APS Workshop 10: Advanced X-ray Capabilities for High-pressure Research

Thursday, May 9, Afternoon

1:30 – 1:35 Workshop Organizers
Introduction

Session Chair: Nenad Velisavljevic

1:35 – 2:05 Guoyin Shen (Argonne National Laboratory)
High-pressure Research at APS-U: Challenges and Opportunities

2:05 – 2:35 Maddury Somayazulu (Argonne National Laboratory)
HPCAT-Upgrade: Upgrading and Adding Techniques

2:35 – 2:50 Break

Session Chair: Stella Chariton

2:50 – 3:20 Vitali Prakapenka (University of Chicago)
Cutting-edge Upgrade of Diamond Anvil Cell Program at GSECARS

3:20 – 3:50 Paulo Rigg (Washington State University)
The Dynamic Compression Sector: Current Capabilities, Representative Achievements, and Future Enhancements

3:50 – 4:20 Jiyong Zhao (Argonne National Laboratory)
New Opportunities of IXS and NRS for High-pressure Studies at APS

4:20 – 4:40 Open Discussion

4:40 Adjourn

Friday, May 10, Morning

8:55 – 9:00 Workshop Organizers
Introduction

Session Chair: Suresh Narayanan

9:00 – 9:30 Wonsuk Cha (Argonne National Laboratory)
New Opportunities with High-energy Coherent X-rays at Atomic and 3DMN for High-pressure Research

9:30 – 10:00 Felix Lehmkühler (Deutsches Elektronen-Synchrotron DESY)
Opportunities for High-pressure XPCS

10:00 – 10:30 Hemant Sharma (Argonne National Laboratory)
High-energy Diffraction Microscopy for High-pressure Research

10:30 – 10:45 Break

Session Chair: Changyong Park

10:45 – 11:15 Daniel Haskel (Argonne National Laboratory)
New Tools for Probing Magnetism at High Pressures at the POLAR Beamline of APS-U

11:15 – 11:45 Naoki Ishimatsu (Hiroshima University)
Applications of Nanopolycrystalline Diamond Anvils to X-ray Absorption Spectroscopy under High Pressure

11:45 – 12:15 Xiaoyi Zhang (Argonne National Laboratory)
Investigation of Excited State Dynamics via TR-XAS under High-pressure Conditions

12:15 – 12:30 Open Discussion and Conclusion

12:30 Adjourn

High-pressure Research at APS-U: Challenges and Opportunities

Guoyin Shen[1]

[1]HPCAT, X-ray Science Division, Argonne National Laboratory, Lemont, IL 60439

Since the beginning of APS, high-pressure (HP) synchrotron research has been an important component in APS science, addressing fundamental questions of matter under extreme conditions. With the orders of magnitude increase in brilliance and coherent flux of the APS-U at high photon energies, together with new developments in advanced detectors, beamline instrumentations, computation capabilities, and novel HP devices tailored for large facilities such as APS-U, the scope of HP research at APS-U will be significantly expanded with a new generation of HP studies not feasible previously. In this talk, I will discuss some characteristics of HP experiments in the previous APS era, and highlight new opportunities of HP research at APS-U. A few examples include how HP experiments at APS-U will be shifted to new approaches from collecting 'averaged' information to obtaining 'individual grain details and their distributions,' from 'observing novel phases' to 'understanding pathways and processes,' from pressure as 'an extreme condition' to pressure (or stress, shear, and their rates) as 'engineering drivers,' from dealing with 'simple systems' to addressing 'complex, heterogeneous materials,' and from measuring 'first-order' properties to obtaining their derivatives for transport properties and thermal expansion data, etc. The challenges associated with the new generation HP synchrotron research will also be discussed.

HPCAT-Upgrade: Upgrading and Adding Techniques

Maddury Somayazulu[1], Nenad Velisavjlevic[2], Eric Rod[1], Kevin D'Amico[3], and the beamline scientists of HPCAT[1]

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The APS-Upgrade is bringing two orders increase in the brightness and concomitant increase in spatial coherence. The decrease in emittance results in smaller focus with better beam profiles due to better focusing elements. All these translate to being able to achieve TPa pressures, at temperatures as high as 5000 K, and study the material response at mSec time scales. Such measurements on materials at extremes of pressure, temperature, and strain rates using several experimental probes is positioned to expand our horizons in materials physics and chemistry. The beamline scientists at HPCAT have embraced this opportunity to upgrade their existing experimental capabilities, deploy new experimental techniques and define the scope of extreme conditions science for the coming decades. They have recently shared their plans with the wider high-pressure community and brainstormed over the defining experiments that will benchmark the upgraded capabilities and the scientific questions that it can answer. This talk will cover the details of the upgrade plans and bridge it to the scientific narrative we hope to construct.

HPCAT is fully funded by the NNSA office of experimental sciences.

Cutting-edge Upgrade of Diamond Anvil Cell Program at GSECARS

Vitali B. Prakapenka[1], Stella Chariton[1], Young Jay Ryu[1], and Zhang Dongzhou[1]

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The Advanced Photon Source is undergoing a complex upgrade replacing its original electron storage ring with a new multi-bend achromat lattice to provide extremely enhanced coherent flux and increased hard x-ray brightness by a hundred times. To take full advantage of these improved capabilities, we have started the process of comprehensive technical improvements and developments of multiprobe techniques across a suite of beamline stations at GSECARS.

The laser-heated diamond anvil cell technique combined with high energy tightly focused x-ray beam and fast detectors for spectroscopic measurements is the workhorse method for exploration of deep Earth mineral physics and chemistry. To provide new constraints on models for planetary evolution and origin, essential properties (melting, structure, phase relation, chemical reactions and kinetics, transport, elastic, electronic and optical properties) of a wide range of minerals must be studied *in situ* at extreme conditions of pressure and temperature. However, existing data sets are often inconsistent or too poorly constrained to provide unique answers. This underscores the need for unique beamline capabilities: high-energy high-flux sub-micron x-ray beam to probe ultra-small samples (micron-sized) in megabar pressure range, high resolution large area fast x-ray and optical detectors for time-domain experiments, sample emissivity and absorption measurements to improve radiative temperature metrology, etc. To accomplish that, we will construct a vibration-free granite-table system at 13-ID-D station, install a pre-shaped sub-micron x-ray focusing system (300 nm) coupled with high precision sample positioners including an air-bearing rotary stage, replace our optical spectrometer, and acquire a new Eiger2 CdTe 9M X-ray detector. We plan to upgrade the x-ray and laser optics (Raman, Brillouin, fluorescence, absorption) at 13-ID-D, 13-BM-C, and 13-BM-D stations to accommodate higher x-ray energies in a tighter focused beam, which grants a significant boost in the reciprocal space explored at high temperatures with increased spatial resolution.

Recent results and details of future developments of the cutting-edge synchrotron and optical techniques for comprehensive characterization of materials *in situ* at extreme conditions will be discussed.

The Dynamic Compression Sector: Current Capabilities, Representative Achievements, and Future Enhancements

Yogendra. M. Gupta[1] and Paulo. A. Rigg[1]

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The Dynamic Compression Sector – a first-of-its-kind user facility – was designed and built to address an important and long-standing need in dynamic compression science: real-time, *in-situ* measurements to gain microscopic/atomic scale insight into condensed matter response during dynamic compression. A diverse suite of experimental platforms capable of producing high stress impulsive loading (peak stresses to > 400 GPa; time durations ~5 ns to 1 μs) were linked to the brightest, high energy (>20 keV) x-ray source in the U.S. – the Advanced Photon Source (APS). A wide range of x-ray and optical detection capabilities routinely permit real-time measurements spanning multiple length scales. To date, the DCS capabilities have been utilized successfully by a diverse user community to address long-standing scientific questions – related to structural transformations, deformation mechanisms, and chemical reactions at extreme thermodynamic conditions – and to make discovery class advances. In conjunction with the APS Upgrade, the DCS will undertake complementary enhancements to ensure that condensed matter scientific frontiers continue to be explored and advanced after the APS-U completion. For example, users will be able to routinely obtain x-ray measurements to 60 plus keV – a unique capability for dynamic compression science. This talk will provide a brief overview of current capabilities, highlight representative scientific achievements, and discuss new opportunities that will be available to users due to the APS and DCS enhancements.

The following colleagues should be viewed as co-authors for this talk: K. D'Amico, P. Das, Y. Li, P. Renganathan, D. Rickerson, A. Schuman, J. Sethian, N. Sinclair, S. Turneaure, Y. Toyoda, X. Wang, and R. Zill.

Work supported by DOE/NNSA. Partnerships with APS, NNSA Laboratories (Los Alamos, Livermore, Sandia), University of Rochester (LLE), and ARL played an important role in the DCS development.

New Opportunities of IXS and NRS for High-pressure Studies at APS

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The high-pressure study has been an important part of user programs at APS, including the momentum-resolved high resolution inelastic x-ray scattering (HERIX) at Sector 30, the resonant inelastic x-ray scattering (RIXS) at Sector 27, and the nuclear resonant scattering (NRS) at Sector 3 and Sector 30. Rich information acquired from these techniques such as dynamics of materials, charge, spin, orbital, nuclear and lattice excitations, phonons, phonon-electron couplings, sound velocities, hyperfine interactions, magnetic phase transitions, and melting under high pressure with either high temperature or low temperature have great impact on scientific research.

With the APS-U, we expect these programs to expand in performance and scope, especially for applications on high-pressure research using smaller beam size introduced by the APS-Upgrade. Detailed upgrades of these beamlines will be described, and future oportunities of the user programs will be given in this talk.

New Opportunities with High-energy Coherent X-rays at Atomic and 3DMN for High-pressure Research

Wonsuk Cha[1]

[1]X-ray Science Division, Advanced Photon Source, Argonne National Laboratory, Lemont, IL 60439

Taking advantage of the high coherence and brightness of the third-generation synchrotron source, Sector 34 of the Advanced Photon Source (APS) provided detailed local structural information and its evolution of crystalline materials in 3D. The usage of modest x-ray optics with relatively long working distance enabled *in-situ* experiments employing Bragg coherent x-ray diffraction imaging (BCDI) and x-ray Laue diffraction microscopy to address scientific questions on the materials under high-pressure.

The high energy coherent flux from the coming upgraded APS (APS-U) will enable a revolution in both BCDI and x-ray Laue diffraction microscopy. Therefore, new Atomic and 3D Micro and Nano Diffraction (3DMN) beamlines at Sector 34, feature beamlines of the APS-U, will take full advantage of the APS-U. We anticipate that 3D imaging at Atomic and 3DMN approaches ultimate spatial resolution in some experimental cases and achieves much higher temporal resolution than what was available at the APS in others. In this presentation, I will introduce scientific contributions from Sector 34 of the APS. I will also discuss the future of BCDI and x-ray Laue diffraction microscopy as well as development at Atomic and 3DMN of the APS-U.

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Opportunities for High-pressure XPCS

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Thanks to its unique signal-to-noise ratio, X-ray Photon Correlation Spectroscopy (XPCS) is the technique that (likely) benefits most from the advent of the newest generation of storage rings [1]. In XPCS, experiments' dynamics are measured in real time via correlation functions from coherent x-ray scattering patterns. The XPCS performance, given by the shortest accessible correlation time, scales with the square of the coherent flux, enabling in principle time resolutions down to nanoseconds for some experiments at the new facilities. Furthermore, the increased coherence at higher photon energies extends the capabilities of XPCS to many different applications. One example is studies at high pressures using dedicated sample environments. In my presentation I will give an introduction into the current and future capabilities of XPCS at modern x-ray light sources [1], ranging from the ultrafast to microsecond time scale at FEL sources [2] to slow dynamics in glasses at storage rings. In addition, I will discuss new concepts of structure-dynamics correlation in soft matter systems [3,4]. The main part of my talk will focus on the results of some recent studies on dynamics of (i) amorphous ices at elevated pressures and (ii) in hyaluronic acid networks under high pressure.

[1] F. Lehmkühler, W. Roseker, and G. Grübel. Appl. Sci. 11, 6179 (2021).
[2] F. Lehmkühler et al. PNAS 117, 24110 (2020).
[3] N.N. Striker et al. J. Phys. Chem. Lett. 14, 4719 (2023).
[4] F. Lehmkühler et al. Sci. Adv. 6, eabc5916 (2020).

High-energy Diffraction Microscopy for High-pressure Research

Hemant Sharma[1]

[1]Computational Sciences and Artificial Intelligence Group, X-ray Science Division, Argonne National Laboratory, Lemont, IL 60439

My talk will describe a suite of techniques available under the umbrella of high-energy diffraction microscopy (HEDM) as applicable to high-pressure research. I will show how different HEDM modalities can be used to study different features of materials at different length scales and pressures. Further, I will describe a recently developed point-focus HEDM technique for studying complex geological samples. The advancements in data processing using high-performance computing resources to enable scientific discovery will be shown.

New Tools for Probing Magnetism at High Pressures at the POLAR Beamline of APS-U

Daniel Haskel[1]

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The POLAR beamline, one of eight feature beamlines of the APS-U project, leverages the enhanced spectral brightness of APS-U and state of the art x-ray optics and instrumentation to advance hard x-ray probes of electronic correlations, chiefly magnetism, using polarization-dependent spectroscopy, scattering, and imaging techniques. POLAR will be especially suited for studies of emergent electronic correlations at extreme high pressures, high magnetic fields, and low temperatures, including imaging electronic inhomogeneity with sub-micron resolution. This talk will highlight the new capabilities of beamline POLAR as well as challenges and ongoing R&D activities aimed at addressing them.

The POLAR beamline project has been in the making for over 7 years, and involves a large number of contributors from engineering, scientific, and support groups at the Advanced Photon Source. Work at Argonne is supported by the US Department of Energy, Office of Science, Office of Basic Energy Sciences, under Contract No. DE-AC-02-06CH11357.

Applications of Nanopolycrystalline Diamond Anvils to X-ray Absorption Spectroscopy under High Pressure

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X-ray absorption spectroscopy (XAS) and x-ray magnetic circular dichroism (XMCD) have long been useful techniques for high-pressure research for both crystalline and non-crystalline materials. Recently, nanopolycrystalline diamond (NPD) anvils have been widely applied in high-pressure research using XAS and XMCD because the nanometer-sized polycrystallization in NPD anvils allows us to obtain glitch-free x-ray absorption spectra. Consequently, quality of the spectrum under high pressure is nearly comparable to that measured at ambient pressure.

My talk briefly reviews technical developments of XAS and XMCD measurements under high pressure, which have been achieved at SPring-8 after the first use of NPD [1]. Furthermore, recent applications of double stage DACs to the XAS measurements of *5d* transition metals are presented as a challenge for the measurements at multi-megabar conditions (> 300 GPa) [2].

[1] N. Ishimatsu *et al*., J. Synchrotron Rad. 19, 768 (2012).
[2] K. Kuramochi, N. Ishimatsu *et al*., High Pressure Res., 40, 119 (2020).

Investigation of Excited State Dynamics via TR-XAS under High-pressure Conditions

Xiaoyi Zhang[1] and Cunming Liu[1]

[1]X-ray Science Division, Argonne National Laboratory, Lemont, IL 60439

Earth-abundant-metal-based photosensitizers as substitutes for their noble metal analogues have attracted increasing attention in recent years owing to their environmental and economic benefits. Despite their promise, the rapid deactivation of their charge transfer (CT) excited state hinders their applications in solar cells and photocatalysis. Here, we demonstrate a novel approach to stabilize the metal to ligand CT (MLCT) state of iron polypyridyl complexes by tuning the potential surface of its main deactivation channel—the metal centered d-d excited states—through the application of high pressure. Our optical transient absorption (OTA) spectroscopy measurements hinted that the MLCT state lifetime increased by 5 orders of magnitude from sub-50 femtoseconds at ambient condition, to 9 nanoseconds under high pressure. However, uncertainties persist regarding the exact nature of the charge transfer excited state and the geometric structure of the photoexcited species under high pressure conditions. To address these questions, we propose the utilization of Time-Resolved X-ray Absorption Spectroscopy (TR-XAS), a powerful tool elucidate the electronic and structural dynamics of photoexcited species. We will discuss the insights gained from our initial efforts to investigate photoexcited species under high pressure, as well as the promising prospects of utilizing this technique with the upgraded source at the Advanced Photon Source (APS). This presentation aims to showcase our findings and to highlight the potential of TR-XAS in advancing our understanding of photochemical processes under extreme conditions.

This research used resources of the Advanced Photon Source, a U.S. Department of Energy (DOE) Office of Science user facility at Argonne National Laboratory and is based on research supported by the U.S. DOE Office of Science-Basic Energy Sciences, under Contract No. DE-AC02-06CH11357.