Directing and Controlling Matter under Extreme Pressures and Temperatures: An integrated approach with at least one order of magnitude improvement in spatial, temporal, diffraction, and energy resolution at HPCAT 16-ID

High-Pressure Science and HPCAT

Pressure, together with temperature, composition, or external fields, alone or in combination, can be used as principal variables for directing and controlling matter as well as for the synthesis of revolutionary new materials. Exploration of the pressure dimension has the potential to at least triple our basis of 300,000 known materials. The sheer number is extraordinary, but more significantly, entirely new classes of materials with intriguing properties may appear, thus opening fertile ground in unprecedented ways to ***produce*** and ***control*** materials with tailored functionalities. In the recent BES Grand Challenges Report *Directing Matter and Energy (DME)*, five Grand Challenges have been identified for *acquiring the ability to direct and control matter all the way down to molecular, atomic, and electronic levels*. A later report on *Basic Research Needs for Materials under Extreme Environments (BES Workshop MUEE, 2007)* has further spelled out the need to design and control revolutionary materials for future energy applications using the controlled extreme pressure environment and advanced analytical probes.

The development of high pressure (HP) techniques and a battery of associated synchrotron techniques have opened a vast new window. HP synchrotron facilities provide crucial x-ray analytical probes to ***characterize*** materials structure and dynamics over broad time and length scales. Designed in 1998, HPCAT has pioneered the third-generation, high-pressure, synchrotron facility in which multiple techniques have been developed and integrated at a single sector for the advancement of multidisciplinary HP research. The HPCAT program has been exceedingly successful in scientific impact, technological advances, and user community development. Within five years of its commissioning, HPCAT has surpassed all high-pressure synchrotron sectors in the world in terms of quality and quantity of scientific publications, and emerged as one of the most productive sectors at APS. In addition, HPCAT has made truly significant contributions to DOE's classified research programs.

Ten year's explorations at HPCAT have revealed clear directions for the next generation of HP synchrotron research. Meanwhile, our prototype facility developed a decade ago is aging and facing steep competitions from newly established HP beamlines in Europe and Asia that adopted the HPCAT innovations. The great demand and shortage of the very limited HPCAT beam time also severely restrict our ability of community outreach. We have reached a critical stage for a major upgrade to stay competitive at the cutting edge. The proposed upgrade will accomplish three goals at the same time: (1) advancing to the next generation HP synchrotron science, (2) optimizing the present main stream HP synchrotron techniques, and (3) resolving the beam time shortage issue. The upgrade will lead to myriad discoveries of novel hydrogen storage materials, superconductors, electronic materials, magnetic materials, superhard materials, optical materials, high-energy-density materials, and stockpile stewardship materials that are central to DOE long-range goals; the upgrade will also have far-reaching impact on fundamental research in physics, chemistry, geoscience, and astrophysics.

Added value of the Mid-term Upgrade

We aim to improve more than one order of magnitude in spatial, diffraction, temporal, and energy resolutions over the current HP frontier. These upgrades can often be coupled and result in multiple orders of magnitude improvements.

1. *Canted double undulator and associated x-ray optics upgrade* will increase the available 16ID beam time by 50%, and more significantly will allow optimization of the 16ID-B x-ray diffraction and 16ID-D x-ray spectroscopy optics, resulting in a total of ten times improvement in efficiency and effective beam time. The undulator and optics upgrade are also a prerequisite of other upgrades.
2. *Optimized submicron incident x-ray beam and x-ray depth probe* discriminate the weak HP sample signals from the very strong background from the pressure chamber materials. This should be the most important consideration for HP experiment, yet least explored and developed. Reducing the incident beam size from 5 to 0.5 µm and depth probe from 300 to 3 µm will undoubtedly revolutionize HP synchrotron science in general, and will increase the counting efficiency by >100 times since S/N ratio only improves with square root of the counting time.
3. *High-resolution HP x-ray diffraction (XRD), time-resolved HP synchrotron experimentation, and optimized medium energy resolution HP inelastic x-ray scattering (IXS)* will each open entirely new branch of HP science which does not exist or is in its infancy.

Detailed Descriptions

Beamline optics upgrade: We propose to upgrade the 16ID x-ray optics train for higher brilliance to keep HPCAT at the most competitive edge of HP research. The anticipated increase in brilliance arising from the beamline upgrade will also allow us to develop novel HP synchrotron techniques as proposed here. We have investigated and identified the key components that need to be strengthened, improved, or replaced for robust operation with maximum brilliance. The current insertion device at HPCAT is a standard APS type-A undulator which was designed conservatively before the establishment of APS. The type-A undulator operates with a magnetic period of 3.3 cm and a length of 2.5 m. We propose to upgrade the undulator to the full length, with two ~2.4 m undulators in canted mode at optimized magnetic periods. The upgrade will free the two 16ID lines for totally independent operation. We have discussed with APS engineers the possibility of bumping the beam so as to allow both undulators to provide beam for a chosen canted beamline, and received optimistic feedback. Such undulator operation options will maximize the brilliance from the undulators for flux-demanding experiments such as IXS and high resolution XRD coupled with sub-micron beams. With the anticipated second undulator at Sector 16 and the overall APS upgrade in the horizon, the source brilliance of HPCAT will be unmatched in the next decade. The undulator upgrade will include the new undulator, modifications to the existing undulator and front end components on the storage ring side of the shield wall. The upgrade also will involve modification or replacement of high heat-load beamline optics, including front end slits, two thermal apertures, and two monochromators. HPCAT currently has a diamond double-crystal monochromator (DCM) to reflect vertically a monochromatic segment of the x-rays at 16ID into the 16ID-C-D-E stations for HP scattering and spectroscopy studies, and a branching DCM to divert horizontally the transmitted x-ray beam (at a higher energy) to the 16ID-B station for HP XRD studies, thus creating two operating, but constrained by energy selection, ID lines. With the proposed two canted beamlines, these two monochromators can be optimized for their individual applications and independent operations. The existing water cooled thin diamond crystals in the DCM will be replaced by cryogenic cooled silicon crystals. The branching DCM will be upgraded for a larger energy range and better efficiency with cryogenic cooling. Upgrading these components will immediately benefit the present operation as well as the proposed new HP x-ray techniques. Moreover, the x-ray optics upgrade will provide sufficient margin for HPCAT to handle the anticipated higher brilliance due to the source upgrades: first, a local enhancement at 16ID by adding the second undulator in the next couple of years, and then a facility-wide upgrade of the APS storage ring and lattice in the next decade.

Beam size reduction from 5-10 μm to 0.5-1 μm: Extreme conditions are achieved at the cost of sample volume. It took three decades to reduce the x-ray beam size from 50 μm to the current standard of 5 μm in HP research. Yet, the impact of this one order of magnitude reduction is tremendously significant. While beam sizes of a few tens of nm have been reached in specialized beamlines, a beam size of 5-10 μm is still typical in all state of the art HP synchrotron beamlines in the world, with an exception of the recent HP beamline upgrade at ESRF, where a beam size of 2-3 μm is now used. We propose to have another order of magnitude reduction to a beam size of 0.5-1 μm in the end station 16ID-E at HPCAT. The upgrade of 16-ID beamline optics, including its high brilliance and minimum distortion optics, will be crucial for obtaining sub-micron beam. We plan to use an ultra-stable support table, K-B mirror systems, and high resolution sample stages. These x-ray optics and controls have been well developed at specialized beamlines (Nano-sector 26, sectors 2, 32, and 34); and we will benefit from their experience. More importantly, the upgrade at HPCAT will focus on optimization for HP vessels and samples in high pressure and extreme temperature environments, in terms of x-ray energy, beam size, flux, diffraction resolution, background discrimination, sample stages, and detector configuration. The end station is ideal for a smaller beam because of its long distance from the source (75 m). HP XRD, HP x-ray spectroscopy (emission, absorption), and HP x-ray imaging setups will be established with the sub-micron spatial resolution in the 16ID-E station. The enabling sub-micron beam, optimized and dedicated for HP research, will allow us to probe small sample sizes which would in turn allow us exceed Terra-pascal static pressures, to investigate grain-to-grain interactions with submicron resolution in all three dimensions, to map out the grain boundary, phase boundary, chemical ordering, local stress/strain and structure evolution within the individual grain, and to study nanoscale single crystals. It will also benefit the study of natural mineral inclusions which are usually submicron in size and embedded in other mineral substrates.

Improving depth resolution: While the spatial resolution in a plane perpendicular to x-ray beam has been effectively improved by reducing a probing size, the spatial depth resolution along the x-ray beam remains an under-developed area. Yet, this is particularly important to HP research because HP samples are always surrounded by chamber materials which often cause strong background signals. Improving depth resolution not only will greatly increase the counting efficiency due to the improvement of S/N ratio, but will make a number of cutting edge projects feasible. The conventional pinhole collimator is often used on the detection side in HP studies, which typically provides a depth resolution of 300-500 μm. We propose to use x-ray lens (e.g., polycapillary x-ray focusing optics) to construct a confocal microscope for collecting scattering signals. Such an x-ray lens can collect a large solid angle (up to 20 degree) with a focus spot as small as 10 μm. The use of x-ray microscope on the detection side will improve the depth resolution by at least an order of magnitude, and will significantly reduce the background from surrounding materials and enhance the detection sensitivity by >100 times. The depth resolution gain through an x-ray microscope is particularly beneficial in HP IXS and HP emission experiments.

Time resolved HP studies: This area is still in its infancy in HP research. A handful of pilot experiments at HPCAT have shown its crucial role in understanding physical and chemical processes at HP. For example, diffraction patterns from single crystal samples under dynamic shock loading have been recorded in real time by performing sub-nanosecond, high-resolution diffraction measurements. Using a dynamic DAC (dDAC), capable of fine control of pressures and (de)compression with a repetition rate of 1 Hz – 1 kHz, phase transition dynamics have been studied at HP. These pilot experiments were performed using user-provided or loaned x-ray optics and detectors. We propose to enable the time resolved capability at the integrated HPCAT facility for the entire HP user community. Proposed equipment for time resolved studies include x-ray shutters, timing electronics, and fast detectors. Different HP devices will be integrated, such as shock wave devices, dDACs, and pulsed laser heating setups. The dynamic DAC technique repetitively applies a time-dependent load/pressure profile to a sample. This capability allows studies of the kinetics of phase transitions and metastable phases at compression (strain) rates of up to 500 GPa/s. On the other hand, pulsed laser heated of the DAC repetitively heats up a sample at a few kHz rate at a given pressure. The laser heated DAC in general has been used for generating extreme pressure-temperature conditions. However, with the traditional continuous-wave laser heating techniques, laser heating at Mbar pressures causes frequent anvil failures. With pulsed laser heating, we expect the shorter time to reduce the exposure of the DAC to extreme conditions and the associated failure modes. There is thus a general need to characterize materials under such extreme conditions by time resolved studies. For time resolved studies in repetitive events, an x-ray shutter (a repetition rate of a few kHz) and a HP device (dDAC or pulsed laser heating) can be synchronized to each point relative to an event. In this case, detectors do not necessarily need the time resolved capability, and existing detectors (e.g., MAR-IP) can be used. For time resolved studies in fast single processing events (shockwave, run-away processes), fast detectors are needed. We propose to have a fast detector having at least 20k fps capability for x-ray imaging studies to monitor chemical reactions or physical processes such as melting at HP. We will coordinate with the APS detector pool to provide faster x-ray detectors to study these processes under HP. Time resolved HP studies will provide important information on structural dynamics, phase transition dynamics, chemical reactions under HP, and materials metastability and local minimum energy configurations.

Higher diffraction resolution: X-ray diffraction has been one of the most basic workhorses for HP research. Currently, instrumentation at all major HP beamlines in the world only allows HP diffraction studies at moderate resolution using monochromatic radiation with 2-dimensional angular dispersive detectors, or low resolution using polychromatic radiation with energy dispersive detectors. Progress at HPCAT has now allowed us to propose the implementation of a high-resolution diffraction system to improve the $\Delta d/d$ resolution by one order of magnitude. We propose to install a high resolution diffractometer with the 2θ arm of at least 1.5 m long, a circular confusion < 1μm, and a 2θ angular resolution of <0.001 degrees. The diffractometer can hold an area detector with a precise stage for changing sample to detector distance. Depending on the experimental requirements, we also plan to employ scanned diffraction capability with a bank of point detectors (e.g. scintillation counters) with slit systems. Pressure-induced electronic and magnetic transitions, such as spin-pairing in transition metals, Mott metallization, and Lifshitz transitions, are often associated with important structural changes that are difficult to detect with the available existing HP XRD resolution, will be clearly resolved with the proposed development of high-resolution XRD. Crucial but subtle HP phase transitions can be quantified and a great deal more similar discoveries will be expected. Numerous controversies arising from limited resolution and accuracy of existing HP XRD technology can be settled.

HP inelastic scattering at 70-200 meV resolution: Pressure has drastic effects on the energy and dispersion of all electronic bands and is an ideal tuning variable to direct and control matter at electronic level. The IXS technique developed at HPCAT has an energy resolution of 0.7-1 eV for investigating fundamental properties of the electron gas, chemical bonding, and high-energy electronic excitations in energy and momentum space. However, many interesting properties occur for electronic excitations within a small fraction of an eV to several eV of the Fermi surface. There is a great need for improving the energy resolution in IXS to obtain more detailed information on electronic excitations and plasmons, and finer features in x-ray Raman spectra. Count rates become a challenging issue because higher resolution means a smaller slice of both the incident spectrum and the scattered photons, in addition to the small inelastic scattering cross-section. With the source and optics upgrade, the increase in brilliance and efficiency allows for developing high resolution IXS in the 100 meV region at HPCAT. We propose to construct a set of medium resolution (ME) monochromators with 70 meV to 200 meV resolution and an analyzer array matching (and slightly higher than) the energy resolution of the monochromator. We will establish a KB mirror system that will collect most of the undulator beam and focuses down to 5 μm. Flat-pixel analyzers, together with the newly developed Pilatus area detector, will be used for high energy resolution, low background, and high efficiency ME-IXS measurements. Particular effort will be devoted to minimizing scattering signals from surrounding materials (anvils and gaskets), by using a confocal x-ray microscope for background rejection.

Partnerships and expected user communities

HPCAT is managed by the Carnegie Institution (CIW) and supported by three other leading HP groups: Lawrence Livermore Nation Laboratory (LLNL), University of Nevada at Las Vegas (UNLV), and Carnegie-DOE Alliance Center (CDAC). The upgrade plan has been thoroughly discussed with and fully supported by HPCAT members. We have also received suggestions and support from HPCAT Technical Advisory Committee (Wolfgang Sturhahn-chair, Chi-chang Kao, Martin Kunz, Mark Rivers, Deming Shu). HPCAT has now more than 200 individual investigators annually, consisting of users from member institutions and through general user proposals. With the one order of magnitude improvement in spatial, diffraction, and energy resolution, the enabled time resolved capability, and the overall beamline optics improvement outline in this proposal, we expect that the demand for cutting edge HP experiments to only grow further.

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
Beamline insertion device and optics **a:** second undulator, high heat load optics, DCM, BDCM, beamline re-configuration; **b:** high resolution monochromators (70, 120, 250 meV)	1,650[a]	650[b]	0	0	0	2300
Sub-micron beam **c:** vibration control table, **d:** KB mirrors and nano stages, **e:** detectors		150[c]	400[d]	300[e]	0	850
X-ray microscopes and control stages One set in year 1, more sets in year 2	300	200				500
Time resolved **f:** x-ray shutter and electronics, **g:** pulsed laser heating and fast detectors	0	250[f]	350[g]	0	0	600
High diffraction resolution diffractometer	0	250	0	0	0	250
IXS at 70-200 meV resolution **h:** bimorph mirrors, **i:** analyzers and detectors	0	0	400[h]	450[i]	0	850
Total	1,950	15,00	1,150	750	0	5,350