**LA-UR 09-07343**

# Research Frontiers and Capability Gaps for Controlling and Designing Functional Materials


**Executive Summary:**

*Scientific Challenge - Control Science.*

Most advances in materials have focused on an observation approach that involves detailed characterization of a new material followed by integration of the material into applications based on the material properties. These properties are further tuned by changes in the processing conditions via a trial and error effort. The notion of predicting the properties of a material and systematically controlling the composition, defects and interfaces is typically viewed as something far off in the future. One exception that stands out is in the computer industry where the systematic characterization, and controlled manipulation of both purity and interfaces at ever increasingly finer resolution has lead to material advances and performance capabilities that not long ago would have been viewed by most people as impossible.

Areas with future scientific challenges requiring the same control approach to realize solutions include:

- Sustainable energy future
  - efficient electrical grid -superconductivity
  - energy conversion - photovolatics
  - energy storage - batteries, capacitors and electrosynthesis
  - solid state lighting
- Information storage and processing
- Advanced optical materials

At the heart of all of the areas is the grand challenge of predictive design of materials to control emergent behavior. To design new materials with predicted properties we must transition away from the historical observational approach to a controlled approach where theory, synthesis and characterization are effectively integrated. The desired property may arise from a variety of interactions involving spin, phonons or electrons. The ability to predict properties in such advanced functional materials requires a detailed theoretical understanding of the correlated interactions between electrons, phonons, spin and charge that lead to the remarkable properties such as colossal magnetoresistance, superconductivity, multiferroicity, and electronic phase separation, and ferroelectricity. Advances in theory will rely on the combination of a synthetic ability to systematically control composition, defects and interfaces and characterization tools to watch how defects and interfaces evolve. Integrating these activities will provide the framework for control science to test and verify new theoretical codes designed to tackle the challenges of highly correlated systems. The focus on interfaces and defects is especially crucial in the area of emergent phenomenon where interfaces between materials with dissimilar properties can lead to the emergence of entirely new properties. The transition to control science will allow for the accelerated design and discovery of new materials with predicted properties.

Currently we rely on an Edisonian approach to materials discovery by serendipity or intuition. Great advances have been made by this approach, but most materials still have a performance or lifetime that falls far short of theoretical limits. There are so many potential combinations of materials that a continued Edisonian approach is unlikely to lead to dramatic improvements rapidly. Synthetic efforts need to be guided by predictive theory. Improved

theoretical approaches are needed with an emphasis on first principle theories designed to predict material properties without requiring material measurements as parameters. Synthesis must be flexible enough to make a wide range of possible theoretical materials and have the capability to control composition, defects and interfaces systematically. Characterization techniques must address the challenges of 3-D non-destructive imaging and dynamic characterization of solid state materials with sub grain size resolution on the time scale of electron-electron and electron-phonon interactions.

The future promises to grow increasing complex as nanoarchitectures, become integrated into composite structures. These new composite materials offer the promise of multi functional capabilities not possible with in single component material, but bring with them a tremendous challenge to manipulate and control interfaces and defects. Simultaneously accelerating materials discovery and advancing 'interface engineering' hold great promise for controlling and designing next-generation functional materials.

*Decadal Challenges in Theory, Synthesis and Characterization*

The primary goal of the workshop was to identify decadal type challenges in each of the sessions. These challenges were then grouped into thematic areas of theory/ modeling/ visualization, synthesis, and characterization. Decadal challenges identified for accelerating materials discovery include:

**Theory/Modeling/Visualization**

- Theories that treat strong correlation dynamically
- Modeling and theory that includes full electronic, ionic relaxation for large systems with interfaces
- Theories that can describe defect interactions at an interface and predict property changes – issue of multiple scales and boundaries
- Visualization -- a large data visualization capability for 3-D tomography data
- Predicting materials properties via learning from database of models/experiments

**Synthesis**

- Rapid synthetic exploration of new materials coupled to rapid characterization
- Synthetic control and characterization of defects, phases and atomic composition to 1ppm
- Synthetic control and characterization of interfaces to 10's of nm in plane and 1 nm perpendicular
- High quality crystalline materials to benchmark developing theories that cannot yet handle many different boundaries
- Control of strain and phase to generate metastable states
- Design of bulk materials with nano – micron inhomogeneities

**Characterization**

- In situ characterization during nucleation and growth of materials
- Characterization of buried interfaces to study electron- electron, electron-phonon and phonon-phonon interactions
- Characterization in extreme environments including magnetic fields to 20 T, temperatures of 10 mK to 300K, and pressures to 20 GPa to tune spin charge interactions
- Characterization of the evolution of defects and interfaces during synthesis, in response to extreme environments and during function of integrated devices

These challenges must be attacked in a concerted manner. A modern materials discovery center must integrate capabilities in each of these areas in order to have the ability to control composition, defects and interfaces in a systematic manner over a variety of length scales. Control of composition, defects and interfaces requires a theory component able to integrate many length scales, a highly flexible synthetic capability that can both rapidly explore a set of new materials and make selected materials with extremely high purity and near monolayer interfacial control, characterization facilities with both the resolution to observe defects and interfaces and the temporal capability to watch them evolve in time in response to different environments, and a close coupling of characterization and synthesis during the critical nucleation and growth process.

**Key crosscutting topics that arise in all three areas are**

- *Interfaces and defects*
- *Control over multiple length scales from nano to micron*
- *In situ techniques to characterize how properties emerge and evolve*

*Interfaces and defects*

The fascinating properties that emerge in solid state materials begin based on elemental composition that can lead to magnetism or superconductivity but these properties are dramatically altered by defects/interfaces that can lead to quenching of excited states or vortex pinning in superconductivity or entirely new physics as competing order parameters come together at interfaces with different materials in intimate contact such as colossal magnetoresistance. The functionality often comes from controlling or tailoring the large response of a material to a small perturbation as a result of competition between nearly degenerate ground states involving coupled charge, spin, and lattice degrees of freedom. Critical to understanding the emergent phenomena is the ability to make single crystal materials with controlled interfaces and as part of composite materials. The US currently lacks a crystal growth resource either in industry or academics and most US researchers buy crystals from other countries such as Japan. A national crystal growth facility is needed for the US to maintain its leadership in materials discovery and subsequent technical advances. High quality single crystals are critical to the understanding of emergent properties that enable many fields of science such superconductivity, ferroics, colossal magneto resistance, and potential future materials base on orbitronics for faster processing. Traditional material discovery centers of the past such as Bell Labs and IBM have completely disappeared or down sized tremendously. This change has the potential to put the US at a disadvantage technologically in the future. The US has always been at the forefront of materials discovery in the past with strong leadership in previous society changing technological advances such as the discovery and technology developments in polymers, semiconductors, and superconductors. Today the landscape is changing as many other developed countries are beginning to devote national resources to extensive research in materials. Workshop participants unanimously concluded that there was a consensus need across all of the areas for national materials facilities in the United States.

*Control over multiple length scales from nano to micron*

Nanotechnology has progressed tremendously over the last decade, in part, through the efforts of the BES nanocenters. These nanocenters combined with university and international efforts have lead to a greater understanding of nanoscale phenomena and an enhanced ability to

synthesize nanowires, and nanoparticles that includes control over dimensions as well as core shell structures in which the composition of the layers is varied to tailor the properties or add multiple functionality. These nanoscale phenomena and materials will play an important role in devices of the future. The next grand challenge is to understand how nanoscale features in a material grow and evolve over micron scales to ultimately affect bulk performance in a material. For example, the nanoscale features that control vortex pinning are known to have significant effects on the meter long superconducting tape. Advanced solar cells are currently being designed with nanoscale architectures designed to efficiently move electrons and holes in opposite directions over micron length scales. Such future devices will involve the manipulation and placement of nanoscale features in an overall architecture with micron and greater length scales.

Hybrid nanostructured materials will provide the key to the future design of multifunctional materials with emergent properties "by design". For example, areas of particular opportunity include energy harvesting and storage, including photovoltaic and photochemical energy conversion, thermoelectric conversion of heat to electricity and visa versa, piezoelectric conversion between strain and electric fields, rapid cycling of battery electrodes to high capacities, electrostatic capacitive charge storage to high energy densities, and electrochemical energy storage and conversion in fuel cells. The control of energy transfer is a key aspect to each of these areas. The ability to exploit the high local fields, large surface areas, enhanced chemical reactivity, short length scales, quantum confinement, local strains, enhanced strength, and the stability of nanoscale arrays in nanostructured materials is opening up new opportunities for advances. An improvement of a factor of 2 to 3 in any of these areas would have dramatic consequences. While incremental increases will almost certainly be achieved over the next few years, an ability to achieve such larger improvements and to do this "by design" in complete devices (rather than empirically) with new understanding of how to control energy transfer by nanoscale structuring is a 5 to 10 year goal requiring a control science approach. The key challenges are the controlled synthesis and fabrication of heterogeneous combinations of materials at the nanoscale and the understanding, through characterization and modeling, of the design of materials performance.

*In situ techniques to characterize how properties emerge and evolve*

Properties in a material both emerge during the nucleation and growth of synthesis and evolve overtime in a real device as it is often subjected to extremes of cycling, temperature, pressure, electromagnetic fields, or chemical environment. In control science, effective synthesis relies on *in situ* characterization techniques to understand what processing parameters control a material microstructure. This can be clearly seen in the case of thin film deposition where RHEED coupled to high vacuum deposition techniques has allowed for the precise growth of thin films with real time feedback. The result in the case of molecular beam epitaxy (MBE) is a digital growth process that can precisely deposit multiple layers of different compositions. This type of synthetic control has helped to advance our understanding of properties such as colossal magnetoresistance (CMR). Fundamental characterization of the nucleation and growth of nanoclusters will also play an important role in furthering the understanding of how and when properties emerge. On the decadal scale, a grand challenge for emergent phenomena will be the integration of multiple synthetic techniques with the same type of *in situ* characterization to control and build up hybrid nano materials. These will include both layered materials and new materials with nanoscale features embedded into larger crystalline matrixes. Such materials will

require non-destructive 3-D *in situ* imaging of microstructure and composition in order to obtain the necessary synthetic control.

Solving the future problems in energy and information technology will be greatly accelerated by designing functional materials based on advanced theoretical models of how interfaces and defects affect material performance across multiple length scales. For these new solutions to be successful the materials must also perform with long lifetimes compete effectively in the market place and to avoid future waste issues. In practice such functional materials are often exposed to some type of extreme condition during their use that eventually results in failure. To extend lifetimes and minimize failure or design self healing materials we will need to understand how these failure mechanisms begin at the atomic scale and eventual manifest themselves at the micron and bulk scale. Such understanding will be greatly accelerated by watching the defects and interfaces as they evolve in the extreme conditions and simultaneously measuring the material performance. In order to achieve this we must be able to non-destructively characterize microstructure and composition while the material is exposed to the extreme conditions. Multi-probe experiments in extreme environments, and the non-destructive 3-D characterization to enable multiple measurements on a single sample represent significant decadal challenges.

*Workshop Goals*

The initial charge for the workshop was to discuss the grand challenges in correlated materials and emergent properties. The sessions were organized to include issues in the areas of synthesis, characterization and theory. The intent was to evaluate the scientific grand challenges of the future and define the state-of-the art and future expectations of the capabilities, developments and innovation in the next 5-10 years with an emphasis how a new facility could be designed to facilitate and accelerate materials discovery. The areas of focus included (a) emergent functional behavior at nanoscale in condensed matter systems and (b) the collective, coherent functional response in single crystal and composite materials.

The workshop was organized into 8 sessions including application of electronic materials, oxide interfaces, high Tc superconductivity/heavy fermion physics, time resolved aspects of functionality, size dependence:quantum and nanoscale effects, ferroics, computational tools and clusters. Participants included both LANL and external speakers with 18 speakers outside LANL from a variety of academic facilities and national laboratories including Harvard, MIT, University of Minnesota, UCSB, UCD, ANL, LBL, MIT, Kyoto University, Rutgers, UCSD, University of Maryland, University of Michigan, Penn State, and Iowa State.