Emerging opportunities in condensed matter research inspired by quantum confinement

Executive Summary

On March 9-10, 2017 science experts met in a workshop with goals to identify:

- How thin films, nanostructured films, interfaces, and heterostructures enable novel function in quantum condensed matter, and
- Specific opportunities for collaborative research in the next five years.

Attributes of quantum condensed matter include one or more of quantum fluctuations (*e.g.*, fluctuations between different ground states including zero point motion), quantization of properties (*e.g.*, property changes by well defined discrete values as a field or temperature is changed), quantum coherence (*e.g.*, a wave function that has a well defined phase) and quantum entanglement (*e.g.*, a collective quantum state of a group of particles that must be described as a whole).

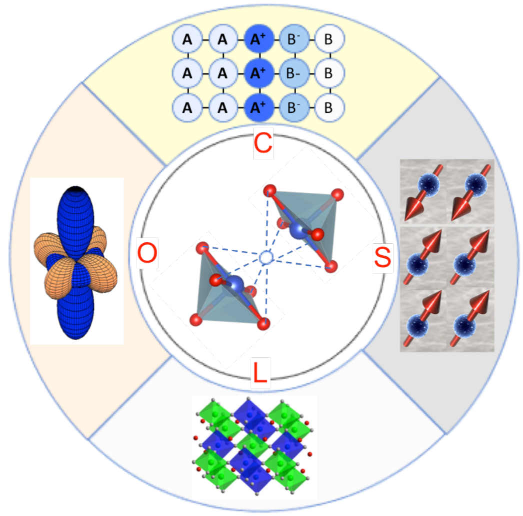
The workshop included scientists from ORNL, UT-Knoxville, and an eclectic mixture from other institutions (approximately a third each, see Appendix 1: Participant list). The workshop's first day was held at the new Joint Institute for Advanced Materials Laboratory on UTK's Cherokee Farm campus. The second day took place at the Clinch River Cabin, ORNL (see Appendix 2: Agenda). The workshop was organized following the protocol used in the Department of Energy's *Basic Research Needs* series. During the first day, participants discussed scientific opportunities and later identified priority research directions (PRD) in one of three science fields:

1. Disorder in quantum materials.
2. Quantum matter far from equilibrium.
3. Topology and coherence in quantum materials.

The PRDs were described in the form of quad charts (Appendix 3), listing a summary of the PRD, the scientific challenge, the potential impact, and the reasons why heterostructures are important (to the PRD). During the second day, the participants evaluated the PRDs from the previous day according to technique-centric panels: synthesis, characterization, and theory. These panels narrowed the focus of the PRDs into actionable tasks, and identified which important problems could be realistically tackled in the next five years. This report consists of narratives from the six panels, and a summary of recommended next experiments, and collaborative opportunities. The overarching conclusion is: *Heterostructures provide an important means to bridge classical and quantum worlds, enabling a fundamental*

understanding of quantum matter and ultimately control of novel states of quantum matter (see sidebar).

The key to achieving this vision is the synthesis of new materials. We recognize that heterostructures combining different materials offer numerous opportunities to realize novel function that is simply not possible in single-phase (homogeneous bulk) materials. Heterostructures and thin films also allow one to exploit quantum confinement or combine materials to create startlingly new states of matter. It has taken several decades of concerted effort to develop semiconductor synthesis to its current level of ~1 imperfection per 10^{13} unit cells. If we are to reach a similar level with quantum materials, which are more structurally complicated, then a large investment of time and effort must be undertaken. In parallel with these efforts, we must develop new methods of introducing specific defect types into as-grown materials in a progressive and continuous manner. Such a development will allow us to explicitly design minute changes to a material's composition and defect structure, beginning from a "clean limit". Defining what the "clean limit" is for a quantum material is in itself an important outstanding task as it will allow us to impose a common language and to establish goals for experiment and theory.



The diverse properties of quantum matter heterostructures arise from the interplay between the lattice, spin, charge, and orbital degrees of freedom (DOF). These DOFs can be modified at interfaces via effects of transitional symmetry breaking, chemical disorder, charge transfer, electrostatic coupling, and strain *etc.*, leading to a wide spectrum of phenomena and opportunities to discover new physics and functionalities. In particular, physical properties of quantum oxides often strongly depend on oxygen stoichiometry, and oxygen vacancies can be controlled due to their high mobility. At the same time, controlling oxygen stoichiometry with solid-state gating is compatible with state of the art semiconductor technology, making it an appealing path to achieve novel functionalities from quantum oxides. (Figure courtesy of Yaohua Liu.)

There is a need to search for new materials amenable to manufacture with (or perhaps only possible with) bottom-up growth techniques such as thin film deposition. Take for instance the intriguing possibilities that topological insulators afford, yet none are presently insulating in the bulk. Further synthesis science is needed to understand the origin of their (bulk) conductivity and to mitigate it for applications. Additional materials choices for quantum spin liquids are also needed. Generally, quantum spin liquids are known to have effective ½ spin, on edge sharing octahedron and honeycomb lattices. Such materials might be stabilized in the Kiteav phase, perhaps through the application of epitaxial strain. Creation of degenerate ground states is a hallmark of quantum materials, with the pyrochlores being one example.

Eventually, technological applications of quantum materials will require integration of the classical and quantum worlds. The development of means to bridge the gap between the two worlds is needed. As an example, manipulation of a quantum material between anomalous quantum Hall and axion insulating states can be achieved by combining quantum and classical materials, *e.g.*, a TI with a room temperature insulating ferro- or antiferromagnet. Bridging the quantum/classical divide will likely require further advances in synthesis science; specifically, integration of different thin film deposition platforms (growers are reluctant to grow anything but chalcogenides in dedicated chambers).

We foresee the need to more closely integrate theory and experiment. Integration means speaking the same language, such as using the same definition of the "clean limit"; modeling and measuring properties and structures at similar length and time scales; and preparation of samples while systematically controlling materials and environmental parameters to test models. Developing this level of integration requires comprehensive theory-experiment matching, including error analysis and multi-dimensional data spaces. As an example, calculations of very fundamental quantities such as the work function of various metals on complex oxides, orientation dependence of the work function, and band alignment of oxide-oxide interfaces are generally unresolved. In other words, interactions between theory and experiment must evolve from the present-day *ad hoc* approach to purposeful, collective interactions between numerous groups. *The sophistication of our research should mimic the complexity of the problems we wish to solve.*

A project worthy of an EFRC-level of investment (*e.g.*, $25M over 5 years) includes the following four tasks:

1. Define the "clean limit(s)" for a myriad of systems using theory and computational modeling.
2. Develop capabilities to synthesize confined quantum condensed matter that realize or closely approach the "clean limit".
3. Develop characterization tools capable of quantifying defects at concentrations at or below the "clean limit".
4. Integrate the tasks above to understand the influence of systematically introduced disorder, and use the understanding to achieve new function in confined quantum condensed matter.

Disorder in quantum materials

Disorder in quantum materials can be beneficial (as in 'homogeneously distributed dopants') or deleterious ('defects'). Moreover, disorder can be present at very different length scales, ranging from atomic scale defects to mesoscopic domain walls. Hence, a general definition of disorder is hard to present, although "the lack of long-range correlations" is potentially sufficient. A bit more specific, describing a defect as "the presence of phase slips in an order parameter" may cover most cases such as the phase slip in an electronic (Bloch) wave function scattering off of a charged defect, the reversal in polarity across a ferroelectric domain wall, or the loss of translational symmetry across a lattice dislocation. The breakout group noted that disorder does not necessarily entail randomness, and may be better described as a problem of local modes that are interacting with each other (*e.g.*, as in a CDW material, where local domains are pinned by defects).

Mesoscale disorder is relatively easier to access due to the length scales involved; however the appeal of mesoscale disorders such as ferroic domain walls lies in their topological character, implying they can often (depending on formation energetics) be created, controlled, and annihilated after synthesis using external probes. The chemistry across these topological defects is invariant, but since this type of disorder arises from specific (symmetry broken) states, control over chemical disorder or (local) chemical manipulation (*e.g.*, as accomplished with heterostructures) does, in principle, afford additional control over this type of disorder. While the study of these mesoscale objects appears to be fairly mature — possibly due to the better accessibility at these length scales — their interaction with atomic scale disorder, the influence of mesoscale confinement, and the potential control resulting from these interactions are subjects where progress is due.

Atomic scale disorder is present in real materials, no matter how perfectly one synthesizes them. For example, high growth temperatures provides the mobility needed for proper crystallization but also results in significant concentrations of point defects due to the larger role of entropy at high temperature. Once crystallized, kinetic limitations may prevent lower concentrations of defects from being reached as the material is cooled to room temperature. The disorder in the energy landscape due to these defects is hard to control; possibly with the exception of the regime where defect concentrations are high enough that they interact. Apart from these entropic deviations from a structure with perfect translational symmetry, many quantum materials are doped or alloyed, and generally a homogenous (random) distribution of dopants over (sub)lattice sites is desired. It is potentially possible to control the relative positions of dopants by suitable manipulation of relevant degrees of freedom (*e.g.*, strain), or by bottom-up film growth (*e.g.*, modulation doping). An example where disorder is always present in the high-T_c cuprates. Here, one could study how the striped phase separation inherent to the superconducting cuprates [1] can be manipulated by the ordering of dopants on the same length scale [2]. In the cuprates, this length scale can be comparable to the coherence length of the superconducting condensate [3], and hence T_c might be significantly affected by such ordering [4,5]. Similar examples could be found in charge ordered phases of complex oxides by manipulating the ordering of A-site dopants to affect the properties of charge-ordered states [6,7].

A major problem for achieving novel functionality through manipulation of atomic scale disorder is a lack of guidance from theory; standard first principles approaches are not able to handle the large cells necessary to approach low and disordered defect concentrations. As a result, multiscale modeling is required, connecting *e.g.*, DFT to effective field models and so forth. In some cases, quantum Monte Carlo methods, capable of handling larger clusters may have more success.

To obtain a description of the microscopic mechanisms underpinning of how (un-) intentional defects interact with the host electronic structure, one has to be able to separate the interaction between these defects from their interaction with the host. Here, simply *co-opting semiconductor knowledge may not be valid. In semiconductors defect states are added to the host band structure, whereas in many transition metal oxides (TMO) the band structure changes upon the introduction of impurities*. This has fundamental consequences for our understanding of the influence of defects on TMO's. The language of percolation theory of overlapping ranges of influence also may not be valid.

Thus, it is imperative to start from the "clean limit" – defined as having a defect concentration low enough to avoid defect-defect interactions. This requires elucidating the "sphere of influence" of defects (in analogy to the Bohr radius of dopants in semiconductors), noting this will likely be specific to configurations of lattice, charge, spin, and orbital order. The boundary of this limit may lie in different regions of parameter space for different materials, and may not necessarily be the same as what is considered clean in semiconductors. One approach would be to decrease defect concentrations until there are no changes to the ground state of the material, and then compare the experimental data to theoretical modeling.

Hence questions arise as to what a single defect looks like electronically, magnetically, and structurally. First principles theory can aid significantly here, but is often imperfect (producing incorrect energies of states in gaps, therefore incorrect spin- and charge states in many cases, which in turn affect the local lattice deformations). Experiments are also challenging, particularly as the clean limit is approached. For example, averaging (scattering) experiments run into sensitivity thresholds, and spatially resolved experiments often require sample preparation procedures that have a high probability of altering the defect states and/or the host. Experimental techniques sensitive to stoichiometry (oxygen vacancies, but also defects compensating the introduction of charged dopants in ionic system), localized electronic states, and paramagnetic impurities (color centers) are necessary.

The next step would then be to study how such local structures (de-)stabilize different, potentially nearly degenerate, ground states. Theoretically, this requires connecting first principles theory to models in which effective potentials of defects are perturb the potential energy surface of the system. This approach may enable separation of 'doping' from 'disorder' effects—see *e.g.*, the cuprates where disorder is always present in the superconducting state because doping is necessary to reach that state. Alternatively electrostatic gating, possible in heterostructures, may be another route towards separating contributions of doping from disorder [8]. Nevertheless, knowledge of the influence of the disorder itself on the phase diagram is still very murky. At the very least, agreement between experiment and theory should be obtained in datasets where the order parameter is plotted as a function of properly quantified disorder strength (or defect concentration).

The approach to systematically change the dopant or defect concentration, x, may allow connection of the 'doping' regime to the regime of 'changed chemistry', *i.e.* to cross a full phase diagram from x=0 to x=1, without describing each (commensurate) phase separately, but casting the observed variety of phases within a single description based on the physics of microscopic interactions between dopants and the host. One should note that entropy may have an important role in the energetics of the system when increasing the level of disorder.

Heterostructures are model vehicles to create control over properties arising from defect-defect and defect-host interactions. The reason for this is that heterostructures allow for direct control over the lattice DOF (through strain and imposed epitaxial symmetries), charge DOF (through gating, interface charge transfer), spin DOF (through exchange coupling across interfaces, as well as the tunability of bond angles through the lattice DOF), and orbital DOF (mostly through the symmetry breaking enabled by the lattice DOF, but also through confinement creating a quantized electronic structure of out of plane dispersing states). Moreover, creating heterostructures with length scales on the order of the defect-interaction lengths, or on the order of post-synthesis perturbation depths (tip induced domain wall formation) allows one to extract signals from affected defects in scattering experiments, rather than having many defects in the bulk of which only a small part is being manipulated. Despite the small volumes of heterostructures, the commensurate influence of defects/doping on properties may therefore be larger than for their bulk counterparts.

Conceptually, the design of heterostructures to systematically explore the influence of disorder on properties is straightforward. However, in practice synthesis of well-defined heterostructures can be a challenge to realize due to thermodynamic and kinetic limitations of synthesis. Thermodynamics and kinetic limitations may produce undesired states of disorder, *e.g.*, intermixing, non-stoichiometry, quenched defects, non-uniform strain, *etc*. A rational "synthesis by design" approach to control disorder is necessary, implying a need for synthesis science.

We see systematic studies of the influence of disorder on thin films as a means to simplify comparison between experiment and theory. Understanding the role of disorder/defects at the atomic scale should provide guidance to how disorder/defects affect the properties (or can be used to manipulate the properties) of mesoscale materials. Being able to harness interface physics in mesoscale systems—systems combining different materials and interfaces—will likely be important for technological applications. Further, creating engineered defects on larger (than atomic) length scales is more amenable to current characterization capabilities. However, in order to be relevant for real materials, a solid connection needs to be made between the concepts of the models describing such structures and the concepts governing the microscopic mechanisms of defects in quantum materials. Ultimately we wish to realize specific Hamiltonians (perhaps underpinned by our understanding of the atomic scale) using synthesis techniques, *e.g.*, self-assembly, amenable to mesoscale materials.

One way to do this is to fabricate in-plane mesoscale quantum dot lattices between leads—but from quantum materials. Depending on how the interactions between the dots in the lattice are mediated and what length scales they have, e-beam lithography might work. For example a Kagome lattice of magnetic dots with intentionally positioned defects can be fabricated to study the influence of disorder on magnetic fluctuations. The concept is similar to what Hari Manoharan [9] did (on a different length scale) using CO molecules to "make graphene" out of the 2D surface state on a noble metal (111) surface. This approach has recently been expanded to quasicrystals as well.

Quantum matter far from equilibrium

Environmental conditions far from equilibrium are of interest for two reasons. First, if the conditions during synthesis are far from equilibrium then atoms do not have sufficient opportunity to achieve thermodynamic equilibrium (*i.e.*, to find equilibrium locations on the growth surface) and chemical reactions do not have the opportunity to proceed to completion. The materials produced by such growth conditions can achieve very unusual architectures such as (but not limited to) vertical architecture networks of nanopillars [10,11], nanobrushes [12], or quasi-stable chemical profiles, *e.g.*, $Lu_2FeO_4/LuFeO_3$ [13], stripes in $LaCoO_3$ [14], and non-uniformity of oxygen (O)-vacancies.

Our understanding of synthesis far from equilibrium (*e.g.*, film growth from a plasma) is woefully incomplete. Generally, we lack the passive *in situ* characterization tools needed to quantitatively monitor a myriad of conditions that vary in space (*e.g.*, within the plasma or across the substrate) and time. Some classic tools for *in situ* characterization, such as reflection high-energy electron diffraction (RHEED), are not as passive as we would like. For example, RHEED is a qualitative diagnostic tool for observing layer-by-layer film growth, yet the tool's potential is untapped. Namely the theoretical and modeling support needed to quantitatively interpret RHEED is lacking. Further, the conditions required for RHEED tend to degrade the quality of some films (growth of III-V semiconductor films is one example [15] or how RHEED introduces O-vacancies into BME-grown WO_3 films [16]). Similarly, O-vacancies are crucial for determining transport properties in oxides, yet we have no way to measure O-vacancy concentrations to the precision we require, nor can we quantitatively control their presence. Obviously the lack of quantitative information about film growth impedes the ability of theory to provide insight. The breakout group identified a priority research direction to move the community from the "Art of Synthesis" to the "Science of Synthesis".

The second issue relating to far from equilibrium conditions is creating transient or metastable states of matter *post synthesis*. An example cited by the breakout group (and echoed by the Topology and Quantum Coherence group) was photo-induced superconductivity [17]. This example is ripe for study as there are two competing theories [18], and the possibility to control superconductivity by light is technologically intriguing. More broadly, the concept is to excite matter at equilibrium into a new state away from equilibrium, but perhaps isolated by an activation barrier that prevents the system from transiting back to the equilibrium ground state. If the excited state were sufficiently long-lived and physically interesting, *e.g.*, a quantum spin liquid, then novel technological applications might be developed. Examples include photo-induced superconductivity, and numerous probes manipulating the ground states of organic molecules [19]. It may be that organic molecules are more compliant (*i.e.*, their ground states might be more easily controlled than inorganic hard matter). In that case, inorganic-organic hybrid materials may be an attractive choice for study. Regardless of the system under study, guidance on what probes (*e.g.*, light, microwaves, stress) and how to use them (*e.g.*, pulse widths, repetition rate) is mostly lacking. Dramatic progress towards driving, manipulating and using materials far from equilibrium will require close coupling of experiment and theory, where both explore the same relevant length and time scales. Once an experimental protocol for generating a metastable state is identified, then we must address whether the state necessarily transient or if its lifetime can be extended for technological purposes.

Achieving a novel state of matter far from equilibrium involves controlling the evolution of competing (or cooperating) degrees of freedom in space and time. Heterostructures offer opportunities to activate and control numerous degrees of freedom through prescribed changes of chemistry that can occur on atomic length scales. Examples include ferromagnetic and ferroelectric order parameters that change and are coupled across ultrathin layers of Lu_2FeO_4 and $LuFeO_3$, respectively. In addition to chemical composition, orbital ordering, charge discontinuity, and atomic reconstruction, there are other degrees of freedom readily controlled in heterostructures. For example, manipulation of O-vacancy non-uniformity has a profound affect on transport and these degrees of freedom compete (or cooperate) at interfaces. If we understand how to exploit this competition, we may be able to navigate a pathway from one ground state to another, and thus harness opportunities afforded by materials far from equilibrium.

The priority research directions identified by the breakout group are:

Understand transient states and structures.

Characterization of transient states and structures of matter, particularly at the extremes of short time scales (fs) and many length scales (Ångstroms to microns), is an on-going challenge. Experimental tools with sensitivity to multiple degrees of freedom (DOF) are required by theory, along with the ability to discern between these DOFs with spatial-temporal resolution. New developments in theory are also required, including advances of first principles approaches for treating dynamics and strong interactions in excited and extended systems. Experiment and theory must be coupled so all the relevant time and length scales are measured and computed.

Manipulate excitations and fluctuations in quantum heterostructures.

We would like to exploit heterostructures to create and couple different degrees of freedom. The goal is then to manipulate these states "coherently" so collective quantum states can be achieved across many nanoscale-sized objects, quasi-particle condensates, and excitations or orbitals and spins.

Synthesize quantum heterostructures.

A comprehensive effort on the synthesis and *in situ* characterization of materials during synthesis is urgently needed. Data collected during synthesis are required to guide theory of synthesis—a nascent field. Advances here will enable us to create samples of controlled quality, ranging from the "clean" to the intentionally disordered/defective limit. Success in this area will not only enable discovery and control of novel phenomena but it will also improve confidence in theory by providing reliable input for modeling.

Create true "hidden" order, states and structures.

While the previous PRDs were focused on producing heterostructures with DOFs that we precisely create and control (at least individually), this PRD is concerned with collectively manipulating the DOFs to achieve a new transient or long lived quantum state. Phase diagrams by definition describe coexistence of different phases in equilibrium. This PRD argues for a new kind of phase diagram that describe how to achieve new states of matter that are not in equilibrium, *e.g.*, superconducting manganites, metallic antiferromagnetic nickelates, and so forth.

Topology and coherence in quantum matter

Quantum states of matter that are protected from unintended alteration may enable disruptive changes to information technology. The 2016 Nobel Prize in physics recognizes the importance of topology in establishing quantum states believed to be protected from backscattering. Topology is ubiquitous in nature, including for example the curvature of the cosmos [20] and knots in DNA [21]. Thus, it should not be a surprise that topology plays a role in quantum condensed matter. Haldane's Conjecture [22] that spin wave dispersion is gapped for a chain (a chain permits only forward or backscattering) of integer spins and not for half-integer spins is one example. The realization of skyrmions [16,23] (initially proposed by the nuclear physics community [18,24]) and which are thought to represent topologically protected spin texture in magnetically ordered materials) is arguably at the forefront of condensed matter research.

Whether skyrmions are a linchpin to next-generation information technology remains unclear pending the outcome of significant challenges. Skyrmion diameters of less than 5 nm may be critical to the development of scalable memory platforms, while skyrmion-based racetrack memories [25] may offer substantial benefits in terms of energy, density, and readout speed [26]. However, a complete understanding of pinning/depinning dynamics at picosecond time scales and nanometer length scales remains an outstanding problem, which is critical to the development of these technologies. Moreover, while currents may be used to interact with magnetic (spin) skyrmions, the development of novel materials supporting oxide (charge) skyrmions may necessitate new interaction paradigms. Skyrmions, or more generally other topological excitations, may become useful as qubits for which topological protection stabilizes steady-state entanglement.

Key challenges in topological materials include:

- Obtaining observational proof of topological protection from backscattering; what are the consequences for other forms of scattering?
- Realizing topologically non-trivial spin textures in materials other than transition metals at room temperature.
- Understanding the origin of and controlling the non-insulating properties of so-called topological insulators.
- Creating heterostructures of ferromagnetic or preferably antiferromagnetic (for increased operational speed) materials with topological insulators.
- Demonstrating operation of devices at room temperature (essential for memory, preferred for computing).

Because topological excitations live at surfaces of topological materials, new functionality may be available at the interface between topological materials and other quantum materials. For example, the electric field induced perturbation of the magnetization of a proximal ferromagnet to a topological insulator is much stronger than were the TI a semiconductor [27]. Recent work has suggested that layers of graphene can be arranged in ways that allows the Berry curvature to be measured with light [28]. Similarly, nanophotonic quasiparticles like plasmons that posses orbital angular momentum of deterministic topological charge [29] may allow for active control when addressing other topological excitations [30]. Coherent light-matter interactions have been increasingly explored as a path toward understanding entanglement in quantum materials and leveraging entanglement for the control of quantum materials. The former may be achieved by characterizing dynamic susceptibilities of quantum materials in order to describe the quantum Fisher information of many-body electronic states [31]. One example of the latter is the generation of entangled quasiparticles, including squeezed phonons [32], and for the optical exploration of non-equilibrium lattice dynamics. However, the incoherent excitation of additional phonon modes has thus far limited the generation of highly squeezed phonon modes, and experiments aimed at generating quantum states of light with strongly correlated electronic materials likewise remain in a very nascent stage as a result of unwanted incoherent interactions. Additional control over light-matter interactions may be enabled by fabricating heterostructures of optically active and strongly correlated materials. Heterostructures of superconductors and topological insulators may enable convenient platforms to entangle quantum states through manipulation of Berry curvature. For example, one can imagine laterally controlling the superconducting state of a surface film to move/control a Majorana Fermion at the interface between the superconductor and a skyrmion [33].

The priority research directions identified by the breakout group are:

Create, measure and manipulate skyrmions at nm length and ps time scales.

The length and time scales are those that should enable technology based on skyrmions to be competitively attractive. The group envisages different ways to create and control skyrmions which included: controlling the Dzyaloshinskii-Moriya interaction across the ferromagnetic/non-magnet interface, using strain to control skyrmion size, combine/couple a skyrmion to a piezoelectric (or superconductor to create Majorana Fermion), or use scanning probe techniques to move skyrmions.

Realize dynamic control of topological states in nanostructures

This theme recognized the large potential topological materials offer, *e.g.*, dissipationless transport, but at the same time acknowledged skepticism of theoretical and experimental claims typical of nascent research. Theory leads the field of topological materials. Synthesis science lags in that the quality of so-called topological insulators, which are neither insulating (in bulk) nor stable (the Fermi level is prone to change with time). Characterization science lags as well in that even if a topological insulator lived up to its reputation, there are few experimental tools available to provide "smoking gun" evidence of topological protection of surface states, and none available to probe topological excitations at buried interfaces. Topologically robust transport should exist at interfaces between materials of differing topological charge. The ability to dynamically tune the topological charge of materials would therefore enable robust active rewriting of information processing elements. Optical analogs to topological insulators, or photonic topological insulators, offer a clear path toward such dynamic control by incorporating phase change materials into photonic plaquettes. The ability to realize such dynamic control in electronic topological materials is critical to the development of topological materials for information processing.

Managing coherence between quantum light and quantum matter

This direction involves managing the light/matter interface. On one hand, we would like to create entangled and squeezed states of light using condensed matter. On the other hand, we would like to use light to interrogate quantum condensed matter. The former leverages nonlinear interactions between photons and phonons, for instance, in order to prepare quantum optical states of light. Quantum correlations in electronic materials may provide a new platform for the generation of quantum correlated photons with the design of appropriate heterostructures to manage the light matter interface. As an example of the latter, far infrared laser light has been used to measure the Berry curvature of graphene layers using the Faraday effect [34], and photon number statistics have been used to describe fluctuations in lattice dynamics [35]. Bell-like inequality violations have been previously observed in neutron interferometers [36], so neutron matter interactions may be mapped by monitoring the quantum state of a neutron probe. Neutron spin interferometry thus provides another potential path toward describing the quantum state of excitations in materials.

Manipulate phonons to minimize decoherence and dissipation

Phonons can be an important part of quantum matter—superconductivity in particular. Or, phonons can compete and often dominate or make inconsequential quantum fluctuations. Thus, the ability to manipulate phonons offers the opportunity to realize new states of matter. An example is driven transient superconductivity. Heterostructures may be a convenient route towards designing the phonon dispersion of materials. The periodicity of a superlattice leads to additional gaps of the dispersion curve. Phonons at the gaps are standing waves, thus, these engineered structures might provide a means to minimize entropy leading to a concomitant increase of quantum coherence. Moreover, control over the photon-phonon interaction Hamiltonian has already been shown to enable the observation of lattice fluctuations below the vacuum noise floor [37] and leveraged to achieve transient superconductivity [38]. Generating squeezed phonon modes may enable descriptions of the lattice dynamics associated with phase transitions in quantum materials that are otherwise hidden in the vacuum fluctuations of the lattice.

Synthesis

Understanding novel function of materials is enabled by growth of suitable samples. But, what does “suitable” mean? In the case of the semiconductor industry, the cleanliness and perfection of semiconducting films was eventually established after an arduous and expensive process. Today, the cleanest Si is achieved using thin film growth of Si films on comparatively dirty bulk Si single crystals.

We envisage a future going beyond devices based on transport of charge through semiconductors. Specifically, we see a future with devices that utilize charge, spin and orbital degrees of freedom to harness quantum mechanics to achieve new function. However, we have yet defined a “clean limit” for quantum condensed matter. Nor do we know how clean or whether cleanliness/perfection is necessary for materials to exhibit new properties, *e.g.*, superconductivity, quantum spin Hall effect, topology, etc. There are two specific fields of thought on how to define the clean limit. On one side, the clean limit is described as limiting defect concentrations to the lowest possible levels similar to what is currently possible with Si films. The other way to consider the clean limit is by defining the pristine state by a desired functional property. This can be particularly attractive in correlated systems, as defects may be a key ingredient to inducing the sought after ground state.

The case of superconductivity is a good example in which the essential metric, the critical temperature, is not as sensitive to the presence of point defects as are quantum (Shubnikov de Haas) oscillations [39]. Optimization of the saturation moment and Curie point of ferromagnetic complex oxides is another example—the establishment of perfect long-range order and the complete elimination of antiferromagnetic, nanoscale, secondary phases of NiO in the double perovskite La_2MnNiO_6 may not be possible [40].

Systematically prepared experimental studies combined with theory may be helpful in regard to defining a "clean limit" and to understand the role of disorder, *e.g.*, as produced by defects, strain, *etc*. Even if some disorder is beneficial, being able to compare function in one material to the clean limit provides valuable insight. We expect thin film synthesis, and heterostructure growth at the nano and mesoscale to be critical in facilitating devices fabricated from quantum condensed matter. Not only do molecular beam epitaxy (MBE) and pulsed laser deposition (PLD) enable growth of materials with chemical structures that are impossible to grow using bulk materials synthesis, these methods also allow a level of perfection that can be unrivaled by bulk materials synthesis (*e.g.,* as in realizing chemical ordering of A-site cations in $La_{1-x}Sr_xMnO_3$). While MBE is able to produce the nearly chemically pure and structurally perfect materials, it should be recognized that even the best ternary and quaternary compounds grown with this technique may possess off stoichiometry of tenths to hundredths of a percent. Moreover, the presence of high-energy particles in the ablation plume can result in the creation of point defects in evolving epitaxial films prepared by PLD. Finally, thin film growth techniques can be combined to grow very dissimilar materials to form composite systems with chemistry and disorder content that vary with unit cell dimension enabling entirely new functionality, *e.g.*, ferromagnetic order from non-magnetic materials, superconductivity from insulators, and so forth.

In the past decade intriguing discoveries of quantum matter in bulk materials, *e.g.*, quantum spin liquids/ice, fractional quantization, quasiparticles, topological materials, spin texture and so on, have motivated scientists to think about how to use the new physics for technological applications. Leveraging techniques to fabricate classical spintronic materials (this includes the gambit of function from exchange bias, electrostatic gating, spin torque, *etc*.), the thin film/heterostructure community is positioned to develop devices that combine classical and quantum matter for new applications. As one example, means to manipulate magnetic films classically can be integrated with quantum matter (*e.g.*, to control time inversion symmetry on the surface of a quantum material) to harness the quantum spin Hall effect. There are significant challenges to overcome before achieving a vision of quantum materials engineering. For example, topological insulators are not very good insulators (defects may be to blame) and robust topological protection is unproven.

To achieve our vision significant investment in synthesis science is needed. The goal is to achieve well-defined samples—meaning systematic control of disorder—that include heterostructures of quantum and classical matter that remain unadulterated by processing, *e.g.*, lithographic patterning. We envisage samples in which strain, defect concentration and spatial position, and the chemistry of interfaces are controlled within tight specifications. Samples may be planar, vertical architecture networks, monolayers, nanotubes, free standing films (implies removal of growth substrate) and so forth. Many films will require designer crystal perhaps with close-packed smooth surfaces (lattice and symmetry matched or better yet nearly matched and tunable to thin films). Acquisition of such crystals is a significant challenge. Means to mitigate introduction of impurities and defects from the substrate into films must be developed. Different characterization techniques have different requirements for sample mass/volume. A goal of state of-the-art synthesis science should be to deliver identical samples regardless of mass/volume.

The preferred method of defining the "clean limit" also determines what experimental approaches can be applied to the investigation of quantum materials. In each case, developing new and better ways of controlling and quantifying defects in a given crystal structure is central to our success.

Characterization

Due to competing charge, spin, and orbital, and lattice degrees of freedom, phenomena in quantum condensed matter can be complex. Samples of the highest quality are required in order to avoid being misled by artifacts. Achieving high-quality samples is a major challenge in synthesis science (a theme of the workshop), and verification of quality is a challenge of characterization science. Although the task of achieving the "clean limit" is common to most condensed matter problems, an intriguing opportunity for correlated electron phenomena is their apparently large tolerance to disorder, *e.g.*, compared to semiconductors. There are both fundamental and practical reasons why this may be the case. Importantly, this unique property enables the disorder itself to become a control parameter, and it makes it possible to achieve reproducible synthesis even with the complex chemical compositions nominally found in quantum materials. Identifying more precisely the boundary of the "clean limit" is a prime task for characterization, theory and synthesis efforts.

Quantum heterostructures present more unique challenges and opportunities for characterization science. Interfaces are the playground for different degrees of freedom to compete through which new function arises, *e.g.*, superconductivity between insulators, ferromagnetism between non-magnetic materials – and they will be the practical embodiments of quantum phenomena for applications. In most instances the interfaces are buried. We aim to examine the static and dynamic atomic, spin and orbital structures and properties of interfaces, and to probe the response of interfaces to stimuli. The challenge for characterization science is to perform measurements covering many length and time scales, ideally simultaneously, from a small amount of quantum matter that is often buried inside some other material that may be an impediment to our tools. For example, inelastic neutron scattering has been the tool-of-choice for studies of quantum spin liquids in bulk materials, but is presently impractical for studies of quantum matter in heterostructures. New opportunities therefore open up for the use and further development of near-surface characterization tools, such as electron spectroscopy (*e.g.,* EELS), optical measurements (Raman, Infrared, near-field microscopy) and many other tools. The case for *in situ* measurements, where material synthesis and characterization are combined on a single experimental set-up is easily made for environmentally sensitive interfaces.

In many cases, the "smoking gun" test for a specific property across multiple length-scales is probably unrealistic. But utilizing a combination of observables toward a specific phenomenon, partially guided by theory is a timely and much needed development for the panoply of emerging quantum materials. For studies of topological materials and spin textures (*e.g.*, skyrmions) electron probes like Lorentz microscopy, angle resolved photon electron spectroscopy (ARPES), spin polarized scanning tunneling microscopy (STM), magnetic imaging (*e.g.*, MFM), resonant x-ray spectroscopy (including resonant x-ray reflectivity and glancing incidence scattering), and neutron scattering can collectively provide insight.

A unique new challenge posed by combined approaches is the sheer size and diversity of the data space. Going beyond a single measurement will require development of specialized data infrastructures, analysis and visualization tools capable of coping with the data volume in real time. Semi-supervised, expert systems can be envisioned for specific techniques. The embrace of data technologies and analytics also opens up a new dimension of theory-experiment matching. This transition involves expansion into many dimensions and with a vastly more comprehensive analysis of statistical variations, noise and other subtle features in the data that are presently averaged into a large error-bar.

More comprehensive utilization of today's measurement techniques for quantum matter heterostructures will naturally form the basis for the development of next generation tools. Specific examples include inelastic neutron measurements on thin films, hard X-ray ARPES for the measurements of electronic structure of interfaces, multidimensional probes, polarization analysis for magnetic crystallography, and coherent techniques for measuring fluctuations. Some of these methods will require a significant time to develop. User facilities will play a key role in not only the development of such tools, but also their broader distribution in the science community.

Ultimately, understanding disorder may lead to new approaches to control quantum phenomena, spin fluctuations, and phonons in order to achieve new states of quantum matter, including the increase of the lifetime of transient quantum states recently revealed by time-resolved techniques. Quantitative guidance from theory on the influence of defects (or disorder) on properties should set achievable goals synthesis science (*e.g.*, to prepare clean samples) and characterization science (*e.g.*, to establish lower limits for measurement of oxygen vacancy concentration). Theoretical guidance is also needed for determining specifications for pump-probe experiments. For example, what frequency light, duration of pulse, *etc*. is required to achieve an excited state? Our vision is with an integrated team of synthesis, characterization and theory scientists highly efficient experiments will be conceived and executed—ones that offer insight into quantum matter in heterostructured materials.

Theory

Developing schemes to uncover hidden states of quantum matter is a task amenable to theoretical guidance. The hidden states may be metastable, static or dynamic. The states may be accessible only by judicious excitation of probes, *e.g.*, optic, sound, spin resonance, *etc*., or perhaps achieved through cleverly applied growth techniques, *e.g.*, to realize heterostructures in unusual configurations (not simply planar ones) or through control of disorder. Theory should be able to tell us how to produce new states of quantum matter.

Clearly confidence in our understanding of materials requires growth of well characterized samples (structure and properties) that can be readily (accurately) compared to theory. Thus, a comparison of theory to samples at the "clean limit" makes good sense. From a theoretical perspective, "clean limit" means samples that are perfect and at zero temperature.

Conceptually, samples realizing the clean limit would be grown and characterized. The results then compared to theory in order to develop reliable theoretical/modeling tools. The next logical step would be to systematically introduce disorder. However, realization of samples at the clean limit may take many years (if possible at all), thus, improvements across the gambit of making, measuring and modeling materials are required to enable the diverse community to meet somewhere between the clean and dirty limits. One step in this direction is for the community to agree on what is meant by the term: disorder.

Disorder can take many forms including quantum fluctuations, thermal fluctuations, spin disorder, *e.g.*, frustration, intrinsic local modes, charge and/or spin domain walls and stripes, defects (impurities, intermixing, vacancies, dislocations, twin and grain boundaries) and so on. An advantage of thin film growth and heterostructures is that introduction of many types of defects can be done in a controlled fashion. For example, the lengths of phthalocyanine chains and locations of a defect(s) in the chain are easily controlled through growth. Disorder created by such defects can be turned on and off with light and is reversible through annealing.

Disorder should be defined for a specific context. For example, isolated defects in a single crystal may have little effect on antiferromagnetic order, but may profoundly affect conductivity. Disorder may be short-range, long-range or exhibit many length scales. Theory and experiment must be sensitive to many length scales. A first step towards understanding how disorder affects function requires an agreed upon definition (by theorists and experimentalists) that is likely specific to a particular problem. The definition drives the protocol and tools needed to pursue understanding and predicting function of quantum condensed matter in heterostructures.

A grand challenge identified by the theory group was to predict new states of quantum matter in heterostructures, to provide insight on how to achieve (*i.e.*, drive to) the new states, and to describe the static and dynamic properties of the new states. Achieving this vision could entail identifying the desired (final) state and then reverse engineering a process from final state to synthesis that provides guidance to the synthesis science theme identified during the workshop.

Recommendations for further research in next 1-5 years

What (is the important task)?	Why (is it important)?	How (do we achieve success)?
Control magnetic order in orbital-ordered crystal through crystal field excitations	Realize/harness novel function.	For example in $LaMnO_3$ with light or phonons where transition from AFM to FM state is dramatic.
Measure Hubbard U of a solid exhibiting photo-induced superconductivity	To resolve conflicting theoretical models of Millis vs. Cavalleri	Auger spectroscopy as was done for C60.
Develop theoretical and experimental means to treat/explore different degrees of freedom.	Realize/harness novel function. Pave the path to next generation tools to achieve new states of quantum matter.	Identify relevant length and time scales. Explore systematic changes to atomic structure (strain, pressure…), chemical, charge, spin and orbital degrees of freedom. Identify systems in which dopants are introduced systematically without adding disorder. Identify and apply new multi-technique (multimodal) approaches utilizing tools not typically applied to quantum heterostructure problems.
Develop theoretical tools to include higher order terms, *e.g.*, in Hamiltonian.	Essential to understand collective modes, correlated phenomena, driven states of quantum condensed matter.	
Develop theoretical and experimental means to model/observe incoherent phonon scattering.	To further advances in superconductivity, relaxor ferroelectrics, thermoelectrics… Advances in photon-phonon interactions to achieve phonon squeezing	
Bring classical modeling tools to quantum matter	To understand disorder in quantum matter.	
Define the "clean" limit for quantum materials	Provides goals for synthesis. Place theory	Calculate region of influence of a defect.

	and experiment on same playing field.	Calculate density at which defect-defect interactions dominate. Recognize that different properties may have different limits. Fabricate systems approaching the clean limit, measure, then systematically introduce disorder. For different properties establish goals for characterization of relevant defects, disorder.
Define what a topological defect is?	Test robustness of topological protection.	Explore phthalocyanine chains as means to test the Haldane Conjecture.
Develop and deploy passive probes of chemistry and structure to monitor synthesis of materials.	We lack data for modeling synthesis.	Be able to measure oxygen vacancy concentrations *in situ* and *ex situ*. Be able to measure intermixing and roughness *in situ*, and distinguish between the two effects.
Broaden familiarity of quantum inorganic and organic materials.	Realize new function.	Use inorganic/organic heterostructures
Develop theoretical tools to identify how quantum states can be manipulated by convenient pumps.	Provide pathways to realized new states of matter that may be transient or metastable.	Use light, strain, microwaves… as pumps
Develop oxygen vacancy engineered materials	O vacancies crucial to properties, transport, magnetism…	

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Appendix 2: Agenda

Quantum Condensed Matter Heterostructures Workshop March 9-10, 2017

On March 9 the workshop will be hosted at the **Joint Institute for Advanced Materials (JIAM) on the UTK campus**. A bus will provide transportation to/from JIAM. On March 10th the workshop will be hosted at the ORNL Cabin.

Thursday, March 9, 2017 **JIAM**

Time	**Event**	**Lead**
7:20-7:30 am	Bus transports Guesthouse visitors to visitor center, departs from the ORNL Guesthouse.	Visitor Center
7:30-7:45 am	Visitor badging	Visitor Center
7:45-8:45 am	***Bus to JIAM, departs from the ORNL Visitor Center as soon as badging is completed but not before 7:45am***	JIAM
8:45-9:00am	Welcome to JIAM Workshop	Veerle Keppens, JIAM Director, Seminar room 147
9:00-9:30 am	Workshop logistics and goals	Mike Fitzsimmons
9:30-9:40 am	Outstanding questions and directions in QCM of Heterostructures	Ho Nyung Lee
9:40-9:45 am	Charges to breakout groups	Mike Fitzsimmons
9:45-10:15 am	**Morning Break**	3rd floor, Mezzanine
10:15-12:00pm	Breakout groups, identify priority research directions (PRD)	Disorder (Johnston, 300) Equilibrium (Chakhalian, 200) Topology (Stemmer, 306)

Time	Event	Lead
12:00-1:00 pm	**Working Lunch:** Down select/consolidate PRD's	Disorder (Johnston, 300) Equilibrium (Chakhalian, 200) Topology (Stemmer, 306)
1:00-3:00 pm	Breakout groups, refine PRD's, prepare Quad charts	Disorder (Johnston, 300) Equilibrium (Chakhalian, 200) Topology (Stemmer, 306)
3:00-3:30 pm	**Afternoon Break**	3rd floor, Mezzanine
3:30-4:30 pm	Finalize Quad charts for PRD's	All
4:30- 5:30 pm	Chair Reports, then adjourn	Breakout group Chairs, Seminar room 147
5:30- 6:25 pm	Bus returns to Guesthouse	Guesthouse stop
6:25- 6:30 pm	Bus returns to Visitor Center	Visitor Center stop
7:30 pm	Car pool to restaurant (tbd)	

Quantum Condensed Matter Heterostructures Workshop
March 9-10, 2017

On March 10 the workshop will be hosted at the **Clinch River Cabin on the ORNL site**. Participants are expected to drive to the Cabin.

Friday, March 10, 2017 **Clinch River Cabin**

Time	Event	Lead
8:30-8:45 am	Charges to crosscut breakout groups	Mike Fitzsimmons
8:30-10:00 am	Crosscut breakout groups, review PRD's and quad charts from Thursday	Synthesis (Chambers, North) Characterization (Freeland, Main) Theory (Rondinelli,South)
10:00-10:30 am	**Morning Break**	
10:30-12:00 pm	Crosscut breakout sessions, Finalize charts	Synthesis (Chambers, North) Characterization (Freeland, Main) Theory (Rondinelli,South)
12:00-1:30 pm	**Working Lunch:** Chair reports and group discussions	Crosscut Chairs
1:30 pm	**Adjourn**	

Appendix 3: Priority Research Directions

1-1 Control defects systematically from the clean limit.

Scientific challenges

Determine what the clean limit is for a quantum material.

Develop and integrate theory, characterization and synthesis to study the same length/energy scales of the charge, spin, orbital, and lattice sectors.

Understand the impact of defects/disorder on selection between degenerate or nearly degenerate ground states.

Find ways to choose specific defects that influence particular materials properties.

Summary of research direction

Starting from the limit of non-interacting defects, we aim to understand the influence of defects on the properties of their parent materials. Moving from this clean limit towards concentrations where defects interact, we aim to connect short- and long-range physics.

Potential scientific impact

Understand the effects of disorder on macroscopic phenomena such as nucleation of phase transitions or enhancing superconductivity.

Move towards rational design of quantum materials and control of defects.

Why heterostructures?

Provide unique control of boundary conditions in all sectors and control over defect parameters.

Can separate “doping” from “disorder” in a systematic way.

1-2 Exploit atomic scale engineering to study correlated disorder.

Scientific challenges

Control disorder beyond atomic length scales. Create interactions between defects to generate functionality.

Identify good materials platforms akin to cold atoms for implementing model Hamiltonians with disorder.

Separate and understand the role of randomness vs. isolated short-range defects

Summary of research direction

Exploit advances in atomic scale manipulation to study correlated disorder and reveal new states of matter. We want to control and systematically study patterned structures on longer length scales.

Potential scientific impact

Develop new capabilities in device functionality. Provide a new control parameter for studying the physics of defects.

Why heterostructures?

Heterostructures already provide a known example platform.

Enables combinations of dissimilar materials where new states form at the interface.

2-1 Understand transient states and structures.

Scientific challenges

To predict the time evolution of quantum materials exhibiting strongly coupled degrees of freedom (DOF) and their selective excitations.

To identify competing transient configurations that are only accessible far-from-equilibrium and exhibit vastly different functionalities than the equilibrium states.

Summary of research direction

Advance ‘first-principles’ theories to treat dynamics and strong interactions in excited and extended systems, which bridge the disparate characteristic timescales of transient states.

Develop tools to capture the nonequilibrium state with sensitivity to each individual DOF and with the requisite experimental spatial-temporal resolution.

Potential scientific impact

Map out landscape of all possible quantum states of matter in a given material.

Usher in deterministic discovery of hidden quantum orders.

Formulate new methodologies that access complex processes involved in energy conversion.

Why heterostructures?

Heterostructures provide the only capability to selectively tune the coupling between DOF thereby identifying the principal driving interactions that govern the evolution transient state.

2-2 Manipulate excitations and fluctuations in quantum heterostructures.

Scientific challenges

To discover and control quantum excitations that extend from nano-to-mesoscopic scales in heterostructures.

Excited magnetic, electronic, orbital, and many body states can span entire objects and are presently at the very limits of experimental control.

Challenges include dissipation, limited quantum coherence of existing realizations, competing low-energy interactions, and poor understanding of the relationship between structure and functionality. Understanding the timescales of competing interactions is particularly challenging.

Summary of research direction

Develop heterostructures that introduce new quantum degrees of freedom that have been previously manipulated incoherently but now provide the prospect of reaching new collective quantum states, including coherent states distributed across multiple nanoscale objects, quasi-particle condensates, coherent orbiton and magnon excitations, and long-lived temporal correlations breaking time-reversal symmetry.

Potential scientific impact

Control over low-energy excitations and fluctuations defines physical responses including electronic transport, magnetism, and optical properties. New excitations can also lead to entirely new properties such as unconventional superconductivity, topological states, qubits, and exotic non-linear phenomena.

Why heterostructures?

Tailor-made materials properties are required to produce and control these novel excitations. Heterostructures provide the means to build materials in which normally competing interactions are deterministically and separately manipulated via size, symmetry and composition. Realizations include ensembles of artificial correlated “atoms,” and designer frustrated magnetic systems such as quantum spin liquids.

2-3 Synthesize quantum heterostructures.

Scientific challenges

To design materials and structures to achieve and manipulate the desired quantum state.

To control composition, structure, shape and size.

To access and manipulate quantum states confined at or defined by the interface.

To decouple the heterostructure from the substrate in order to remove the effect of the substrate.

Summary of research direction

Comprehensive effort on the synthesis and *in situ* characterization of advanced heterostuctures.

Studies for heterostructures made with materials using different growth methods (chemical and physical deposition). Example – chalcogenides and oxides with properties tuned by strain, interface structure, symmetry, gating, 3D elastic states, interface curvature dimensionality…

Potential scientific impact

Flexibility to move beyond initial choice of elements and structures.

Deep understanding of the dependence of electronic and magnetic properties on material properties.

Gaining understanding of metastable states.

Deciphering the intrinsic quantum phenomena from that are induced by coupling to the substrate.

Why heterostructures?

Realization of "quantum science". i.e., the science of extended quantum states.

Overcoming obstacles to designing quantum materials.

New quantum materials for low-power, fast electronics, energy conversion within a structure/device.

2-4 Create true "hidden" order, states and structures.

Scientific challenges

To create new states of matter with novel properties that do not appear in the known phase diagram.
Examples include:

- Superconducting manganites
- Metallic antiferromagnetic phase in nickelates
- Ferroelectric metals in chalcogenides

Summary of research direction

Multimodal stimuli to drive materials into unexplored and unexpected regions of phase space. Requires detailed understanding of the phase diagram and tools to map out these regions.

Potential scientific impact

Nurture vs. nature in materials. Is the future of a material pre-defined by its genetics or can it be driven to wholly new places with the right external stimuli?

Why heterostructures?

Need new knobs to tune states: nanoscale structural control (e.g. quantum confinement), new symmetry breaking landscapes, build in proximity of phases…

3-1 Create, measure and manipulate skyrmions at nm length and ps time scales.

Scientific challenges

Realize, observe, and move sub-10 nm small skyrmions at room temperature.

Robust skyrmions that do not interact with each other.

Increase response time to ps.

Summary of research direction

Create Dzyaloshinskii-Moriya interactions at interfaces.

Form skyrmion/superconductor hybrid heterostructures to create Majorana fermions at interfaces, and then move the Majorana fermions.

Exploit thin films of AFMs: skyrmions at surfaces and domain walls.

Use strain to control of skyrmion size by combining piezoelectric and skyrmion materials

Potential scientific impact

New routes to Majorana fermions

Spintronics

Low power computing and nonvolatile memory

Why heterostructures?

Readily amenable to strain engineering.

Manipulate superconducting order parameter locally to turn the Majorana fermions on and off or to move them.

Readily amenable to use of scanning probe techniques for local manipulation.

3-2 Control dynamically topological states in nanostructures.

Scientific challenges

Enhance topological effects and signatures.

Room temperature operation.

Verify topological magnetization current and distinguish from competing effects.

Robust, lattice-matched (to topological insulators, …) ferromagnetic insulators at room temperature.

Mobile topological domains.

Summary of research direction

Improve spin-orbit coupling (SOC) using heterostructures (proximity effects).

Exploit wire (cylindrical) geometries.

Leverage advances in high-throughput theory for heterostructures.

Richer phases with strong correlations.

Exploit internal interfaces (domain boundaries,…) and topological domains.

Understand dominant interactions in heterostructures.

Create and destroy degeneracy in the quantum states: artificially create and tune across non-trivial states.

Determine robustness of topological states.

Potential scientific impact

Dissipationless devices.

Control topological effects for information storage and computing.

Beyond spintronics: Non-Abelian physics, quantum computing, RF, valleytronics, chiral currents

Why heterostructures?

Basis for technology and devices

Materials with properties not possible in bulk

Control with external parameters

Exploit stoichiometry to test topological protection

3-3 Manage coherence between quantum light and quantum matter.

Scientific challenges

Leverage quantum correlations in electronic materials to generate entangled and squeezed states of light, i.e. single qbits.

Probing electronic correlations within quantum states in matter with light.

Description of quantum materials in position or momentum space beyond classical resolution.

Summary of research direction

Develop coherent optical readout of quantum electronic states.

Use quantum material to generate an optical state.

Enable quantum coherence to be seen at higher temperatures.

Potential scientific impact

Leveraging quantum correlation in electronic materials for quantum state preparation

Light-matter interfaces for quantum information technologies (quantum repeaters etc.)

Better squeezed light sources for gravitational waves interferometry to magnetometry.

Why heterostructures?

Manage the photonic/solid state interface

Coupling between optically active and electronically active materials

3-4 Manipulate phonons to minimize decoherence and dissipation.

Scientific challenges

Maximize the coherence of quantum states or fluctuations.

Identifying relevant phonons and means to impact them.

Summary of research direction

Measure phonon gaps and phonon quantization in superlattices.

Transient or driven superconductivity (e.g., pump-probe)

Phonon squeezing

Potential scientific impact

Quantum thermodynamics

Achieve non-disparate gaps (e.g., superconducting and quantum spin liquids)

Reduce decoherence that comes with thermal fluctuations.

Minimize entropy contribution with phonon-driven states or heterostructures.

Improved thermoelectrics.

Why heterostructures?

Heterostructures for manipulation of phonons