A micro-focused, multi-probe high energy x-ray beamline
for measurements at extreme conditions

Science case.

This proposal relates to two recent BES workshops "Materials under Extreme Conditions" and "Basic Research Needs for Geosciences". The conclusions of the first report identify one common denominator in all types of extremes considered; the importance of *"atomic and nanoscale defects"*. That is to say, understanding the role of atomic disorder is vital for understanding and enhancing a materials properties and functionality. Both reports also emphasize the importance of *in-situ* characterization tools to probe structure at the atomic level and the use of multiple probes to identify structure/property relations. With this in mind, the penetrating power of high energy x-rays makes them well suited to experiments at extreme conditions involving bulky sample environment equipment; such as magnets, cryostats, furnaces and high pressure cells. In order to achieve high pressures (>15 GPa), precise temperatures, better spatial resolution as well as enable dual capabilities such as x-ray diffraction plus micro-Raman or calorimetry, micro-focused high energy x-ray beams are required.

It is well known that phase transitions often show strong structure/property relations with varying pressure, temperature or magnetic field. In recent years the scientific community has started to tackle more difficult problems in this area which relate to the inherent local disorder, rather than just the ordered crystalline component of the materials structure. Such emerging phenomena include pressure induced amorphization, the existence of abrupt and reversible liquid-liquid transitions and the formation of transient metastable states. Amorphous to amorphous and amorphous to crystalline transitions are provide the latest 'knob' for controlling a materials properties and are becoming a major research focus area in both academia and industry. A prime example of a recent technology built on an amorphous to crystalline transition in a chalcogenide glass is that of the DVD Blu-ray, the next generation optical disc format. The new idea that unique structural forms (and hence properties) are intimately related to the formation of reluctant or "fragile" glass formers around the glass transition temperature T_g, brings again into question the structural nature of the glass transition itself. This together with the recent notion that an analogous glass transition pressure Tg also exists, provides the possibility of tailoring existing materials with a plethora of new and unique properties.

In condensed matter physics, it is becoming increasingly recognized that the lattice degree of freedom plays a pivotal role in many intriguing physical phenomena, ranging from metal-insulator transitions, superconductivity, colossal magneto-resistance, piezoelectricity to multiferroics. The complex phase diagrams with novel physical properties, originating from the interplay between different interactions in the systems, have many structural manifestations, including spin/charge/orbital-order-induced structural modulations, polar nanoregions, nano- to micro-scale phase separations, heterogeneities. *In-situ* investigations on the structural responses to the external stimuli (temperature, pressure, fields etc.) will provide key information for better understanding many fundamental issues of the underlying physics. The proposed micro-focusing capacity will allow structural studies of emerging materials under multiple sample environments.

Future increasing energy demands require novel energy technologies, which will rely on materials with better performance with respect to extremes. Often materials fail at one-tenth or less of their intrinsic limits, which is not well understood. It is known however, that the failure in the bulk materials starts from multi-length scale defects at surfaces and/or interfaces. High penetration capacity of high-energy x-rays allows determinations and characterizations of those defects in bulk samples, without destructive sample treatments. Without a micro-focusing beam, the bulk samples would provide large “background” in addition to the diffraction patterns of those defects. Micro-focusing high-energy x-rays will provide the spatial resolution needed to investigate the local structures of defects and their relation to bulk properties.

Multi-probe approach

The structural determination of materials over a wide range of pressure and temperature is necessary to evaluate structure/property relations, and whenever possible combinations of different experimental techniques are required to solve a particular problem. High energy experiments are well suited to accommodate other probes since most of the useful scattering is in the forward direction and only occupies a small solid angle suitable for the use of both area detectors and arrays of single point detectors.

For example, *in-situ* microbeam high energy x-ray Pair Distribution Function (PDF) measurements and micro-Raman capabilities at high temperatures and pressures using diamond anvil cells would provide complimentary information on both the local structure and Q-speciation. Similarly, high temperature calorimetry combined with highly penetrating microbeam high energy x-ray diffraction can give a direct link between the thermodynamic heat capacity and structural re-arrangements at the atomic level. In this regard, the feasibility of microbeam high energy x-ray diffraction with NMR should also be explored in the longer term. High pressure acoustic measurements used to determine bulk moduli have recently been shown to strongly correlate with local structural arrangements in a chalcogenide glass. Similar capabilities, if developed at sector 13 using a multi-anvil device would provide complimentary property information to the structural measurements made using a diamond anvil cell at high pressures.

Analysis software and modeling

Our ability to measure x-ray scattering data far outweighs our capability to provide realistic atomic scale structural models of the problems we are trying to address. In this modern age of computing, we argue that any significant investment in an experimental upgrade should be accompanied by a modest investment in data analysis and atomic to nano-scale computer modeling of the scattering data. Since the nature of this upgrade proposal is to cover a wide range of states or densities and to use multiple probes, an atomic simulation code able to make use of all the data and predict trends would be extremely beneficial. Computers dedicated to running available data analysis software for all the probes, atomistic Molecular Dynamics and Monte Carlo codes for user and in house simulations of the phase transitions, together with atomic data visualization software, would provide the first step toward the ideal goal of producing not only unique measurement capabilities, but the unique answer(s) to a specific problem.

Added value to the medium term upgrade

Probably the most important application of micro-focused high energy x-ray beams is the use of high pressure diamond anvils cells. Considerable progress in this area has already been made at sector 1. A fixed high energy micro-focused beamline of ~100 keV is a natural home for this new field of research, and will provide a means of investigating phase transitions relevant to both condensed matter physics and the geosciences. The combination of a micro-focused high-energy x-ray beam with a high-resolution capacity would also provide a unique experimental tool to study phase transitions at high pressure and low or high temperatures. One recent example of this performed (with difficulty) on 11-ID C using an unfocussed beam is the origin of morphotropic phase boundaries in ferroeletrics.

Expected User Communities

One of the mains goals is to extend the application of high-energy x-rays to understand novel phenomena and materials with unique properties, which occur at extreme environments. Another goal is to align our scientific program with planned complimentary neutron scattering instrumentation developments at SNS to study materials at extreme conditions and to further exceed these capabilities. The user communities are primarily those from high pressure, geosciences and condensed matter physics and materials science.

Enabling technology and infrastructure

Components required:

- Microfocusing optics.
- GE a-Si area detector.
- Multiple analyzer and point detector arrays for high-resolution and fast data collection.
- Online pressure determination setup and pressure cells.
- Micro-Raman for combined spectroscopy and diffraction studies of phase transitions.
- Calorimetry for combined thermodynamic and diffraction measurements e.g. through glass transition temperature.
- Computers, MD and MC simulation codes and visualization software.

Partnerships and user interest

Many professors and scientists in the US have expressed strong interest on the micro-focusing capacity for high-energy x-rays. A strong interaction with complimentary neutron facilities at the SNS is desired by many users who are a part of both the neutron and x-ray communities:

Dr. Ho-kwang (David) Mao, Geophysical Laboratory, Carnegie Institute of Washington;
Prof. Takeshi Egami and Dr. Wojtek Dmowski, University of Tennessee;
Prof. Jianshi Zhou, Texas Materials Institute, University of Texas at Austin;
Dr. Chris Tulk, High pressure instrument scientist, SNS, Oak Ridge National Laboratory
Dr. Joerg Neuefeind, Disordered materials instrument scientist, SNS, Oak Ridge National Laboratory
Prof. Jeffery L. Yarger, Professor of Chemistry, Arizona State University
Prof. Sabyasachi Sen, Professor of materials science, University of California at Davis
Prof. John B. Parise, Professor of Geosciences, Stonybrook University
Dr. Muhtar Ahart, Dr. Maddury Somayazulu, Geophysical Laboratory, Carnegie Institute of Washington ;
Dr. Malcolm Guthrie, Dr. Yang Ding, HPSync, APS, Argonne National Laboratory

Industry and technology transfer

Possible industrial links with GM motor company (high temperature liquid alloys), California Cement Company (spatial resolution of amorphous cements) and Corning glass (low T_g glasses).

Estimated Budget :

	Year 1	Year 2	Year 3	Year 4	Year 5	Total
M&S	$50K	$50K	$30K	$20K	$10K	$160K
Capital	$480K	$370K	$120K	$60K	0	$1030K
Total	$530K	$420K	$150K	$80K	$10K	**$1190K**