Structural Science Using Tunable High-Energy Synchrotron X-Ray Source

Science

The knowledge of the structure of materials is the fundamental of understanding of their properties, activities and functionalities. The information of different characteristics of materials provides important clues of discovery of new superconductors, thermoelectics, catalysts, hydrogen storage, or battery materials. It is even more important to understand how structures vary under different environmental conditions such as temperature, pressure, gas flow, magnetic or electric field. This structural information will give us a unique insight of the behavior of materials under real application environments. High-energy x-ray provides the unique capability to probe through the environmental devices and obtain crucial structural information. The proposed high-energy x-ray facility will employ various x-ray diffraction techniques, such as high-resolution powder and single crystal diffraction, diffuse scattering, resonance scattering, and charge density measurements, to study detail fundamental structures and structural changes under various environmental conditions that relate to properties, activities and functionalities of materials. There are many research programs using high-energy x-ray scattering techniques extensively studying structural behaviors of materials in areas such as condensed matter physics and chemical materials. This structural information provides evidence of many fundamental characters of materials and their potential applications. Below are a few examples of scientific applications.

Single crystal and diffuse scattering studies Using single crystal crystallography and diffuse scattering techniques is a powerful way to study the detail structures at atomic level. By employing high-energy x-rays for diffuse scattering measurement, the advantages are extension of the accessible diffraction space to higher wavevectors, the ability to use crystals of irregular shape without the need for absorption corrections, less need to prepare sample surfaces. There are many studies taking advantage of the high energy capability. Most notably are the studies of doped bilayered Manganite, $La_{2-2x}Sr_{1+2x}Mn_2O_7$, because they exhibit colossal magnetoresistance (CMR)[1]. Recently, the diffraction results present clear evidence for a first-order transition from a charge or orbital odered insulator to an in-plane metal at low temperatures in the layered manganites with $x = 0.6$[2].

Magnetic-field-driven structural transitions The fact that an electron is a tiny magnet has long been ignored in conventional electronic devices. Recently the spin degree of freedom of the electron is being utilized to play a key role in the next generation electronics, the so-called spintronics. In this science and technology frontier, exploration of the interplay between spin and other (e.g., charge, orbital and lattice) degree of freedoms in condensed matter and material sciences is of both fundamental and practical importance. During last two decades, giant and colossal physical and mechanical responses induced by magnetic field have been discovered in many systems, ranging from transition metal oxides, to intermetallic compounds to magnetic alloys. The colossal magneto-physical, caloric and mechanical phenomenon will lead to smarter and more energy-efficient applications. Our in-situ high-energy x-ray diffraction study of materials under magnetic fields have demonstrate the significant interactions between the spin and lattice, that the magnetic-field-manipulations of the spin degree of freedom often give rise to atomic-level structural reconfigurations, as evidenced by lattice modulations and/or structural transitions. The observed colossal magneto-responses are in fact the manifestations of the magnetic-field-induced structural transitions, involving phases with drastically different property.

High temperature laser heating The developments of using the laser heating levitator system to study materials at extreme high temperature have performed at high energy beamlines, 6-ID-D and 11-ID-C. The combination of this containerless sample environment at extreme high temperature and high energy x-ray allows the simultaneous investigation of structures and properties at high temperatures while avoiding problems of environmental contamination. This unique capability has enabled the study of pioneering studies of short and medium range order in liquids above and below the liquidus temperature; liquid-liquid phase transitions; phase-transformation kinetics during solidification and their effect on materials processing; evolving order in systems undergoing structural transformations; and the detection and characterization of metastable phases and their structures. Recent study of SiO_2 melt process is one of examples of high temperature oxide melt studies[3]. A $Ti_{39.5}Zr_{39.5}Ni_{21}$ liquid alloy showed a preferential nucleation of a metastable icosahedral quasicrystal instead of the stable C14 Laves crystal phase, indicating a lower nucleation barrier for the metastable phase[4]. Understanding the formation of structures far from equilibrium has been identified as an important current area of research. Supercooling and glass formation in viscous liquids is also becoming a subject of considerable study; especially interesting is the approach of supercooled “fragile” liquids (reluctant glass formers) towards the glass transition where the temperature dependence becomes super-Arrhenius. The ability to determine *in-situ* both the structure and properties of these metastable phases, their formation with composition and temperature, and their role in the formation or inhibition of stable phases at lower temperatures will add tremendously to our understanding of stable phase formation in complex systems.

Atomic distribution/phase determination using resonant scattering Using resonant scattering to generate diffraction contrast to study neighboring elements distribution is a very powerful x-ray diffraction technique. Most of the resonant scattering experiments are using the absorption edge in the lower x-ray energy range due to the availability of the radiation. However, in many cases absorption from the sample and/or environmental device becomes a major barrier to the acquisition of high quality resonant scattering data at low-energy absorption edges. There are many elements' K-edges above 40 keV that may minimize the absorption issues and be useful for the resonance scattering studies. Recently, a study using resonant scattering at very high energy K-edge on Pb/Bi distribution in $Pb_5Bi_6Se_{14}$ [5] is an excellent example that using high energy K-edges to avoid absorption problems at lower energy L-edge have provided a viable alternative route to obtain this crucial information. Another example is a recent study using high energy Ho K-edge resonant scattering to perform SAD experiment to solve the phase problem for determining the protein structure[6]. By using high energy x-ray, the radiation damage to the protein will be reduced; therefore, the phase information from the experiment will be determined more accurately for structure solution.

Charge density study of superconductor The major challenge for charge density studies is measuring highly accurate structure factors and recording them as higher resolution, higher Q, as possible. The high energy synchrotron x-rays offer the ability to minimize the systematic errors such as absorption and extinction that hinder the accuracy of the data and the opportunity to measure more high resolution data. In addition, the high photon flux from synchrotron radiation will enable the measurement of the high resolution weak reflections more accurately. The ability to obtain high quality high resolution data allows the study of heavier elements with more complicate structures. The charge density analysis of $YBa_2Cu_3O_{6.98}$ high Tc superconductor[7] is one of the important achievements of utilizing high energy x-ray for charge density studies.

Add value of the medium upgrade

We propose to upgrade a beamline to a tunable high-energy x-ray user facility dedicated for structural science researches. This facility will offer high-energy x-rays at a tunable range of 40 - 100 keV with high energy resolution and micro focusing capabilities. Various dedicated instruments provide unique opportunity to conduct experiments such as high-resolution single crystal or powder diffraction, diffuse scattering, resonance scattering, and charge density studies obtaining structural information for materials researches. The facility will also provide opportunity of using high-energy x-ray penetrating the environmental devices to study structural behavior under extreme condition such as high temperature, high pressure, magnetic or electric field with dedicated environmental devices designed and constructed for the beamline instruments.

Expected user communities

A very sizable user communities currently using high-energy x-ray to study structures of various important materials. This ever growing user base has reach and beyond the current high energy x-ray capacity at APS. This new upgraded facility will relieve the pressure of current users and allow the continue expansion of the user communities to utilize the unique capability at APS.

Enabling technology and infrastructure

The tunable high-energy x-ray facility will equip with two inline undulators, one standard APS undulator A and one special high-energy undulator, consist of two optical hutches and two experimental hutches. The first optical station will house the high-energy monochromator with a tunable energy range of 40-100 keV. Focusing optics and a high energy resolution monochromator will be located in the second optical station. The experimental stations will be located down stream of the optical stations. The first experimental station will have two permanent experimental set up, a high-resolution powder diffractometer and a diffraction set up using 2D detector that accommodate various environmental devices. An experimental set up for magnetic diffraction studies and a permanent laser heating levitator system will be located at the second experimental station.

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