Research Activity: **Ultrafast Science and Instrumentation**

Division: Materials Sciences and Engineering

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Portfolio Description:

This activity supports basic research in condensed matter physics and materials physics using ultrafast electron, optical (terahertz to extreme ultraviolet), and x-ray techniques and sources. Basic research to observe, control, and understand material dynamics at the "atomistic" or "quantum" level, ultimately approaching the fundamental length (Angstroms) and time (attoseconds) scales of matter, as well as to develop and apply state-of-the-art laboratory scale advanced imaging techniques exploring dynamics at the ultrafast and ultrasmall, are supported. Projects aligned with unique capabilities at Office of Science facilities are of particular interest, as are ones that can leverage, or otherwise help advance related investment areas within BES. While conventional time-averaged probes such as time-integrated spectroscopy, DC transport, or thermodynamic measurements have provided significant insight into the microscopic properties of complex materials, many novel physical properties arise from excitations out of the ground state into energetically higher states by thermal, optical, or electrical means. Applications of ultrafast science based approaches often enable a material's degrees of freedom to be temporally separated, and this is a key aspect motivating this program that gives access to cause-effect relationships in complex materials exhibiting dynamic competition between, for example, charge, lattice, spin, and orbital degrees of freedom. Quite generally, this advances new understanding of ground state properties, coupling parameters, degrees of freedom influencing transport, the nature of phase transitions (reversible and irreversible), and non-equilibrium dynamics. Thus research in this activity helps underpin the most fundamental understanding and identification of detailed mechanisms responsible for the electronic, magnetic, optical, and physical behavior of materials. A focus of this program will be placed upon new materials science (e.g., nanoscience, condensed phase molecular processes, and condensed matter physics), enabled with ultrafast techniques and technologies in this rapidly developing field. Understanding complex material properties at a predictive discovery basis level for the control, design, synthesis, fabrication, and development of new materials possessing desired properties and functionalities is the broad goal of this program.

The activity also supports ultrafast research motivated by the largest BES construction projects in recent years - the Advanced Light Source (ALS), Advanced Photon Source (APS), and the Linac Coherent Light Source (LCLS). Basic Energy Sciences is a major supporter of both the research and the instrumentation at these and other facilities. With the advent of ultrafast x-ray beams produced by such fourth generation synchrotron radiation facilities, a number of unique capabilities will become possible including real time imaging of complex material dynamic behavior. Outside of specific ultrafast instrumentation, this program also supports activities for probing the detailed nature of materials using capabilities that are more subtle, sensitive, and precise. Basic Energy Sciences recognizes that many of the sophisticated tools for materials science research are expensive and complex, often limiting an individual investigator's ability to procure operate and maintain such instrumentation. In some such cases resources will be available for small to midsize, multiuser instruments primarily located at user facilities. Here, material science inspired midscale instruments and tools to innovate and advance creative research as well as promote education and training will be supported, with an emphasis on unique capabilities to enable discovery and understanding in emerging topics.

Unique Aspects:

Conventional diffraction techniques such as x-ray, neutron, and electron diffraction are all powerful probes of the equilibrium atomic arrangement of material systems, but they are unable to follow the evolution of transient nonequilibrium structural states. The promise of ultrafast techniques in materials dynamics is to elucidate the changes in the atomic and electronic configuration of materials along dynamical pathways while they are occurring. Ultrafast techniques provide a path towards investigating phenomena approaching the fundamental timescales of electronic and atomic motion, offering a unique snap-shot into the properties of cooperative condensed matter and material systems. Such investigations can often be used to complement time-integrated spectroscopic, neutron, and x-ray based studies. For example, capabilities such as ultrafast x-ray diffraction and absorption spectroscopy, electron diffraction, and terahertz and photoelectron spectroscopy are currently being developed. A key characteristic of dynamical techniques is that the system investigated is no longer in strict thermodynamic equilibrium (this deviation might be marginal or great). The material under study may be either in an excited state whose decay into other degrees of freedom is being probed, yielding information unavailable to conventional time-averaged frequency domain spectroscopies, or in a metastable state with fundamentally different physical properties. We envision a path towards an imaging capability at the frontier of ultrafast science, enabling measurements which begin to simultaneously approach the intrinsic length (Å) and time (10^{-18} s) scales of matter. The resultant detailed understanding of the properties of materials on an ultrafast time scale enables a robust rational basis for the design and development of new materials possessing desired properties and functionalities.

Relationship to Other Programs:

The Ultrafast Science and Instrumentation program is coordinated with BES activities under Mechanical Behavior and Radiation Effects, Physical Behavior, Synthesis and Processing, X-Ray and Neutron Scattering, Condensed Matter Physics, Atomic Molecular and Optical Sciences, Chemical Physics and the Nanoscience Centers; with other DOE offices, including the National Nuclear Security Agency; and with other federal agencies through the National Nanoscience Initiative (NNI). This activity also crosscuts several recent National Research Council (of the National Academies) studies assessing opportunities in science and technology disciplines where aspects of ultrafast science can provide impact and leadership. Some such topical activities are included in two monographs from the Decadal Survey of Physics: *Controlling the Quantum World – The Science of Atoms, Molecules and Photons* and *Condensed Matter and Materials Physics – The Science of the World Around Us* as well as in *Midsize Facilities - The Infrastructure for Materials Research.* Some research in the Ultrafast Science program is aimed at a fundamental understanding of the behavior of materials that underpin DOE technologies. Improving the understanding of the physics of materials on the nanoscale is technologically significant as these structures offer enhanced properties and could lead to dramatic improvements in energy generation, delivery, utilization, and conversion technologies.

Mission Relevance:

The increasing complexity of energy-relevant materials currently of interest to the Office of Energy Efficiency and Renewable Energy such as superconductors, semiconductors, and magnets, especially at the nanoscale, requires ever more sophisticated experimental tools to extract useful knowledge and to develop new theories for the behavior of these materials. Hence one primary strategic investment impact from the program is the ultimate ability to control the phases and structures of materials, including those which can advance nanoscale device development. New and powerful predictive understanding of materials would be enabled to advance the discovery and rational design of systems with new and desired functionalities. As examples, such a design toolset could accelerate the synthesis and fabrication of materials and devices that include high efficiency and ultrastable photovoltaics, advanced photonic and magnetic platforms, extreme radiation hardened materials, new sensors, cradle to grave green technologies, and even the exotic possibility of materials announcing fatigue and impending failure and/or self repair. Such discovery helps sustain our nation's innovation infrastructure in support of the America Competes Act. The program is also relevant to the National Nuclear Security Administration in its activity on the behavior of matter in extreme environments, especially high strain rates, pressures, and temperatures, as well as in nonproliferation through research facilitating discovery of new sensor modalities.

Scientific Challenges:

Our ability to investigate the fundamental properties of materials, for the most part, has been outpaced by the facility with which these materials can be grown. Aside from scanning nanoprobes such as the scanning tunneling microscope and the atomic force microscope (both transformational capabilities first enabling ultrasmall scale imaging), studies of nanosystems such as wires, tubes, and dots have been carried out on ensembles exhibiting a relatively wide distribution of lengths, widths, chiralities, etc. As a result, a great deal of the detailed bulk and surface electronic structure is ultimately lost in the average values such measurements produce. A Scientific Grand Challenge in this topical area is to fully exploit the promise of new materials, especially at the nanoscale, which requires an integrated and innovative design, fabrication, measurement, modeling, and theoretical effort to achieve a predictive understanding of such complex materials. Developing the ability to better understand, predict, and use the significant differences of energy states and exotic scaling laws due to surface to volume ratio effects, together with discovering how to design novel electromagnetic response(s) with enhanced transport properties, is required to accelerate the use and integration of nanostructured materials. Research to discover materials possessing emergent properties could positively impact many applications including, superconductivity, sensors, transducers, magnetic and microelectronic devices, photovoltaics, and photonic based devices using bandgap engineered structures or metamaterials.

Coherent Control - A major theme in atomic and molecular science is: If we now understand the basics of how atoms and molecules work, can we manipulate and control them in useful ways? The control of matter on the sub-nm, sub-fs, distance- and time-scales explores the limits of fundamental atomic and molecular processes, as well as the limits of optical technology. Since atoms and molecules interact with their environment entirely by means of electromagnetic fields, the preceding question specifically asks: Is it possible to "design" a pulse of light that, when incident on an atom or molecule, can precisely control its evolution in any desired way? During the past decade, as laser technology has progressed, light pulses can now be manipulated on a cycle-by-cycle basis, and the topic of "coherent control" of atoms and molecules has drawn increasing interest. Control of coherent behavior in condensed matter systems has been difficult primarily due to the fast dephasing times of electronic or excitonic states. Advances in ultrafast lasers may soon enable the application of coherent quantum control techniques, already demonstrated in gas phase atoms and molecules, to more complex condensed matter systems. From a fundamental scientific perspective, entirely new physics emerges when the quantum states of solids are coupled and manipulated by intense optical fields. Our understanding of tightly coupled light-matter systems is, however, hampered by our lack of in-situ microscopic probes. Recent and foreseeable advances in high-brightness x-ray sources create an unprecedented opportunity to micro-probe the primary event in how optical radiation manipulates matter. When we ask a light pulse to manipulate matter it accomplishes this task by driving atomic scale distortions in a material's valence charge density. These initial microscopic distortions then propagate out to drive subsequent material evolution. To date there has been no way to directly probe these transient light induced distortions to the valence charge. New experimental capabilities are required to provide fundamental insights important for learning how coherent quantum control techniques might be used to synthesis novel materials.

Studies of Correlated and Complex Materials – One of the most important aspects of contemporary condensed matter physics involves understanding strong Coulomb interactions between the large numbers of electrons in a solid. Electronic correlations lead to the emergence of unique materials properties, such as superconductivity, magnetism, heavy Fermion behavior, Bose-Einstein condensation, the formation of excitonic gases, metal-insulator transitions, and the integer and fractional Quantum Hall effects. Such materials often exhibit competition between the charge, lattice, spin, and orbital degrees of freedom, whose cause-effect relationships are difficult to ascertain. An important characteristic of these materials is the existence of several competing states, as exemplified by their complicated phase diagrams. Such electronic complexity has consequences for applications of correlated electronic materials, because not only charge (semiconducting electronic), or charge and spin (spintronics) are of relevance, but in addition the lattice and orbital degrees of freedom are active, leading to giant responses to small perturbations. Moreover, several metallic and insulating phases compete, increasing the potential for novel behavior. The electronic correlations lead to unique materials and device properties such as high-temperature superconductivity, colossal magneto-resistance, coupled electric and magnetic functionality, and large thermopower. Potential real world applications include dissipationless wires and devices from high temperature superconductors, superior thermoelectrics, enhanced memory with states stored in magnetic and electric polarization made from multiferroic manganites, and smaller, faster, more efficient electronics. A major challenge in correlated materials is to realize this enormous potential in functionality to discover, understand, and develop new multifunctional materials and devices which perform more functions, more effectively than conventional semiconductor devices. To succeed in this challenge a more complete understanding of the underlying mechanisms and phenomena in correlated materials is required to ultimately control this complexity to achieve desired functionality. The discovery of high-TC superconductivity in particular was a watershed event, leading to dramatic experimental and theoretical advances in the field of correlated-electron systems. Yet, after one of the largest research efforts ever in physics, involving hundreds of scientists, even basic properties of the cuprate high temperature superconductors, such as the pairing mechanism, linear resistivity, and pseudogap phase, are still only poorly understood. In the past, it was expected that suitably modified theories of ordinary metals would explain the unusual properties of the cuprate's normal state, but a true understanding of this complex phenomenon remains elusive and certainly will not be incremental. Just as the historical discovery of antiferromagnetism occurred in 1949, only with the advent of neutron scattering, new and improved tools must be developed to probe "hidden" correlations in these complex materials. Ultrafast spectroscopy, consequently, is playing an ever increasing role to provide insight into cause-effect relationships associated with many-body interactions in complex materials. Quite generally, important insight into ground state properties, coupling parameters, degrees of freedom influencing quasiparticle transport, the nature of phase transitions, and nonequilibrium dynamics can be obtained as ultrafast experiments naturally probe the dynamics of systems with interacting quantum degrees of freedom. From a theoretical point of view, calculating even the ground state of correlated systems, for example the simple Hubbard model of interacting electron systems, has proven essentially intractable. It would appear that computing the full quantum dynamics of correlated and complex systems far from equilibrium would be even further out of reach. The natural impulse when faced with such a challenge is to make drastic approximations at the outset, for example to treat a problem using mean field theory, or to recognize that phonons are slow compared to electrons and to treat them semiclassically or in the adiabatic approximation. In a small molecule, where electronic excitation energies are large compared to phonon energies, the adiabatic approximation, or the assumption that the total wavefunction factors into a product of an electron piece and a phonon piece, is often appropriate. However, when the molecule becomes large, for example a long carbon nanotube or a full 3D solid, the electronic excitation energies become arbitrarily small and dense, often much smaller than a typical phonon energy. It would seem that the adiabatic approximation may fail in this regime, as in fact it does. Efforts to bridge experiment, simulation, and theory, especially leveraging information accessible at the ultrafast and ultrasmall scales, will help to fill this gap.

Studies of Systems Far From Equilibrium – Dynamic material behavior that would benefit from study with high temporal resolution, especially high-fidelity, laboratory scale, single-shot approaches, include advanced diffraction and measurement techniques to identify and quantify ultrafast strain-driven processes including phonon dynamics, phase transformations, plasticity, melting and solidification, deformation twinning, solid-state chemical reactions, radiation damage, and shock propagation processes. Such observations will allow the direct measurement of certain quantities used in models and would help serve to validate various materials simulations at atomistic scales. A broader challenge is to linking across length and time scales to achieve multi-scale understanding requiring computational and experimental coordinated approaches to define materials from atomistic to bulk scales, especially under conditions far from equilibrium. For validation, the materials community needs to search for experimental techniques and modeling approaches that link scales, where directly connecting experimental and simulation spatial and temporal scales remains a critical and pivotal need. As an example, predictions of phase transformation kinetics active during shock and dynamic deformation events will ultimately follow from prediction of the atomic motions during phase transformations. Similarly, surface reactions leading to corrosion and radiolysis effects during radiation damage require linking of spatial and temporal scales. Probing the ultrafast and ultrasmall enables a direct means to benchmark predictive models at the atomistic level, as the preferred approach is to validate such models at each length scale, from the electronic and atomic-level dynamics in single crystals, to the mesoscopic interactions between grains, and finally to the bulk polycrystalline response of engineering samples. Since predictability flows from microscopic models to higher length and time scales, experimental validation at the micro-scale is required to confidently predict material properties under rapidly varying and extreme conditions at the engineering scale, where the design and simulation of a system or device is required for engineering applications.

Funding Summary: N/A

This activity is part of the Neutron and X-Ray Scattering Research Area.

Projected Evolution:

Advances in ultrafast science will be driven by the scientific opportunities presented by improved source performance and instrumentation optimized to take advantage of that performance. The ultrafast science activity will support emerging capabilities at the DOE facilities by providing instrumentation and research support. A challenge to the program is to identify and provide support for the researchers and instruments to keep the program and facilities at the forefront of science in this technologically rapidly advancing field.