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**Research Frontiers and Capability Gaps for  
Controlling and Designing Functional Materials  
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## 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 led 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 - photovoltaics
  - 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 from observation 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.

Prediction of emergent properties will not happen overnight. This is a multi decadal challenge that will be significantly accelerated by effective integration between theory, synthesis and characterization. Initial advances in theory will rely on databases of materials properties in which parameters such as composition and architecture are systematically varied. Such databases will provide the statistical basis set for new theoretical directions in material property predictions and allow new theoretical codes to be effectively benchmarked for validity. Rapid synthesis that can explore a large suite of possible materials combined with characterization techniques that can effectively probe both structure and material properties will play an essential role to develop such databases. To be effective the material databases must be designed such that the compositional changes and the materials properties to be measured match well with the input and outputs of the theoretical codes. The ability to predict material properties fundamentally means that we must connect theoretical codes across scales as one starts with the atomic composition and adds defects and interfaces to ultimately predict the bulk properties of interest such as superconductivity, band gaps, magnetism, and electron transport. Likewise, synthetic techniques must bridge scales from small well defined single crystals to bulk materials that incorporate defects and interfaces and characterization tools must be able to determine both long range structure and local atomic in homogeneities.

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 clustered 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 crystals and crystalline composites to benchmark theories
- 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
- Non destructive 3-D characterization
- 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*

Across all three areas of theory, synthesis and characterization there are crosscutting topics that emerge as key areas that must be addressed to solve the decadal challenges:

- **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 as superconductivity, ferroics, colossal magneto resistance, and potential future materials based 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 led 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 strongly correlated electron materials, application of electronic materials and oxide interfaces, time resolved aspects of functionality, size dependence: quantum and nanoscale effects, ferroics, clusters and computational tools. 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.

## Detailed breakout results:

### *Applications of Electronic Materials and Oxide Interfaces*

*Chairs: Michael Fitzsimmons (LANL), Darryl Smith (LANL)*

*Speakers: Christopher Hammel (Ohio State), Paul Ruden (U. Minn.), Christopher Palmstrøm (UCSB), D. Basov (UCSD), A. Bhattacharya (ANL), E.D. Dahlberg (UMN), W. Pickett (UCD), I.K. Schuller (UCSD)*

Composite materials have long been important constituents of commercial products; however, with improvements in the synthesis and characterization of materials enabling nanometer-control of length-scales, the importance of interfaces becomes preeminent. Materials with nanometer structures, or so called nanoscale materials, necessarily mean that interfaces play a decisive role in determining the response of composites to their environment. Many materials systems are composites of materials with dissimilar properties. The resultant behavior of the composite can be simple leverage sums of its constituents, e.g., hardness and toughness of ceramic-metal composites, or produce new behavior due to competing order parameters.<sup>1</sup> In comparison to interfaces in metals and semiconductors, the electronic (spin and orbital) and magnetic character of the cation and anions that comprise oxide interfaces lead to complex behavior. For example, in ferroelectrics, interfaces are sinks for defects that pin the motion of ferroelectric domains,<sup>2</sup> affect the anisotropy of ferroelectricity through interfacial strain, or, as sites for discontinuity in electric polarization, may lead to improper ferroelectricity through proximity effects. Oxide interfaces also exhibit some of the largest figures-of-merit for thermoelectric response,<sup>3</sup> which is believed to be a consequence of high charge conductivity and low thermal conductivity. The structural perfection of interfaces is believed to play a crucial role in the performance of acoustic cavities<sup>4</sup> and phonon lasers,<sup>5</sup> which provide new opportunities to couple photons, phonons and electrons. Some of the most promising applications of materials that couple electric fields and magnetism involve composites of materials that utilize exchange bias across an interface to amplify magnetoferroic response.<sup>6</sup> In all these examples, the novel behavior is often *attributed* to interfaces, but the actual role of interfaces is often not actually known, e.g., the phonon dispersion of an interface has never been measured.

Perhaps the most dramatic example of how the future might be influenced by oxide interfaces lies in the potential to use the orbital spin degree of freedom to convey (or manage) information. The unique opportunity of oxide electronics vis à vis semiconductor electronics is the degeneracy of orbital occupation that could be controlled using electric field, strain etc. with unprecedented speed. This arises from the fact that the orbiton frequency—between 10 and 100 THz—is much higher than the electron spin precession frequency of 1-100 GHz.<sup>7</sup> Presently, off-the-shelf computers have clock speeds around 3 GHz. If clock speeds increase at a rate that double every 18 months (Moore's law), then in ten years' time, clock speeds will approach ~300 GHz. The higher speeds might be realized using the concepts of spintronics that are presently

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<sup>1</sup> R.E. Newnham et al., *Mat. Res. Bull.* **13** 525 (1978).

<sup>2</sup> M. Dawber et al., *J. Phys. : Condens. Matter* **20** 264015 (2008).

<sup>3</sup> H. Ohta et al., *Nature Mater.* **6** 129 (2007).

<sup>4</sup> J. Chen and J.B. Khurgin, *Appl. Phys. Lett.* **81** 4742 (2002).

<sup>5</sup> P.A. Fokker et al., *Phys. Rev. B* **55** 2925 (1997).

<sup>6</sup> N. Spaldin and R. Ramesh, *MRS Bull.* **33** 1047 (2008).

<sup>7</sup> E. Dagoot and Y. Tokura, *Mat. Res. Bull.* **33** 1037 (2008).

being developed (and indeed are in place for certain products). Yet, what about the time beyond 10 years—the time frame of MaRIE? To achieve clock speeds in the THz regime, orbital engineering or orbitronics, appears attractive. Thus our view is that rather than focusing on the transitional technology of spintronics, MaRIE should focus on the dramatically new technology of orbitronics—a field that would require a radical change from semiconductor to oxide electronic technology (since the orbital occupancy of semiconductors is much less easily manipulated compared to oxides).

#### *Making, Measuring, Modeling Materials*

New materials can be discovered in one of two ways—either through serendipity or through use of predictive methods. Regrettably, in practice, interesting materials have been discovered primarily through serendipity. This argues for new approaches that rely on “serendipity-by-design”. Serendipity-by-design endeavors to discover interesting materials by synthesizing large numbers of samples using parallel synthesis methods (as opposed to serial methods of the past), and applying parallel characterization methods (discussed later) to discover the interesting ones. An example of the approach is to grow many chemically different interfaces using phase spread alloys<sup>8</sup>—an alloy obtained from co-deposition of materials from two or more sources. The composition of the deposited material—covering the entire phase diagram—varies in a controlled fashion across the substrate. Combining parallel synthesis and characterization approaches, uninteresting compositions can be rapidly discarded to discover those that are interesting. Once discovered, traditional serial growth techniques can be employed to optimize sample structures.

One such traditional approach is molecular beam epitaxy (MBE). Typically MBE yields compositionally disordered films (much like those grown using bulk techniques) with a minimum of defect structures. In many cases, compositional disorder also means strain disorder, since cations in oxide films are not necessarily the same size. However, a new form of MBE, called digital growth MBE (DG-MBE),<sup>9</sup> offers a dramatic new opportunity to grow compositionally ordered films (and interfaces) with the same average composition of traditional MBE without the variance in strain disorder. For example, in the case of  $\text{La}_{0.5}\text{Sr}_{0.5}\text{MnO}_3$ , a digitally grown structure consists of alternating layers of  $\text{LaMnO}_3$  and  $\text{SrMnO}_3$ . Since the thickness of each layer is less than the critical thickness for strain relaxation, the strain is uniform; thus, its variance is zero throughout the film.

The ability to synthesize an ordered film with zero strain variance can have dramatic consequences on materials properties. In the case of an LSMO film, an A-type antiferromagnet, the Néel temperature of the chemically ordered film is 65 K greater than the chemically disordered film.<sup>10</sup> It is no small wonder that modeling would have difficulty predicting the Néel temperature of a disordered film. In fact, synthesis and characterization of ordered films may reduce the complexity of modeling (discussed later). The ability to reduce strain and compositional variation to zero will enable us to push the average composition of samples to the extreme limits of the phase diagram.

#### **Grand challenges in making:**

- (1) Intelligent approaches (e.g., parallel growth methods, information-science-guided methods) towards materials discovery that seek to enhance chances of serendipitous discovery. Having discovered an interesting system...

<sup>8</sup> I.K. Schuller, private communication.

<sup>9</sup> H. Luo et al., *J. of Electronic Mater.* **22** 467 (1993).

<sup>10</sup> A. Bhattacharya, private communication.

- (2) Growth of the highest quality samples (e.g., using DG-MBE) with a minimum of defects, e.g., less than one part in  $10^6$  (dictated by the size of a wave function in an oxide),
- (3) zero or alternatively well controlled variation (much less than 0.1%) of strain (0.1% strain can change anisotropy, etc.),
- (4) control of rumpling (to length scales less than a dimension of the wave function of an electron) and reconstruction of interfaces, and
- (5) zero or alternatively well controlled variation (much less than 0.2%) of composition (dictated by the desire to be as close as possible to the transition between different phases) that is comparable to what is obtained by the semiconductor industry.
- (6) (2)-(5) need to be applied to grow samples with macroscopic dimensions that are inhomogeneous on length scales of tens of nanometers. In other words, we are not simply interested in thin films and planar interfaces, but arrays of stripes, dots, etc. in two-dimensions that may also be stacked in three-dimensions. To achieve this goal, improvements in the polydispersity (much less than 10%) of bottoms-up (self-assembly) approaches and the throughput (goal of  $1\text{ cm}^2$  samples) of top-down (lithography) approaches are required.

#### *Making. Measuring. Modeling Materials*

Strategies to characterize materials fall into two classes. The first “parallel characterization” aims to quickly discard samples that do not exhibit a property of interest. For example, measurements of temperature dependent microwave absorption from large samples (even ones that are inhomogeneous on nanometer length scales) can identify those that are not superconducting.<sup>8</sup> These are discarded. Specialized techniques—the 2<sup>nd</sup> of the two classes—are then brought to bear on the remaining samples.

One goal of MaRIE should be to develop tools to rapidly identify samples as interesting or not according to the property du jour, e.g., superconductivity, ferroelectricity, thermoelectricity, etc. A second goal should be to refine specialized characterization techniques aimed at determining atomic structures with sub nanometer precision and the static and dynamic (DC to 100+THz) properties (electronic, magnetic, acoustic etc.) of individual interfaces. This implies that specialized techniques must have the ability to distinguish the structure and properties of the interface from the adjoining material. In other words, the probes must be spatially discerning (to 1-nm length scales, or its equivalent in reciprocal space). In the time domain, electronic transitions (fs) are of interest. Measurements of the dispersion (wave vector) in the response of the system to excitations are needed. Indeed, many bulk structures (and presumably interface structures) are energetically equivalent (or nearly so) at 0 K, and it is primarily the entropy of the structure at higher temperature (introduced through phonons) that determines the most stable structure. To date, there are no experimental measurements of phonon dispersion for interfaces. Such measurements are crucial to understanding electron-phonon and photon-phonon coupling found for multiferroic, ferroelectric, thermoelectric and acoustic phenomena.

Many of the requirements for characterization we seek (imaging, spatial and dynamic specificity) can be fulfilled using the coherent and pulsed characteristics of a free electron laser. An outstanding challenge is to tailor the x-ray wavelength from the FEL to provide the element specificity presently obtained using x-ray resonance techniques as practiced today at synchrotrons.

Additional emphasis on (or thought about) computing is warranted. Some of the characterization techniques envisioned for MaRIE (e.g., reflectometry, resonance probes that are

highly non-linear, etc.) require computation of recursive algorithms. Recursive algorithms are not amenable to parallel computation. The speed of the Roadrunner is due to its massively parallel architecture.

**Grand challenges in measuring:**

- (1) Develop tools to rapidly distinguish samples exhibiting interesting properties from the many that do not.
- (2) Develop techniques to measure vacancy concentration (1 ppm) and stoichiometry with tens of nanometer lateral resolution and 1 nm resolution perpendicular to an interface.
- (3) Develop techniques to measure magnetization from ordering of 10's of orbitals or spins.
- (4) Develop *in situ* characterization of samples during growth that include quantitative measurement of vacancy concentration, stoichiometry and atomic structure of an interface.
- (5) Develop techniques to measure the dispersion of phonons at interfaces in  $\mu$ g samples.
- (6) Develop techniques to measure orbital occupancy with 10 nm lateral and 1 nm perpendicular resolution.
- (7) All of the above techniques need to be performed in high (20 T) magnetic fields, low (10's mK) temperature and applied stress (producing  $\sim$ 1% strain) along prescribed directions preferably at the same time! (For thin films and interfaces, application of stress is the interesting control parameter as opposed to pressure for bulk materials.)

*Making Measuring Modeling Materials*

The modeling community can point to several successes where modeling has provided reasonably compelling explanations for interfacial phenomena, e.g., conductivity at the LAO/STO interface, collapse of magnetism as a driver of the Mott transition in MnO, and origin of the metal-insulator transition in VO<sub>2</sub>. Beginning with the framework of interatomic potentials, modeling can predict lattice relaxations in response to electrostatic (i.e., polar) discontinuities that yield band structures from which the conductivity (though not yet magnetism?) of interfaces can be predicted. A specific example in hand is the case of the predicted shift of surface states towards the Fermi level in LAO/STO bilayers that increases with LAO thickness in response to an increasingly strengthening Coulombic potential. Eventually, the LAO layer becomes conducting.

Modeling will be a significant benefactor of improved sample growth proposed as a MaRIE goal. The ability to grow compositionally ordered films with little or no strain variance should greatly increase the relevance of model calculations (model assumptions of perfection in imperfect samples—to make modeling tractable—can be realized with better samples). Modeling will also benefit from faster computers; however, it should be noted that for some instances, particularly in highly correlated systems, the calculations may not benefit from highly parallel computing. For example, the landscape of magnetic and insulating texture—a consequence of competing order parameters—is highly correlated.<sup>11</sup> The very existence of a magnetic/conducting region in one part of the sample stabilizes non-magnetic/insulating regions in other parts. Thus, in these instances it is not yet clear that parallel computing is necessarily advantageous.

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<sup>11</sup> K.H. Ahn et al., Nature **428** 401 (2004).

**Grand challenges in modeling:**

- (1) Full electronic, ionic relaxation (including rumpling, rotating, reconstructing) for large systems modeling isolated interfaces, and for thin slabs where interfaces are still coupled to the surface.
- (2) To treat isolated defects near interfaces realistically [including relaxation as in (1)] will require at least many hundred atoms, with codes that should include the developments mentioned below.
- (3) Strong correlation treated dynamically, since *ab initio* calculations do not properly incorporate strong electron-electron interaction.
- (4) Full charge self-consistency including correlations.
- (5) Include atomic dynamics (phonon dispersion) in (1)-(4).
- (6) Obtain and include entropic effects, especially spin entropy. Properly done, would require dynamic quantum spins; less precisely, statistical averages of classical spins.
- (7) Calculate electron transport, thermodynamics. Key issues to understand include defect energetics and kinetics, many-body physics, transport in and about interfaces in systems that are spatially non-uniform.
- (8) Predict critical temperatures, e.g., for superconductivity, magnetism, etc.
- (9) Computational materials design particularly pertaining to interfaces. Develop physically based guidelines as well as computational algorithms.
- (10) Predict occupancy of orbitals at interfaces. [This is implicit in (1), (3), and (4) above.]
- (11) Include and/or refine models, incorporating experimental information on atomic structures and dynamical effects at interfaces.

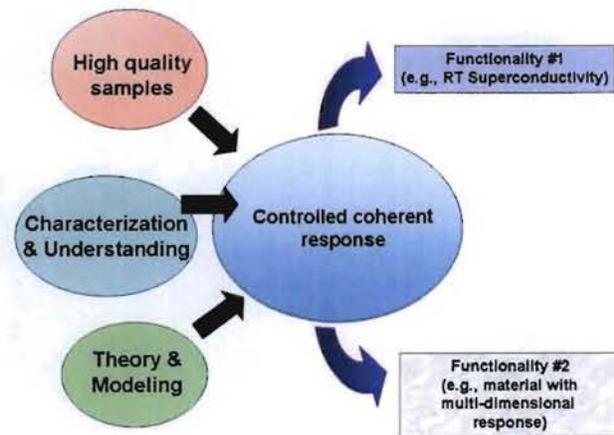
*Strongly Correlated Electron Materials*

*Chair: Eric Bauer (LANL)*

*Speakers: Sasha Balatsky (LANL), Eric Hudson (MIT), Takasada Shibauchi (Kyoto Univ.), Joe Thompson (LANL), Rich Martin (LANL)*

Functionality comes from controlling or tailoring the large response of a material to a small perturbation. In strongly correlated *d*- and *f*-electron materials, new states of matter and phenomena emerge from the competition of nearly degenerate ground states involving coupled charge, spin, and lattice degrees of freedom. These new and often unexpected states and phenomena are the (large) response to competing interactions of the system. Control of these responses in correlated matter, which are inherently quantum mechanically derived, defines an important decadal challenge for the scientific community and MaRIE. Unconventional superconductivity formed from an antiferromagnetic state in cuprate, heavy fermion, and organic materials is a prototypical example of a (phase) coherent quantum mechanical response to the underlying nearly degenerate interactions, while charge stripes in LSCO and the (possible *d*-density wave) pseudogap in both electron- and hole-doped cuprates are examples of coupled quantum states out of which superconductivity emerges. The control of even one (or a select few) of these collective and emergent quantum phenomena is a promising route to new functionality and could lead to the development of room temperature superconductivity or a multi-functional (e.g., multiferroic—ferroelectric—superconducting) material that displays a large coherent response near room temperature in small magnetic/electric fields.

As history has shown with Bednorz and Muller who set out to make a better ferroelectric material and ended up discovering high temperature superconductivity, unexpected and



**Figure 1** Schematic diagram of the approach to control of the quantum mechanical response of strongly correlated electron materials to competing interactions

potentially much more interesting phenomena in strongly correlated electron materials can be uncovered in a research effort with a well-defined scientific agenda. Once the scientific agenda has been defined, three focus areas (Fig. 1) applied to correlated electrons are necessary for progress towards control of their inherent response to competing states and subsequent functionality: 1) novel routes of complex electronic materials growth, 2) understanding the fundamental interactions and associated energy scales at the microscopic level that comprise the complex and competing phases through basic characterization, atomic-level imaging techniques, and specialized spectroscopies, and 3) new microscopic theories and theoretical/modeling techniques that provide a

thorough understanding of the fundamental interactions and a predictive capability of materials properties.

A renewed emphasis on materials synthesis and crystal growth is essential for a solution to any aspect of the control of strongly correlated electron materials. Several BES/DOE (Ames 2003, CMMP 2010), NAS (2009), and popular (Physics Today 2007) reports lament the decline of crystal growth infrastructure and expertise, largely supported by industry (Bell, IBM, etc.) in the past, and all make strong recommendations to reestablish U.S. superiority in novel materials synthesis. The high- $T_c$  cuprates illustrate the difficulty well. Nearly a decade passed from their initial discovery to a point where the samples were of sufficient quality (and quantity), primarily produced by the optical float-zone technique in Japan, that sophisticated measurements could obtain a deeper understanding (e.g., intrinsic inhomogeneity, Fermi surface), and that were generally consistent. Even today, almost a quarter-century later, most large-scale single crystals of cuprates still come from Japan. A U.S.-based crystal growth facility that included the following capabilities would address this national need and provide the means to achieve the chosen scientific mission in correlated electron materials: 1) synthesis of exceptionally high-quality single crystals to minimize extrinsic effects that obscure intrinsic behavior and to uncover emergent states only observed in ultra-clean samples, 2) production of large-scale single crystals suitable for neutron scattering and other spectroscopies to understand both the underlying competing interactions and the manifest coherent responses, 3) novel synthesis routes such as high-pressure synthesis to produce metastable phases, and 4) (intermetallic) molecular-beam-epitaxy (MBE) or other growth techniques for thin films for control of crystal growth at the atomic level, production of metastable phases, and multilayer arrangements to tune/control dimensionality and important energy scales.

The coupled of charge, spin, and lattice degrees of freedom in strongly correlated systems pose a significant challenge to understand the essential interactions responsible for their physical behavior. These interactions produce a delicate balance between competing states and give rise

to emergent phenomena (e.g., charge-ordered, magnetic, ferroelectric and superconducting states) with properties quite distinct from the original states. The necessary understanding of all of these complex states of matter will only come from the application of a suite of experimental techniques. We envision the following decadal-scale needs for this characterization and understanding: 1) measurements of the physical properties under the extremes (or a combination) of pressure (20 GPa), temperature (50 mK), and magnetic field (>20 T) to both access emergent states of matter and to tune relevant and competing interactions to elucidate their influence on the physical behavior; 2) the development of advanced spectroscopies, including spin-resolved atomic-scale imaging (e.g., STM, MRFM), spin-resolved, angle-resolved photoemission (ARPES) (<1 meV resolution) to probe charge and spin degrees of freedom; 3) inelastic neutron scattering and NMR capability development in a variety of extreme sample environments to provide the necessary probes to understand the intertwined fundamental interactions involving spin and charge. In some cases, competing interactions in correlated matter yield electronic, magnetic and structural inhomogeneities at the nano-to-micro scale that exhibit nontrivial spatial and temporal dynamics. The combination of atomic-scale imaging techniques and ultrafast spectroscopies, such as the proposed free-electron laser, would provide unique insight into the competing interactions between the correlated system's constituents (and associated inhomogeneities), such as electron-phonon, electron-electron, and spin-lattice interactions, which are necessary for a complete understanding of the emergent phenomena.

Strongly correlated electron materials also pose serious hurdles for theory and modeling. While Local-Density Approximation (LDA), Dynamical Mean Field Theory (DMFT), and hybrid Density Functional Theory (DFT) approaches show promise for accounting for strong correlations, the community still lacks a microscopic theory. Furthermore, many "effective" theories are not able to predict physical properties. Proposed theory/modeling advances to provide a theoretical underpinning of the mechanisms of strong correlations, a predictive capability of properties, and a guide for a directed search for new materials include: 1) band structure calculations on unknown materials, 2) a density functional for hybrid DFT that allows calculation/prediction of relevant energy scales and physical properties with sufficient precision, 3) development of the Random Phase Approximation (RPA) for a DFT approach to correlated metals, 4) adequate modeling of competing interactions with spatially inhomogeneous states, and 5) a theory of time resolved probes and electronic, magnetic, and lattice states far from equilibrium.

### *Time Resolved Aspects of Functionality*

*Chair: Toni Taylor (LANL)*

*Speakers: Keith Nelson (MIT) Robert Schoenlein (LBL) Roberto Merlin (U. Mich.)*

The study of non-equilibrium phenomena in condensed matter has become over the past decade a major field of research. Ultrafast techniques provide the means to investigate 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, complementing techniques such as time-integrated optical spectroscopy or inelastic neutron scattering. It is now possible to routinely generate and detect sub-100 femtosecond pulses across the electromagnetic spectrum enabling a broad portfolio of novel time-resolved spectroscopic investigations. For example, capabilities such as ultrafast x-ray diffraction and absorption

spectroscopy, electron diffraction, terahertz and photoelectron spectroscopy are currently being developed and applied providing complementary dynamical information to conventional ultrafast spectroscopy. A key characteristic of these 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. To fully exploit the promise of new materials, especially at the nanoscale, requires an integrated design, fabrication, measurement, modeling and theoretical effort to achieve a predictive understanding of complex materials. By developing the ability to better understand, predict and utilize the significant differences of energy states and transport, novel electromagnetic response, as well as the differing scaling laws due to surface to volume ratio effects, we will better guide materials development and discovery for emerging applications including superconductivity, sensors, transducers, microelectronic devices, photovoltaics, bandgap engineering and photonics and metamaterials.

### 1. Ultrafast dynamics in correlated materials:

- *Quasi-particle dynamics in superconductors, colossal magneto-resistance materials, low-dimensional charge and spin density wave compounds, insulators, Kondo systems and heavy fermion compounds*
- *Dynamics of structural, electronic, magnetic and orbital degrees of freedom*
- *Photon-induced phase transitions in complex materials*

From a materials science perspective, femtosecond temporal resolution combined with spectral selectivity enables detailed studies of electronic, spin, and lattice dynamics, and crucially, the coupling between these degrees of freedom. In complex materials there is no dominant energy scale with the implication that the charge, lattice, orbital, and spin degrees of freedom couple to determine their functional, and often emergent, properties. This exciting possibility enables, for example, nonequilibrium phenomena such as the selective photo-doped creation of a metastable state that would not, necessarily, be thermally accessible. A vast array of potential couplings leads to rich macroscopic and mesoscopic behavior including colossal magnetoresistance, superconductivity, multiferroicity, and electronic phase separation. Hence, there is a need, and opportunity, to explore and develop this approach to better understand the optical and electronic properties of complex material systems from the dual perspectives of fundamental phenomena and potential functionality.

### 2. Ultrafast materials science:

- *Electron-phonon interactions, non-equilibrium electronic structure, vibrational energy dissipation*
- *Structural dynamics during phase transformations, such as the atomistic mechanisms for grain boundary and interface changes*
- *Ultrafast magnetic and electronic phenomena*
- *Structural rearrangements in crystalline solids*
- *Ultrafast induced irreversible changes in materials in both perturbative and non-perturbative (extreme conditions) regimes*
- *Solid-liquid and solid-solid structural phase transitions*

- *Electron-phonon interactions, non-equilibrium electronic structure, vibrational energy dissipation*

Conventional diffraction techniques such as X-ray, neutron, and electron diffraction, are all powerful probes of the equilibrium atomic arrangement of material systems, but 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. Relevant dynamic material behavior benefiting from study with high temporal resolution, especially high-fidelity single-shot approaches, include advanced diffraction and measurement techniques range from fundamental strain induced electronic and magnetic effects, to the identification of and 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 serve to validate materials simulations across varying length scales.

### 3. Ultrafast processes in nanoscale materials:

- *Excitation and relaxation processes in 0-, 1-, and 2-D materials*
- *Charge separation, charge transfer and transport, exciton transport, spin dynamics*
- *Phonon dynamics, thermal dissipation, and phase transitions*
- *Interactions and energy flow on the nanometer spatial scale*

Nanoscience, through materials synthesis, self-assembly, and advanced characterization, seeks to produce emergent functionality in materials systems, via design of their nanoscale structure. As might be expected when considering dynamics of nanoscale systems, ultra small dimensions equate to ultrafast phenomena such as energy and charge transport, energy relaxation, and excitation mechanisms. In fact, understanding and designing emergent phenomena in nanoscale materials requires an understanding of nanoscale interactions in materials that fundamentally occur on ultrafast timescales. As in the study of strongly correlated materials, as described above, understanding and manipulating the fundamental properties of nanoscale materials, along with the creation of emergent functionality in these materials, requires tools which enable detailed studies of spatial and temporal (or dynamic) correlations over a range of length and time scales. Ultrafast spectroscopic tools, which enable measurements at the fundamental timescales of electronic and nuclear motion, can therefore be expected to play an increasingly important role in the quest to understand and tune fundamental interactions in nanoscale materials. Recent activity in the topics outlined above is emphasized providing insight into important and fundamental processes in nanoscale materials occurring on ultrafast time scales.

### 4. Ultrafast materials chemistry:

- *Ultrafast dynamics of energy transfer*
- *Coherent control in condensed phase materials*
- *Production of new states of matter via ultrafast interactions*
- *Control of energy flow, charge separation, charge transfer and transport, exciton transport, spin dynamics*

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? Ultimately this often requires understanding the essence of energy transfer amongst various degrees of

freedom and is a multiscale problem. Ultrafast techniques provide a means to directly interrogate such processes. Here, the study and control of matter on the sub-nm, sub-fs, distance- and time-scales is exploited to explore the limits of fundamental atomic and molecular processes, as well as the limits of optical technology. In the realm of understanding and coherently controlling more complex material systems, a very recent result hinting at the promise that this area holds involves controlling a phase of matter by coherently manipulating specific vibrational modes. Complex solids with strongly correlated electrons, in which subtle crystallographic distortions result in colossal changes of the electronic and magnetic properties, are an ideal class of materials for exploiting this approach to achieve important new insight to the physics underlying these remarkable materials. 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. With advances in ultrafast lasers it is now possible to contemplate extending coherent quantum control techniques, first applied to gas phase atoms and molecules, to 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. Numerous experiments have now demonstrated the concept of optimal control to emphasize certain quantum trajectories over others, selectively excite given vibrational modes, or drive dissociative reactions to a prescribed set of final products. However, to date, coherent control has been demonstrated in simple bulk samples, primarily in the gas phase, generally employing amplified and programmably shaped ultrafast coherent radiation pulses to control dynamics in matter. The objective in such schemes is to prescribe a new quantum state of the physical system, such as a new chemical, a special kind of light, or an excited state of a molecule with special properties. Perhaps the most exciting development in quantum control research is the learning control algorithm. In a learning algorithm, the quantum system under study actually runs the experiment itself. The apparatus interrogates the atom or molecule, which provides direct feedback to the laser. The laser system and the quantum system work together through an evolutionary optimization approach to find the optical field that can produce the desired dynamics. Recent breakthroughs helping to advance this field are identified and described.

#### 5. Novel experimental techniques and diagnostics for ultrafast materials science:

- *Femtosecond x-ray techniques*
- *Ultrafast electron pulses and diagnostics*
- *Ultrafast terahertz science*
- *Towards the attosecond regime*

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. Advanced, high-brightness x-ray sources offer new opportunities for direct probing of the lattice, as well as the valence charge density on ultrafast and ultrascale scales. This new experimental capability provides fundamental insights important for learning how coherent quantum control techniques might be used to synthesis novel materials. We will review recent remarkable advances and will present a vision for a path an imaging capability at the frontier of ultrafast science, enabling measurements which begin to simultaneously approach the intrinsic length ( $\text{\AA}$ ) 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.

#### *Size Dependence: Quantum and Nanoscale Effects*

*Chair: S. T. Picraux (LANL)*

*Speakers: E. Garfunkel (Rutgers U.), V. Klimov (LANL), E. Yu (UCSD), G. Rubloff (U. Maryland)*

#### 1. Nanoscale materials for multifunctional control

Nanoscale materials offer new opportunities to advance our fundamental understanding of the design and control of emergent properties of materials. The recent focus on nanomaterials research has revealed a wide variety of new approaches to achieving enhanced materials performance not previously imagined. These concepts are enabled by two things: 1) the new properties of materials that emerge at the nanoscale, and 2) the ability to integrate these properties into hybrid nanomaterials. These ideas point the way for a future of multifunctional designer materials where properties are combined and optimized to reach new levels of performance.

A key opportunity is to achieve new materials combinations that control and exploit the high structural quality (crystalline or amorphous), well defined interfaces, large strains, enhance fields, and quantum effects at the nanoscale. Quantum well heterostructures that localize coherent light generation and provide built-in strain to tailor valence and conduction band matching are design elements that provide increased efficiency of light output and are used in essentially all semiconductor lasers manufactured today. The above advances in electronics and photonics are based on the synthesis of hybrid nanoscale materials and the ability to control fabrication of ordered 3-dimensional nanoscale architectures.

The benefits of hybrid nanostructured materials are not limited to the above electronic and photonic examples. Rather this approach provides 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” (rather than empirically) with new understanding of how to control energy transfer by nanoscale structuring is a 5 to 10 year goal requiring a MaRIE-level effort. 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 this control makes possible.

## 2. Nanoscale Synthesis and Assembly

The controlled synthesis and assembly of materials and their heterogeneous combinations at the nanoscale is a key gap. Without control of materials fabrication little progress can be expected. The current state of the art enables relatively good control of 2-dimensional (2D) layered nanostructures for crystalline semiconductors, amorphous dielectrics, and polycrystalline metals. This control is achieved by top-down methods of layer deposition (MBE, CVD, ALD, vapor deposition, sputtering, PLD). However for other classes of materials such as crystalline oxides, ferroelectrics, organics, superconductors, even 2D control is limited. Further, to advance this area 3-dimensional (3D) structures need to be synthesized and hybrid nanostructures fabricated. This area of 3D heterogeneous fabrication is still greatly lacking at the nanoscale. Selected materials such as gold or II-VI nanoparticles by solution phase processes and semiconducting nanowires by vapor phase processes can now be synthesized with some degree of control, but formation of hybrid structures, even for the best studied cases, is very limited. Research needs to be extended across the range of semiconductors, complex metal oxides, metals, and organic materials where multifunctional nanostructured systems hold great promise.

Emerging bottom-up approaches to synthesis, in combination with top down processing, show much promise but improved experimental control and in situ methods to monitor and understand growth are essential. We must learn to exploit the nanoscale forces that lead to self-assembly of nanostructured materials and arrays, which provide self-limiting reactions for size control, and which can exploit natural templates for assembly of nanoscale architectures. To do this the role of surface and interface energies, growth catalysts, kinetics, and low temperature growth processes need to be much better understood. As examples, catalyst-controlled chemical vapor deposition is enabling the growth of electrical doped nanowires with 3D hybrid structure control, and atomic layer deposition is providing self-limiting growth of conformal films with single atomic layer control over 3D structures for aspect ratios exceeding  $10^3$ . Polymer-assisted synthesis and other mediated growth methods also offer new opportunities for low temperature growth of inorganic materials. The use of surfactants, catalysts, and differences in interfacial energies has made it possible to achieve liquid phase growth of hybrid materials with selective shapes and core-shell structures. Such new synthesis methods need to be greatly extended in terms of new materials, their combinations, doping, interface structure, and morphology control. Cluster chambers that allow sequential growth of different types of materials (semiconductors, metals, dielectrics, organics, etc.) as well as low temperature solution-based approaches need to

be exploited to explore nanoscale heterostructure properties. Hybrid materials assembly at the nanoscale is also essential and methods for the assembly of nanocomposite materials must be expanded. Examples motivating such assembly include high density nanowire arrays for Li ion battery storage, nanoparticles in a semiconducting matrix for efficient, tunable light emission, and nanowire-organic matrix structures for improved light harvesting. Advances in nanomaterials integration into micro and macro length scale structures is a crucial area to advance. Low temperature methods to self-assemble or fabricate such nanostructure composites are limited and poorly controlled at present. Focusing these hybrid nanoscale materials studies on energy detection, harvesting, transfer, and storage, and on radiation and chemical detection and sensing will provide many opportunities for advancing DOE mission areas as well as addressing a broader set of national needs.

### 3. Real-time Nanometrology

Dynamic observation, such as by optical scattering, electron microscopy, and x-ray scattering, can greatly accelerate progress in understanding nanomaterials synthesis and stability. High resolution/analytical electron microscopy and ultrafast optical spectroscopy are premier approaches to gain microscopic structural and electronic information about nanomaterials. Fast time-resolved electron and optical in situ characterization during growth can revolutionize progress in hybrid nanomaterials fabrication and assembly, as well as understanding of the response and stability of these materials during energy harvesting, conversion, and storage.

### 4. Nanoscale Properties and Modeling

An essential need for nanoscale multifunctional materials development is to establish the microscopic properties on a length scale comparable to the heterogeneous tailoring of the materials and to learn how interactions allow one to manipulate hybrid materials performance. For example, how do electronic, optical, structural, and magnetic interactions in nanostructures with their associated large field and strain gradients, and structural variation on length scales comparable to characteristic excitation wavelengths change the transport of electrons, photons, plasmons, phonons and other excitations? How can we manipulate and control surface and interface states, and carrier and energy flow across interfaces? How can we understand and predict the collective effects of regular nanostructured arrays in establishing the performance and emergent properties of nanosystems (for example in the control of energy flow by photonic lattices and metamaterials)? To address these questions we require probes with high spatial and time resolution. Electron and optical probes have continued to rapidly advance. A state of the art high resolution analytical electron microscopy facility must be established for structural characterization and ultra fast optical spectroscopy techniques continue to be exploited for electronic structure and energy transfer characterization. Scanning probes with high spatial resolution are the third area which needs to be pursued. Such probes include those based on optical excitation (absorption and excitation spectroscopies, Raman, local nanoprobe field enhancement), conducting, potential, electron tunneling, force, and thermal measurements. Additional new nanoprobe tools such as atom probe tomography, photoemission electron microscopy, and the full suite of scanning probes need to be considered. Establishing local structure, properties, and time response of energy transfer processes in combination with theoretical modeling will provide the basis for exploiting the new properties available with hybrid nanostructured materials and high density nanostructure arrays.

Theoretical techniques must be developed to treat the interactions and composite response of multi-domain systems. Due to the small dimensions, the high density of interfaces, and high field and strain gradients new approaches to deal with nonlinear responses are needed. Both materials growth kinetics and materials performance needs to be addressed. The small dimensions lead to quantum confinement, strong interactions between charges, reduced dielectric screening, and relaxation of conservation rules. For example, in nanowires it has been observed that thermal transport (phonon flow) can be greatly reduced relative to electrical transport (electron flow), and that the effects are not predicted by conventional theory. Such properties could provide large enhancements in thermoelectric performance by increasing the ZT parameter by up to a factor of 3, completely revolutionizing the harvesting of waste heat for electricity and refrigeration without moving parts. Other examples include the understanding of energy transfer effects in high fields, for example in field-enhanced energy absorption, carrier multiplication by multi-exciton formation, and plasmon flow and energy loss within nanostructures. Control of charge separation and radiative versus non radiative energy flow are at the heart of nature's approach to solar energy harvesting and fuel production. Methods to understand and optimize the relative flow of energy and minimize energy-loss channels, for example between radiative, Auger and charge transfer processes, could have far-ranging impacts, from new generation high efficiency solar cells, to ultra-efficient solid state lighting, to Si-based lasers. To exploit the opportunities afforded by nanoscale 'materials engineering' given the wide range of possible hybrid nanostructured systems, progress in microscopic characterization of energy transfer processes together with advances in theoretical modeling and simulation of these processes is essential.

### *Ferroics*

*Chair: Turab Lookman (LANL)*

*Speakers: Dan Dahlberg (U. Minn.) Wenwu Cao (Penn State) Cristian Batista (LANL)*

The study of multiferroics (and in particular magnetoelectrics) and associated heterostructures is amongst the fastest growing and most cited research areas in condensed matter physics. The scientific challenge entails understanding the mechanisms of the coexistence of magnetism, polarization and strain on the one hand, and learning to increase and control the effect for applications. Although many classes of single phase magnetoelectrics have been discovered, the coupling is often too weak or at extreme temperatures ( $< 20\text{K}$ ) or magnetic fields ( $> 5\text{T}$ ). The possibility of magnetoelectric memory, that could be realized in the next decade, will dramatically increase the speed and stability of read/write devices, compared to today's technology that depends on either ferromagnetic materials based on reversal of magnetic dipole or ferroelectric random access memory that depends on electric dipole direction.

The aim of this session was therefore to evaluate the current science and future developments in magnetism, ferroelectricity and magnetoelectricity.

#### 1. Nanomagnetism frontier (Dahlberg):

It is clear that the next frontier in magnetism requires dealing with response in the presence of defects. The role of defects in both statics and dynamics of magnetic behavior is not really

understood, yet they seem to dominate key properties in many technologies. Having a long exchange interaction length can help in overcoming this, but there is a need to have a fundamental understanding of how surfaces, and point defects affect magnetic properties. As an example, the crescent feature seen in Ni dots was stable in the real experiments but micro-magnetic simulations show very slow transients. It is very likely that there is a defect effect which stops the slow evolution, leading to trapping in the crescent shape.

*Characterization:* With advanced probes the spatial resolution has been demonstrated to be on the order of 30 nm. With commercial tips the resolution is limited to between 100 and 200 nm and the moment sensitivity observations have lower limits of  $3 \times 10^6$  Fe atoms, with a signal to noise of at least 10 to 1. It is likely that sensitivity could be reduced to somewhere on the order of  $1 \times 10^5$  Fe atoms. Inherent difficulties are that the MFM tip can produce a magnetic field which can influence observations. This can make quantitative MFM measurements difficult but could also be used as a microscopic susceptibility probe by doing two scans with opposite tip magnetization and subtracting the images.

The MFM is sensitive to the field gradients at the tip. This means one is sensing ALL the fields from the specimen at the tip position. On the positive side it implies that the probe is not merely limited to the surface magnetization. The technique of magnetic STM is only sensitive to magnetism for a specific atom, which can be good or bad. There is always the question of separating magnetism from possible artifacts. In many respects, the exchange length is comparable to the MFM resolution so the added resolution of Magnetic STM may not be needed.

*Micromagnetic simulations:* These have been extensively developed with open software widely available. This has become a standard tool for interpreting experimental results. Its based on solving the Landau-Lifshitz-Gilbert equations with various interaction energy terms, including exchange, anisotropy and magnetostatic. In most cases, there is excellent agreement between MFM images and micromagnetic simulations. This agreement implies that the micromagnetic simulation's much better resolution can be relied upon for atomic scale states. The real problem, however, is that the simulations are sensitive to the specific damping values used in the simulations. These damping values have to be obtained using other measurements. Other problems include that the coercive field calculated by the simulations is always greater than experiment. Moreover, the simulations do not reflect the effects of defects either in the bulk or at the surface. Although not generally accepted, energy minimization techniques provide an easy first step in determining the likely magnetic state. Developments in dealing with various magnetic heterogeneities represents an outstanding challenge in theory. Applications of pure magnetic systems include sensors, advanced memory elements, and fundamental magnetic research MFM. Simulations have also proven to be excellent tools.

## 2. Ferroelectrics (W, Cao):

Ferroelectric materials are by far the most studied over the last 50 years of any of the ferroics. Experimental measurements on well characterized samples, together with the use of a well parametrized Landau theory, and the need for sensors and fast memories are factors primarily responsible for the progress.

The emphasis will now be on finding Pb free ferroelectrics. Currently, the polarization

susceptibility,  $d_{33}$ , of these lead free candidates is  $\sim 230$  pC/N, about half of PZT (lead titanate) ceramics which are the most prevalent. Moreover, the ferroelectric phase transition temperatures are also lower than PZT, so significant issues related to temperature stability as well as limitations on not having high enough temperatures, remain. This is a central materials challenge in the field for next decade as there are environmental considerations that also come into play.

Domain engineering is a future strategy for designing ferroelectrics. Here the idea is to use appropriate polarization domain inclusions to provide access to regions in the field  $E$  vs temperature  $T$  phase diagram where  $d_{33}$  can be maximized. Scattered studies have been conducted but this will be a dominant strategy in the future. In addition, there is a need for design tools to minimize search times and parameter space in the search for materials with desired specifications.

The science questions relate to understanding why Pb is so important to piezoelectric activity. These questions include: Can we find substitute elements that have this property and are non-toxic? Why do good piezoelectric materials have perovskite structures? Can we stabilize some meta-stable crystals structures that can have better piezoelectric effect using extreme processing methods? In addition, there is need for characterization methods that can directly probe the ferroelectric domain walls. These are functional interfaces that control the microstructure and these can be hidden in bulk. Hence, a need for very refined structural work and polarization probing at nano-meter scale within domain walls.

The use of piezoelectric and pyro-electric materials as efficient energy harvesting materials is a future challenge. Most piezoelectric electricity harvesting produce power on the order of mW, which is too small for system applications. However, this output may be enough for hand-held devices such as some commercially-available self-winding wristwatches, monitoring sensors and mobile phones. For better efficiency of energy harvesting there is a need for more intelligent designs. Finally, we require devices that will be robust enough to endure long-term exposure to hostile environments (in space and extremes of temperature, pressure) and have a broad range of dynamic sensitivity.

### 3. Magnetolectric multiferroics (C. Batista)

The decadal and central challenge of the field is to design room temperature magnetolectrics with small switching fields. Progress will depend on innovation in synthesis and characterization techniques coupled with an understanding of the mechanisms that couple polarization and magnetization. This understanding and associated theoretical developments are the key to optimizing and controlling the effect for applications.

Single phase magnetolectrics, such as the family of Tb based ( $TbMnO_3$ ,  $TbMn_2O_5$ ) and hexagonal Ho based ( $HoMnO_3$ ) perovskites, have been instrumental in the revival of interest in multiferroics. However, the size of the magnetolectric coupling is two orders of magnitude less than what is desired for applications. Moreover, the energy scale is such that the temperature where the effect occurs is very low ( $< 25K$ ). Soft organic systems are potentially candidates for

larger effects and there are on-going efforts at NHFML, LANL in this direction. Charge density wave insulators potentially could have strong polarization and magnetoelectric effects as well. These are induced by competition between the super and double exchange and could be a route to applications. Low dimensional multiferroics, such as quasi one-dimensional chains and two dimensional sheets, also represent possible frontier candidates.

From an applications point of view, nanocomposites of ferroelectric (e.g. PZT ) and ferromagnetic (e.g. BiFeO<sub>3</sub>) epitaxial films offer the best prospects of room temperature multiferroics. One suggestion is to increase the magnetoelectric coupling by rotation of the polarization by the action of a magnetic field. The rotation would take place by the action of strain on the ferroelectric that results by applying a magnetic field. Developments in sputtering techniques, high resolution x-ray diffraction and piezoresponse force microscopy would accelerate the search for these composite layered magnetoelectrics.

### *Computational Tools*

*Chair: Frank Alexander (LANL)*

*Speakers: Krishna Rajan (Iowa State), Ivan Schuller (UCSD), James Ahrens (LANL),  
Christopher Stanek (LANL)*

This session discussed the need for a new breed of computational tools, both software and algorithms, for materials science and condensed matter physics suited to work with petascale and exascale computational hardware. This set of tools must accelerate discovery and analysis processes via informatics approach and must maximize information acquisition from each experiment -- Make every photon count! There is a need to develop high-throughput screening for materials problems similar to what's been/being done for biological systems -- namely a data-driven Materials Science -- like genomics, proteomics, metabolomics, etc. One area of focus discussed how statistical/combinatorial, learning methods can build in expert knowledge to improve performance.

Several topics were discussed in the session included:

- 1.) A Combinatorial Approach / Quasi Monte Carlo / Sampling Parameter Space Search/Coverage to experiment design and search
- 2.) The application of Statistical Learning / Machine Learning to predicting materials performance.
- 3.) Work flow management for collaborative efforts in large scale experiments.

A specific focus, important to MMMM is the understanding of defects. Defects often alter (inhibit or enable ) performance of functional materials. Their role and importance for scintillator design and engineering was discussed. In particular, the goal is to identify deleterious defects and subsequently remove or minimize those defects. A pathway to achieve this is to combine modeling and experiment for defect identification.

Grand Challenges in Making:

- 1.) Can one predicting materials properties via advances in machine/statistical learning ?

- 2.) Can one design a superconductor or a material with other desired characteristics?
- 3.) Can one achieve optimal (or near optimal) Experiment Design?

#### Grand Challenges in Measuring:

One of the key challenges in measuring is the ability to absorb the information coming out of the experiments. Visualization is a key to this challenge and was discussed extensively. In particular,

##### 1.) Visualization -- need a 3d large data visualization capability

Understanding the scientific results of MARIE simulations, experiments and theoretical models is at the heart of the MARIE endeavor. Key to MARIE's success will be using advanced visualization and analysis techniques to rapidly analyze and understand results. Real-time visualization of experimental results will also help steer the experimental process. The process envisioned for MARIE includes three phases:

1. An experiment planning/design phase
2. The running of the experiment phase and
3. A post-experiment analysis phase.

Requirements that direct our visualization effort include:

1. The size of data being generated during each phase
2. the speed at which the data needs to be processed
3. the type of data being generated (specifically detector results and large-scale simulation results) and
4. the type of visualization approaches (such as comparative visualizations and custom visualizations).

Additional requirements are driven by the locations the results are delivered to. On the LANL campus, these locations include:

1. offices and a theater in the new science complex building,
2. the M4 materials facility and
3. the MARIE four experimental halls located at LANSCE and
4. external collaborators around the world.

Modern science is team-based and results will be delivered to all these locations supporting collaborative science.

Given the multi-year time horizon for MARIE instantiation, it is critical to think about technology trends and what future technology will be available to meet these requirements. To address these requirements the following four efforts are required:

1. Real-time large data visualization algorithms - software and hardware will need to be developed to handle the massive data output from sensors during each MARIE experimental run.
2. Multi-disciplinary visualization and science teams – Based on our previous successes, we believe the best way to enable effective visualization is to deploy teams composed of both visualization scientists and materials scientists to address the visualization and analysis needs of each phase of the MARIE process.

3. State-of-the-art facilities – We can improve the way we do scientific work by creating state-of-the-art facilities (especially offices) as an integral part of the MARIE infrastructure. The facilities will effectively deliver visualization and analysis results. Collaborative support for multiple stakeholders to interactively explore, view and discuss their scientific results is a key part of our vision.

- 2.) Recording and following a workflow and collaborating
- 3.) Verification and Validation --comparing simulation with experiment
- 4.) Provide scientists tools which greatly reduce the need for human intervention
- 5.) High-speed networks
- 6.) Desktop supercomputers
- 7.) Software for fusion of disparate data to create knowledge
- 8.) Multidimensional Data analysis
- 9.) Data Provenance

Modeling can benefit greatly from enhanced experimental capabilities.

Grand Challenges in Modeling:

One of the significant challenges in modeling is to develop models which also come with prediction guarantees. This requires a rigorous and yet practical form of uncertainty quantification. Support Vector Machine classification for establishing phase diagrams was described as one such principled way to carry this out.

*Clusters*

*Chair: Roland Schulze*

*Speakers: Joel Parks (Harvard), Bogdan Mihaila (LANL), Milan Sykora (LANL),*

In the broader view of scientific knowledge and advancement, the scientific community believes it has a good understanding of the principles governing atomic and molecular behavior, and a good understanding of the principles governing much behavior in condensed matter. However, it is in the transition region between these two extremes that we are only beginning to explore. This transition region, from the regime of stable molecules composed of a few to a few hundred atoms, to the regime of condensed matter composed of extended, ordered structures of tens of thousands of atoms and larger, is largely unexplored except for the situation of special cases such as large biomolecules and perhaps soft matter systems of polymers. The initial foray into the transition regime has produced many new surprises regarding physical and chemical behavior of small ensembles of atoms in the regime where quantum effects couple with classical continuum properties, and materials (and ensemble sizes) with new properties are discovered almost every day. Examples here include quantum dots and quantum structures where new physics is manifest due to scaling effects and where quantum processes may dominate.

Current state of the art in the field of clusters is synthesis and fabrication of materials through synthetic chemical methods. In general, with some exceptions, these methods produce not a select composition and size of clusters, but rather produce a distribution. As such, the measured properties present an average, or at best a distribution, making analysis and interpretation difficult. Nonetheless, these methods can be very powerful for materials discovery in this

transition, cluster regime of materials. Many of these discoveries are made in an Edisonian mode by trial and error coupled with more comprehensive study of significant results. What is required for a systematic study of new materials through this transition regime are new tools for rapid production of clusters of carefully controlled composition and carefully controlled size coupled with direct methods for measurement of physical and chemical properties of these assembled clusters. Furthermore, the experimental measurements indicated here have the real possibility of coupling directly to theory, as the phase space is limited in size, and so full theoretical models, as well as mean field theories, are computationally within reasonable grasp.

The goal of this discussion was to foster ideas regarding the role cluster science should have in defining or influencing activities and capabilities in future material facilities. In this context, cluster science is defined as the careful study of small elemental or alloy clusters ranging from a few atoms to a few 10-thousand atoms in size. Cluster creation, cluster characterization and properties measurement, and cluster theory are examined. The intention here is to determine how cluster science can contribute to the advancement of new functional materials. This discussion includes possible contributions to fundamental condensed matter science, the science of nano-scale materials, and clusters in non-equilibrium situations (non-ground state, metastable, and extremes), as well as providing ideas for methods for a foundry of materials discovery. It is clear that success in this area will lead to new understanding of the assembly of matter through the nano-structural regime.

It is important to have the ability to create clusters rapidly, characterize and separate them on a real-time basis by composition and size or mass, and then interrogate them to measure their physical and chemical properties in an isolated state (gas phase or perhaps isolated in rare gas matrix) where there are no possibilities of correlations beyond the surface limits of the individual cluster. New methods must be defined and developed to produce clusters in rapid, combinatorial fashion, with the ability to select and filter these by exact size and composition. Gas phase molecular beam methods are an obvious choice, although there may be other viable approaches. The further modification of these mass and composition segregated clusters through gas phase surface reactions or perhaps annealing processes is also important. The important specific measurements to make on these clusters will include atomic structure, electronic structure, phononic structure, magnetism and spin momentum, correlation effects, surface effects including chemical reactivity, excited state properties, and others. The types of measurements are virtually unlimited and will be tailored to answer specific questions regarding the clusters, in addition to the basic fundamental set. The collection of these clusters on a surface with the intention of building condensed matter materials under specific and definable conditions then comes essentially free, and must also be considered as a fundamental mode of materials synthesis. The direct ability to attain both equilibrium and metastable energy states in these atomic clusters is possible through various modes of production. This can be done by thermal variation of the clusters during formation and afterwards through collisional cooling and/or annealing. This will allow modification of both atomic structure and symmetry of the cluster as well as concurrent modification of the specific electronic structure

Equally important, and vital to materials discovery and design, is the need to understanding the transition of properties from individual atoms and molecules to the solid-state in a theoretical framework. Coupling the measurement and calculation of clusters will foster this goal. For the first time, modern theoretical, computational, and experimental techniques are available to solve this problem in an accurate and efficient way. This is a tremendous opportunity and direct many-body theory is called upon to provide leadership and support. The development of a predictive many-body computational capability will transform the understanding of the fundamental properties of condensed-matter systems and materials. It will also provide the much necessary linking across length and temporal scales leading to successful extrapolations of existing models to regimes of temperature and pressure that are not currently accessible in the laboratory.

New methods to study the properties of finite-size many-body systems (e.g. clusters of atoms) are required. One example consists of using a new approach to solving the coupled-cluster expansion (CCE) of the many-body Schrödinger equation. The lofty goal of developing a predictive many-body theory can be achieved by taking advantage of the progress in the algorithm development surrounding the CCE and by exploiting the LANL high-performance computational capabilities. In particular, the hybrid architecture of Roadrunner is a perfect fit for addressing the problem of finite-size many-body systems.

The presentations by Joel Parks, Bogdan Mihaila, and Milan Sykora, which all discussed cluster science methods at the forefront of science, technology, and application, were aligned with themes of current and future general materials scientific needs and requirements to make MaRIE/M4 a “signature” concept. Joel Parks of the Rowland Institute at Harvard spoke of gas phase cluster production and structural analysis using electron diffraction methods, of size (mass) selected clusters. Key here was the ability to measure the cluster structure with size differences consisting of a single atom: the transition of a basic structural configuration (and properties) of these clusters took place in the span of adding a single atom, and can be basically thought of as a phase transition in the cluster with concurrent changes in physical properties. The ability to examine fundamental transformations at the energetic ground state of materials, such as a phase transition in a limited size cluster, is a powerful tool to understand the basics of condensed matter behavior. Dr. Parks also spoke of changes in the chemical reactivity of the cluster as a function of size, and it became clear that this methodology also is a powerful tool for discovery of new materials with tailored properties such as catalytic activity. Bogdan Mihaila from LANL spoke of the direct link between accessible theoretical methods in this size regime and the experimental tools now possible in cluster science – the calculation is *exactly* what is being measured in the experiment. The power and excitement of this area comes from the ability to calculate and predict exactly cluster structure and properties of the sizes of interest in this transition region using coupled cluster methods within many-body theory. It is fair to predict that the carefully controlled experimental cluster methods for making and measuring properties of clusters described and suggested here will become a test bed for new theoretical developments. Theoretical frameworks and understanding, and computational methods such as used in Roadrunner now make it possible to forgo mean field theories and calculation, and perform exact calculations in reasonable time frames for the cluster sizes or interest (a few 10s to a few 10000 atoms). It is imperative that the theoretical basis for exact computation and understanding in the material ground state be comprehensively complete in order to develop predictive theories for

use in understanding the behavior of matter in extreme environments or far-from-equilibrium cases. This direct coupling between theory and computation and experimental measurements gives us the opportunity to do so. Milan Sykora of LANL spoke of more conventional chemical methods of producing larger clusters in the form of quantum dots and making measurements on them to discern their specific properties and characteristics. This highlighted the great advances in synthetic methods to producing narrow size and composition distributions of useful clusters, but also emphasized the importance of producing exact size and composition materials so that measurements are on isomorphous materials rather than an average over a distribution of composition and size. Furthermore, the control required to do this means finding methods of producing clusters where the interface – the outer surface of the cluster – is carefully controlled; either through a direct termination of the primary composition or tailored ligands or surface layers. This extreme degree of compositional control is necessary in order to insure specific measurement of known materials rather than averages over a distribution.

A variety of general and specific points were made in this discussion of clusters. These included concepts required to carefully study clusters and cluster properties:

- 1) ultra-careful synthesis and characterization/verification of cluster solids produced
- 2) parallel synthetic method across size and composition ranges – combinatorial approach
- 3) rapid, direct measurements of fundamental properties of clusters in situ
- 4) New theory method(s) including coupled cluster concepts to improve viability of performing exact calculations (in addition to mean field methods) with systems involving many particles, and to take advantage of new computational hardware and approaches (Roadrunner)
- 5) direct coupling of experimental and theoretical methods (requirement/desire that modeling is exactly what is measured)
- 6) aspects in line with “materials discovery” concepts
- 7) collection of clusters (having desired property) to enable building in a controlled manner, bulk solids/thin films with desired properties

Functional needs for future development of methods to achieve careful control of cluster size and composition as well as tools for promoting guided materials discovery include:

- 1) Stable super high-intensity gas phase cluster sources to produce mass selected cluster beam – production of highly controlled cluster materials
- 2) New synthetic routes to mass produce bulk amounts of high quality cluster materials in specified size and composition space
- 3) New methods and ideas to collect clusters in order to build materials
- 4) New tools (instruments) to relatively rapidly synthesize and examine properties (electronic structure and others) across broad size and composition scales – linking length scales across the nano-regime and covering broader compositional phase space
- 5) New theoretical methods to perform full computation and treat correlations exactly

### **Summary:**

Prediction of material properties has been a lofty dream for some time. As computational tools have progressed to current supercomputer systems that can operate at petaflops, the theoretical codes that were at one time severely constricted to idealized systems with no boundaries can now

perform atomistic modeling with cubic micron volumes. In the upcoming decades computational power will continue to expand and theory will be able to incorporate interfaces in composite systems. Such modeling progress offers great potential for realizing the goal of designing functional materials. Composite materials can drive functionality both in terms of combining known material properties and discovering new properties at interfaces. The role of these composite systems is rapidly growing and will impact materials solutions in areas including sustainable energy, information storage and processing and advanced optical materials.

There are so many possible combinations of material types and architectures that rapid advancement will require the ability to effectively design functional materials across many length scales. Composition, interfaces, nanostructures and 3-D micron architecture are all important. Predictive design of functional materials is a grand challenge that will require decades of work. Linked to this grand challenge is a transition from an observational approach to control science. Control science relies on an integration of theory, synthesis and characterization. Synthesis and characterization tools are required to systematically vary and measure both defects and interfaces over a variety of length scales from atoms to microns. *In situ* techniques play a key role in enabling synthetic control through understanding nucleation and growth process and in watching how functionality evolves in time and in extremes. Well defined samples with systematic variations combined with characterization measurements of both structure and properties will provide the material database to develop and validate new theoretical codes capable of predicting functional properties. There will be significant challenges in many areas including *Applications of Electronic Materials and Oxide Interfaces, Strongly Correlated Electron Materials, Time Resolved Aspects of Functionality, Quantum and Nanoscale Effects, Ferroics, Computational Tools and Clusters*. The greatest challenge will be the effective integration of people and the wide range of synthetic and characterization tools. New materials based discovery centers in which integration occurs at a single facility and focuses on a class of materials will play an important role in accelerating the path to materials by design.

**Appendix 1**  
**RESEARCH FRONTIERS AND CAPABILITY GAPS FOR**  
**CONTROLLING AND DESIGNING FUNCTIONAL**  
**MATERIALS**

**Los Alamos Research Park**  
**Rooms 203A and 203B**

**January 20-22, 2009**

**Tuesday, January 20**

8:30 – 9:00 Welcome, MaRIE Initiative  
M4 Concept

John Sarrao (LANL)  
Mark McCleskey (LANL)

***Applications of Electronic Materials***

9:00 – 9:30

*Darryl Smith (LANL)*  
Christopher Hammel (Ohio State)

9:30 – 10:00 Modeling of Electron Devices Fabricated  
from Novel Materials

Paul Ruden (U. Minn.)

10:00 – 10:30 TBD

Christopher Palmstrøm (UCSB)

10:30 – 11:00 Breakout Session / Break

***Oxide Interfaces***

*Michael Fitzsimmons (LANL)*

11:15 – 11:45 New frontiers in infrared spectroscopy of complex materials:  
gated structures and nano-scale spatial resolution

Dimitri Basov (UCSD)

11:45 – 12:15 State-of-the-art Electronic Structure Calculations

Warren Pickett (UCD)

12:15 – 12:45 Superlattices, MBE

Anand Bhattacharya (ANL)

12:45 – 1:30 Breakout Session /Lunch

1:45 – 2:00 Emergent Behavior at Nanoscales

Sasha Balatsky (LANL)

***High T<sub>c</sub> Superconductivity/Heavy Fermion Physics***

*Eric Bauer (LANL)*

2:00 – 2:30

Eric Hudson (MIT)

2:30 – 3:00 Heavy Fermions in Flatland

Takasada Shibauchi (Kyoto)

3:00 – 3:30 Break/Snack

3:30 – 4:00 Superconductivity in Strongly Correlated Electron Materials  
(LANL)

Joe Thompson

4:00 – 4:30 Hybrid Dynamic Field Theory Approaches for  
Strong Correlations

Rich Martin (LANL)

4:30 – 5:15 Breakout Session / Close



***Clusters***

11:15 – 11:45	The Evolution of Structural Order with Metal Cluster Size	<i>Roland Schulze (LANL)</i> Joel Parks (Harvard)
11:45 – 12:15	Quantum Many Body Theory for Atomic Clusters	Bogdan Mihaila (LANL)
12:15 – 12:45	Quantum Dot Synthesis and Characterization	Milan Sykora (LANL)
12:45 – 1:30	Final Breakout Session / Lunch / Close	