X-ray Scattering

Portfolio Description

This activity supports basic research on the fundamental interactions of photons with matter to achieve an understanding of atomic, electronic, and magnetic structures and excitations and their relationships to materials properties. The main emphasis is on x-ray scattering, spectroscopy, and imaging research, primarily at major BES-supported user facilities. Instrumentation development and experimental research in ultrafast materials science, including research aimed at generating, manipulating, and detecting ultrashort and ultrahigh-peak-power electron, x-ray, and laser pulses to study ultrafast physical phenomena in materials, is an integral part of the portfolio.

Unique Aspects

The DOE history and mission have played important roles in BES' current position as the nation's steward of major x-ray facilities. As part of its stewardship, BES maintains strong fundamental research programs at these facilities in materials and related disciplines. This includes the research that has motivated part of the largest expansion of BES construction projects in recent years - the Advanced Light Source, Advanced Photon Source, the SPEAR III upgrade, and most recently, the Linac Coherent Light Source and NSLS II. The unique properties of synchrotron and free electron laser radiation – high flux and brightness, tunability, polarizability, and high spatial and temporal coherence, along with the pulsed nature of the beam – afford a wide variety of experimental techniques whose development and early application to materials science are supported by this program.

Ultrafast materials science involves time domain investigations examining, for example, the early stages of chemical reactions, bond breakage and formation in catalytic reactions, the nucleation of defects in materials that result in the degradation of their properties, and the differences in electronic configurations and transport mechanisms that govern the flow of energy in devices engineered with attention to novel nanoscale property effects. Potential applications involve the coherent control of surface chemical reactions and structures, switching and control of magnetic spin and electrical polarization domains, and non-equilibrium optical processing during material synthesis.

Relationship to Other Programs

This activity interacts closely with the research instrumentation programs supported by other federal agencies, especially in the funding of beam lines whose cost and complexity require multi-agency support. Within the various DOE programs, x-ray techniques play a key role in the investigation of materials and processes related to energy conversion and use by providing atomic- and molecular-level information on the structure of nano-particles and catalytic surfaces under in-situ realistic chemical environments and in realistic device structures. Extending into the ultrafast regime, there is the promise of expanding understanding across the full range of chemistry and materials sciences by allowing femto-second stroboscopic investigations of the earliest stages of dynamic phenomena critical to energy conversion.

The scattering program interfaces with other programs in BES dealing with scattering theory and models; soft matter and biophysical materials interrogation through techniques such as grazing incidence small angle scattering and resonant soft x-ray scattering; geosciences research through high pressure x-ray scattering techniques; and spectroscopy applied to heavy element chemistry.

Approximately one third of the recently established BES Energy Frontier Research Centers (EFRCs) benefit from the significant involvement of synchrotron x-ray researchers and their techniques. *In situ* characterization and nanoscale tracking of active materials in realistic energy conversion environments enhances the activities of several EFRCs. Advanced synchrotron characterization supports EFRCs involved in catalysis, electrical storage, superconductivity, gas separation, high pressure extreme environments, nuclear reactor materials, and photovoltaics.

Significant Accomplishments
The program supports groups that have contributed to the development of such powerful techniques as inelastic x-ray scattering, x-ray absorption structural spectroscopy, x-ray microscopy, nanoscale focused beam diffraction, time-resolved spectroscopy, and resonant x-ray scattering providing specific chemical, magnetic, and excitation contrast.

Recent accomplishments include sensitive measurements of surface segregated atomic and electronic structure in new catalysis alloys, as well as measurements of distortions in the atomic ordering resulting from the interfacial constraints on perovskite oxide films which exhibit unique magnetic and electron transport behavior. Progress in understanding the rich magnetic and electronic structure of correlated electron materials continues in terms of mapping out phase boundaries and determining the nature of the competing quantum interactions behind transitions in physical properties. Refined *in situ* techniques have become more adept at probing small samples, surfaces, and interfaces under extreme processing environments of temperature, pressure, and reactive gases. When a material is excited by light or thermal energy to non-equilibrium states, different pathways back to equilibrium often have different time scales. Recent experiments in ultrafast science have employed multiple probes with different sensitivity to various relaxation mechanisms. Fresh results are beginning to tease out the faster dynamics of electronic structure from the slower recovery of atomic motion and lattice strain.

Mission Relevance
The increasing complexity of DOE mission-relevant materials such as superconductors, semiconductors, and magnets requires ever more sophisticated scattering techniques to extract useful knowledge and to develop new theories for the behavior of these materials. X-ray scattering probes are some of the primary tools for characterizing the atomic, electronic, and magnetic structures of materials, including in extreme environments such as high pressure.

Scientific Challenges
<u>Correlated Electron Systems</u> – The effects of strong electron-electron interactions give rise to a remarkable range of anomalous behavior in condensed matter systems, producing phenomena as varied as metal-insulator transitions, colossal magnetoresistance, and high-temperature superconductivity. Such cooperative macroscopic phenomena result from the interplay of charge, orbital, spin, and lattice degrees of freedom that can arise in correlated electron behavior. Techniques such as inelastic and resonant x-ray scattering and angle resolved photo-emission, among others, have enabled scientists to unravel the crystallographic and microscopic electronic

structure of these materials.

The ultrafast excitation and exploration of dynamic pathways to metastable states provides another knob to explore the subtle energetic phase space of correlated electron materials, (much like ultra-high pressure techniques access new states along that not fully explored dimension.) Optically pumped excited states may be far from equilibrium and short lived, but the probe measurements are ultrafast and capable of capturing the elusive physics in a unique regime of matter. Recent and foreseeable advances in high-brightness x-ray sources create an unprecedented opportunity to image the primary event at nanometer spatial dimensions and ultra-fast time scales. Understanding how ultra-fast coherent radiation can manipulate condensed matter and how matter relaxes back to its unperturbed state may ultimately lead to novel materials synthesis techniques, especially at the nanoscale.

Matter Under Extreme Conditions **–** Opportunities in high pressure research address a broad range of new scientific problems involving matter compressed to multi-megabar pressures. Extreme pressures provide a fertile ground for the formation of new materials and novel physical phenomena as compression changes the chemical bonds and affinities of otherwise familiar elements and compounds. Highly collimated and intense synchrotron beams provide the ideal source for ultrafine and sensitive x-ray diffraction microprobes necessary to measure high stresses in a very small area. Scattering experiment performed in the presence of magnetic fields can be used to study materials during magnetic, structural, and superconducting phase transitions and allow researchers to segregate magnetic field effects from those observed via doping.

In situ Studies of Complex Materials – Recent advances in both sources and instrumentation have yielded gains in intensity on sample, facilitating rapid experiments and *in situ* configurations. Smaller samples can be probed with unprecedented temporal and spatial resolution, accuracy, and sensitivity under various parametric conditions. Such information aids the development of novel processing techniques and the search for new exotic materials. In-situ studies are entering the ultrafast time domain through coupling laser excited ultra-fast electronic excitations to atomic strain driven processes. There also exists the possibility of selectively studying the dynamics of such phenomena through the photo-doped creation of metastable states that would not, necessarily, be thermally accessible.

Projected Evolution

Advances in x-ray scattering and ultrafast sciences will continue to be driven by scientific opportunities presented by improved source performance and optimized instrumentation. The x-ray scattering activity will continue to fully develop the capabilities at the DOE facilities by providing support for instrumentation, technique development and research. A continuing theme in the scattering program will be the integration and support of materials preparation (especially when coupled to *in situ* investigation of materials processing) as this is a core competency that is vital to careful structural measurements related to materials properties. New investments in ultrafast science will focus on research that develops and uses radiation sources associated with BES facilities and beam lines but also includes ultra short pulse x-ray, electron beam and THz radiation probes created by conventional tabletop laser sources.