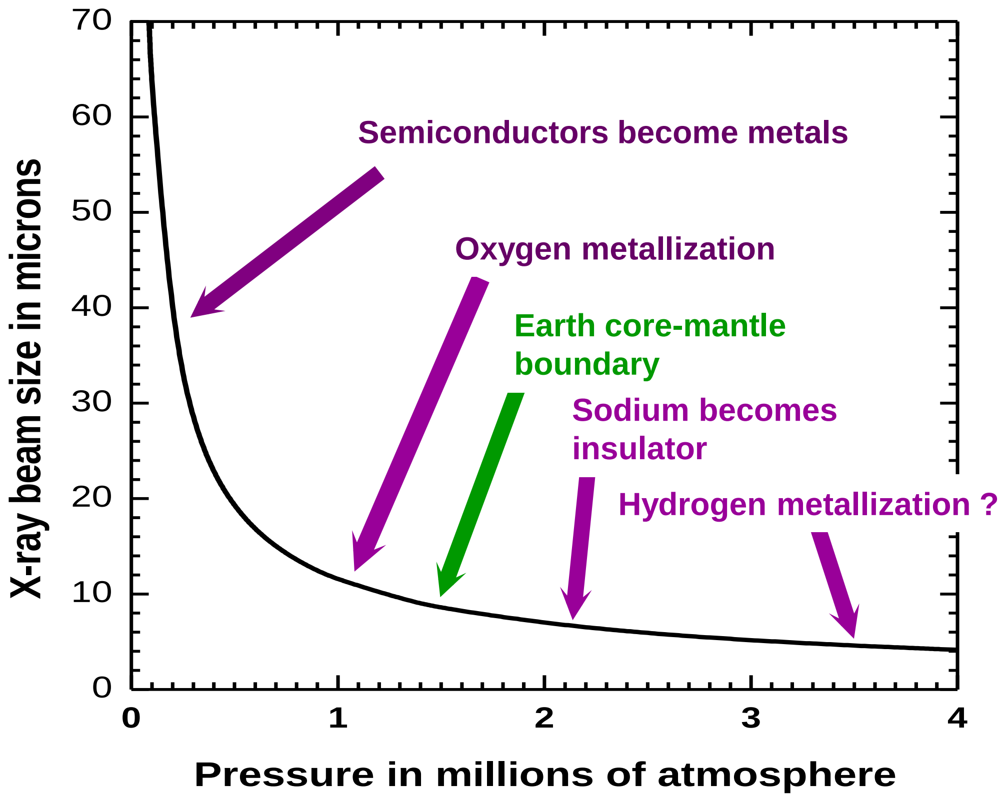
HPCAT: HIGH-PRESSURE SYNCHROTRON X-RAY CAPABILITIES ADDRESSING NNSA MISSIONS

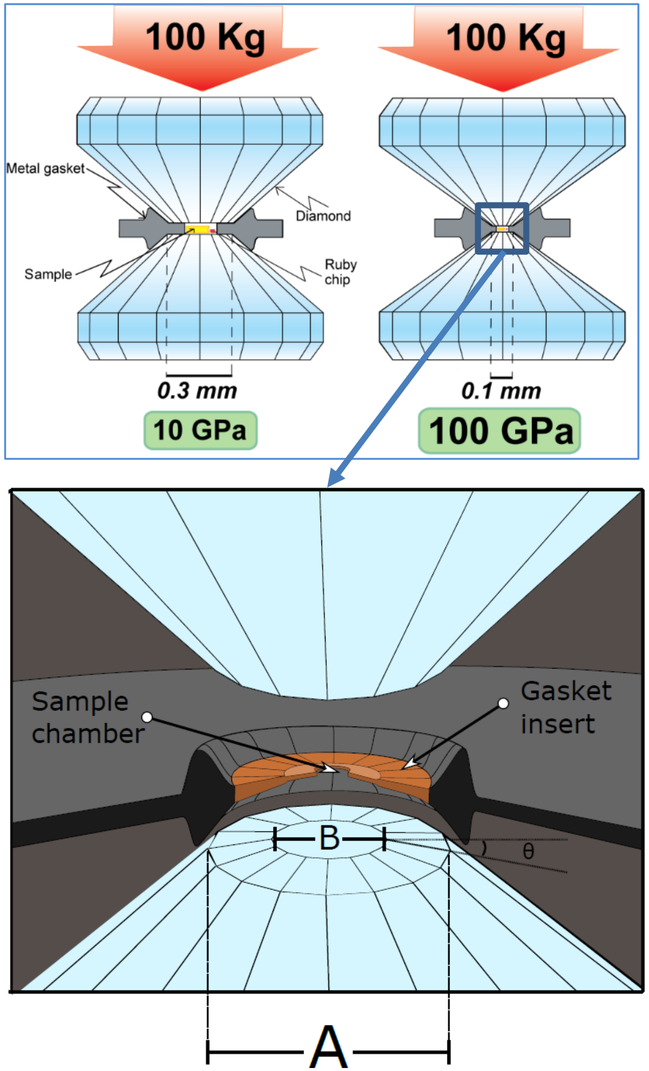
G. Shen & S. Sinogeikin (HPCAT, Carnegie)
R. J. Hemley & S. A. Gramsch (CDAC, Carnegie)
W.J. Evans (LLNL)
N. Velisavljevic (LANL)

Outline

- What/where is HPCAT?
- HPCAT current capabilities and user programs
- HPCAT opportunities on the horizon of APS-Upgrade

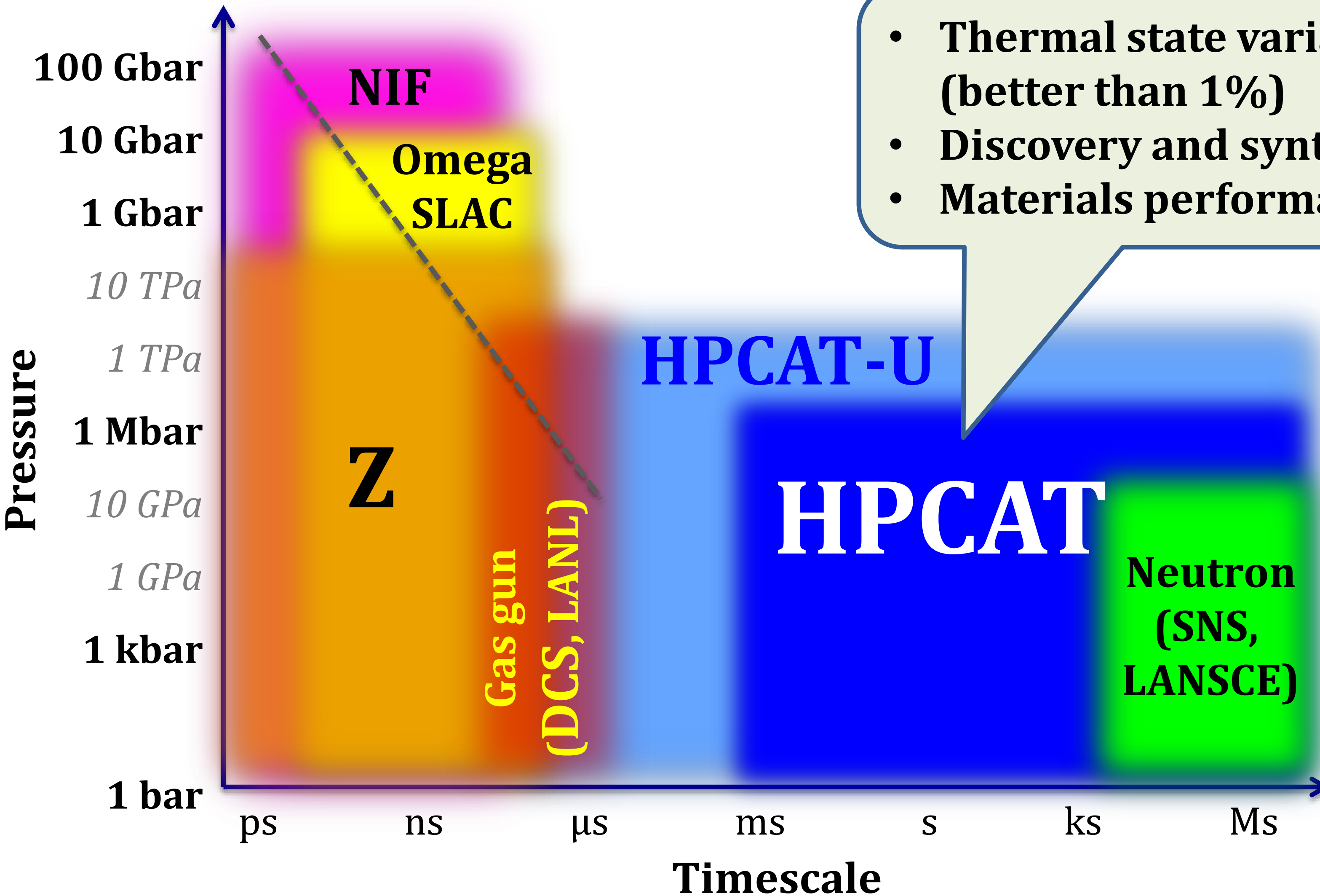
Why High-pressure Synchrotron X-ray Capabilities?





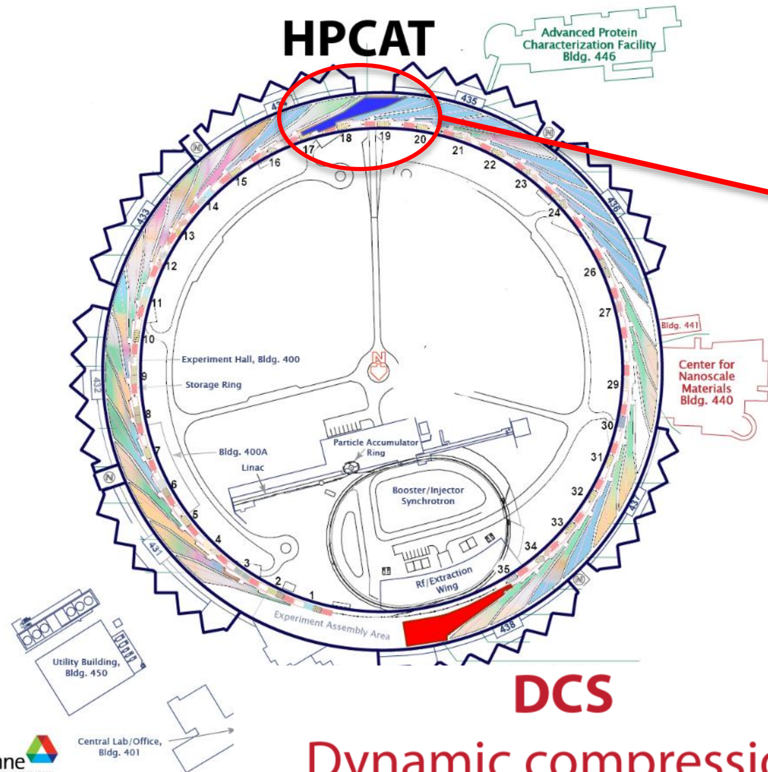
- Small and bright x-ray probes
- High energy penetrating power

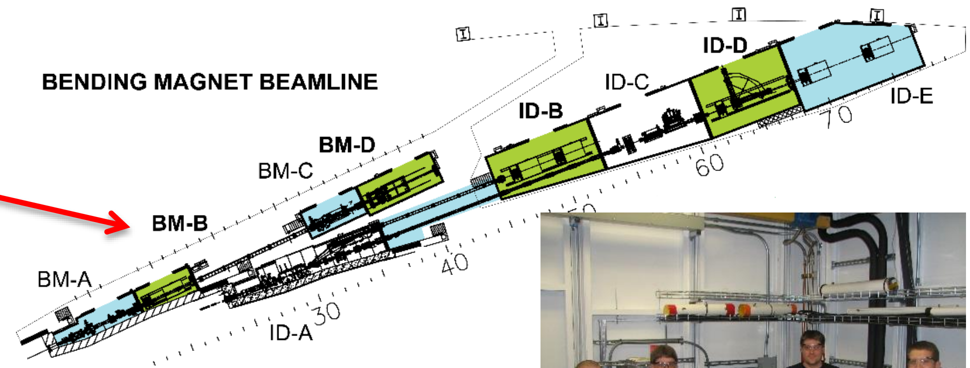
HPCAT Among Other National Facilities

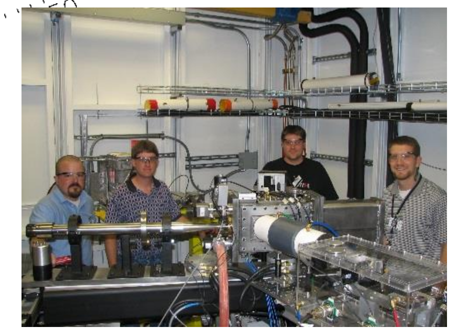


- Thermal state variables (better than 1%)
- Discovery and synthesis
- Materials performance

HPCAT at Advanced Photon Source







Shock experiments at HPCAT by Washington State group in 2007

DCS
Dynamic compression

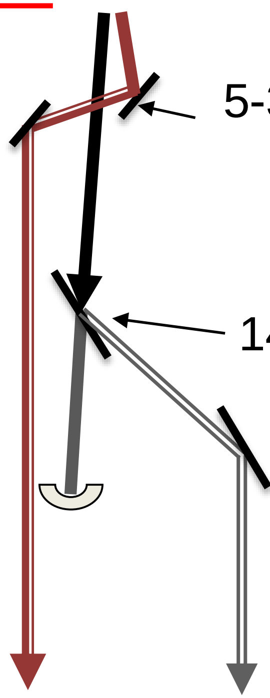
HPCAT
"Static" compression

Laser schock | Kolsky bar, Gas gun | Pulsed LH, Dynamic DAC | Diamond Cells, LVP

fs — ps — ns — µs — ms — s — ks

HPCAT – Four Beamlines

Sector 16
Newly canted undulator beams

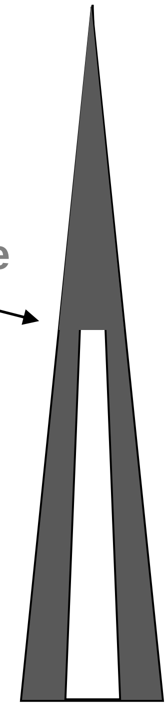


5-36 keV

14-42 keV

Sector 16
Bending magnet beam

Split in space



ID-C/D/E:
Spectroscopy
Scattering
XES,IXS – 1eV
NRIXS – 2meV

IDB:
Micro-diffraction
(high photon flux)
Laser heating (>3000K)
Cryostat (<4k)

BM-C/D:
Micro-diffraction
XANES

BMB:
White Laue
Paris-Edinburgh Press
(fast x-ray imaging)

HPCAT – Four Beamlines

Sector 16

Sector 16

- **The largest synchrotron facility dedicated for static high pressure (HP) research**
- **The most versatile HP synchrotron techniques in a single sector, covering x-ray diffraction, x-ray spectroscopy, and x-ray imaging**
- **Each individual technique at the state-of-art**
- **Comprehensive set of advanced on-line support equipment, sample preparation, and complementary characterization devices**

e
burgh
Press

XES,IXS – 1eV
NRIXS – 2meV

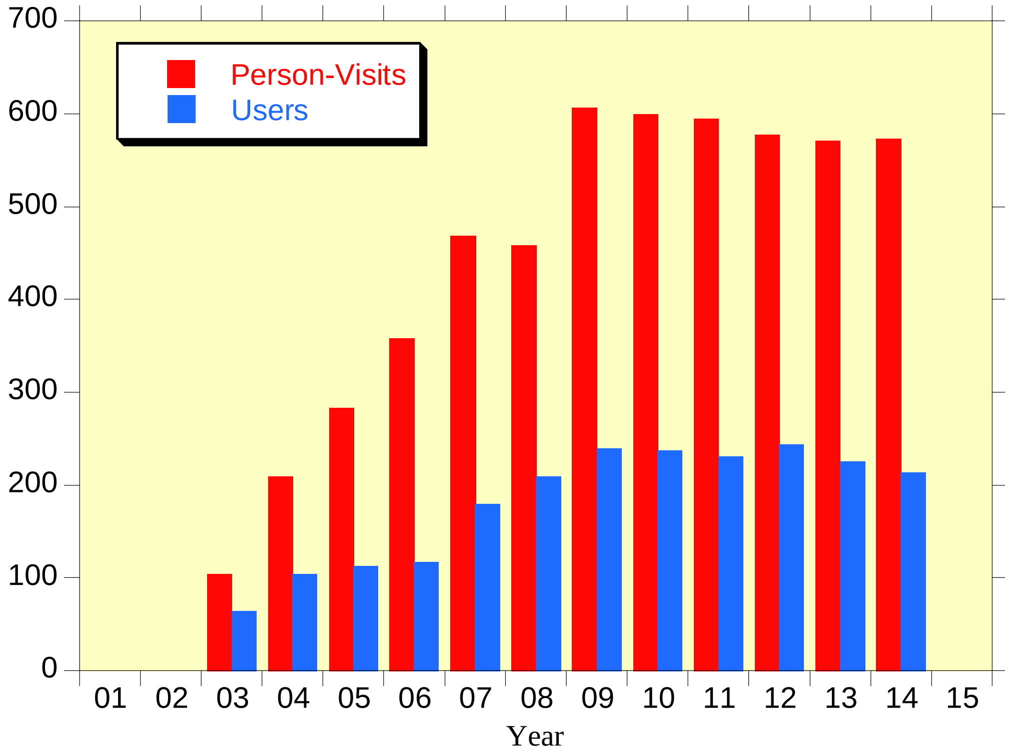
Cryostat

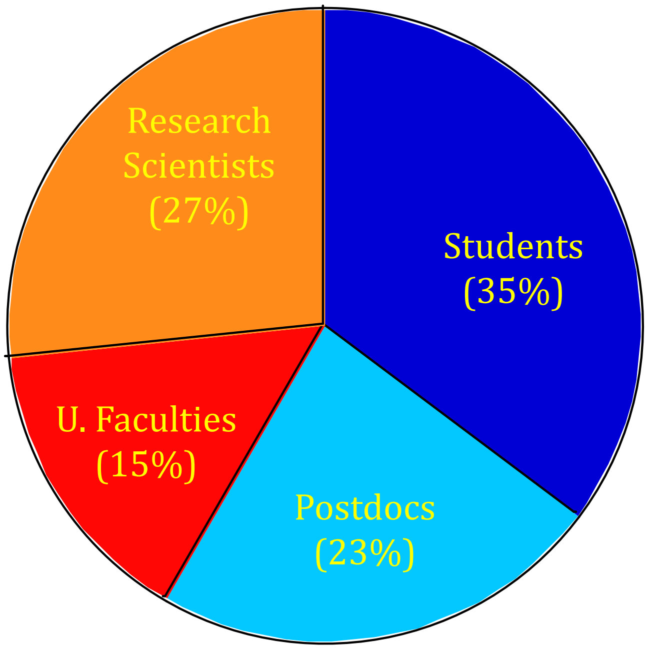
Addressing Programmatic and Technical Needs for NNSA

- **Structure at extremes**
  - Crystal structure: symmetry ⟶ atomic position ⟶ electron density topology
  - Amorphous - nano-structure – micro-structure – grain boundaries
- **Electrons at extremes**
  - Valence electrons, conducting electrons
  - Strongly correlated electrons, (de)localization
  - Hybridization, spin transitions
- **Equations of states and phase relations**
  - P-V-T EOS at extended range
  - Phase diagram at extended P-T range
- **Kinetics under rapid (de)compression**
- **Strength and rheology at extremes**
- **Liquid properties at extremes**

$\Longrightarrow$ **Materials performance, materials synthesis**
Basic science, novel materials discovery

HPCAT Users and Publications





User Distributions in 2014

- >2 papers/week in peer reviewed journals, one of the most productive sectors
- >23% appear in journals with impact factors ≥PRL
- 11 student or postdoc users who later joined National Labs as staff
- 61 Ph.D. theses since 2003

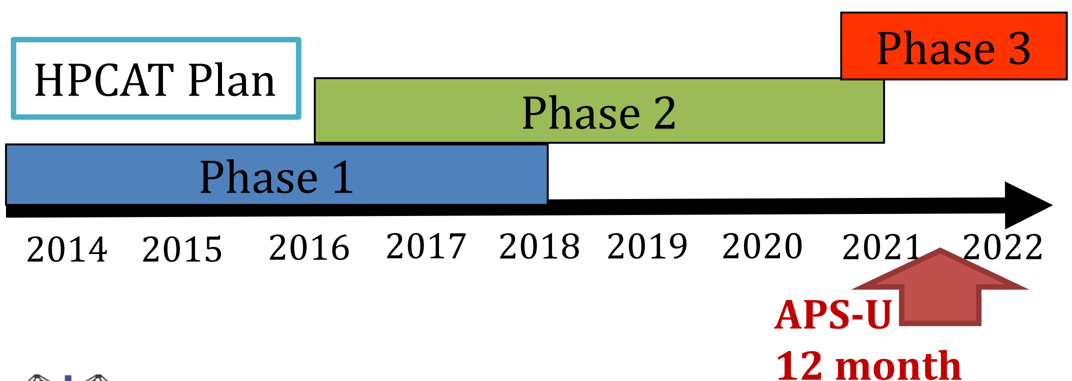
What are the Technical Challenges?

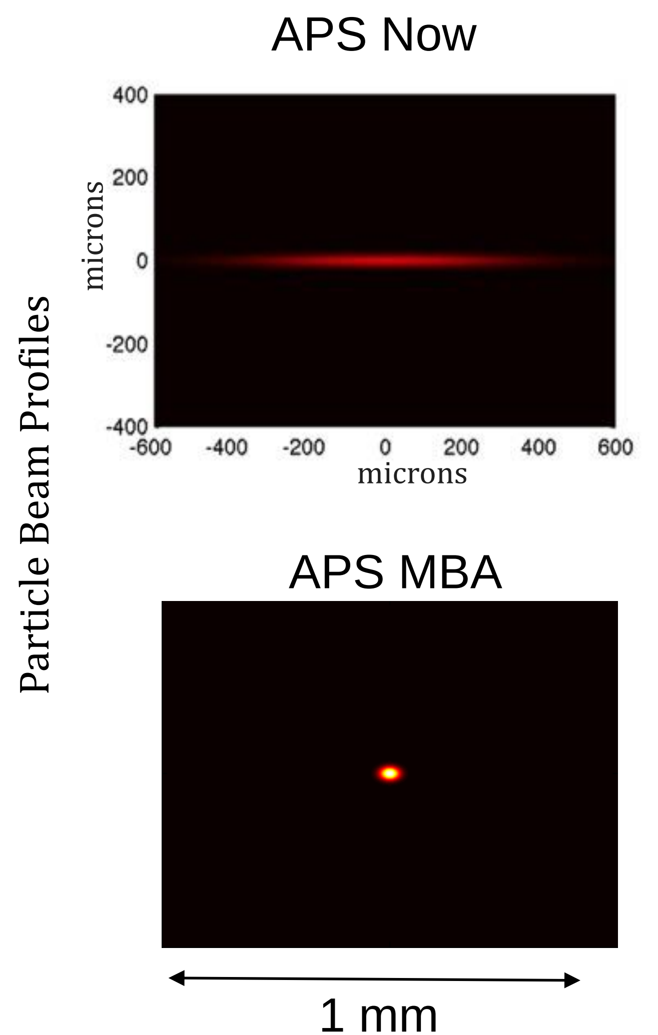
- Going to pressures beyond 400 GPa
  - Pressure device and anvil designs, sub-micron probes
- Hydrogen at Mbars
  - Higher flux, better collimation
- Thermal EOS – challenging precision and accuracy
  - Gradients, calibration
- Complex and heterogeneous materials
  - Microstructure, meso-scale structure, interfaces
- Software
- Theory
- Detectors

APS Upgrades / Changes

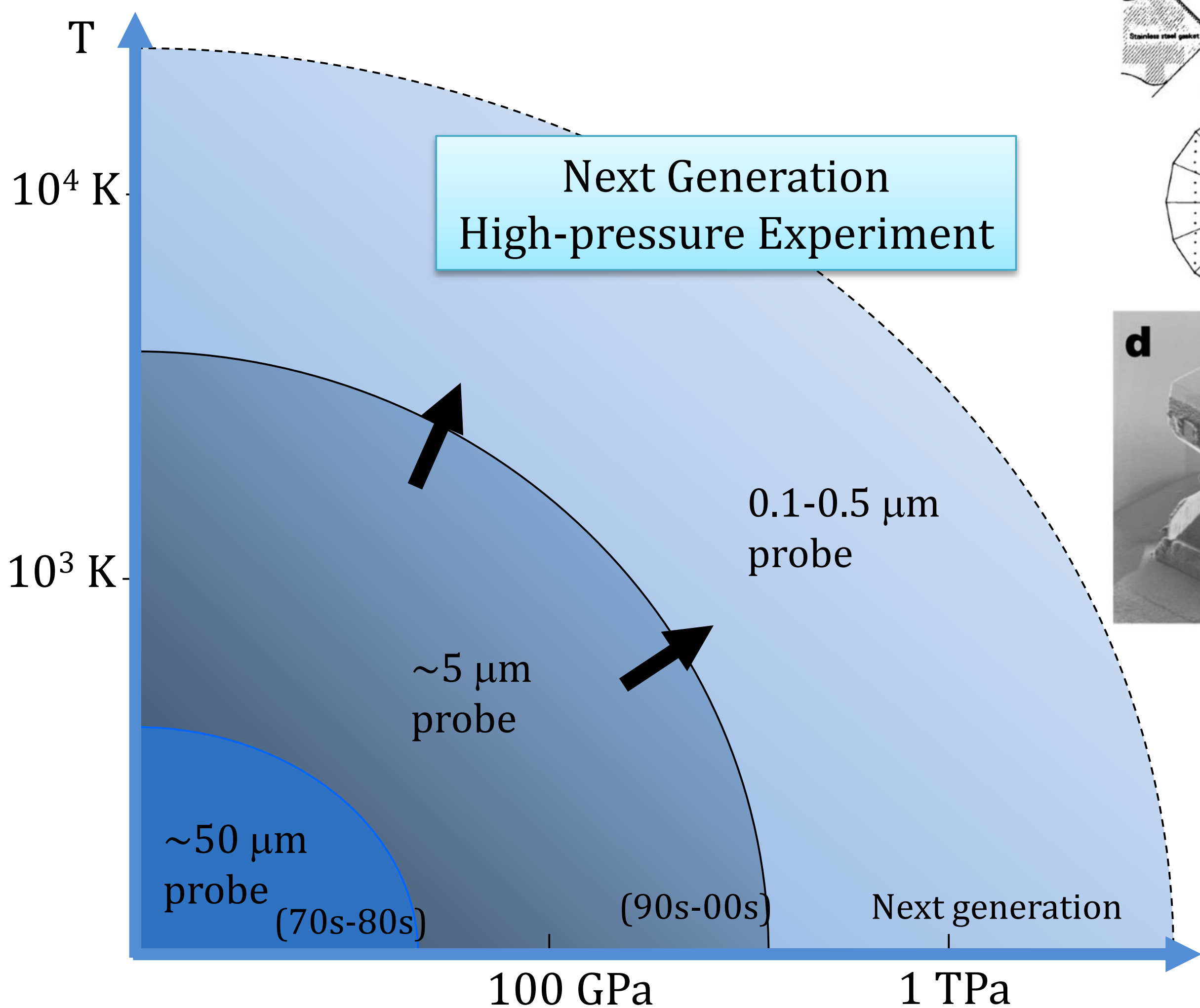
- Higher brightness (~x100)
- Higher coherent fraction (~x100)

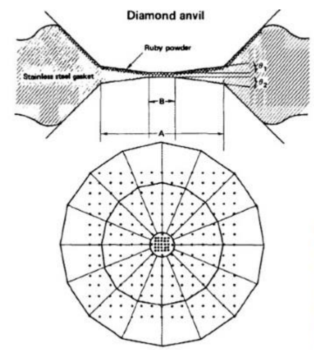
- 3-Pole Wiggler will replace Bending Magnet Source
- Superconducting undulator
- High quality focusing x-ray optics
- Detectors

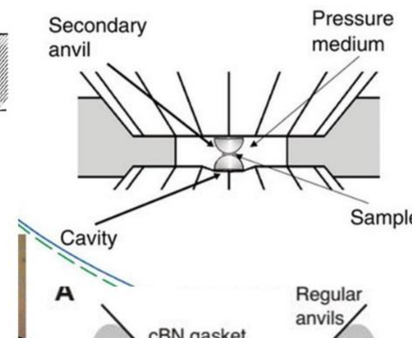


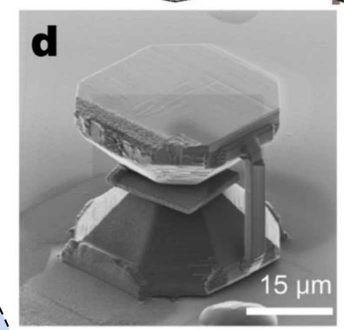


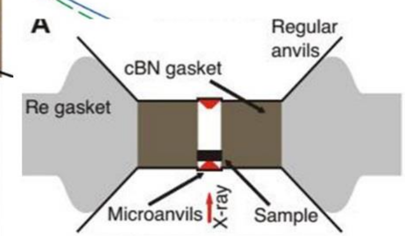
Opportunities for HPCAT

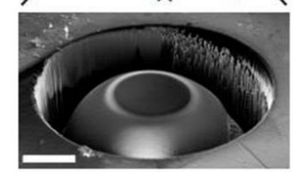


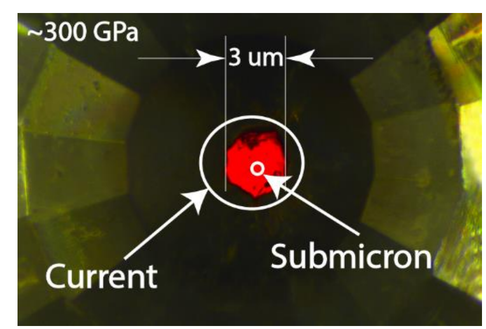






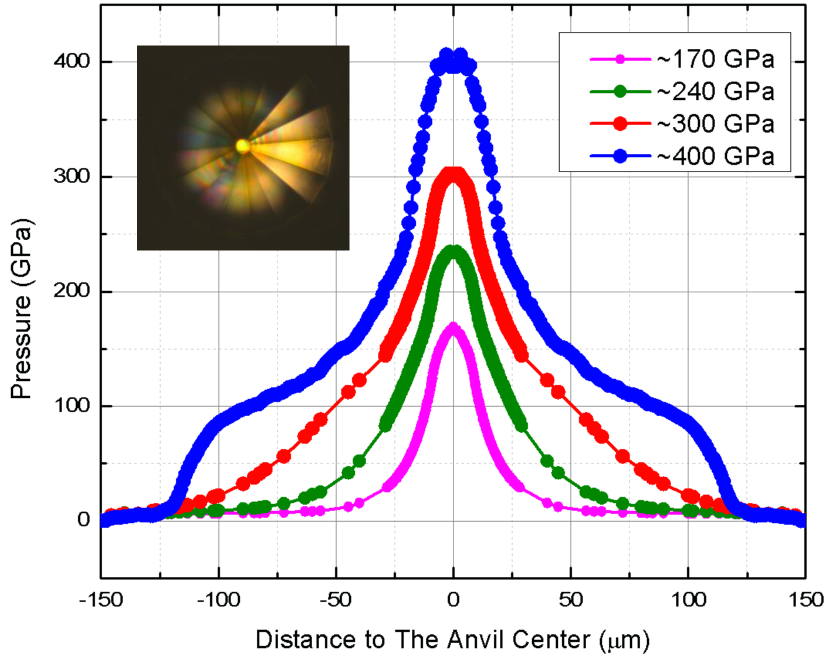


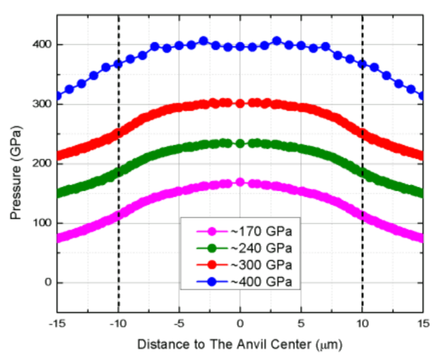


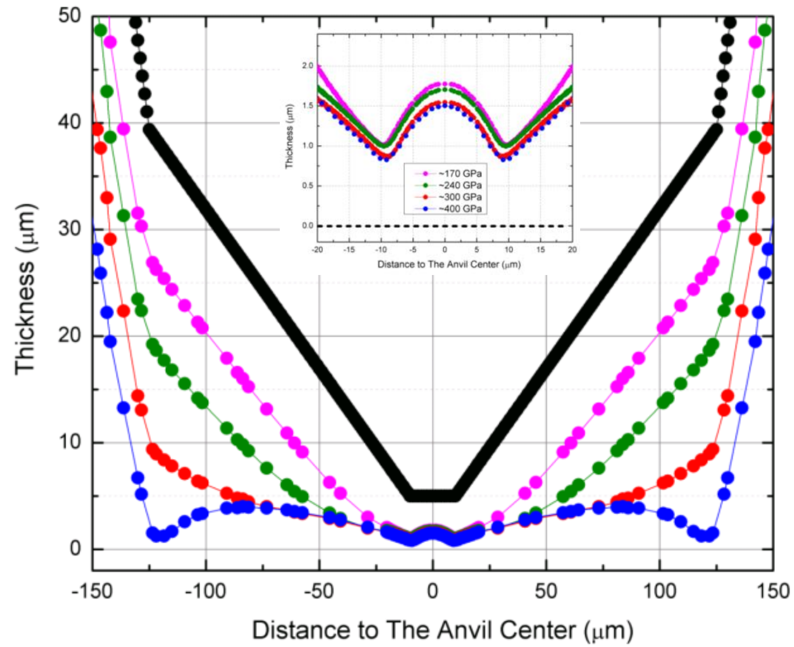


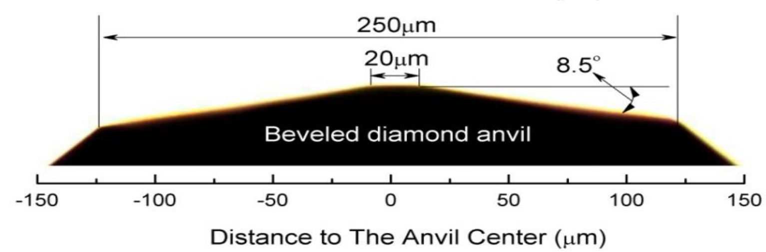
Opportunities for HPCAT

Going Higher Pressures



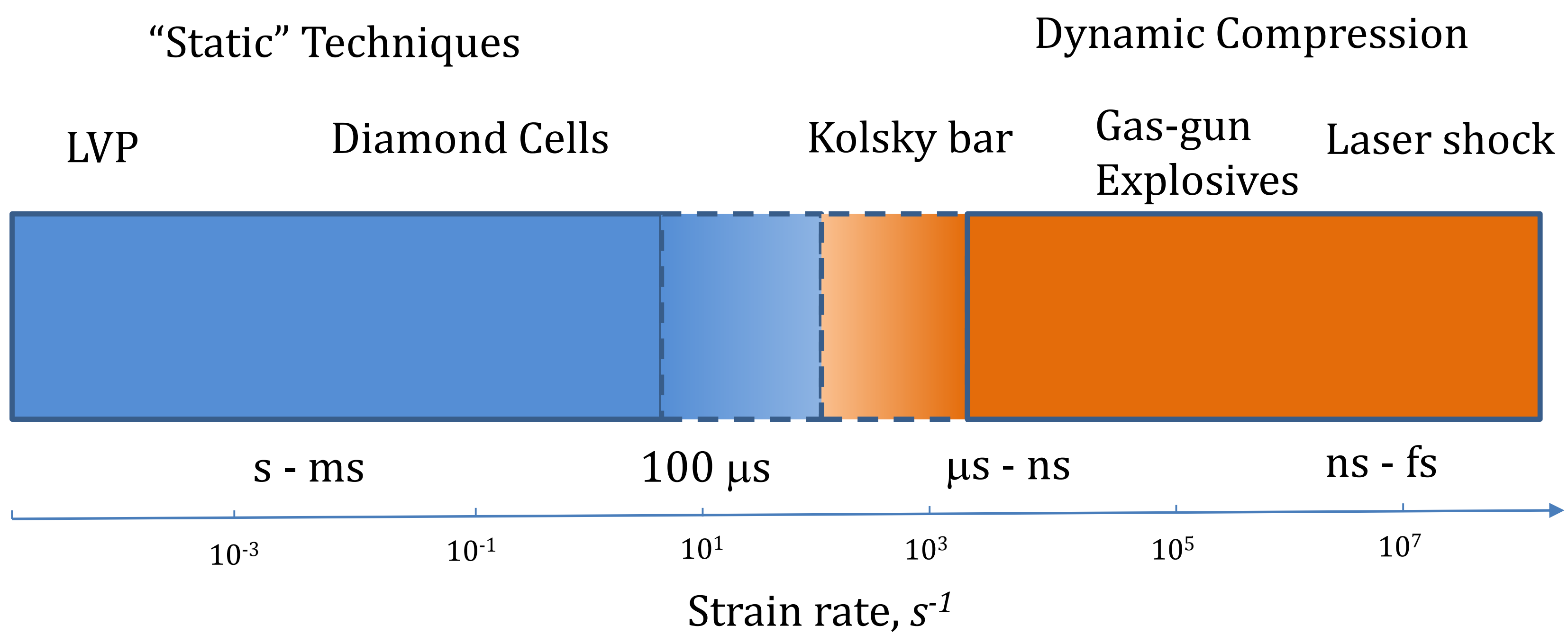






Opportunities for HPCAT

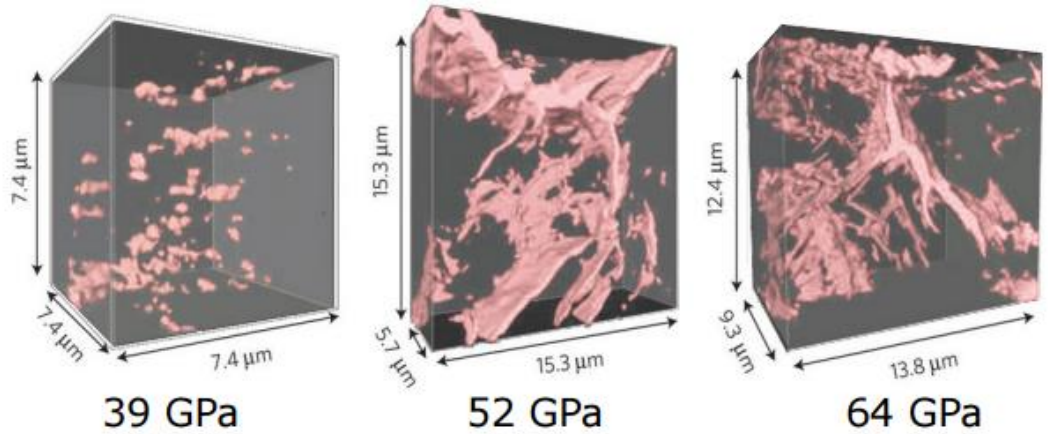
Filling the strain rate gap between "static" and "dynamic" and study the dynamic properties



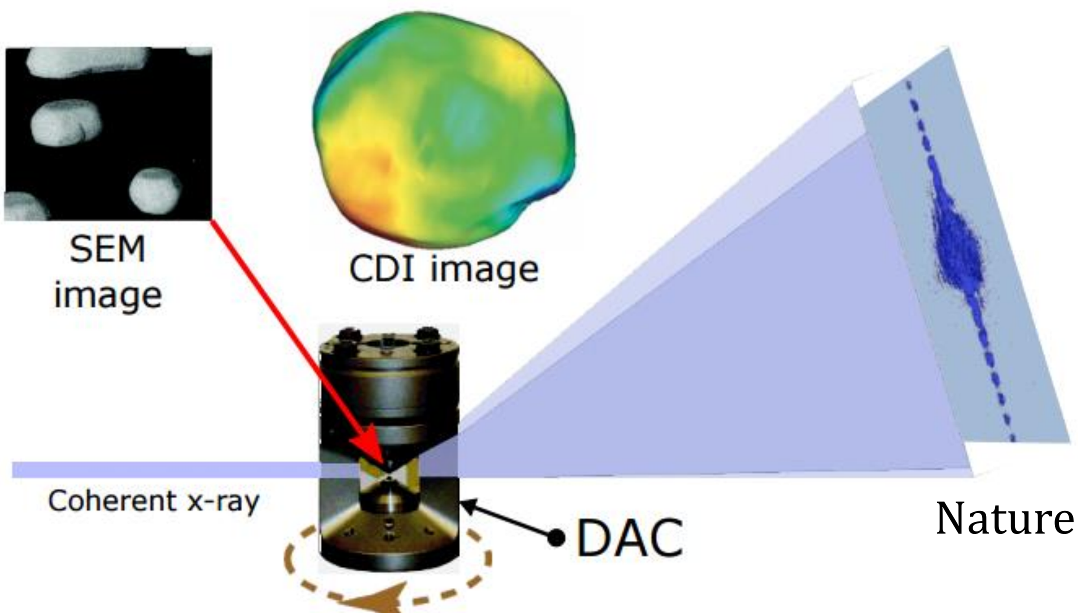
Opportunities for HPCAT

Complex and heterogeneous materials

Obtain 3D imaging at sub-micron scale for multi-scale information at extremes



Nature Geo. (2013)



Nature Comm. (2013)

Goals of HPCAT Upgrade

- To establish a set of sub-micron x-ray probes for multi-Mbar experiments
- To reach static pressures beyond 5 Mbar, even above 1 TPa

  Designs, routine operations
- To obtain precise thermal EOS and determine HP melting at multi-Mbar
- To address polyamorphism in liquids
- To fill the strain rate gap between “static” and “dynamic”

  Kinetics, metastability, transition and reaction mechanism
- To obtain 3D imaging at sub-micron scale for multi-scale information at extremes

  Heterogeneity of materials, micro- and meso-scale structure

Acknowledgment

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

Sponsors

NNSA
National Nuclear Security Administration

U.S. DEPARTMENT OF ENERGY
Office of Science

Partners

CARNEGIE INSTITUTION FOR SCIENCE

CDAC
CARNEGIE/DOE ALLIANCE CENTER

UNLV
UNIVERSITY OF NEVADA LAS VEGAS

Lawrence Livermore National Laboratory

Los Alamos NATIONAL LABORATORY EST. 1943

SUPPLEMENTAL SLIDES

HPCAT Beam-time Distribution

- 25% to APS general users
- 60% to HPCAT partners

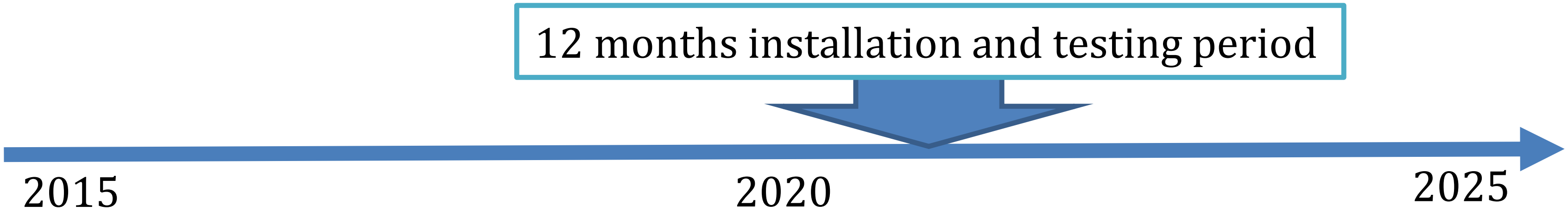
  Within the 60%

  – 75% NNSA (30% CDAC, 20% UNLV, 20% LLNL, 5% LANL)

  – 25% BES
- 8% beamline operation (typical)
- 7% R&D (HPCAT staff or via collaboration with partner users)

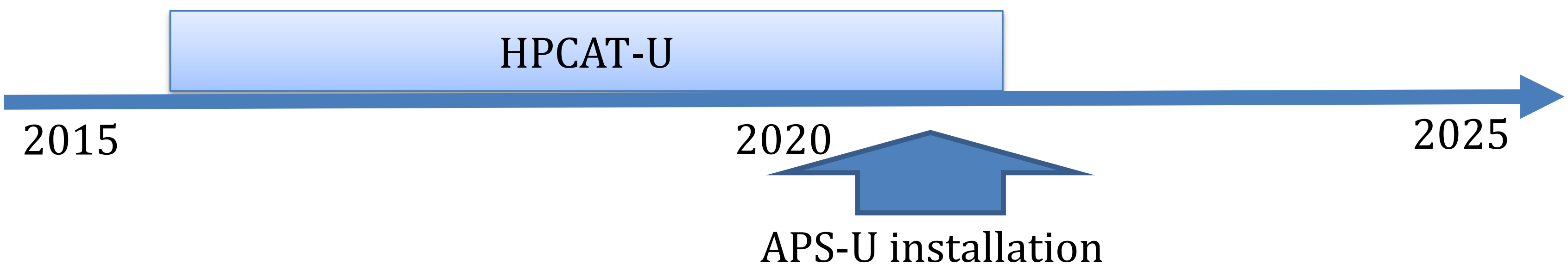
APS-U Scope

- New, 4th generation multi-bend achromat storage ring lattice in the existing tunnel
- Doubling of the ring stored beam current
- Replacements of all Front-Ends
- A suite of new beamlines designed for best-in-class performance with MBA source properties
- Optics for remaining beamlines to take full advantage of MBA source properties
- Improved beam stability
- **All beamlines will realize significant benefits**
- **Well-defined** installation and testing period is a **key deliverable**

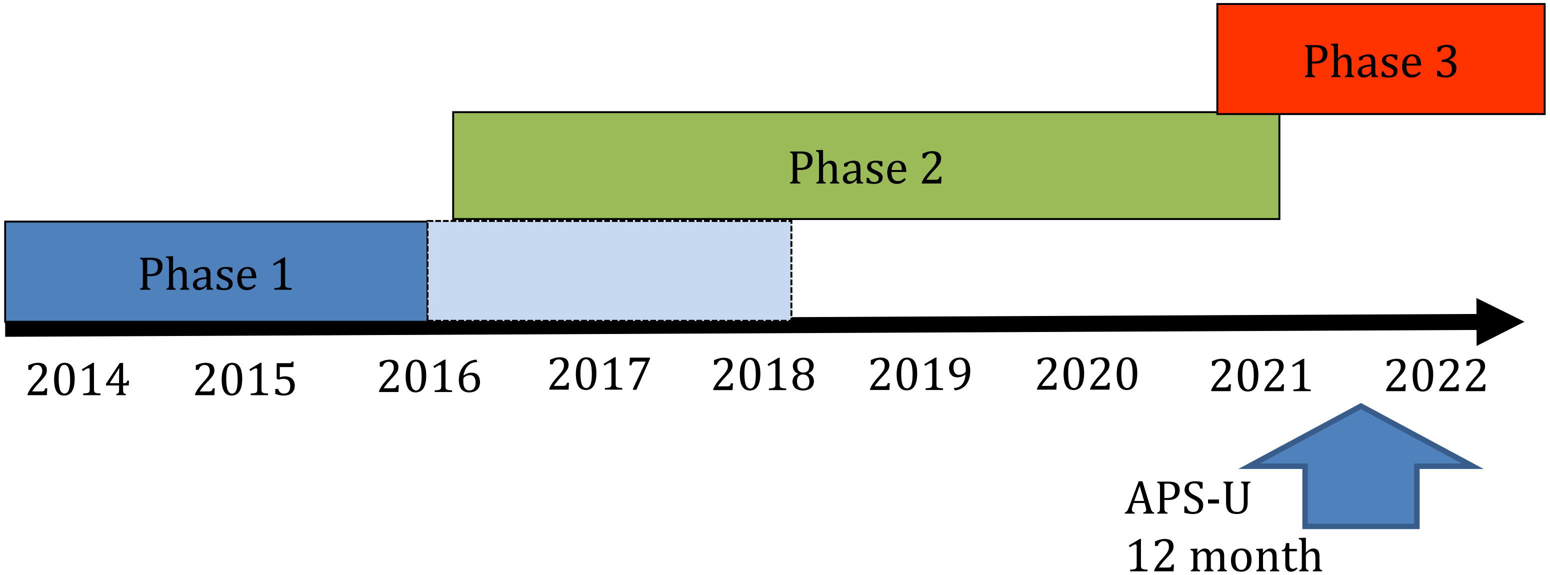


HPCAT-U Scope

- Enabling sub-micron x-ray probes, including diffraction and spectroscopy
- Replacements of x-ray optics matching the APS-U
- Reconfigurations of bend magnet beamlines for 3-pole Wiggler source
- On-sample flux increase by >100 times in all four beamlines
- Improved beam stability
- Advanced detectors
- Installation and testing period consistent with APS-U



HPCAT-U Timelines



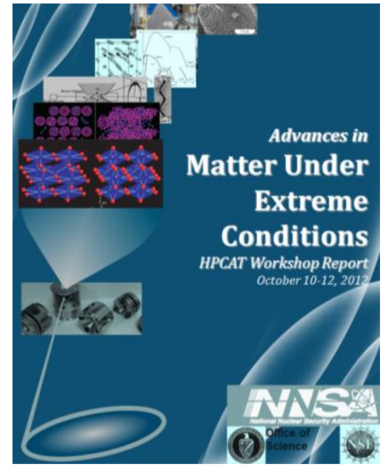
- Phase I: *Near future projects*
- Phase II: *Sub-micron probes, hutch reconfiguration, collimation optics, advanced detectors*
- Phase III: *New x-ray imaging techniques (coherence, diffraction tomography), advanced detectors*

Student and Postdoc Users at HPCAT

Who later joined	Number of people
National Lab as a staff	**11**
National Lab as a postdoc	**10**
Faculty member in U.S. universities	**24**
Ph. D. Thesis since 2003	**57**

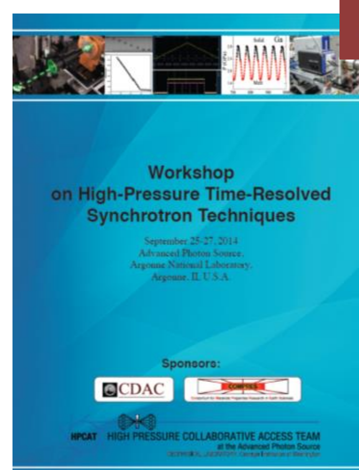
Working with Communities

- Workshops and summer schools
  - Once a year in average
- Scientific meetings
  - Users and staff
- Monthly meetings
  - HP-SR
  - HP interest group (APS and beyond)
  - Student/postdoc/visitor



2012

2013



2014

Recent Upgrades and New Capabilities

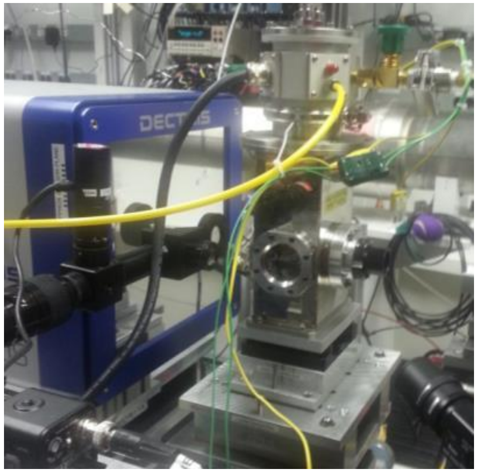
- **Beamlines**
  - Canted undulator operation - two completely independently operational undulator beamlines
  - Improved x-ray focusing optics - beamsize reduction by 5-10 times
- **HP X-ray Diffraction**
  - Rapid (de)compression - kinetics
  - Modulated (pulsed) laser heating – HP melting
  - Multigrain crystallography – charge densities
- **HP X-ray Spectroscopy**
  - Resonant x-ray spectroscopy – electronic structures
  - Advanced collimation techniques – measuring weak inelastic scattering from hydrogen
- **HP X-ray imaging**
  - Fast optical and x-ray imaging – viscosity, miscibility, transition kinetics
- **Integrated**
  - Comprehensive studies of liquids at high pressures - density, structure, viscosity, sound velocity
  - XRD and XANES; XES and XRD, NRIXS/NFS and XRD – better overall science picture
- **Support equipment**
  - Fast and sensitive x-ray detectors
  - Various cryostats, various heating techniques
  - Advanced sample preparation instruments

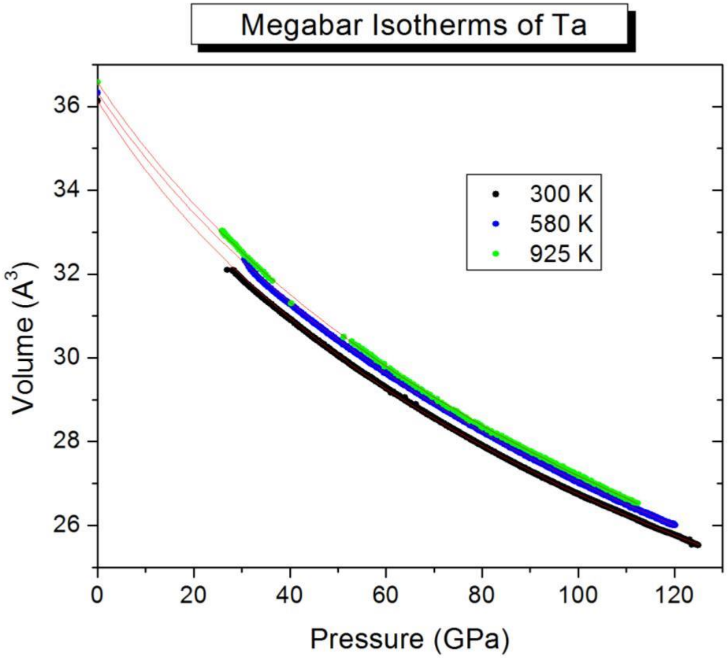
Examples of Data Obtained at HPCAT

- Thermal equation of state (EOS)
- Dynamical properties using rapid (de)compression
- Charge density at HP
- Liquids at HP
- Band structures of hydrogen and alkali metals
- Micro-structure and heterogeneity

High Precision of Equations of State

One isotherm in a few seconds





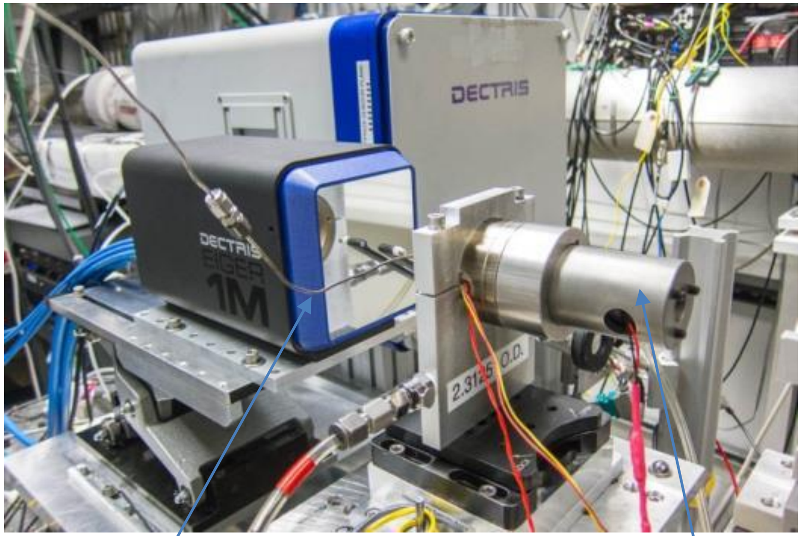
Experiment	V_0 uncertainty	B_0 uncertainty	B_0' uncertainty
Cynn and Yoo, 1999	None (fixed)	0.025	0.029
Dewaele et al, 2004	None (fixed)	0.017	0.046
HPCAT (recent)	10^{-4} (refined)*	0.005	0.006

*important, and it *should* be refined (see, for example, Ross Angel)

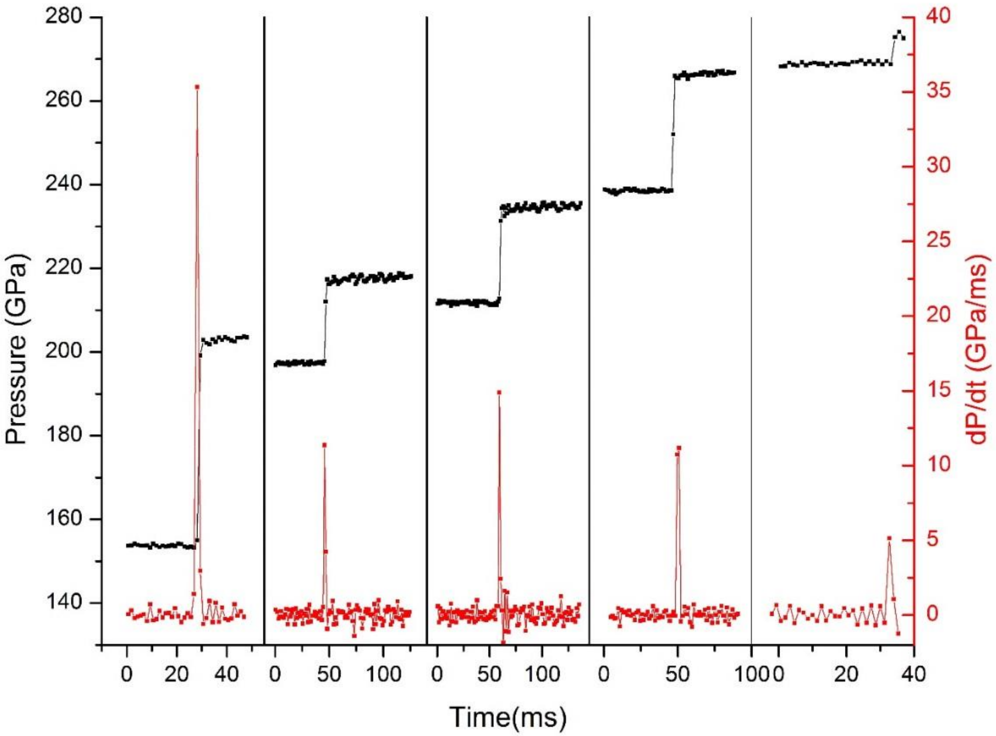
Rapid Compression and Decompression

Filling the Strain Rate Gap between Static and Dynamic Experiments

Time-resolved full-frame experiments



Eiger 1M **d-DAC**

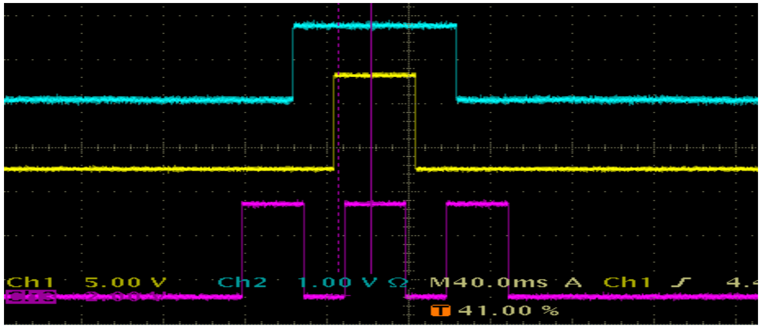


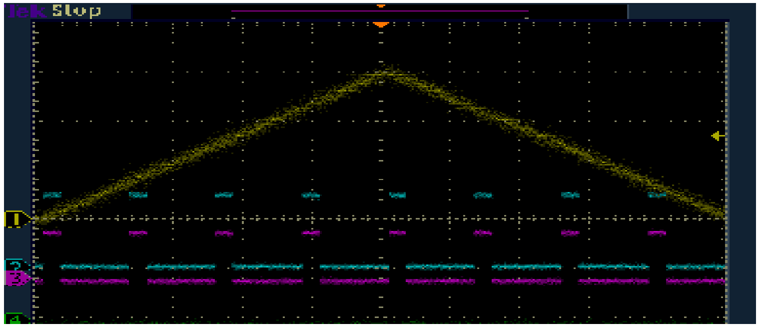
Anvil: *50-250 μm bevel*
Sample: *Mo+MgO*

Rapid compression rate at multi-Mbars:

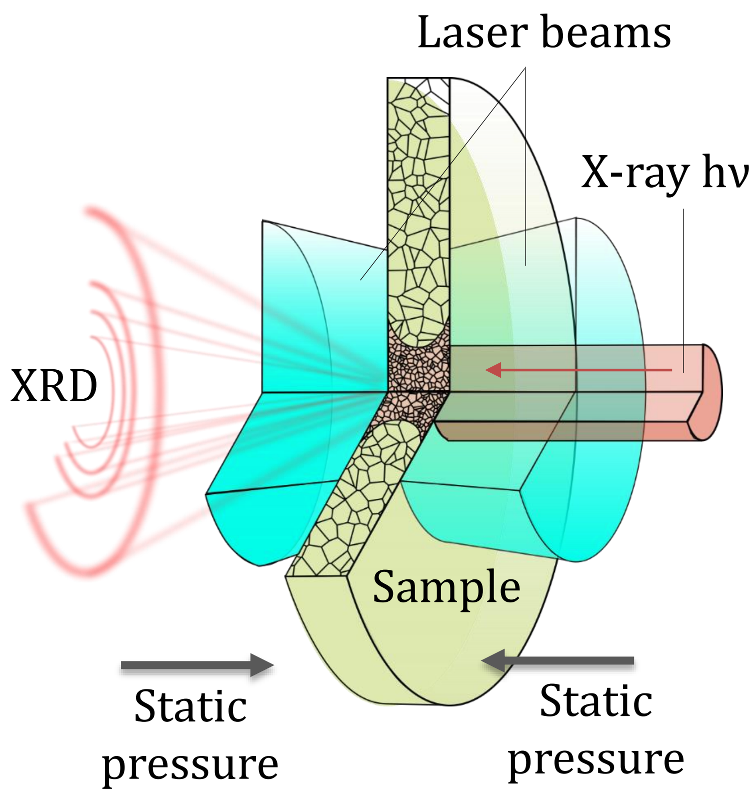
> 35 TPa/s (or 350 Mbar/s)

Modulated Laser Heating
addressing HP Melting and Thermal EOS

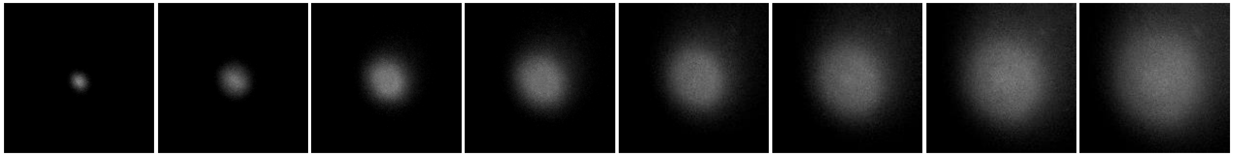




Modulated heating to control kinetics

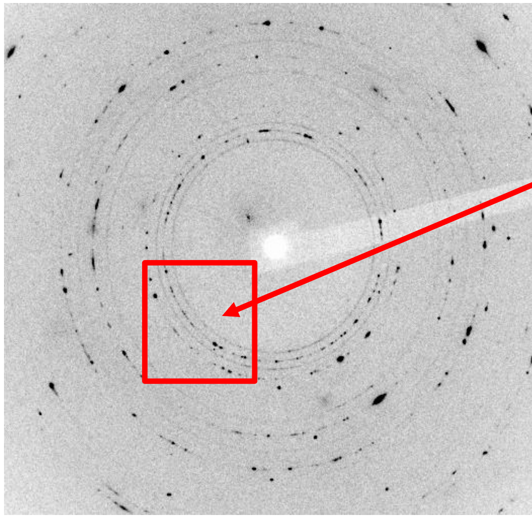


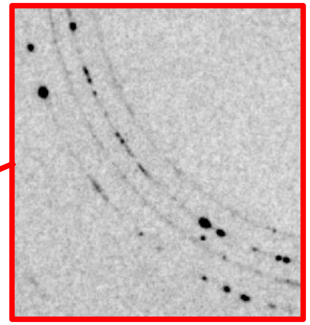
Double sided heating to minimize axial T gradient



Heating spot > Ø120 μm
to reduce radial T gradient

Electron Density Distribution from Multigrain Crystallography

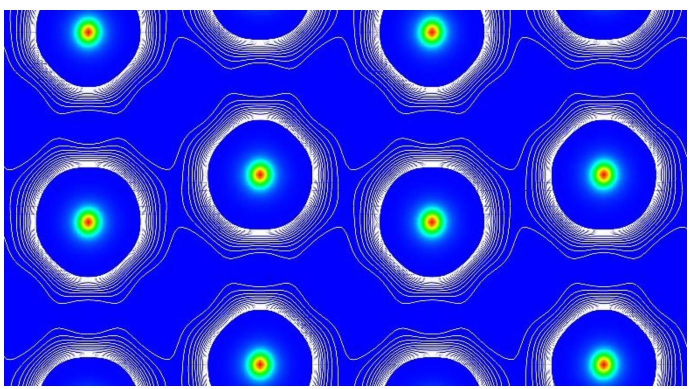


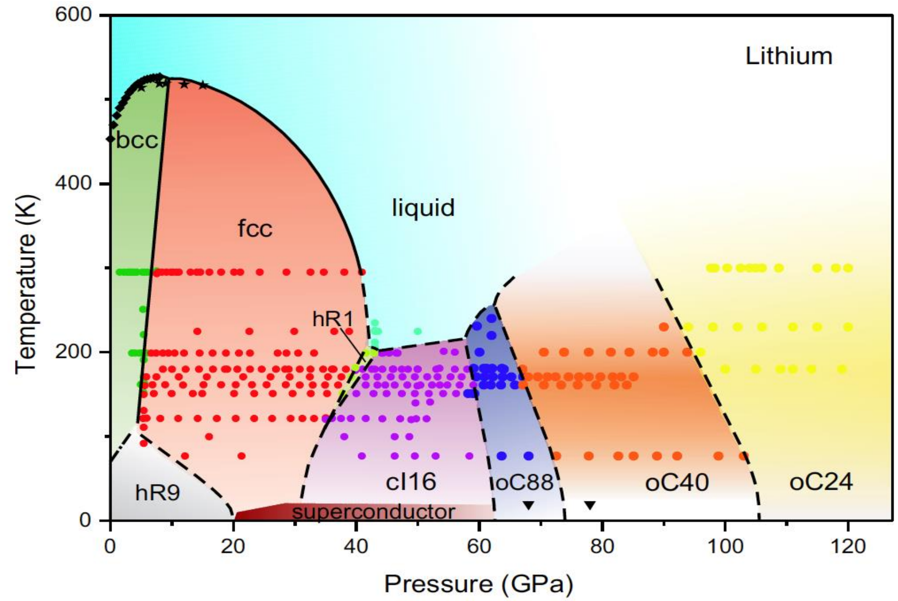


Single crystal XRD information from course powder patterns

High pressure
α-Ce at 5.7 GPa

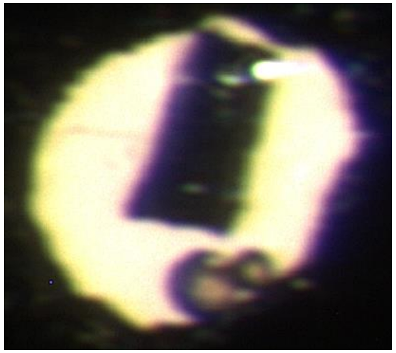
< 3% R-factor
100% Completeness!

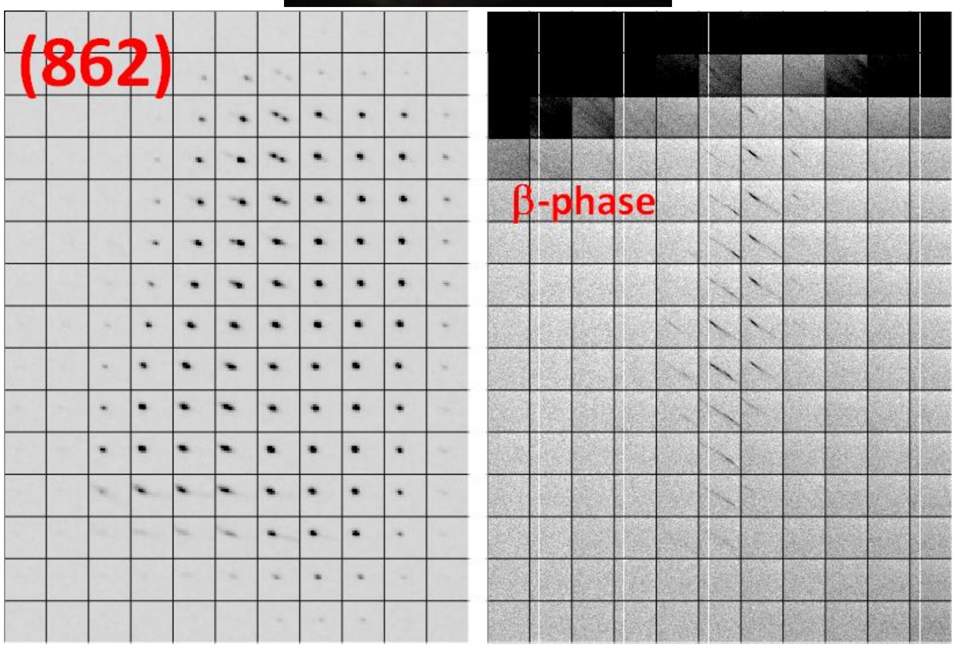




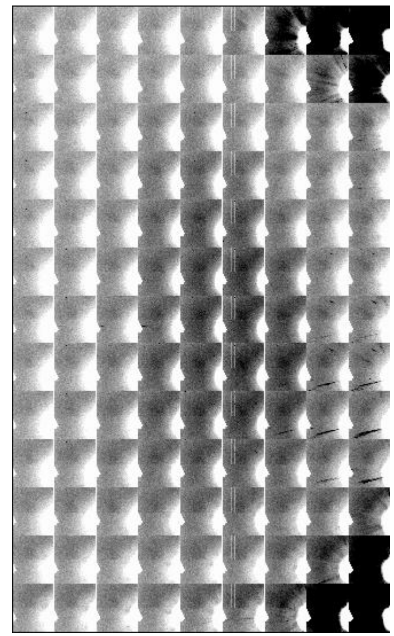
Complex HP crystal structures are being determined

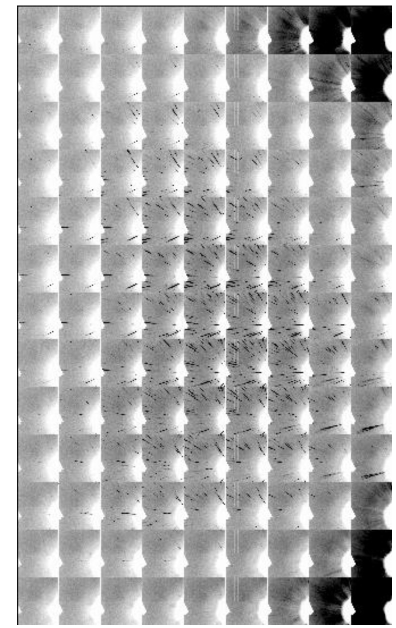
Time Resolved Microstructural Changes





Strain, mosaic, and orientation of Si at 12.8 GPa

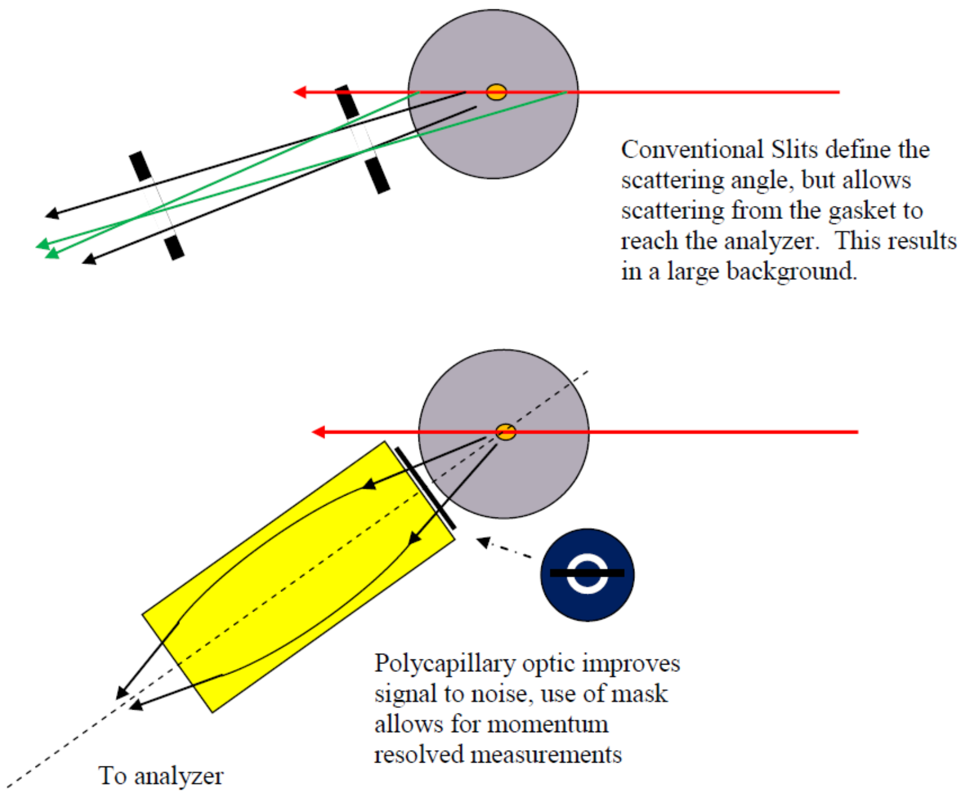

Zr at 4.3 GPa

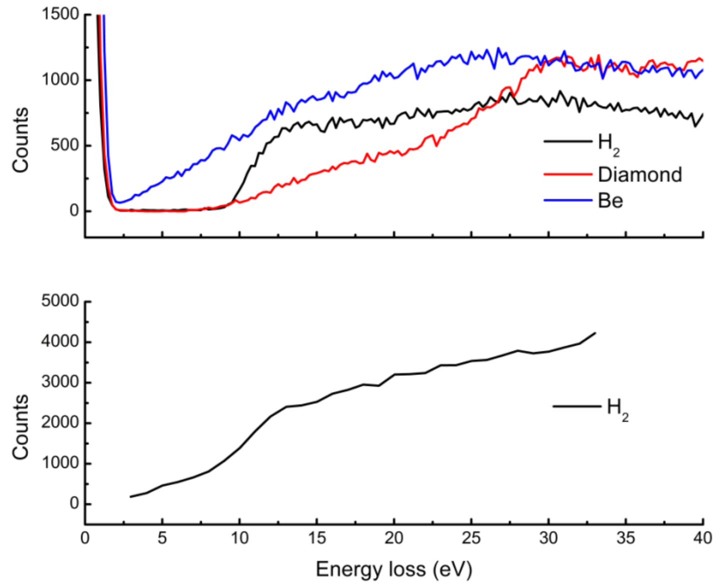

Zr ω-phase at 5.0 GPa

Reducing beam size for high spatial resolution

Monitoring the Band Structure of Hydrogen at HP

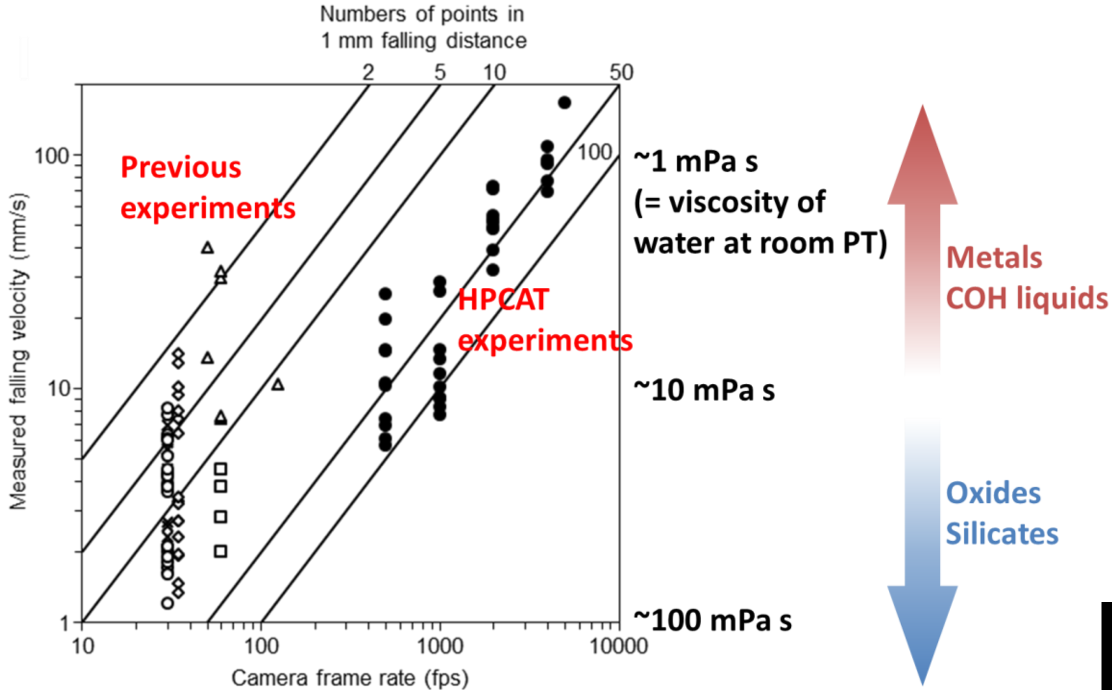
Eliminating background scattering from massive surrounding materials

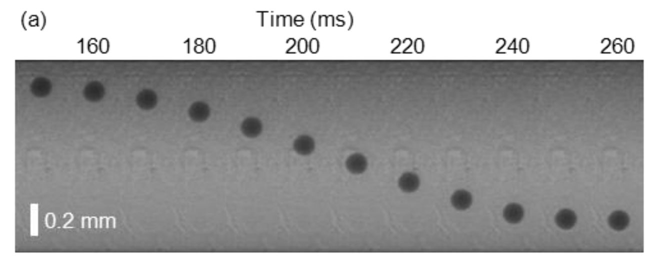




X-ray inelastic scattering of hydrogen. *Top* – using a polycapillary optic significantly suppresses the background from Be gasket and diamond anvil. *Bottom* - hydrogen signal using slit collimation for comparison.

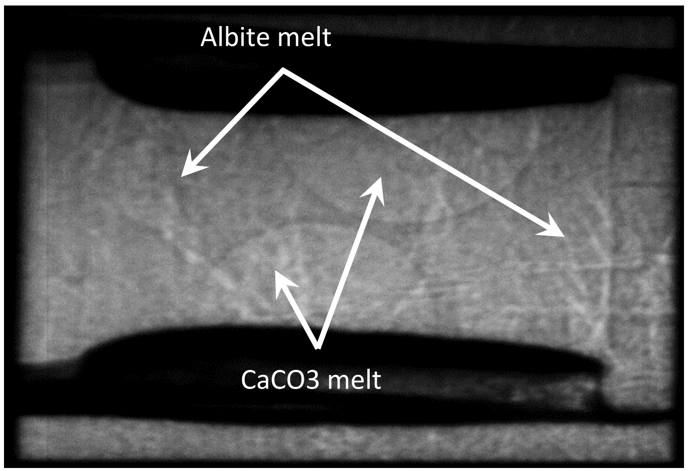
Studies of High Pressure Liquids





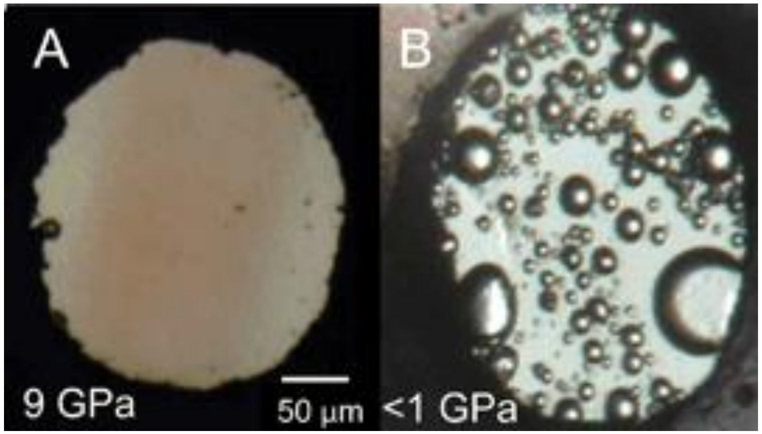
HPCAT fast imaging capability allows for measuring viscosities of low viscos liquids

Phase contrast imaging for studying immiscibility of liquids

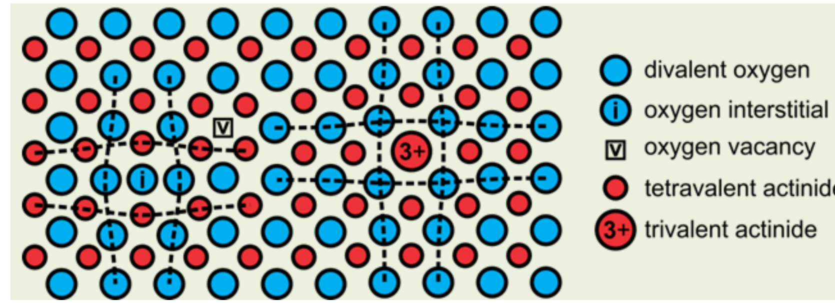


Structure ↔ Density
Viscosity
Sound velocity
Miscibility

HP Photochemistry "Radiation Damage"



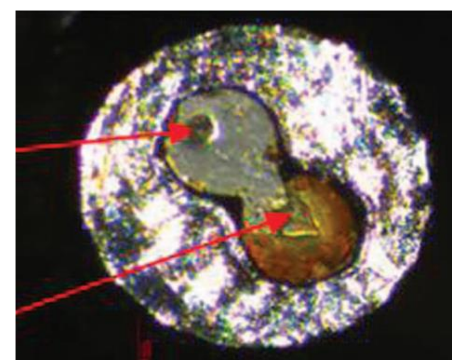
The breakdown of H_2O with radiation at HP, forming a molecular alloy $(H_2)_2O_2$. (Mao et al, 2006)



The **radiation tolerance** of actinide materials is found to depend on the efficiency of redox reaction, thus can be enhanced by altering grain size and cation valence variability. (Tracy et al, 2015)

Decomposition of explosives (a PETN sample) with radiation at HP

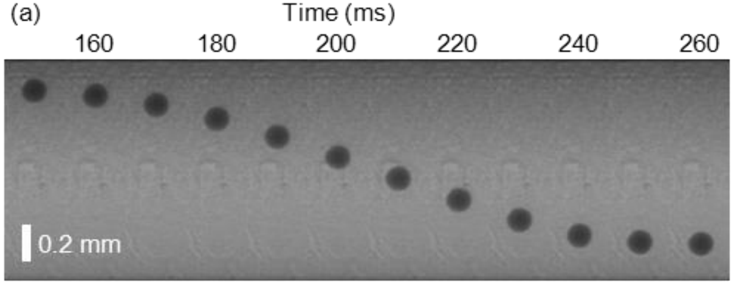
Generating molecular mixtures (H_2, O_2) under HP from NH_3BH_3 and $KClO_4$



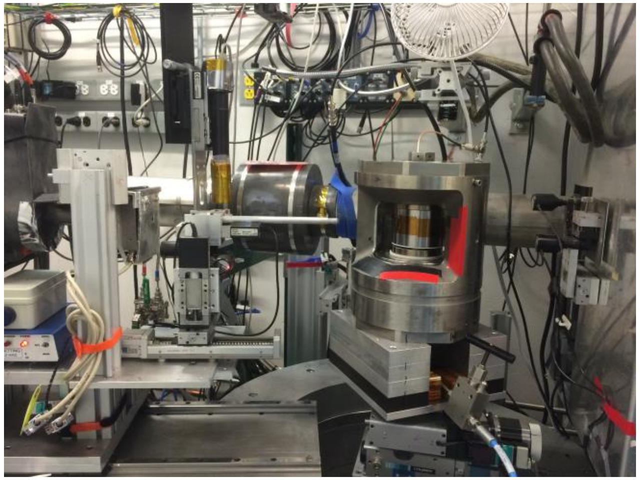
Pravica et al (2014)

16-BM-B: Paris Edinburgh Press and White Laue

- White beam Laue (under commissioning)
  - Fast, no sample rotation
  - Microstructure: morphology, orientation
  - Deformation, dislocation, etc
- EDXD (PE Press)
  - Amorphous/liquid structure
  - Fast radiographic images or PCI for viscosity, immiscibility, density (mono, under development)
  - Acoustic measurement

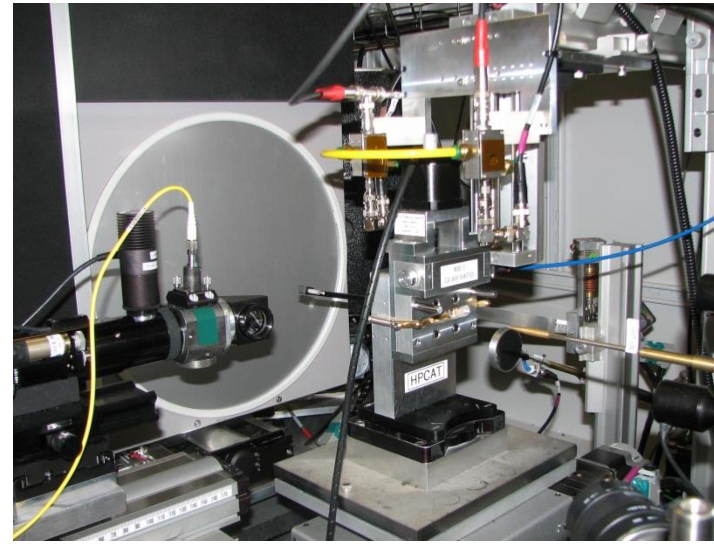


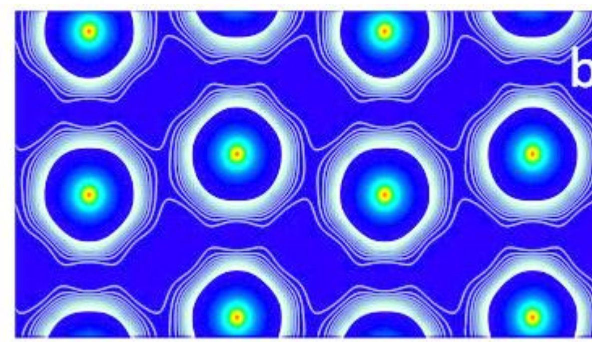
A Pt sphere in carbonate melt



16-BM-D: HP μ-XRD + μ-XANES

- XRD
  - General μ-XRD
    an important addition for off-loading the IDB (flux two orders of magnitude less than that in IDB)
  - Single crystal diffraction
    Mbar, multigrain crystallography
- XANES
  - Inter-changeable between XANES and XRD
  - High energy XANES (>20 keV)

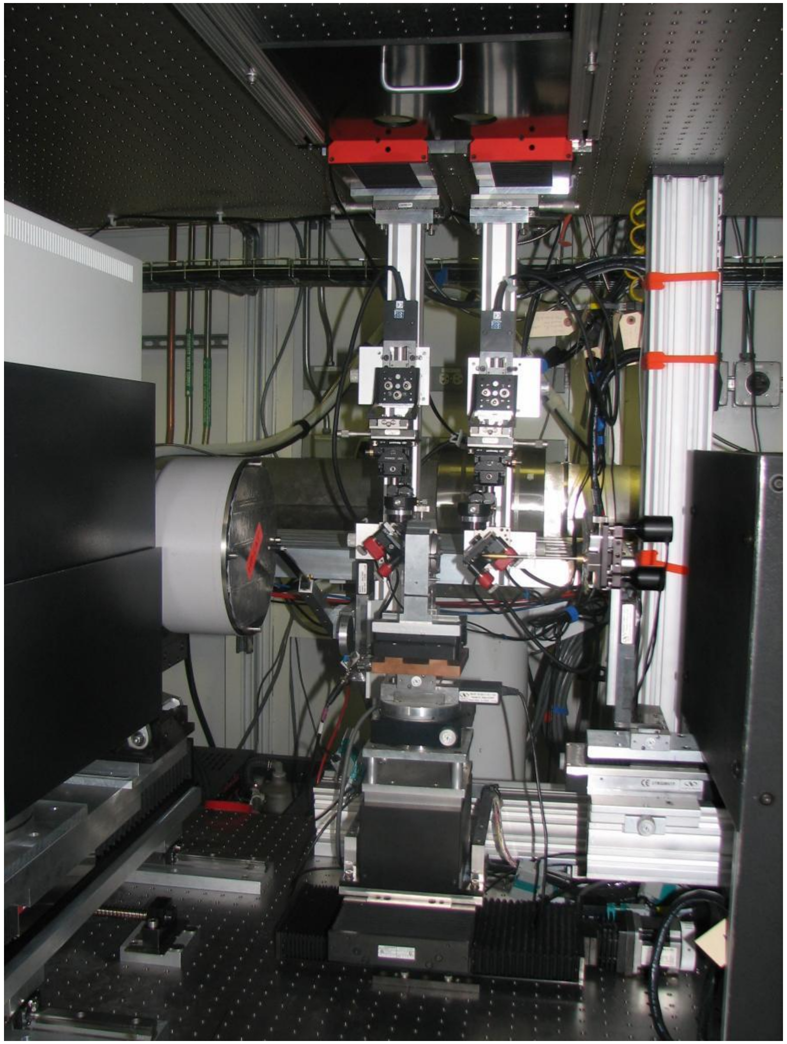




Electron density distribution of β-Ge at 12GPa

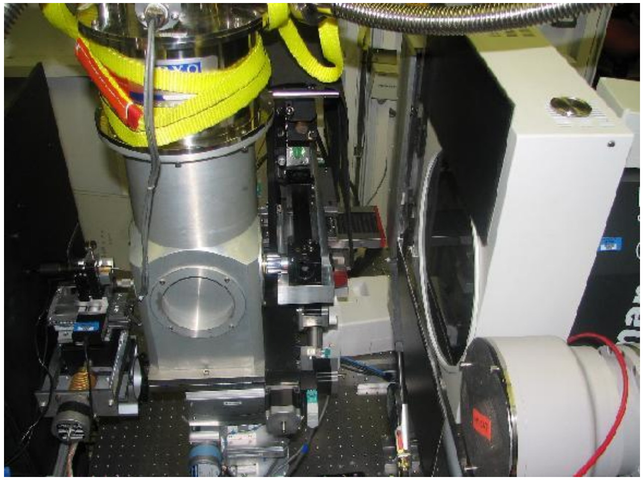
16-ID-B: Laser Heating Table

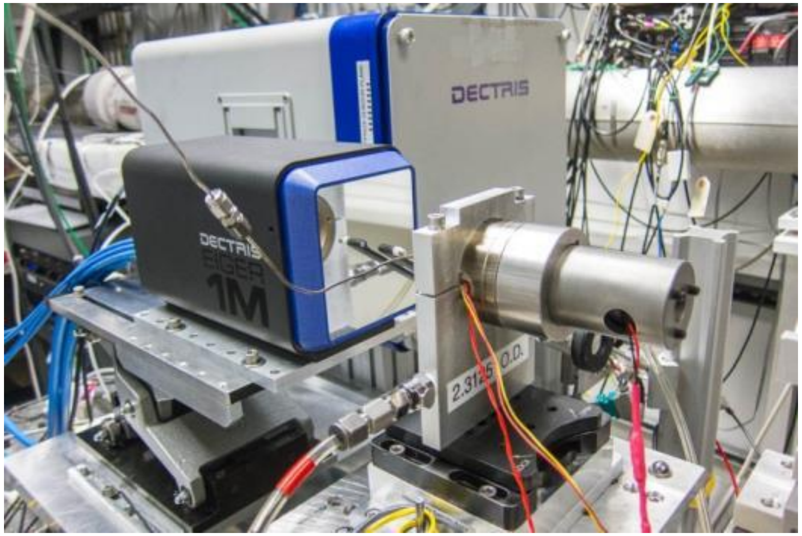
- On-line laser heating for in situ XRD measurements
- Modulated heating/cooling
- Fast T measurement
- HP melting
- Thermal EOS
- Phase relations



16-ID-B: General Purpose Table for μ-XRD

- HP μ-XRD
- Open space for cryostat and other bulky devices
- Time resolved XRD with rapid (de)compression



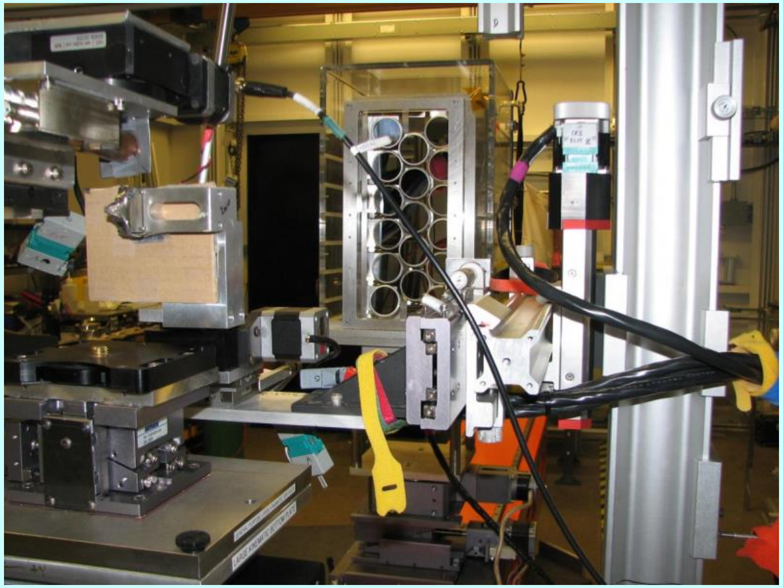


e.g., filling the strain rate gap

16-ID-D:

HP X-ray Scattering and Spectroscopy

- **1-2 eV – IXS**
  - **Chemical bonding**
  - **Electronic excitations**
- **(R)XES**
  - **Spin transition**
  - **Valence band electrons**
- **Nuclear IXS (2meV)**
  - **Phonon DOS**
  - **Mössbauer effect**



X-ray inelastic scattering/x-ray Raman setup using multi-element analyzer

Support Equipment

- On-line
  - Cryostats for XRD, XES, NFS, IXS
  - Ruby/Raman systems/Laser heating
  - P controls (diaphragm, gear box, piezo, doubled sided)
- Off-line
  - Preparation lab, new micro-manipulator
  - Laser drill, (EDM, EDM-Be)
  - Off-line laser heating, Raman, ruby
- Software and Controls
  - Data evaluation on site
  - More macros for specific setups and controls