X-ray DAC Program at NSLS-II

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Integration of synchrotron x-rays and diamond-anvil cell (DAC) technology has fostered a number of scientific breakthroughs in mineral physics such as the discovery of post-perovskite, the influence of the iron spin-state transition, redox on mantle minerals and, most recently, the possible presence of FeO_2 in the lower mantle. Increasing demands of such state-of-the-art facilities from the Earth Science community are currently far beyond their availabilities. This project aims to promote scientific research by expanding and advancing a community facility to fulfill such demands.

Introduction

NSLS-II began operations in October 2014. It is currently rapidly ramping up its science and user programs, while continuing the development of new beamlines and associated scientific capabilities. The high beam brightness enables the study of material properties and functions with unprecedented spatial and energy resolution and with the ultra-high sensitivity required to probe materials at extreme conditions. A diamond anvil cell (DAC) is the most popular and productive tool for the study of material properties under extreme conditions, especially relevant to the Earth's and other planets' interiors. The COMPRES DAC program at the original NSLS had been a workhorse for synchrotron-based experimental high-pressure research in the COMPRES community, and was the most productive COMPRES facility based on the data in the COMPRES 2017-2022 renewal proposal to NSF. While x-ray diffraction (XRD) is still the primary method for in-situ high-pressure x-ray studies, other emerging techniques such as inelastic scattering, tomography and coherent scattering have opened new windows to study materials under extreme condition. Here we propose to establish a DAC program at NSLS-II not only to enable a permanent DAC platform for XRD and PDF routine experiments at the XPD beamline, but also to facilitate integration of the DAC with inelastic scattering at the IXS beamline, nanoprobe imaging at the HXN beamline, coherent scattering at the CHX beamline, x-ray emission at the SRX beamline, and full field x-ray imaging at the FXI beamline. While the x-rays from NSLS-II are only possibly brighter in the energy range below 23keV over other synchrotron sources (e.g., APS), key features of the project include: *1)one project enables multiple (above) techniques; 2)emphasis on complementary techniques (i.e. multi-grain XRD and PDF) in the high energy range; 3)emphasis on more challenging techniques (i.e. IXS, nanoprobe and coherent scattering) in the lower (but still acceptable to DAC) energy range*.

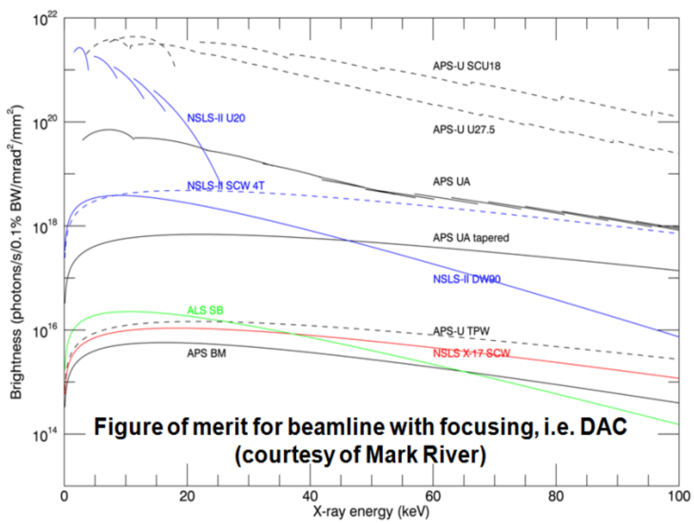

Figure of merit for beamline with focusing, i.e. DAC (courtesy of Mark River)

Community Input and Planning for this Proposal

A collection of community input regarding this proposal began at the 2017 COMPRES Annual Meeting. A workshop for the proposal planning was held on October 28-29, 2017. Thirty five people attended the workshop, with introductions to COMPRES by Carl Agee and to worldwide as well as COMPRES DAC projects by Mark Rivers. Science highlights were given by potential NSLS-II users: Dave Mao (GL), Kanani Lee (Yale), Jie Li (U Mich), Alex Goncharov (GL), Jin Zhang (UNM), Sean Shieh (UWO), June Wicks (JHU), Jing Yang (GL), John Tse (U SASK), and Xinguo Hong (HPSTAR). The Deputy Director for Science of NSLS-II, Qun Shen, and Lead Scientists of the XPD, HEX, IXS, HXN and CHX beamlines (Eric Dooryhee, Zhong Zhong, Yong Cai, Yong Chu, Andrei Fluerasu) also addressed the attendees with their enthusiasm about DAC applications at NSLS-II and at each of the individual beamlines. HPSynC manager, Wenge Yang, COMPRES NSLS-II IR project PI, Zhenxian Liu, and multi-anvil project PI, Don Weidner, presented the importance of synergy and infrastructure at a synchrotron facility for high pressure research. Additional input from individuals who were not able to attend the workshop [Andy Campbell (Chicago), Rebecca Fischer (Harvard), Jennifer Jackson (Caltech), Wendy Mao (Stanford), Wendy Panero (Ohio State), Sang-Heon Dan Shim (Arizona State), Michael Walter (Bristol, soon to be Geophysical Lab), Heather Watson (Union College) and Quentin Williams (UC Santa Cruz)] was solicited by telephone or email. The following project description reflects the collective ideas and addresses some of the concerns.

Science Motivations

Multi-grain XRD: Our knowledge about Earth's lower mantle mineralogy continues to advance as our ability to recognize subtle changes of phases in a complex system under lower mantle pressure and temperature conditions. While the multi-grain XRD technique has not yet been widely adopted by the majority of the COMPRES community, application of this technique has demonstrated its amazing power in identifying new phases and/or intermediate phases during phase transition (*e.g.*, Hu *et al.*, *Nature*, 2016). This project will offer an experimental routine for multi-grain XRD at the DAC platform of XPD beamline including data analysis software (FABLE). Popularizing such a state-of-the-art technique among the COMPRES community will enable more discoveries of mantle phases in Earth's deep mantle, as well as better insight into well known ones (*e.g.*, Zhang *et al.*, *PNAS*, 2013). The setup will also allow EOS and phase diagram determination from typical XRD experiments (*e.g.*, Gu et al., *Nature Geosci*., 2016).

Pair Distribution Function (PDF): Mining data beneath Bragg peaks offers unique opportunities to understand short-range atomic ordering in crystalline and non-crystalline materials that play key roles in determining physical properties of interest to Earth science, such as compressibility and conductivity. In Earth's interior, melts exist not only in the outer core, but also in the mantle as partial melt fractions. Melts, often present as small fractions of mineral aggregates, disproportionately influence the bulk properties (*e.g.*, Liu *et al*, *PNAS*, 2016). While properties of solid minerals have been extensively studied, properties of melts have gained increasing attention for comprehensively understanding Earth's interior. Additionally, the study of melts will help to further our understanding of the magma ocean and early Earth evolution. This project will offer an experimental routine for PDF data collection at the XPD beamline.

Sound Velocities: Measuring bulk acoustic wave velocities, Vp and Vs, and elastic properties of opaque metallic samples at the core P-T conditions has been extremely challenging and nearly impossible using common light scattering or ultrasonic methods. The project establishes a high-pressure synergy infrastructure to facilitate the application of DACs at the IXS and SRX beamlines. The enabled science includes the study of core composition and identifying the light elements in the core by measuring and comparing sound velocities of candidates for core materials at high P-T conditions with seismic velocity models of the core (*e.g.*, Fiquet *et al. Science*, 2001). The study of the spin state of iron in mantle minerals under high pressures, through the use of x-ray emission, is also a priority.

Ultra-High Pressure and Nano-Imaging: Expanding the static pressure range in a DAC beyond Earth's core routinely has been a dream of the high-pressure community. As the achievable pressure increases towards 1 TPa and beyond, the sample volume at such ultra-high pressures decreases dramatically down to a few microns or less (*e.g.*, Dubrovinsky *et al*. *Nature Comm*, 2012). The infrastructure of this project will facilitate the application of a nanometer-sized x-ray beam to a DAC at the HXN beamline not only to accurately map the pressure with the small peak-pressure area inside a double-stage DAC, but also to enable a measurement of the sample located in such an area without large pressure gradients. Other scientific research that can take advantage of the integration of a DAC and nano-imaging capability at the HXN beamline is to study the melt-solid dihedral behavior inside a DAC during partial melting to understand the process of the core formation early in Earth's history (*e.g.,* Shi *et al. Nature Geosci*, 2013). While the DAC/nano-imaging is highly challenging, DAC/micro-imaging at SRX is more routine offering considerable flexibility for experiment geometry and transmission X-ray microscopy at FXI offers high-throughput with 30-nm resolution imaging for DAC recovered samples.

Transport properties of melts and glasses: The measurement of diffusion coefficients at high pressure is of great interest for understanding the dynamics of Earth's interior. X-ray photon correlation spectroscopy (XPCS) provides a great tool to detect slow dynamic events. By taking a series of speckle patterns at small angle or Bragg angle with coherent synchrotron illumination, the characteristic relaxation time can be determined from sub-millisecond to thousands of seconds (Sutton *et al*. *Nature* 1991). It is particularly useful to study the diffusion coefficient in glass materials, and viscosity change during melting or solidification. The CHX beamline provides both small angle (SA) and wide angle (WA) XPCS with incident energy up to 18 keV, which makes it a unique one-stop beamline to study motion of nano-particles or clusters at low viscous state with SA-XPCS, and the atomic motion in the lattice using WA-XPCS. The infrastructure of this project will facilitate coupling these techniques with a DAC. It provides a unique technique for studying melts and glasses at conditions of Earth's deep interior.

User Community

The proposed facility will be open to the entire COMPRES community. A broad range of users from not only the east coast (*e.g.,* Fei, Goncharov and Walters from GL, Lee from Yale, Saxena and Chen from FIU, Parise from SBU, Walker from Columbia, Wicks from JHU, Watson from Union College, Rebecca Fischer from Harvard, Sean Shieh from UWO) but also central and west coast (*e.g.,* Li from U Michigan, Zhang from U New Mexico, Panero from Ohio State U, Jacobsen from Northwestern, Campbell from U Chicago, Shim from Arizona State U, Lin from UT Austin, Mao from Stanford, Tse from U Saskatchewan) have expressed strong interest in and demands for a DAC facility at NSLS-II for new science opportunities. Considering the average oversubscription rate of 3 at GSECARS and HPCAT, many expressed the importance of a DAC facility at NSLS-II for even just *in situ* laser-heating XRD and one-stop shopping for both IR and XRD. Additionally, the proximity to the Center for Functional Nanomaterials (CFN) at Brookhaven National Laboratory offers the possibility of ex situ characterization of quenched DAC samples. The proposed facility is very important to the newly hired faculty members (*i.e.* Wicks, Watson, Fischer and Zhang) whose career development heavily relies on synchrotron-based research. During the community discussion regarding the proposed facility, some non DAC/synchrotron faculty members (*e.g.,* Peter Heaney from Penn State) expressed strong interests in becoming a user of the facility. The project will fulfill these demands from the community and foster new users of DAC/synchrotron facilities through workshops and hands-on training activities.

Management Team

The management team of the project includes Jiuhua Chen (FIU), John Parise (SBU) and Kanani Lee (Yale), who are all active COMPRES members. Chen has been working at high-pressure facilities (both multi-anvil and DAC) at NSLS for more than two decades, during which he was involved in beamline development, user support and facility management. In addition to overseeing the operation and supervising beamline scientists of this project, he also serves as a coordinator for collaboration between COMPRES and HPSTAR (a partner sponsor of the project). Parise is a renowned DAC and synchrotron expert. He initiated the very successful high pressure facility SNAP at the Spallation Neutron Source (SNS) at Oak Ridge National Laboratory (ORNL) as the project PI and currently serves as the Director of

Joint Photon Science Institute (JPSI), a joint Stony Brook University - Brookhaven Lab initiative. He will supervise the project interaction with NSLS-II and local support (from SBU and BNL) to the project. Lee is a longtime APS and ALS user and has a strong science agenda for synchrotron-based Earth science research. Her laser-heating experience, not only as a user at national facilities, but also as one who has designed her own lab laser heating setup, will help shape the laser heating capabilities at NSLS-II. She will oversee the facility daily operation, mentor the beamline scientist(s) and ensure a close interaction with the community for feedback and suggestion. The management team integrates the strengths of each member in scientific research, beamline development, user support, project management and community communication. In addition, the management team also includes an infrastructure development specialist, Dr. Wenge Yang. He is currently the program manager of HPSynC at APS and will work part-time (to be paid by FIU) for this project to develop the infrastructure at NSLS-II integrating DAC capability and the beamlines that do not have a permanent DAC platform (*i.e.* IXS, HXN, CHX and SRX). He will also advise/assist COMPRES users for project initiation and beamtime application at these beamlines.

The Facility

Proposed DAC facilities at NSLS-II

Beamline	XPD	IXS	HXN	CHX	SRX	HEX
Access (P/I)*	P	I	I	I	I	P**
Technique	ADXRD/PDF/ multi-grain diffraction	Inelastic X-ray scattering	Nano-XRD/ tomography	Coherent scattering	X-ray emission	ADXRD/EDXRD/PDF/multi-grain diffraction/full field imaging

* P: permanent platform; I: Infrastructure enabled; ** Future plan

Permanent DAC platform beamline: The project will establish a permanent platform at the XPD beamline for in situ DAC XRD experiments with *in situ* laser-heating. The XPD beamline is optimized for powder diffraction and pair distribution function (PDF) x-ray total scattering measurement at the photon energy up to 70 keV (https://www.bnl.gov/ps/beamlines/ beamline.php?r=28-ID-2). In addition to adoption of these techniques for DACs, this project will establish an experimental routine for multi-grain diffraction techniques at this beamline. NSLS-II has agreed to accept this project to install such a permanent DAC platform at the XPD beamline.

Infrastructure enabled beamlines: Lead Beamline Scientists at IXS, HXN, CHX and SRX at NSLS-II agreed to work with the project to facilitate the application of DACs at their beamlines. These beamlines (e.g., HXN and CHX) require either special sample stages or specialized DACs that are light weight and/or of small dimensions. The project will design and provide the sample stage and specialized DAC as well as training to COMPRES users to enable such experiments. The project will also work with the SRX beamline scientist to develop the XES capability for DAC experiments.

Infrastructure for DAC operation: The project will establish and maintain a suite of essential equipment to ensure high quality DAC experiments to be conducted at NSLS-II. This equipment includes a gas-loading system, laser drilling system, ruby pressure-calibration system, micro-EDM tool, offline DAC alignment system as well as integrating the existing COMPRES infrastructure at FIS facility (*e.g.,* offline Raman, IR) and multi-anvil facility. This project will offer COMPRES users comprehensive support from cell preparation, sample/pressure-medium loading to data processing.

Beamtime allocation: Initially, the beamtime allocation plan of this project was to regain the 20% Partner User Proposal (PUP) at the XPD beamline to share with the COMPRES multi-anvil project, on top of which COMPRES users will obtain additional beamtime through General User Proposal (GUP) at the XPD and other beamlines. At the workshop, there were extensive discussions on beamlime allocation for the DAC project as a whole. For example, all the related beamlines may take more than one DAC experiment each cycle if enabled by the infrastructure; some may take as much as 50% beamtime for

DAC experiments through GUP program if the proposals are of high quality. On the other hand, a suggestion was made by one of the lead beamline scientists (Yong Cai) that it is better to consider establishing one collective PUP for the DAC project with multiple related beamlines instead of *e.g.*, five individual PUPs with each related beamline. The Deputy Director Qun Shen supported the idea and further elucidated the reality. He also mentioned another potential access mode *i.e.* through a Block Allocation Group (BAG), for the DAC project to access any of the related beamlines/ techniques, and agreed to work with COMPRES to develop an access mode along one of the methods or a combination of them. As the proposal is being written, the discussion is underway.

Future development: At the workshop, the Deputy Director Qun Shen emphasized that COMPRES is a very organized and scientifically promising community and NSLS-II is open to such a community with various opportunities to host specialized instrument at its future beamlines, *e.g.*, the HEX beamline. Then, staff scientist, Zhong Zhong, who was the spokesperson of the previous DAC beamline, X17 of NSLS and is currently in charge of design and construction of the HEX beamline of NSLS-II, presented the audience an exciting blueprint housing DAC stations at the upcoming HEX beamline. The HEX beamline is a high-energy, high-flux x-ray source from a superconductor wiggler (2-3 orders of magnitude higher in brightness than the X17 of NSLS). The beamline will be developed using the funding that is already committed by New York State and is expected to be complete in five years (late 2022). It will offer three branches delivering x-rays simultaneously. The NSLS-II strategic plan for this beamline is to focus on the key science areas of "*structural analysis of the chemical reaction fronts and their evolution in real battery systems during charge and discharge cycles, <u>materials microstructural changes under external extreme conditions such as high mechanical stress, high pressure, high temperatures, and corrosive environment</u>, and phase contrast hard X-ray imaging with a large field of view in real space for studies of micro-crack formation and propagations under realistic operating loads*." As of now, only the center white branch is planned to be instrumented, the other two branches (one white branch and one monochromatic side branch) are open to competitive science programs. The DAC high pressure project is identified as a potential program for these branches. This project will work closely with the scientist in charge to develop a plan for a future dedicated high pressure sector at NSLS-II, like GSECARS/ HPCAT at APS.

Budget Requested

$2.2M for five years (equipment to be installed, DACs for infrastructure and technical details are included in the budget justification).

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