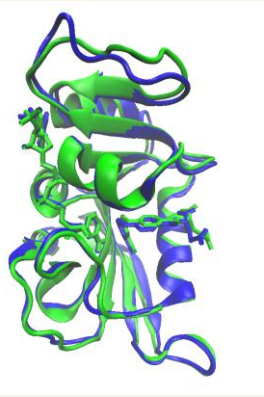

Nenad Velisavljevic, Dan Dolan, and Chris Seagle

LANL-SNL-Northestern-HPCAT

Biomolecules under pressure: computational extreme biophysics

Biophysics of a protein



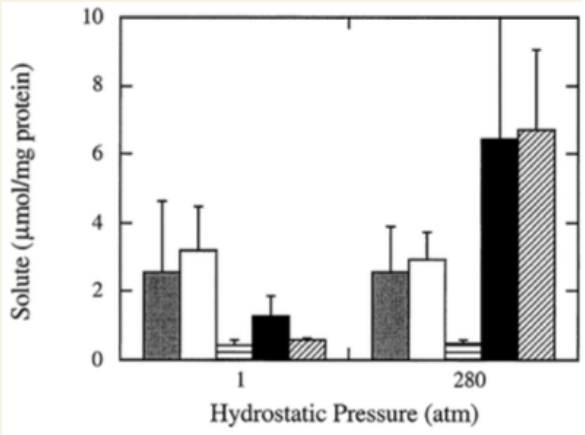


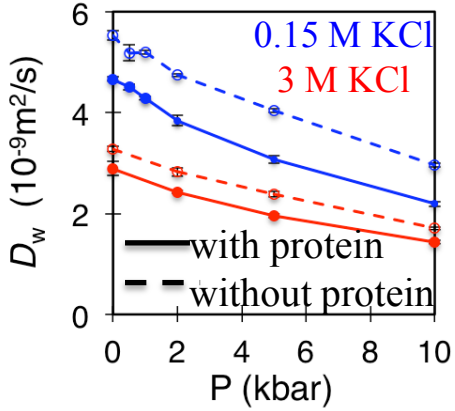
DHFR

How do intrinsic protein structure and fluctuation couple with intracellular solution properties to yield functioning proteins at high P?

- Very different enzymatic activity for proteins with highly similar structure. (Ohmae et al. in *BBA* 2012 and *BBA* 2013)

Chemical physics of the solution





Toshiko Ichiye

Jocelyn Rodgers

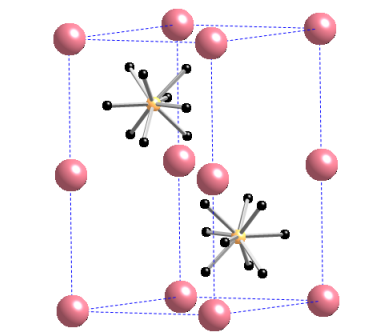
- Piezolytes. Different solutes accumulate in cells at high pressures (Bartlett et al. in *Extremophiles* 2002) Why?
- Preliminary simulations imply strong effects of pressure and solutes on solution viscosity.

CDAC

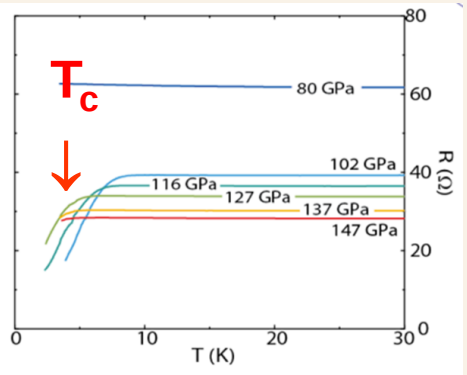
Georgetown-Carnegie

NNSA

Characterization of $BaReH_9$ superconductor



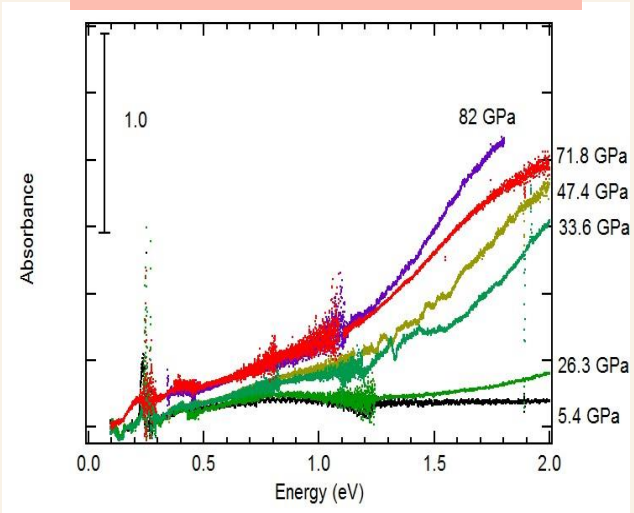
[Markapoulos et. al., *JACS*, 132, 748 (2010)]



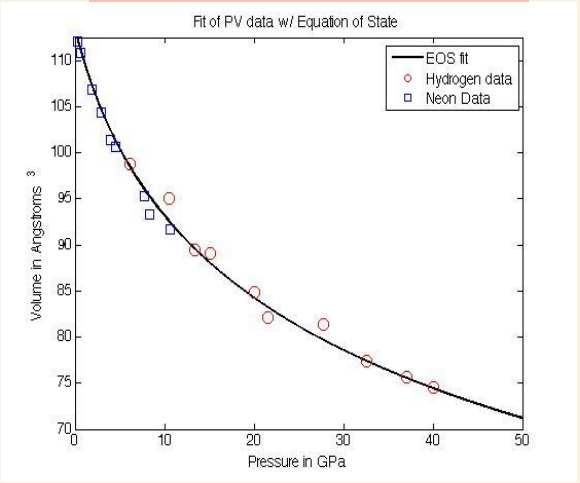
[Muramatsu, et al., *J. Phys. Chem.* 119, 18007−18013 (2015)]

We have successfully synthesized and characterized $BaReH_9$, Na_2ReH_9, Li_2ReH_9 and their deuterides and have conducted extensive XRD, Raman, Synchrotron FTIR and UV-VIS measurements on $BaReH_9$ [Vinitsky et al., *in preparation*]

UV-VIS-NIR Transmission



EOS in Ne and H_2



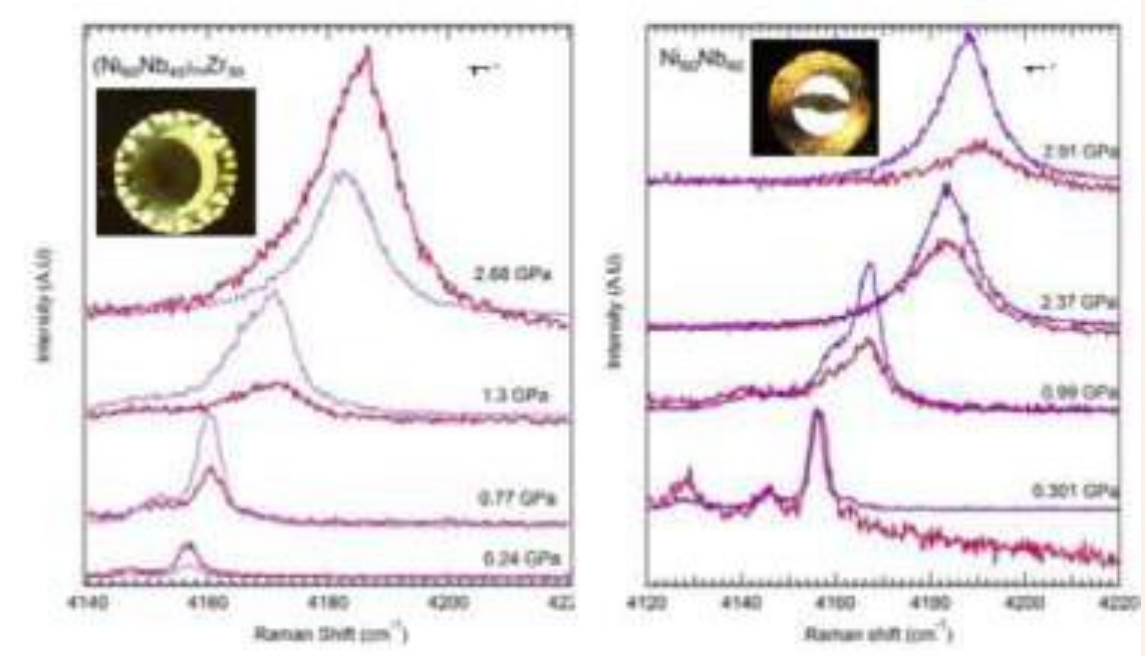
Disorder in Re-H bonds under pressure causes formation of a 'poor' semi-metal that slowly transforms to a metal under annealing paralleling other hydride superconductors.

Caltech-BNL-HPCAT-Carnegie

Eugene Vinitsky

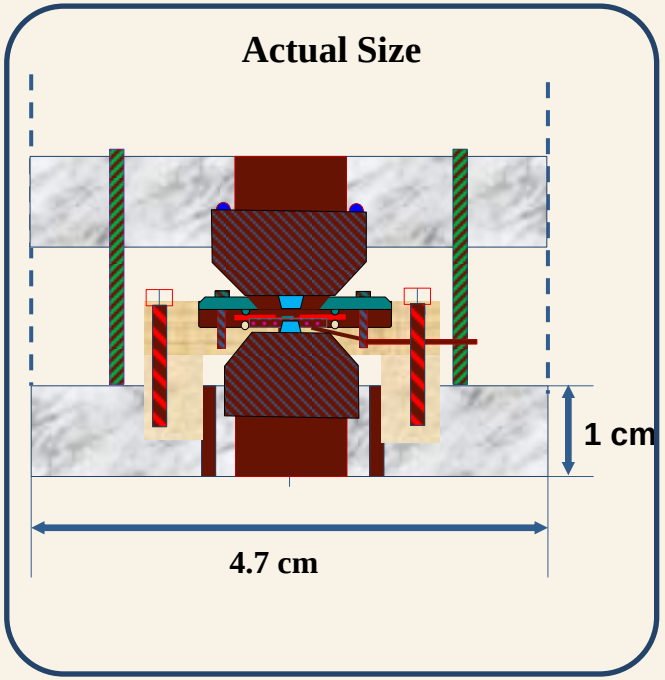
M. Somayazulu

Ni-Nb-Zr alloy gas permeation membrane ribbons at extreme pressures

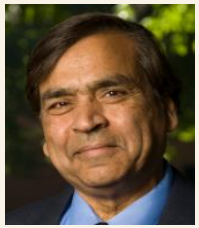


Raman spectra of $(Ni_{60}Nb_{40})_{30}Zr_{30}$ and $Ni_{60}Nb_{40}$ in H_2

Modifying DAC for gas diffusion studies – SSAP Collaboration with UNR



Suchismita Sarker



Dhanesh Chandra

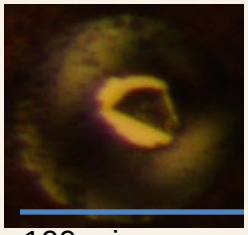
M. Somayazulu

CDAC

UNR-Carnegie

NNSA

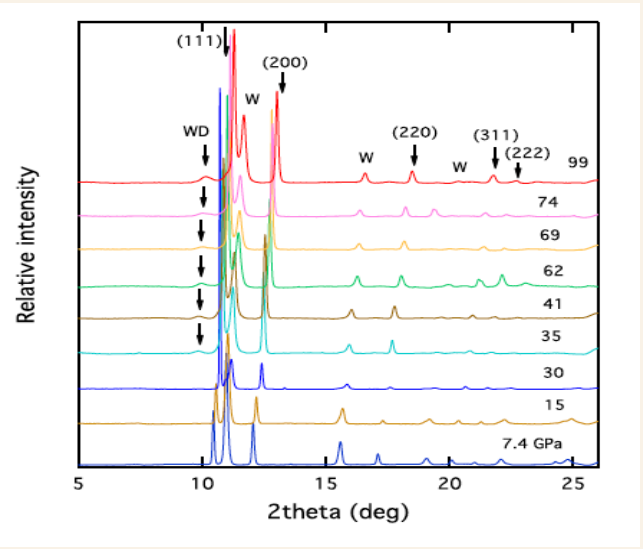
Synthesis and compression of PdH_x and PdD_x to megabar pressures



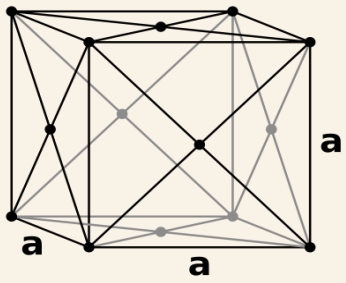
100 microns

Pd Foil / Deuterium at 10 GPa at 300 K

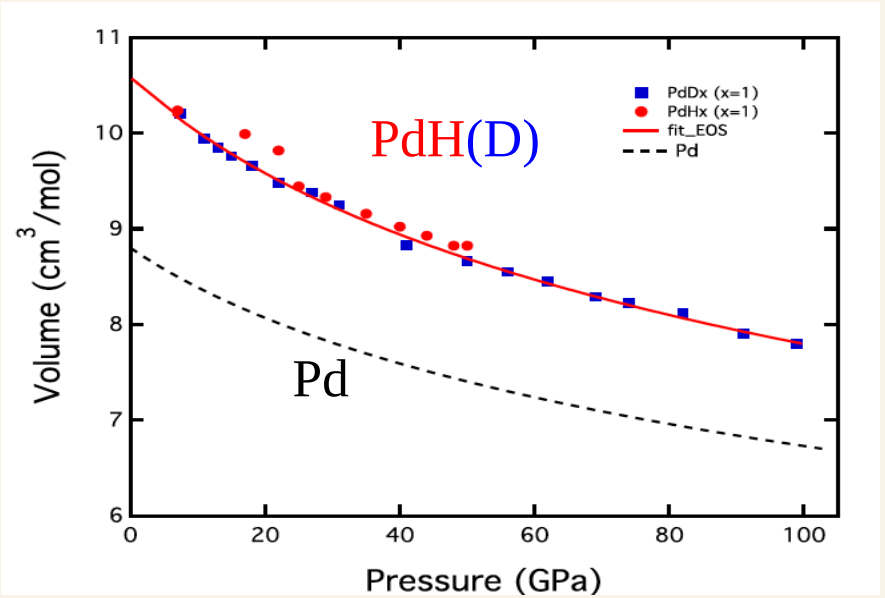
- **High-pressure behavior of PdH(D), a known superconductor?**
- **PdH_x where x > 1 is predicted to have a much higher T_c.**
- **Can we create PdH_x or PdD_x where x > 1 under pressure ?**



Structure



FCC Pd and PdHx



Diffraction patterns of PdD_x show that PdD persists up to 100 GPa

[Ahart *et al.*, *in progress*].

Keenan Brownsberger Muhtar Ahart

Whitworth-Carnegie

- **Synthesis of PdD_x and PdH_x with x=1**
- **No structural transitions o bserved up to 100 GPa**

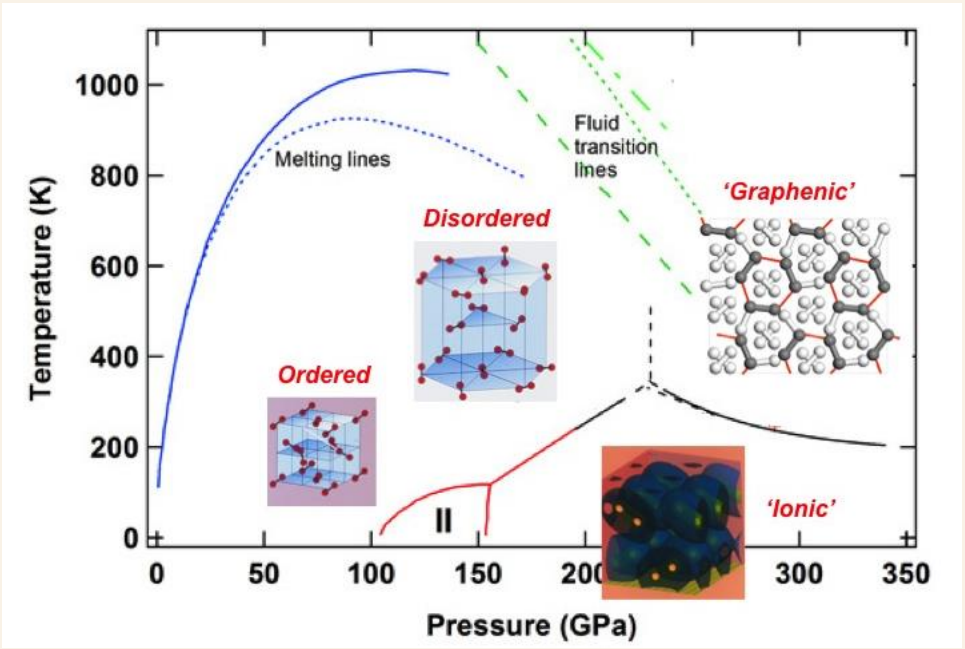
CDAC

NNSA

Continued studies of dense hydrogen

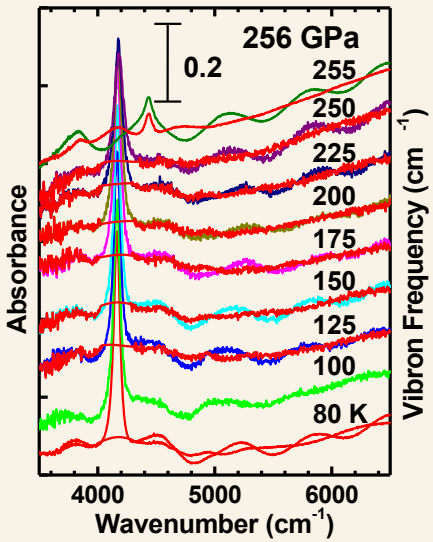
- **High energy density material?**
  (400 kJ/mole: 35 x TNT)
- **High-T_c superconductor?**
  [N.W. Ashcroft, *Phys. Rev. Lett.* 21, 1748 (1968)]
- **Superconducting superfluid?**
  [E. Babaev, A. Sudbo, and Ashcroft, *Nature* 431, 666 (2004)]
- **Path to inertial confinement fusion?**

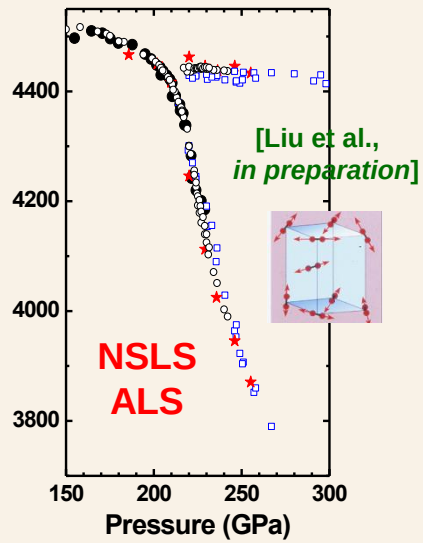
Neil Ashcroft



[Zha et al., *Phys. Rev. Lett.* (2013)]

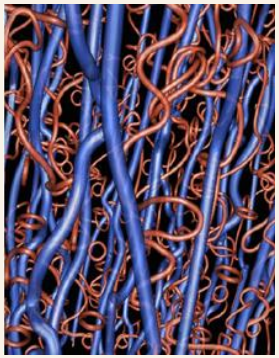
Synchrotron Infrared





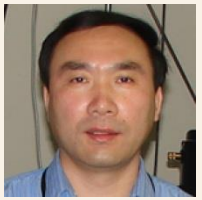
?

Superconducting Superfluid > 400 GPa



[E. Babaev *et al.*, *Nature* 431, 666 (2004)]

- 'Molecular' to 360 GPa (<300 K)
- New phases
- Semiconducting/ semimetallic

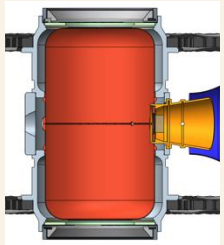


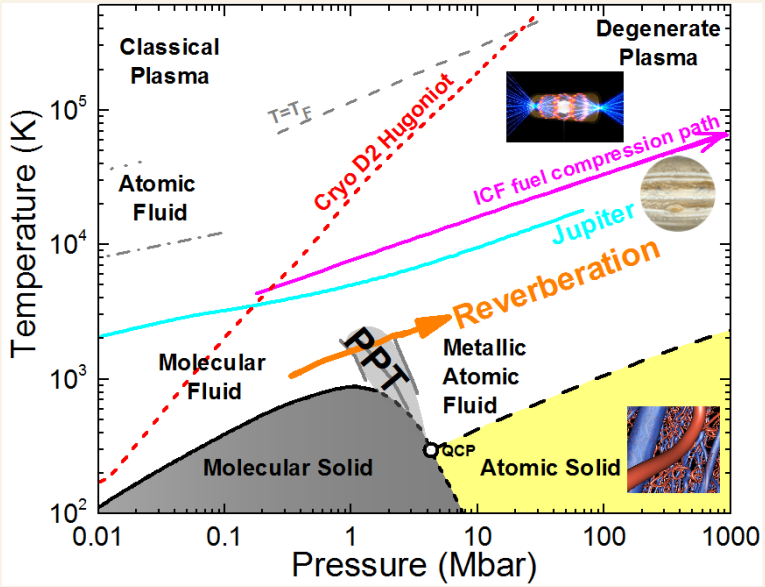
Zhenxian Liu

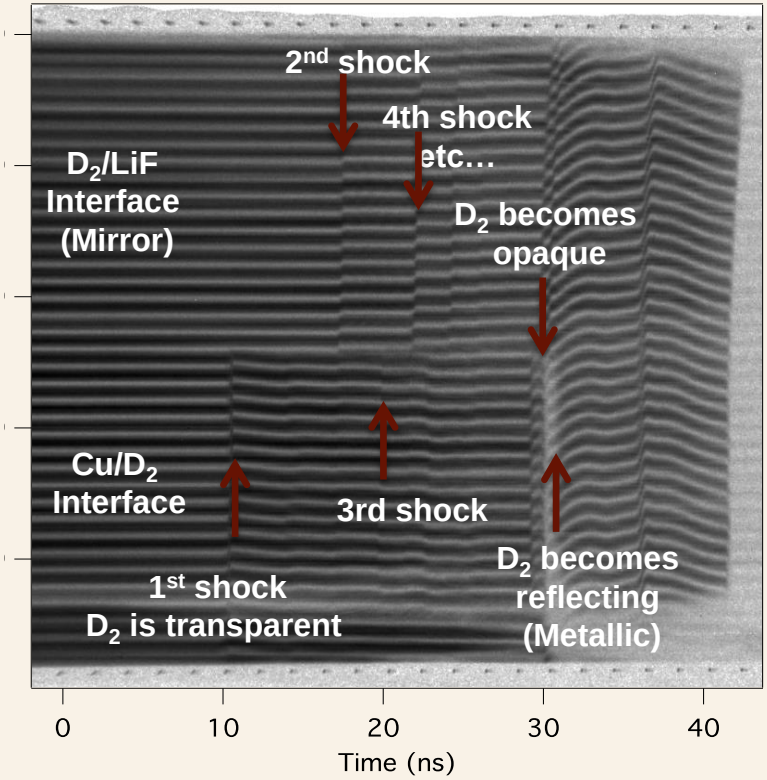
CDAC supports NIF Discovery Science experiments to explore the higher *P-T* behavior

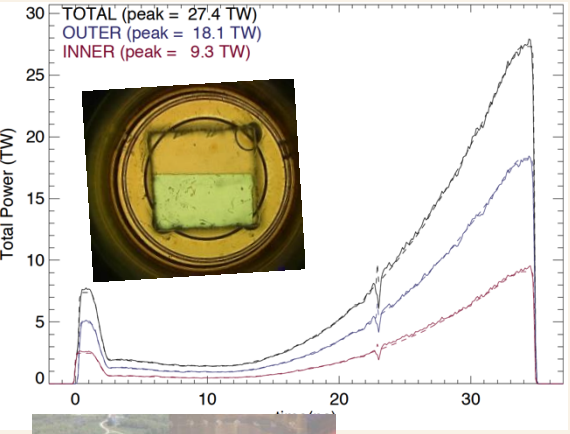
Metallization of fluid hydrogen near 2 g/cc

Marius Millot









Exploring similar P-T than Knudson et al Science 2015
~10x faster timescale

- ✓ 4 cryogenic shots
- ✓ Successful reverberation compression to >300 GPa
- ✓ Excellent quality VISAR data
- ✓ Signatures of changes in the hydrogen optical properties

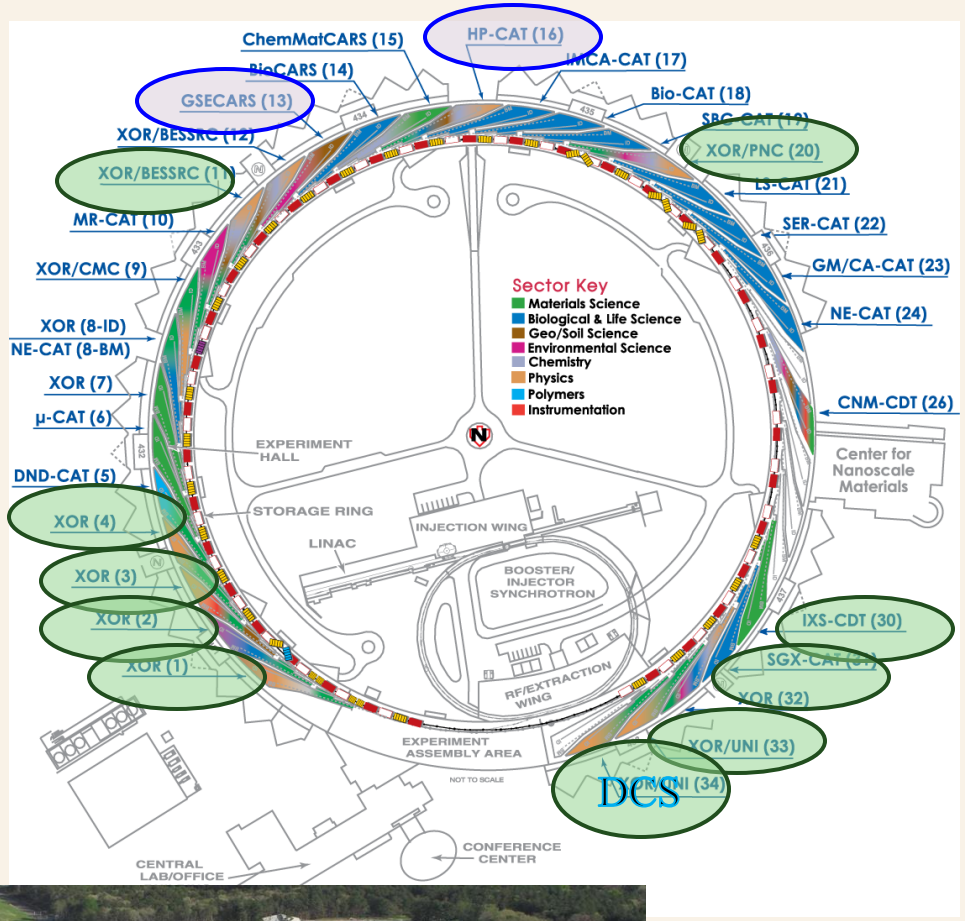
CDAC

LLNL-Berkeley-Edinburgh-CEA-Carnegie

NNSA

Facilities developments provide new opportunities for extreme conditions science

- APS Upgrade
- HPCAT Upgrade
- DCS Operations
- Other APS beamlines
- FIS Beamline at NSLS II
- NIF Discovery Science
- **Other Facilities (LCLS, Omega, MaRIE, …)**



CONCLUSIONS AND OUTLOOK

1. Education and Training

- Diverse student program with a large group of university partners
- Continued placement of personnel in NNSA labs

2. Science Program

- Continued growth in number of high-profile publications
- Novel phenomena over a broad range of extreme conditions
- New opportunities for materials dynamics under extremes
- *Opportunities at APS for the NNSA labs*

3. Technique Development

- Continued technique developments to support NNSA labs
- New x-ray techniques (imaging, time-resolved, static/dynamic)
- *Need to take advantage of APS upgrade*
- *Opportunities for coordination across DOE facilities*