OUTLINE

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   EDUCATION
   TRAINING
   OUTREACH
   SCIENCE
   TECHNIQUES

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   NNSA OPPORTUNITIES
   NEW PARTNERSHIPS
Studies of extreme environments have opened up a new world of materials.

1. OVERVIEW

- Physics
- Chemistry
- Materials
- Geoscience
- Planetology
- Astrophysics
Components of the Center

MISSION

Develop techniques and training to examine the full complement of high P-T materials science problems essential for Stewardship Science.

Academic Partners

CARNegie INST. (Hemley)
ALABAMA - BIRMINGHAM (Vohra)
CALIF. - BERKELEY (Wenk&Jeanloz)
ILLINOIS (Dlott & Cahill)
CALTECH (Fultz)
YALE (Lee)
UCLA (Kavner)
NORTHWESTERN (Jacobsen)
WASHINGTON-ST. LOUIS (Schilling)
HAWAI’I (Dera)
WASHINGTON STATE (Yoo)
SUNY-UNIV AT BUFFALO (Zurek)
UTAH (Miyagi)

Academic Collaborators

FACILITY USERS

NNSA Laboratory Partners

ALL HIGH P-T GROUPS AT LLNL, LANL, SNL; STEERING/ADVISORY COMMITTEE MEMBERS
CDAC manages and coordinates activities at major facilities for high $P-T$ research

- **Carnegie/Partner facilities:**
  - High $P-T$ technology
  - Spectroscopy labs
  - Diffraction and microanalysis
  - Computational resources
  - CVD diamond growth

- **Technique development/training** at unique facilities at NNSA Labs

- **High $P-T$ synchrotron IR beamline at NSLS**
1. OVERVIEW

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

- 1988 ANL/Chicago Workshop
- GSECARS (Sector 13)
  - High-pressure geoscience
- HPCAT (Sector 16) launched 1998
- Dedicated high-pressure facility
  - Physics, chemistry, materials
  - Advanced techniques
  - Programmatic work (NNSA Labs)
- > 5600 person visits (1/15)
- 843 peer reviewed publications
- Training and education
  - Approx. 60% users are students and post-docs
- Enhanced capabilities

- 2012 Trilab (LLNL, LANL, SNL)
- Upgrade APS-U and HPCAT-U

75%: CDAC/Carnegie, LLNL, UNLV
25%: DOE-BES

DOE NNSA/SC Partnership

- 9 hutches
- 4 independently operating stations
- support laboratories
High $P-T$ IR synchrotron beamline
NSLS-FIS is an important CDAC facility

1. OVERVIEW

Fully upgraded U2A end station with new Raman systems

Hydrothermal DAC for high-P and high-T IR experiments

Standard symmetric DAC for high-P and low-T IR experiments

U2A side station with additional far-IR microscope

Off-line CO$_2$ laser heating system

Complete sample preparation facilities

The facilities will be moved to NSLS-II as one of the NxtGen beamlines
CDAC HIGHLIGHTS 2014:
Education, training and outreach

1. OVERVIEW

• Growth of users at HPCAT
  • (800+ to date)

• CDAC supported 18 PhD students

• 42 PhDs awarded with CDAC support

• 14 Students/Postdocs to NNSA Labs

• 4 undergraduate/high school interns

• Workshop/symposium sponsorship
  • Ferroelectrics 2014 (Carnegie)
  • Neutron and X-Ray Scattering School
  • Workshop on Time-Resolved Techniques

• Presentations at national meetings
  • 2014 APS March – 33 presentations
  • 2014 Fall AGU – 47 presentations
CDAC HIGHLIGHTS 2013-2014:
Education, training and outreach

1. OVERVIEW

- Student-Focused
- Research Highlights
- People News
- Announcements
- Resources
Summer Enrichment at NNSA Laboratories

- Eloísa Zepeda-Alarcón
  UC-Berkeley
  Advisor: Hans-Rudolf Wenk
  LANL Sponsors: Ricardo Lebensohn, Carlos Tome
  *ViscoPlastic Modeling of Two-Phase Materials: Periclase + Silicate Perovskite Aggregates*

- Andrew Shamp
  University at Buffalo
  Advisor: Eva Zurek
  LLNL Sponsors: Sebastien Hamel, Tadashi Ogitsu
  *Theoretical Predictions of the EOS of Boron Carbide Under Extreme Conditions*

- John Lazarz
  Northwestern University
  Advisor: Steven Jacobsen
  LANL Sponsors: Cindy Bolme, Kyle Ramos, Dan Hooks
  *Determination of the Elastic Tensor of Acetaminophen, an RDX Analogue*
CDAC investigates a broad range of fundamental problems in high P-T science

- STRUCTURES AND PHASE RELATIONS
- EQUATIONS OF STATE
- PHONONS AND ELASTICITY
- RHEOLOGY AND STRENGTH
- TRANSPORT PROPERTIES
- EXTREME CONDITIONS CHEMISTRY

2. SCIENCE

18 POSTERS AT THIS MEETING

2014-: 136 Publications (including in press)

Since 2003: 1465+ Publications (200 Student Publications)
(128 Student First Author Papers)
Boron Carbide Under Pressure

- XtalOPt Evolutionary Algorithm
- CBC\textsubscript{p} Dynamically Stable
- Thermodynamically Unstable

Unstable with respect to pure C and B for P > 50 GPa.

Andrew Shamp
University at Buffalo
Small Molecules at Extreme Conditions

- Paracetamol, $\text{C}_8\text{H}_9\text{NO}_2$
- Raman + Angle-dispersive XRD (HPCAT)
- Four phases below 15 GPa


- N2 in $I\bar{2}3$ (cg) and $Pba2$ (LP)
- Laser heating + Raman spectroscopy
- 3D to 2D (symmetry lowering) at HP


2. SCIENCE
Dehydration Melting in Earth’s Mantle

- \((\text{Mg,Fe})_2\text{SiO}_4\) at 30 GPa, 1600 °C, 1% H\(_2\)O
- Laser heating, synchrotron IR spectroscopy (NSLS-U2A)
- Transformation to \((\text{Mg,Fe})\text{SiO}_3 + (\text{Mg,Fe})\text{O} \): Intergranular melt
- Dehydration melting consistent with seismic velocity decrease below 660 km
- Suggests hydration of a large part of the mantle, with H\(_2\)O trapped in transition zone

OH in quenched glass
Hydrous Ringwoodite
Intergranular melt

Steve Jacobsen, Northwestern

High Pressure Studies on Ferroelectrics

- Polycrystalline Pb(Fe_{0.5}Nb_{0.5})O_3
- Raman scattering at high P and T
- Refined phase diagram

- Single crystal Pb(Mg_{0.33}Nb_{0.67})O_{3-x} – PbTiO_3
- Raman scattering at high P and T
- XRD at HPCAT 16-BM-D (High Energy)
- Monoclinic-Rhombohedral at 3 GPa
  Octahedral tilting
- Soft optical phonon at 9 GPa
- Drastic changes at 27 GPa
  Orthorhombic or monoclinic?

Brandon Wilfong
Washington College

Muhtar Ahart, Carnegie

Magnon-Phonon Interactions in bcc-Fe

- Phonon dispersions from NRIXS
- Large non-harmonic phonon softening caused by magnon-phonon interactions
- Magnon-phonon vibrational entropy stabilizes bcc-Fe above $T_C$

Thermal Transport at High Pressure

• Extension of time-domain thermoreflectance measurements to high pressures.
• Tests of theoretical models of thermal energy transport in materials and across interfaces.
• Pressure is used to systematically vary phonon and electron densities of states, and interface bonding.
• Measured thermal conductance is well above the phonon radiation limit.
• Unexpectedly high thermal conductance consistent with two high frequency diamond phonons interact with a low-frequency metal phonons.

G. Hohensee et al., *Nature Communications*, in press.
Defects and the Elastic Properties of Materials

- At Northwestern, GHz interferometry is used to determine the elastic properties of materials.
- Acetaminophen (RDX analog), vitreous silica, B-doped diamond

- Complete optical indicatrix
- Completing elastic tensor (Brillouin + ultrasonics)
- Applied to modeling elastic properties of composites in energetic materials

- Poisson’s ratio from elastic tensor as measured by GHz interferometry
- Boron reduces elastic Moduli and elastic anisotropy of diamond


- Anomalous compressibility in v-SiO$_2$
- HAD:LDA ratio set at $T_f$, fixed below permanent densification
- Compression behavior consistent with floppy modes in a mixture of LDA and HDA domains
Deformation of Polyphase Samples at High Pressure

- Studies of deformation in MgSiO$_3$ – MgO at high pressure allow interpretation of seismic anisotropy and infer flow patterns in the Earth’s mantle.

- Collaboration with LANL on simulations with VPSC and VPFFT codes allows modeling of deformation microstructures including intersite interactions.

Laser heating

Diffraction cone

X-rays

rDAC

Diffraction image

Microstructure

Blue tones = Perovskite
Orange tones = MgO

Stress Field

Normalized units

Strain-Rate Field

Normalized units

Jesse Smith, HPCAT and Eloísa Zepeda-Alarcón, Berkeley
Deformation of Polyphase Samples at High Pressure

- B1 NaCl exhibits different textures as a pure phase under deformation as compared to 75% NaCl-25% MgO.

Mike Jugle and Max Gianetta, Utah

Laser heating in radial geometry
ALS 12.2.2
Magnetic Ordering in Rare Earth Metals: Dy

- $T_o$ rises rapidly in Dy metal at $P > 70$ GPa reaching 400 K at 157 GPa.

- Surpasses $T_o = 292$ K at 0 GPa in Gd metal $T_o$ rises slowly with pressure.

- Suggests that high ordering temperature in Dy metal is a highly correlated electron effect—pressure destabilizes the magnetic state.

Superconductivity in the CaFe$_2$As$_2$ System

- Sr substitution into CaFe$_2$As$_2$ decouples volume collapse from the Fe moment.
- Superconductivity develops out of the paramagnetic normal state.
- Pr substitution into CaFe$_2$As$_2$ yields $T_c = 51$ K at 1.9 GPa.
- Resistance measurements suggest two superconducting phases at 0.5 GPa.

Shocked Polymers and Fluorescent Probes

- Laser-launched cold, intact flyer plates
- Embedded dye + PMMA
- Time-resolved monitoring of dye emission spectrum: variation of red shift with time.
- At ~ 10 ns, change from elastic to viscous compression
- Next: materials that absorb shocks

Aromaticity and Closed-Shell Effects in Hydrogen

- H$_2$ in molecular clusters, 2D and 3D crystals investigated using quantum chemical and solid state physics approaches.
- Stability of dense hydrogen structures at > 200 Gpa arises from the intrinsic stability of 6-membered rings with properties similar to carbon graphene.
- Atomic and electronic properties are controlled by closed-shell effects.
- Closed shell effects are critical to understanding how hydrogen becomes metallic.

Nanodiamond Research at Carnegie

- MPCVD process results in nanodiamond formation at atmospheric pressure (3 mm quartz tube in CVD chamber cavity).
- SEM, Raman measurements show a pure diamond phase.
- Adamantane at HP-HT conditions results in nanodiamond formation through dehydrogenation.
- Abrupt change at ~600 °C indicates loss of hydrogen.
- HRTEM shows developing periodicity.
Flash Heating and the Melting of Refractory Transition Metals

- Issues to overcome: chemical reactions, sample instability, thermal runaway
- 20 ms rectangular heating pulse, increase laser power to give 100 K increase in temperature for each new sample area—8-10 runs per sample loading
- SEM, EDS, FIB analysis of heated sample spots
- Re to 48 GPa, Mo to 45 GPa, melting curves reproduced well compared to other methods
- Ta to 85 GPa and 4320 K—discrepancies with previous methods are significant

Amol Karandikar
Carnegie

Temperature and Emissivity Mapping: Melting of SiC

- Carbon-rich exoplanets: Interior Details?
- Flash heating, temperature, emissivity mapping
- SiC appears to decompose upon melting
- Suggests a mantle with alternating Si and C layers

Kierstin Daviau, Yale

β-SiC: 22 GPa

Temperature

Emissivity
New Developments at HPCAT BM-B

- Simultaneous diffraction, radiography, resistance, and thermal measurements in the Paris-Edinburgh Cell

- Feasibility demonstrated to 6 GPa and 1000 °C

- White-beam Laue diffraction applied to spatially resolved strain mapping during α-β transition in Si

- Opens possibilities for studies of kinetics, mechanisms of phase transitions

- 2-3 orders of magnitude faster than with a monochromatic beam
Recent progress at CDAC presents new opportunities

1. **Education and Training**
   - Expanded student program with a large group of partners
   - Continued placement of personnel in NNSA labs
   - Summer schools/workshops and other outreach
   - Student visits to 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 for the NNSA labs (beyond SSAA)*

3. **Technique Development**
   - Continued high $P-T$ device developments
   - New x-ray techniques (imaging, time-resolved, static/dynamic)
   - Need to take advantage of APS upgrade
Broader Science Challenges

1. Structure and bonding at high compression
2. New physics in ‘cold’ dense matter
3. Fundamental thermodynamics
4. Time-dependent transformations
5. Strength, plasticity, rheology
6. Optimized new materials
7. Synthetic chemistry frontier
8. Radiation-induced high-pressure chemistry
9. Earth and planetary science, astrophysics
10. Life in extreme environments

OUTLOOK
Technical Challenges

1. Reaching 1 TPa and beyond
2. Multiprobe ‘intelligent’ devices
3. Stress-strain and P-T calibration
4. Advancing x-ray methods
5. Real time x-ray imaging with nm resolution
6. Filling the strain-rate gap: static to shock
7. Transport/constituitive properties
8. Liquids and amorphous materials
9. Thermochemical & magnetic measurements >100 GPa
10. Other techniques, including neutron scattering
1. Science Opportunities
   There are numerous opportunities to address major scientific questions that both span the sciences and cut across static and dynamic compression research.

2. Compression Science and the APS
   Research at APS has led, and continues to lead, many of the advances in the field, and is poised to lead an integrated and coordinated program in materials in extreme environments for basic, applied, and programmatic science.

3. Role of CDAC
   CDAC will serve as a bridge to help grow the academic and broader user communities.
Opportunity for a coordinated effort for extreme conditions science at APS

Other facilities (LCLS, NSLS-II, Omega, NIF, MaRIE, ...)

Opportunities for coordination at APS