Frontier science by adding pressure as a new dimension at APS beamlines
HPSynC upgrade

Science

In recent decades, we have witnessed an unprecedented surge in high-pressure (HP) research at synchrotron facilities that has greatly improved our fundamental understanding of materials and the processes that govern their behavior - at the electronic, atomic, and molecular levels. These activities have benefited greatly from the development of dedicated HP beamlines, where primary tools, such as diffraction, are optimized for HP environments. However, many *other* communities continue to develop specialized, cutting-edge synchrotron techniques at their own beamlines, which remain an untapped resource for HP science. The High Pressure Synergetic Center (HPSynC) at APS provides a new approach to HP science that complements the dedicated HP beamlines by building a framework encompassing the full diversity of scientific activity around the entire synchrotron ring. This approach permits HPSynC to tailor dozens of non-HP beamlines for the HP community while simultaneously opening a route for HP users and non-HP users to explore the potential of the pressure variable in multi-disciplinary science.

The importance of this approach was underscored in the 2007 DOE report "*Basic Research Needs for Materials under Extreme Environments (MEE)*". Here, four key science priorities are outlined in furthering our understanding the electronic, atomic, and molecular processes that occur within the bulk and at surfaces in materials in extreme environments: *(1) achieving the limits of performance; (2) exploiting extreme environments for material design and synthesis; (3) characterization on the scale of fundamental interactions, and (4) predicting and modeling materials performance*. Pressure, together with temperature, composition, and/or external fields, is a principle variable that can be manipulated for directing and controlling matter as well as for the synthesis of revolutionary new materials. In order to achieve these goals, it is imperative that we look beyond the focused, but localized, perspective of the dedicated HP beamlines. The holistic approach of the HPSynC initiative, facilitated by the requested upgrades, will open previously unexplored avenues for conducting cutting-edge research as highlighted in the following examples:

Magnetic interactions under HP: The research in magnetism and magnetic materials impacts a wide range of areas including superconductivity, magneto-resistivity, low-dimensional electron systems, and spin-polarized electron transport. At present, routine magnetism research depends mainly on high-magnetic fields (Zeeman effect), low temperatures (thermal effect), or dimensionality-reduction (including nano-scale size effects) to induce changes in the magnetic systems. Unlike these methods, *pressure*, which can directly alter the magnetic interactions via modification of orbital overlaps, has uniquely profound effects on the magnetic structure and properties. Therefore, pressure provides an important complimentary variable in the study of magnetism. Its implementation depends on the coupling of HP technology and modern synchrotron based magnetism probes, such as x-ray magnetic circular dichroism (XMCD), nuclear forward scattering (NFS), (non)resonant x-ray emission (R/XES), and (non)resonant x-ray inelastic scattering (R/XIS). HP research on magnetism offers the promise of not only observing the effects of pressure on materials properties but also increasing the fundamental understanding of matter, which will undoubtedly lead to the discovery of materials with novel properties and interesting new physics.

Liquid structure and dynamics: The liquid state is, in general, the most conceptually challenging manifestation of matter. While the atomic or molecular constituents of a liquid lie in very close proximity, and therefore interact strongly, they have no fixed structural relationship to their neighbors, difficult to explore fully the inter-species potentials. As such, the *structure* of liquids – as a function of density – gives direct information on the fundamental forces at play in the condensed phases of matter. Recently, a surprising complexity of behavior has been uncovered, including first-order liquid-liquid structural transitions at HP and strong maxima in the melt lines of several elemental systems (e.g., sodium and carbon) that indicate changes in the relative densities and entropies of the liquid and solid state. Direct information on the structures of these exotic, dense liquids is almost entirely absent at this time. We propose achieving world-leading capabilities for characterizing the liquid structures of fundamental materials (H_2O, C, Na, H_2). Our ultimate goal is to achieve a quality of data comparable to that existing for the crystalline state. Our primary technique will be x-ray diffraction utilizing high energy beams (>60 keV) - which are not generally available at dedicated HP beamlines - to obtain quantitative structure-factor determinations across wide momentum transfers. The use of high energy beams will require novel adaptations of existing diamond-anvil and specially-tailored collimation. In addition, it will also be essential to use optimal synchrotron infrastructure, in particular, detector and focusing technologies. A *portable* laser-heating capability will be an essential part of migrating to new beamlines. The ancillary development of sample containment techniques will be necessary to address the challenges of the, often severely corrosive, nature of liquids under these extreme conditions. We also propose to study the dynamics of liquids at HP. So far, liquid dynamics is described only on two length scales: the continuum limit (hydrodynamics) and on the scale of the interparticle distance (kinematic theory). On the intermediate scale of a few interparticle distances, no straightforward approach exists in statistical physics. High resolution inelastic X-ray scattering (HRIXS) can provide a deep insight in the dynamic properties of liquids. The application of tunable pressure variable in HRIXS can provide essential data for testing various models and our better understanding of liquid dynamics at HP and transportation properties, such as viscosity and diffusivity.

Hydrogen at HP: Understanding the behavior of dense hydrogen, the most abundant element in the universe, is perhaps the most profound and fundamental challenge in the field of HP physics and astrophysics. Outstanding problems include accurate determinations of the density, phase transitions, electronic transitions, and thermodynamic properties of hydrogen, in both the solid and fluid state to multimegabar pressures (>300 GPa) over a wide temperature range, and ultimately to the high-density plasma. Since the early years of quantum mechanics, it has been predicted that hydrogen molecules will dissociate into a dense alkali metal-like atomic form at sufficiently HP. Subsequent calculations indicated that the unique quantum character of the lightest element will impart dramatic and equally unique properties to the material at very high densities, including novel superconductivity and singular high energy density. Before its dissociation, solid molecular hydrogen has been predicted to exhibit various novel behaviors, including orientational ordering in a crystalline lattice, band-overlap metallization or, alternatively, the conjectured excitonic insulator state. After decades of HP experimental efforts, optical measurements have been extended in solid hydrogen to the 300 GPa range, which exceeds the originally predicted metallic transition at 25 GPa by more than an order of magnitude. However, the original goal of alkali-like metallic hydrogen remains elusive: the proton-paired state persists with the enhanced Friedel structure up to 300 GPa. The upgrade of HPSynC, together with a series of specialized beamlines, will enable key diagnostic experiments and extend the maximum P beyond 500 GPa which is the currently prevailing estimate for the transition to the ultimate atomic metal. New HP diffraction with sub-micron beams will extend the maximum pressure and yield direct crystallographic and orientational information on the ordered crystal structure of hydrogen II and III as well as the atomic metal. In the past, due to the limitation of the diamond window bandgap at 5 eV, the hydrogen bandgap could only be implied indirectly through measurements of refractive indices. HP experiments at optimized spectroscopy and scattering beamlines will enable access to the rich electronic information above 5 eV, to follow the change and closure of the hydrogen band-gap as a result of compression and phase transitions, to probe the plasmon if hydrogen becomes a metal and, alternatively, to detect the hydrogen exciton if indeed hydrogen becomes the first example of an excitonic insulator. The “holy grail” of metallic hydrogen is finally within the realm of a practical goal.

Added value of the Mid-term Upgrade

Establishing key equipment is critical for cutting edge HP research and for bridging distinct scientific communities. This upgrade will greatly enhance HPSynC capability to develop novel HP synchrotron techniques and make them available to a wide user base. The proposed equipment includes state-of-art sample preparation facilities, experimental apparatuses for specific beamline applications, as well as portable or fixed beamline optics and equipment to accommodate HP apparatuses.

Micro-machining with femtosecond laser: The diamond anvil cell (DAC), coupled with various modern synchrotron radiation techniques, is anticipated to retain its leading role in the next-generation HP science. Successful HP experiments depend critically upon the quality of sample preparation. Next-generation DAC based HP research will require focused effort on micro-machining techniques in sample preparation. For example, we are now facing grand challenges to fabricate single crystals of various materials to meet stringent requirements of shape, size, and crystal perfection. Techniques such as high pressure single crystal diffraction, inelastic x-ray scattering and charge/spin-density-wave x-ray diffraction, all require perfect single crystals without background scattering from damaged surfaces. This constitutes a unique challenge when the ideal HP sample is typically plates of several microns thickness and several tens of microns width. Unfortunately, the commonly accessible micromachining techniques by mechanical polishing, EDM, and Q-switched YAG laser cutting cause several microns surface damages and are typically limited to larger samples, while focused ion beam (FIB) nanofabrication, on the other hand, is too small and too time consuming. So far, only femtosecond (FS) laser micromachining plus ion-milling surface cleaning shows promise, since it provides high quality micromachining of many materials and abilities for minimal damage and precise processing. We propose to establish FS laser micro-machining technique at APS. This will also benefit the strain-free cutting of silicon and diamond single crystals for the Optics Fabrication and Metrology Group at APS.

Portable systems: In addition to a few portable systems (ruby fluorescence, small focusing mirror system, DAC adaptors) under construction, we propose the following key devices.

Portable laser heating system: Adding the temperature variable in combination with pressure is particularly valuable because P and T are two independent dimensions, and their multiplicative effect greatly widens the scope of materials base and physical states. While the HP DAC is a portable and flexible component, the conventional laser heating (LH) systems with its meter-length laser, bulky power supply and cooling system, rigid optical train and optical table, are very difficult to move and time-consuming to align, and must be regarded as fixed instrument to be installed at individual facilities. With the development of fiber laser technology, we propose to construct portable laser heating systems that can be used flexibly in many beamlines for performing specific high pressure-temperature experiments. Besides its portability, the proposed LH system will extend the accessible temperature range up to 10,000 K and down to 500 K. The system will provide stable laser heating on a target material in DAC in terms of both time and pointing position. The heating spot size will be 30-150 μm in diameter to match various analytical probe sizes.

Portable cryostats: To add the temperature variable down to sub-Kelvin, we propose to build three cryostats suitable for DAC and matching specialized synchrotron techniques including x-ray diffraction, x-ray scattering, and x-ray absorption. These cryostats will have both membrane and motor driven controls for pressure tuning of the DAC inside, thus proving large tunable high-pressure low-temperature conditions.

Portable bimorph KB mirror system: Extreme pressure is reached at the cost of reducing sample size. Small probing size is thus essential for conducting HP experiment. For specialized beamlines that do not offer small beam, a portable x-ray focusing system is necessary for HP experiments. The proposed system will consist of two 300mm long mirrors, since they would provide the best compromise between a large beam acceptance and a small tabletop set up. Both mirrors will be equipped with the maximum number of electrodes to offer the greatest versatility and a beam size less than 5 μm.

HP sample preparation laboratory: We propose to equip the HPSynC sample preparation laboratory with the state-of-the-art devices including micro-machining facilities, micro manipulators, and a gas loading system. These added systems will put HP sample preparation to a new generation, and will yield a significant competitive edge to the APS HP programs.

Micro-machining devices: In addition to the above proposed micro-machining with femtosecond laser, we also propose to have an in house laser ablation facility. These micro machining devices not only provide capabilities for machining super HP samples, but are important in developing novel HP synchrotron techniques. For instance, for weak scattering samples, development of a new generation of pressure cells is clearly needed with two primary foci: to achieve a minimum amount of illuminated anvil material and to maximize the sample volume for a given pressure. Both of these could be addressed by micro-shaping of the anvils. The DAC has been hugely successful as a tool for HP science, but its design has remained relatively unchanged since original inception in the 1950's. We propose to develop an extensive program of tailor-profiled diamond culets.

Micro-manipulator: We propose to have micro-manipulators for precise sample loading. This facility will ensure quality sample loading and enable new sample configurations for cutting edge HP research. It is especially beneficial when combined with a glove box for environment sensitive samples.

Gas-loading system: Many important scientific problems require the loading of samples or pressure media that are gaseous under ambient conditions into pressure cells. GSECARS has already successfully developed a sophisticated gas-loading set up. Their design is optimized for both safety *and* usability, making it ideal for visiting users. However, due to existing construction materials, the current system is limited to inert gases such as He, Ne and Ar. While this supports the loading of "pressure transmitting media" to ensure hydrostatic conditions for a solid sample, it means studies of samples that themselves are gaseous is not possible. We propose a new system, using the same highly versatile GSECARS design, but to adapt for use with specialized gases. Of particular importance will be construction from hydrogen proof steels to facilitate studies of hydrogen storage materials and hydrogen itself. In addition, we will develop specialized gas-handling systems for hazardous gases: such as O_2, CO_2, CH_4, H_2S etc. This capability will provide a unique facility at the APS and will unlock significant advances in our scientific endeavors.

Partnerships and expected user communities

HPSynC is co-managed by APS and Carnegie Institution of Washington (CIW). The main mission is to develop novel HP synchrotron techniques and make them available to user communities. The proposed upgrade items are all essential equipment for HP research and are, thus, beneficial for all HP users. For example, the preliminary survey on micro machining with femtosecond laser has immediately received strong support from 15 groups, including Abby Kavner (UCLA), Elizabeth Cottrell, (Smithsonian Institution), Hongwu Xu, (LANL), Jay Bass (UIUC), Jie Li (UIUC), Jingzhu Hu (BNL), Kanani K. M. Lee (Yale), Kurt Leinenweber (Arizona State), Malcolm Nicol (UNLV), Murli H. Manghnani (U Hawaii). Robert Liebermann (Stony Brook), Thomas Duffy (Princeton), William J. Evans (LLNL), Yanzhang Ma (Texas Tech). The development of non-crystalline diffraction capabilities has received enthusiastic support from John Parise (Stony Brook), Chris Tulk (Oak Ridge), Jeff Yarger (Arizona State), Chris Benmore (APS), Dan Shim (MIT), and Alex Goncharov (Carnegie)

Enabling technology and infrastructure

See *Added value of the mid-term upgrade*

Estimated budget ($ in thousands)

	Year 1	Year 2	Year 3	Year 4	Year 5	Total
Micro machining with femtosecond laser	500	100	100	100	100	900
Portable laser heating system	160	200	100	0	0	460
Portable cryostats	0	100	100	150	0	350
Portable bimorph mirror system	450	0	0	0	0	450
Laser ablation facility	0	300	50	0	0	350
Micro manipulator systems and glove box	50	100	0	0	0	150
Gas loading system	200	20	20	20	20	280
Total	1,360	820	370	270	120	2,940