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# Midscale Instrumentation for Quantum Materials



**NSF sponsored workshop report**

# Workshop Report on Midscale Instrumentation to Accelerate Progress in Quantum Materials

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Mid-scale Research Infrastructure from NSF's 10 Big Ideas  
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**Cover image:** Two different quantization conditions - imposed by magnetic field up to 45 tesla and by a super lattice - interfere to create a fractal energy landscape in graphene. Image adapted from reference [1] and also discussed in section 3.1.2.2.

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## Executive Summary

A broad category of “quantum materials” has taken center stage in condensed matter physics. Their exotic physical properties are stimulating fundamental progress in our understanding of electrons in solids and offer opportunities for transformative technological impacts[2, 3].

Essential to progress is the development and availability of powerful new experimental tools to synthesize quantum materials and elucidate their unique behaviors across a wide range of length and time scales and under a wide range of thermodynamic conditions. After a brief introduction to the main scientific challenges, this workshop report describes instrumentation to dramatically improve our understanding of quantum materials and help to harness their technological potential.

At the atomic length scale, matter exhibits wave-like characteristics: electrons tunnel through barriers, pursue multiple trajectories, and form standing waves when bound by nuclei within atoms. But while quantum mechanics ultimately determines materials properties at the macro-scale, matter wave characteristics generally fade from view through de-coherence in a quantum to classical cross-over regime. In quantum materials on the other hand, the veil is lifted on the quantum world and the deeply confounding features of quantum mechanics including entanglement, coherence, and quantum fluctuations directly impact materials properties and thus can be harnessed to create exotic and/or powerful new phenomena.

This report describes fundamental challenges in our understanding and ability to control quantum matter and the instrumentation needed for a *new* quantum revolution. We note there is not a single quantum materials problem, and as outlined in this report, dozens of unsolved states of matter remain, with, to date, only conventional superconductivity and the quantum Hall / fractionalized quantum Hall effect seemingly at a mature level of understanding. Let’s compare with the astounding success of LIGO in detecting gravity waves: For the first time, we are able to study the universe with means beyond photons and mass. Will this help us to understand Dark Energy and Dark Matter? In this report, because of the diversity and breadth of quantum materials, we cannot define one detector, but lay out a suite of midscale instrumentation techniques, that combined with the existing revolution in computation and materials growth, will transform our understanding and ability to design and control quantum matter. Phase coherence is key to our modern view of quantum materials. So, in discussing future instrumentation for a *new* quantum revolution, we emphasize tools that explicitly probe both wave function phase and coherence.

### Superconductivity (see section 2.1)

The phenomenon of superconductivity is a prime example of quantum physics holding sway beyond the atomic scale. Each holding identical electric charge, electrons repel each other and are obliged by the Pauli exclusion principle to occupy progressively higher energy states when confined within a metal. But in superconductors, a lower energy collective state is achieved when electrons form a quantum mechanical wave of “Cooper pairs.” It brings features normally consigned to the atomic scale to the macroscopic scale through unfamiliar physical properties including resistance-free electrical conductance and perfect magnetic screening (Meissner effect). A technologically viable superconducting material would transform major technological sectors including energy, quantum information, and transportation. However, controlling and enhancing

superconductivity presents enormous fundamental challenges. We must understand the interplay between charge, spin, and lattice degrees of freedom and their interactions and instabilities across multiple length and time scales in the presence of disorder. This work is of great fundamental importance and impacts many areas of condensed matter physics. The report describes targeted investment in specialized instrumentation needed to expose interacting electrons within solids and accelerate progress towards enhanced, practical superconductivity.

### **Quantum Magnetism (see section 2.2)**

Magnetic materials have drawn the attention of natural philosophers and driven technological progress for millennia. We now find there is a radically different type of magnetism driven by quantum fluctuations and entanglement with properties as thought provoking as the lodestone. Just as helium fails to solidify at ambient pressure, interacting atomic moments in quantum magnets form a perpetually fluctuating spin liquid within the crystalline solid. The apparent fabric of exotic alternative universes, such quantum fluids support collective quasi-particles described by quantum field theories analogous to the standard model of quarks, leptons, gauge bosons, and neutrinos that make up our own universe. As we learn to control and manipulate artificial collective quasi-particles within solids, these might become the basis for alternate quantum computational platforms.

### **Topology in electronic physics (see section 2.3)**

Classification of solids and their physical properties by symmetry was a cornerstone achievement of twentieth century solid state physics. In this century, it has expanded to the exploration of the powerful impacts of topology – loosely speaking “twist and connectedness” – upon physical properties. For example, when the topology of the electronic wave functions of an insulator is distinct from that in the volume outside of the solid, the result is a metallic surface layer “protected” by the topologically distinct character of the solid. The importance of topology in physics was discovered in studies of strongly correlated electronic materials and phase transitions that led to the 2016 Nobel Prize in physics. The study of topological materials is now a major focus that is bringing forth a range of unusual electronic transport and materials response functions that could have transformative technological impacts. For example, it is believed that topological superconductors could enable new types of decoherence-protected quantum information/quantum computing schemes with dramatic advantages over other forward-thinking schemes.

The variety of exotic properties associated with quantum physics at the macroscopic scale brings a scientific and technological quantum leap into view.[4] However, our understanding of interacting electrons in solids must substantially progress to accomplish the quantum leap. In this effort, midscale instrumentation will play essential roles that we classify and describe in 3 categories.

### **Forming Quantum Matter (see section 3.1):**

The richness of quantum materials science rests on the vastness of the materials space but our ability to identify and synthesize inorganic materials with specific physical properties is based on empirics and serendipity and is decades behind organic synthesis. To accelerate progress, a concerted effort and investments are needed in the science of inorganic synthesis. Beyond chemistry, extreme conditions of pressure and electromagnetic fields extend the variety of

quantum matter that can be formed in a given material. Examples of midscale instrumentation to realize radically new quantum-dominated states of matter include:

- X-ray, neutron, photon, and mass spectrometry probes for atomic scale real time monitoring during quantum materials synthesis.
- Solid state synthesis under extreme conditions including immersion in supercritical fluids at hundreds of bar.
- A national 2500+ ton multi-anvil high pressure synthesis facility.
- Crystal MBE where synthesis conditions are controlled and modulated to achieve artificial layering in the bulk single crystalline form.
- Coordinated nationwide network of MBE systems dedicated to distinct materials classes so individual scientist can form nanostructures of a wide range of materials through local and remote access to element specific MBE systems. Transparent sharing of growth conditions automated robotic sample growth and rich real time characterization during synthesis will be key elements of a successful program.
- A broader variety of tools for probing materials at static pressure extremes including optimized neutron and x-ray beam lines for rich characterization of small quantities of material under multi-extreme conditions of pressure and temperature (including low temperatures and high pressure).
- Reaching for more extreme pressure conditions. Static pressure systems reaching the TPa ( $10^7$  bar) regime.
- Dynamic compression systems with ultra-fast sub Å microscopy to access and characterize cold condensed matter.
- Facility for large volume synthesis and recovery of high pressure materials combined with metastable low-pressure growth by CVD.
- 135 tesla pulsed field system with time scales beyond milliseconds to explore quantum materials with functionality near room temperature including topological semimetals and high temperature superconductors.
- 60 tesla DC magnetic field and 40 tesla superconducting magnet system for complete characterization of equilibrium state of quantum materials under ultra-low T conditions and high pressure including by scan probe techniques.
- Ultra-high field magnets (DC and pulsed) at x-ray and neutron scattering facilities to expose the structure and dynamics of quantum matter that only exist at high fields.

### **Probing Quantum Matter (see section 3.2):**

Quantum materials present an array of new experimental challenges. They have multiple competing degrees of freedom that require the coordinated use of multiple experimental probes each placing conflicting demands on the sample shape, size, and quality. Quantum matter can be extremely sensitive to low level disorder so that the response and properties vary through the sample volume. This calls for spatially resolved advanced characterization. The quantum state of interest may only exist under extreme conditions of sample purity, electromagnetic fields, pressure, or temperature. This requires high sensitivity and the ability to focus on minute sample volumes and on short times scales where such conditions can be sustained. Finally, distinguishing the

unique properties of quantum matter from classical behavior requires novel probes and radical progress in use of existing probes. Unique solid-state properties that current instrumentation do not probe effectively include Berry curvature, quantum entanglement, multipolar correlations, and non-local quasi-particles. Magnetic field is a vector probe that explicitly couples charge and spin and can help disentangle them in the case of strong spin-orbit coupling. Perhaps most importantly, fields provide a bridge from quantum materials to quantum information science applications by providing a measure of coherence (e.g., orbital quantization) as magnetic fields directly tune electronic phases via the vector potential. Below are examples of midscale instrumentation to expose, understand, and control the quantum realm:

- The quantum microscope: Laser based real-time 3D imaging of quantum materials with multiple electronic contrast mechanisms.
- Transient hyper-spectral scan probe imaging of quantum materials and photoinduced phase transitions.
- The quantum factory: synthesis and fabrication of quantum materials and the associated devices. Based on scan probe technology made agile and capable of high throughput.
- Hybrid scan probe tools based on infra-red optics and atomic force microscopy to probe transient electronic phenomena in the femto-second to pico-second range using mixing and lock-in techniques.
- Scan probe spectroscopic imaging at magnetic fields beyond 20 tesla.
- Scan probe instrumentation to separately access intertwined superconducting and magnetic order parameters.
- Multi-spectral microscopically resolved spectroscopy: Time and space resolved nano-ARPES combined with time and space resolved resonant inelastic x-ray scattering.
- Scan probe microscopy and micro-wave spectroscopy coupled with x-ray tools.
- High efficiency fully polarized neutron spectroscopy on 100 mg samples of quantum materials spanning the energy range from 0.05 meV to 200 meV.
- Ultra-high DC magnetic field, fully superconducting, split coil magnet systems for neutron scattering and light scattering (FIR to x-ray and THz) experiments starting at 25 tesla and eventually reaching beyond 35 tesla.

### **Controlling Quantum Matter (see section 3.3)**

Quoting Ernest Schrödinger whose equation governs the quantum realm: “the phenomenon of entanglement is the essential fact of quantum mechanics, the fact that makes it so different from classical physics.” Entanglement can be exploited in quantum computers that one day might achieve “quantum supremacy” the ability to carry out certain key types of computation that classical computers practically cannot. While there is much ongoing research in quantum computation, there are also significant challenges. Many different possible realizations of the essential qubit are being pursued without clarity as to whether quantum coherence can be sustained long enough and for a large enough number of qubits to achieve quantum supremacy. The report describes the fundamental challenges to be overcome and outlines two examples of midscale instrumentation to accelerate progress.



- Integrated multi-modal in situ UHV materials analysis and processing system relevant for qubit technologies.
- Research-grade foundry providing hybrid quantum photonic integrated circuit technology to a broad set of users in quantum information processing research.

# 1. Introduction

This document resulted from a workshop that convened top scientists and engineers of the field (Participants in Appendix A) with the aim to expose the needs and opportunities for instrumentation in the midscale range to greatly accelerate progress in the formation, characterization, and applications of quantum materials.

The workshop on Midscale Instrumentation for Quantum Materials (MIQM) took place near the National Science Foundation during 2.5 days of December 2016 and was attended by approximately sixty leading scientists and engineers by invitation. The workshop focused on instrumentation in the midscale budget range from \$4M to \$100M, which has previously been underserved by NSF instrumentation programs. This focus notwithstanding, accelerated progress in quantum materials will require sustained, balanced investments in instrumentation spanning the full spectrum from the individual investigator scale to midscale to the facility scale.

Most plenary talks were recorded and are available along with slides at the workshop website: <http://physics-astronomy.jhu.edu/miqm/>. The workshop agenda is in Appendix B. The first day of the workshop was devoted to developing an overview of opportunities for new instrumentation to transform quantum materials. After the charge from NSF and plenary presentations by Dmitri Basov (Columbia), Darrell Schlom (Cornell), Russell Hemley (Carnegie and GWU), and Matthias Steffen (IBM) on instrumentation for quantum materials and quantum computing, the workshop separated into technical panels:

- (1) Beamline instrumentation
- (2) Microscopy, spectroscopy, and scanning probes
- (3) Extreme sample environments
- (4) Synthesis and materials discovery
- (5) Quantum structures and devices

Two leads for each panel organized the discussions and were responsible for reporting the deliberations in the panel reports that make up chapter 3 of this document.

The second day was devoted to identifying major scientific and technological goals in quantum materials and how these can be furthered by midscale instrumentation investments. Following a stimulating plenary presentation about the interplay of topology and interactions by Piers Coleman of Rutgers University, the instrumentation panel leads presented the outcome of their discussions. In the afternoon, the workshop participants were organized by scientific themes to discuss and prioritize how the advanced instrumentation they specialize in can contribute to accelerated development of

- (a) Superconductivity
- (b) Quantum Magnetism
- (c) Topological Materials
- (d) Applied Quantum Materials

Two leads for each panel organized the discussions and were responsible for writing the panel reports that make up chapter 2 of this document. The main workshop concluded with a summary from the scientific panel lead.

During the morning of the third day the panel leads, the co-chairs, and NSF program managers met at the NSF to discuss their findings and organize the report. Most of the report writing was done remotely after the workshop by panel leads and co-chairs.

## 2. Quantum Materials Research Objectives

Their great variety of electronic, magnetic, thermal, and mechanical properties and their responses to a variety of stimuli make quantum materials transformative from a fundamental and engineering perspective. This chapter provides an overview of quantum materials research, highlighting the major questions that continue to attract top talent as well as private and public funding worldwide.

### 2.1 Superconductivity

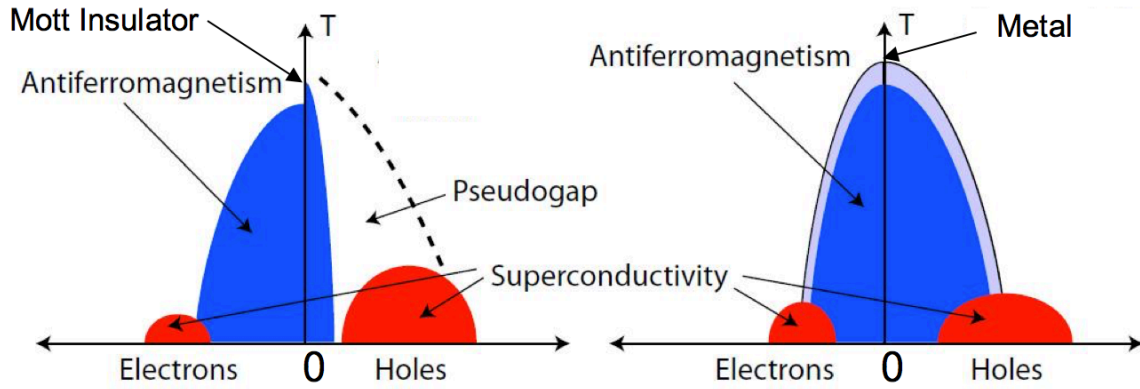
High-temperature superconductivity (HTS) holds the promise of revolutionary applications in electricity generation, delivery and storage, in energy efficiency, in information technology, and in medicine. By enhancing the correlations between electrons so that superconductivity can be achieved at even higher temperatures, quantum materials hold the promise of enormous advances in all of these fields. The interplay between superconductivity and correlated states of spin, charge, and the lattice is a rich area of condensed matter physics with profound intellectual links to other fields of science where new experimental tools are vital for progress. Current highest temperature superconductors have a complex atomic structure and are highly anisotropic. Practical applications may ultimately be made more viable with the discovery of more isotropic and less complex structures.

#### 2.1.1 Decadal Objectives

The phenomenon of superconductivity is associated with a phase transition wherein electrons pair up and condense into a single quantum state. There is no single theory of superconductivity that encompasses both conventional (“BCS”) superconductors mediated by interaction with the underlying lattice and unconventional superconductors, the latter generally characterized by an anisotropic pairing strength or order parameter. The vast majority of known superconductors lie outside the BCS electron-phonon mediated paradigm for a variety of reasons, including strong correlations, nontrivial order parameter symmetry, anomalous normal state properties, the existence of local magnetism, and other orders.

Nearly all unconventional superconductors exhibit similar phase diagrams that reflect competition between the different degrees of freedom associated with the electron, namely its spin, charge and orbital moment. The materials are nearly always characterized by some secondary order parameter, such as antiferromagnetism or charge/spin density wave order and superconductivity residing in a dome-shaped region of the  $p$ - $T$  or  $x$ - $T$  phase diagram, where  $p$ ,  $x$ , and  $T$  refer to pressure, doping and temperature, respectively. The phase diagrams associated with the two most studied unconventional superconductors, the cuprates and the Fe-based superconductors are shown in Fig. 2.1.1. Both of these systems display a myriad of complex phases dependent on whether they are doped with holes or electrons. A major focal point for superconductivity research over the next several years will be to obtain a detailed understanding of the universal physics at play behind these phase diagrams, ultimately allowing the control and identification of new superconducting materials.





**Figure 2.1.1** Schematic diagrams of on the left the cuprate superconductors and on the right, the Fe-based superconductors. In both cases the effect of electron doping is shown on the left and that of hole doping on the right.

### 2.1.2 Impact

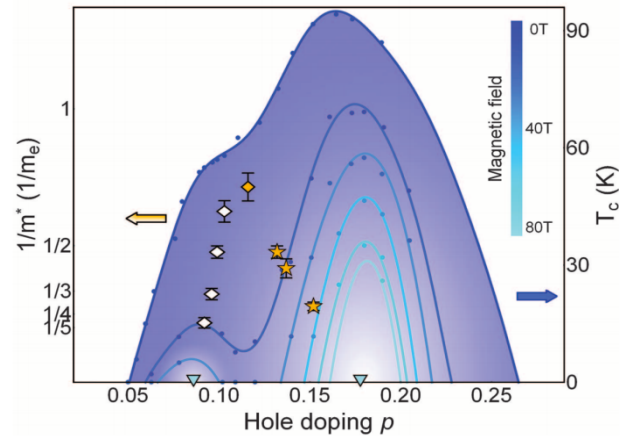
The requisite “quantum leap” in our understanding of superconductivity now seems likely to ultimately be driven by parallel innovations in multiple experimental probes that can expose the electronic interactions and/or intertwined orders underlying the complex phase diagrams. When enriched by theoretical work, such progress could well provide a pathway to the discovery of new superconducting materials. New classes of superconductors may in turn lead to new and revolutionary applications in electricity generation, delivery and storage, in energy efficiency, in information technology, and in medicine.

### 2.1.3 Challenges

Several key experimental developments are envisioned that will greatly expand our understanding. These include:

#### *In-situ probes in high magnetic fields.*

Strong magnetic fields are central to experimental studies of superconductivity. One of the primary uses is to suppress diamagnetism, providing access initially to the mixed state, in which vortices penetrate the superconducting state in the form of quantized vortices, and finally to the normal state, where superconductivity is completely suppressed. Access to the normal state allows measurements to be made of the ground state electronic structure or magnetic properties in the absence of superconductivity, which are necessary to ascertain the microscopics of the

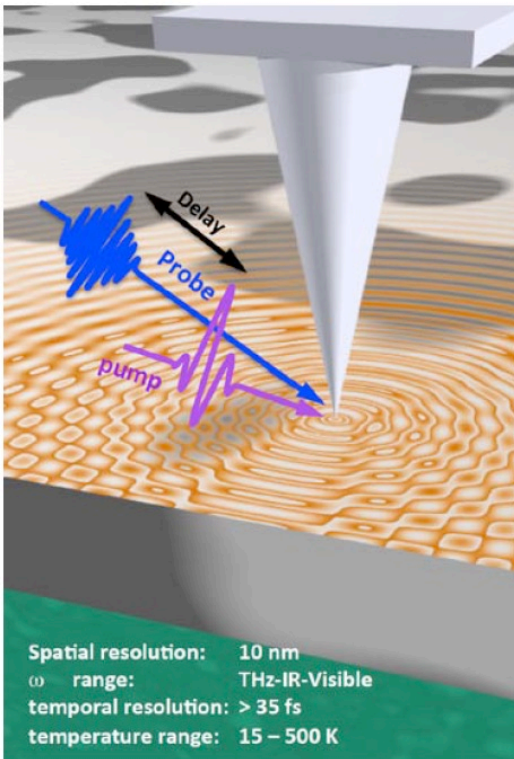


**Figure 2.1.2:** Contour plot of the hole doping dependent superconducting transition temperature of the high-temperature superconductor  $\text{YBa}_2\text{Cu}_3\text{O}_{6+p}$  under magnetic fields of varying strength. As the magnetic field is increased, superconductivity is suppressed and is found to persist only for doping levels close to  $p = 18\%$  at 80 tesla. The inverse effective mass (stars) is seen to fall linearly with doping, indicating a divergence in the effective mass and the existence of quantum critical points (triangles).

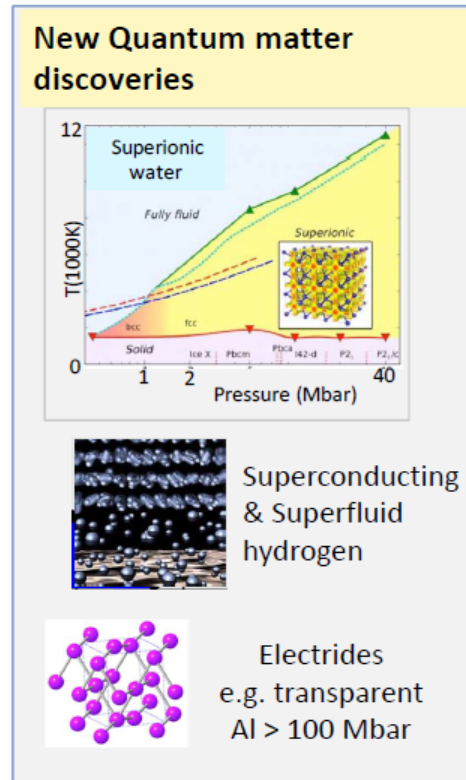
pairing interaction. The thermodynamic critical magnetic field at which superconductivity is destroyed provides a robust estimate of the superconducting coherence length, which is a key parameter for understanding the superconducting wavefunction. However current high field capabilities do not allow experimental access to the hidden quantum critical points as indicated in figure 2.1.2. Indeed next-generation pulsed high magnetic fields of 120 T to 150 T will be required to completely quench the superconducting state for detailed studies of the phase diagram within the superconducting dome at  $T = 0$ . A further area of need is to combine strong magnetic fields with sensitive thermodynamic measurements, as well as neutron and x-ray scattering techniques, for measurements on competing states that appear (or are revealed) only under a strong magnetic field. Competing broken symmetry states typically require magnetic fields in the range of 20 to 60 T, which are significantly beyond those provided at scattering facilities. The present world record magnet is 45 T for DC magnetic fields. Neutron and synchrotron beam experiments in such high field phases could produce a quantum leap in our understanding of strongly correlated electrons and high temperature superconductivity.

***New scattering and spectroscopy techniques:*** Strongly correlated complex quantum materials are often characterized by collective behavior that manifests on a wide range of length and time scales. This necessitates the development of new spectroscopic probes—based on photons, neutrons, and electron, for examples—that can interrogate these materials in previously inaccessible ranges of momentum and energy or, equivalently, distance and time. Such techniques should strike a balance between national facilities with supreme capabilities, such as free electron laser (FEL) and synchrotron light sources, neutron sources, and “table-top” techniques that provide more flexibility and easier access for a larger number of scientists. Laboratory-based Compact x-ray FELs also represent an exciting new development.

Key examples here will include capabilities in nanoscale angle-resolved photoemission spectroscopy (ARPES) that reveal information on single particle excitations in heterogeneous materials, near-field optical techniques capable of measuring frequency-resolved susceptibilities with nanoscale resolution (Fig 2.1.3), meV-resolved electron energy-loss (EELS) spectrometers that exploit aberration-correction technology for unprecedented energy resolution, new concepts in resonant inelastic x-ray scattering (RIXS) spectroscopy that employ dispersion compensation for high throughput, new nonlinear spectroscopy techniques capable of probing higher-order correlation functions revealing exotic symmetries, and high intensity polarized neutron scattering techniques to expose the spin texture of exotic quasi-particles in small single crystals. These techniques, and others still to be proposed, will provide a more complete picture in both space and time of the elementary excitations of unconventional superconductors and provide critical information about the inner workings of quantum materials. A continued theme will be increasing the energy resolution and the range of thermodynamic conditions (including low temperatures), so the most relevant low energy states may be directly accessed. Quite often these probes will be combined into a single facility. Thus, nanoscale ARPES providing information on the single particle excitations may be combined with nanoscale RIXS to provide complementary information on the collective modes of the very same sample volume.



**Figure 2.1.3.** Schematics of a nano-IR experiment. The infrared nanoscope is a hybrid of an atomic force microscope (AFM) scanner and an IR laser. Ultra-fast lasers allow pump-probe experiments at the nanoscale, limited only by the radius of the AFM tip.

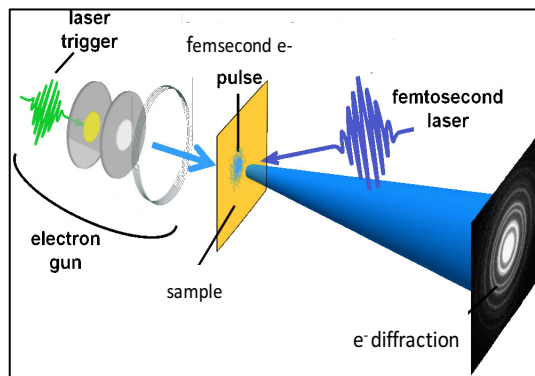


**Figure 2.1.4.** New states of matter achieved in high pressure environments.

**Improvements to high-pressure techniques.** The phase diagrams shown in figure 2.1.1 reflect different levels of chemical doping. Phase diagrams that report the effects of hydrostatic pressure on electronic phases of high-temperature superconductors are also prevalent in the literature. The recent discovery of superconductivity induced in hydride materials under high pressure has given considerable new impetus to the development of both improved high-pressure technologies and probes capable of exploring the properties of materials in this extreme environment (Fig. 2.1.4). Pressures in excess of 200 GPa are needed to study the next generation of hydride superconductors, predicted to achieve superconducting transition temperatures in excess of 250 K. Instrumentation that enables advanced materials characterization at extreme pressures have great potential for discovering new phases of quantum materials.

**Non-equilibrium studies.** Studies of materials beyond thermodynamic equilibrium have proliferated and are noticeably shifting the research agenda. Optically pumping materials far from equilibrium allows for the investigation of new states of matter (Fig. 2.1.5) and provides exciting opportunities to explore the interaction of excited electrons with collective modes, including phonons, CDWs and SDWS. Pumps combined with time resolved ARPES (trARPES) allow

detailed studies of electronic relaxation processes. Pumps combined with ultrafast electron diffraction can expose structural reorganization and relaxation. Soft x-ray FELs offer the possibility of time resolved studies at higher energies but there is a need to develop high rep-rate laboratory sources that operate at higher photon energies. Such developments will extend the momentum range accessible in trARPES.

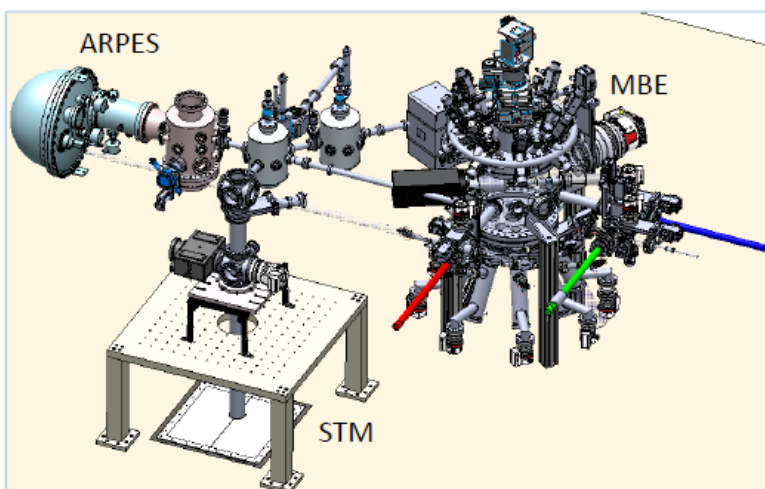


**Figure 2.1.5.** Ultrafast electron diffraction combined with pumps in the mid-IR provides direct access to time resolved atomic displacements providing direct access to discrete phonon modes.

**Improved techniques for materials synthesis:** A crucial component of these investigations will be improved materials synthesis including 3D crystal growth, molecular beam epitaxy (MBE), and pulsed laser deposition (PLD) techniques, which are well-suited for precision composition control needed to elucidate the physical origin of complex phase diagrams. Such developments will result in improved material quality, increased control over extrinsic effects such as disorder, precision tuning of composition to control material properties, and even to predictive materials design. These new capabilities should be combined with a range of characterization techniques; both single-investigator based capabilities and measurements that require the use of major light and neutron sources. An example is in figure 2.1.6, which shows the OASIS facility at Brookhaven National Laboratory (BNL) that combines MBE capabilities with ARPES and scanning tunneling microscopy (STM). Facilities of this type will allow the fabrication, characterization, and design of the functional materials of the future. Combining MBE and PLD synthesis capabilities with the characterization capabilities offered at the major facilities will greatly enhance our ability to understand, control, and discover new quantum materials.

## 2.2 Quantum Magnetism

Research into the field of quantum magnetism is of fundamental importance for understanding the effects of topology and entanglement in many body quantum wave functions. The 2016 Nobel Prize in Physics [5] is emblematic of this fact, where the new quantum states of one- and two-dimensional spin systems opened up fundamentally new paradigms to understand quantum electronic matter. The



**Figure 2.1.6.** The OASIS facility at BNL brings together synthesis in the form of MBE with the in-situ characterization tools of ARPES and STM.



spin systems at the core of the Nobel Prize winning discoveries constitute examples from a rich universe of exotic quantum matter that must be explored as modern technology reaches the limits of manipulating conventional electronic states. Expanding our understanding to the entire “periodic table” of quantum phases, to “molecules”, and more complex constructs made of exotic entangled states requires a quantum leap in experimental capabilities—one not dissimilar in nature to the transition from the Geiger-Marsden apparatus used in Rutherford’s gold foil experiments to the modern neutron scattering spectrometers needed for confirming and exploring the ramifications of the Nobel Prize winning theories[6, 7]. Achieving breakthroughs in understanding and exploiting emergent behaviors of highly entangled quantum systems requires great advances in instrumentation and scientific infrastructure. Such a leap in our ability to expose what is now prohibitively difficult to observe, or entirely inaccessible, requires developing experimental capabilities that allow precise manipulation and visualization of quantum wave functions with the goal of eventually scaling this control as needed for real world applications in materials science and electronics.

Quantum magnetism is an umbrella frontier under which a broad range of opportunities for both exploring and utilizing quantum matter reside. These opportunities range from establishing new concepts in many-body entangled quantum states to their deployment in devices[8-16]. Specifically, investigating the spin wave functions of quantum magnets is an essential first step towards manipulating and understanding large-scale entanglement in ensembles of a greater number of quantum constituents. By probing the dynamics of highly entangled states, exploring thermalization, ergodicity, and freezing in the quantum limit (where the electron cyclotron energy is much greater than the Fermi energy), we will be able to explore, tune, and tailor responses of exotic quantum phases for applications [11-24]. These studies of quantum magnetism advance our abilities to manipulate ensembles of quantum states needed for future technologies. In the far term, understanding entanglement in an extended network of quantum elements is a critical lynchpin for developing scaled quantum computing technologies, where such problems will inevitably arise upon scaling from a few-qubit system to massive qubit arrays. Exploring materials with tailored entanglement of quantum spins can help us to apply quantum coherence and entanglement across broad technological sectors including energy and information.

### **2.2.1 Decadal objectives**

Major advances are sought in both designing and understanding the responses of quantum magnets. Within a decade we can aspire to master the detection, creation, and manipulation of entangled states. Central goals are to understand how entanglement forms in magnetic systems, how it scales, and how we can exploit it for high-density qubit arrays. This involves manipulating spin wave functions at both the designer small- $N$  limit as well as exploring emergent states in bulk materials as a path towards understanding the universe of many-body entangled quantum states.

The ability to probe and tailor dynamics, responses, and the robustness of quantum-entangled spin states in complex networks should be developed. Additionally, efforts exploiting the availability of inherently large- $N$  quantum magnets in nature and phenomena in highly entangled quantum

spin systems should be harnessed to anticipate and understand emergent states/responses as quantum networks are scaled. Methods and experimental probes for identifying and probing the topology of quantum magnetic states will be developed as a means of exploring the expanding landscape of entanglement. To this end, an overarching goal will be to create and exploit emergent composite quasiparticles, fractional quantum numbers, and novel entangled states in magnetic materials [14-23].

Possible milestones include creating and understanding large-scale quantum entanglement of wave functions and excitations in quantum spin liquids [20-23] as well as the development of novel experimental techniques to identify and classify these states. A further goal includes developing avenues for entangling spin states with itinerant electrons. For instance, understanding the entanglement of itinerant electrons and topologically protected (Dirac, Weyl) charge carriers with quantum spins in strongly correlated materials is an emerging frontier (e.g. recently identified topological Kondo materials)[14-19, 25]. Additional tools for probing Berry phases and nonlocal order via spectroscopy and other techniques therefore must be developed. Understanding new emergent symmetries, chirality, and topologies that stem from spin-orbit coupling (SOC) is essential; and quantum magnetism, which arises from the spin-orbit entangled states of materials exhibiting both strong SOC and electronic correlations is a fertile realm for progress. Current mastery of interface states in designer heterostructures must also be expanded to realize and understand manifestations of quantum entanglement of spin and charge at interfaces and ultimately to explore emergent behaviors in devices[26].

### **2.2.2 Impacts**

We are on the cusp of a revolution in information storage and processing. Beyond the limitations inherent to using binary charge states for data storage, technologies that leverage nonlocal and protected quantum states can underpin a new arena for economic growth: the “quantum economy.” By developing the means of realizing and mastering entangled or topologically protected magnetic states, new avenues for quantum technologies will be opened.

Based on newly generated knowledge about quantum spin and charge entanglement, an array of technologies and materials impacting everyday life are emerging—from new sensors, to new superconductors, to computer memory and information storage systems. For instance, by exploiting new types of quantum charge and spin transport sensitive to quantum phase and topology, novel modes of dissipation-less charge transport[17, 18] can be developed. New spintronic devices[9, 10] that exploit spin entanglement in both bulk materials and at interfaces may be developed for enhanced information transport or for magnetically re-configurable devices[26, 27]. Topological defects in quantum magnets, such as magnetic skyrmion states, present new avenues for energy efficient memory elements. The entropic responses associated with the self-organization of these defect states and, more generally, in the formation of delicate many-body spin states, can be manipulated via small external fields, presenting new opportunities for low field magnetic cooling and sensor development. More broadly, insights gained via studies of entangled states in quantum magnets are essential for the realization of high-density quantum

computation where quantum many-body effects may eventually become relevant at the system level.

### **2.2.3 Challenges**

The most interesting and important insights into the fundamental properties of many-body entangled magnetic states such as quantum spin liquids requires measurements that cannot be undertaken with current instrumentation. This is due to a combination of dramatically insufficient experimental sensitivities to these states, undeveloped experimental capabilities needed to interrogate them, and a deficit in our capabilities to discover and synthesize the needed materials. Spin liquids and quantum glasses generally lack long-range order so that Bragg peaks and resolution limited spin waves are supplanted by broad, diffuse continua of fractional spin excitations[21-23]. Detecting and interrogating these fractional excitations is complicated by the suppression of measurable magnetic moment fluctuations by anisotropic g-factors and form factors associated with spatially delocalized magnetic states[28] and accessing the underlying quantum entanglement presents a further profound challenge. To realize the transformational opportunities inherent to mastering quantum magnetism, the following challenges must be met:

Enhanced abilities to control and perturb entangled quantum spin states must be developed. This involves developing sample environment systems for new extremes in low temperatures, high magnetic fields, and high-pressure capable of reaching currently inaccessible regions of phase space. This also crucially involves developing new ideas and infrastructure for tuning and probing entangled magnetic systems. Examples such as combined spectroscopy and diffraction using entangled probes of quantum-entangled spin states: spectro-interferometry needs to be explored. New means of performing spectroscopy and diffraction measurements on materials perturbed by short, pulsed magnetic fields, electromagnetic drives, and other sources will be required to explore the dynamics of many-body quantum phases, out-of-equilibrium entanglement, and the thermalization of quantum-entangled states. Magnetic fields play a dual role here – they are critical not just to form quantum states but also to probe the underlying properties of the quantum systems relevant to their functionality at zero magnetic field. Thus, higher DC and pulsed magnetic fields, are needed such as the 135 T pulsed and 60 T DC magnets, and magnetic fields need to be combined with spatially-resolved measurements, x-ray, neutron and other spectroscopic techniques.

To explore fractionalized and entangled spin states where real space correlations are often very short ranged and the corresponding measurement signal is exceedingly weak, an orders-of-magnitude leap in data collection rates for spectroscopy and diffraction is required. This would transform an impossible, year-long measurement into a feasible, few-days experiment. Invention, development, and deployment of novel multiplexing data collection systems for neutron and synchrotron x-ray techniques are needed, and new ideas for advanced scattering instrumentation have to be developed for this new class of magnetic materials. Examples include the development of new wide-angle spectrometers for cold neutron spectroscopy of quantum spin liquids and advanced x-ray polarization analysis capable of probing broad regions of momentum space in

resonant x-ray scattering studies at synchrotrons. There is a need for high-throughput polarized neutron capabilities to isolate the magnetic response of a much broader range of materials and ultimately make techniques like neutron reflectometry a transformational tool for exploring quantum magnetism in heterostructures[28]. This could enable frontier device characterization, including in-operando studies of technological devices that harness entangled magnetism.

A substantial bottleneck in the exploration of quantum materials stems from our limited access to the vast materials space. This limitation has been particularly significant in studies of quantum spin liquids,  $(4,5)d$ -electron, and  $(4,5)f$ -electron systems with strong SOC, and correlated topological compounds such as magnetic Weyl and topological Kondo systems. Exploratory synthesis of these materials is particularly challenging because small amounts of disorder can obscure experimental signatures. As a result, ultrahigh purity crystals are often required both from a materials and measurement standpoint to provide the needed experimental windows for discovery. Developing new capabilities for synthesis under extreme conditions of high-temperature and high-pressure synthesis, and pushing towards the extremes of actively controlled crystal growth techniques will add a transformative boost in the rate of novel quantum materials discovery. With new investments in the national materials synthesis infrastructure, the essential pipeline of new magnetic materials hosting exotic many-body quantum states can be broadened, allowing access to fundamentally new realms of metastable and currently hidden electronic states.

### **2.3 Topology in Electronic Physics**

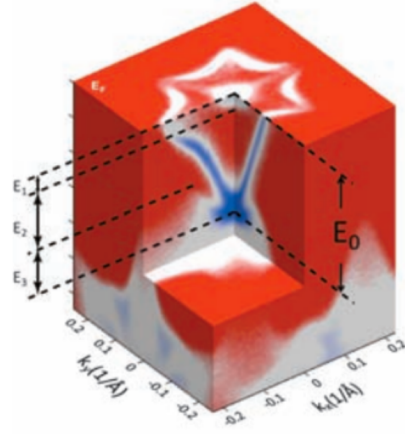
Beginning in the 1980's, physicists started to realize the key role that topology can play in electronic transport properties. Topology is for example essential for understanding the quantum hall[29] and the fractional quantum hall[30-32] effects as well as in the superconducting transitions[33-35] of ultrathin materials. While progress continued through the next few decades, the next truly major step came in the mid-2000's, with the discovery that materials with heavy elements (and hence strong spin-orbit coupling) can create their own very strong internal fields that can invert the valence and conduction bands of an insulator [36, 37], to create 2D[38] and 3D[39, 40] topological insulators. Soon many new and exotic types of properties and new quasiparticle excitations were theorized and/or discovered (Weyl points, Fermi arcs, Dirac loops, axions, triplons, Majoranas, etc). Interestingly several of these types of particles were proposed as fundamental particles but only discovered as quasi-particles within the confines of condensed matter. The corresponding new materials and new types of quasiparticle excitations are rapidly defining and advancing our basic understanding of a key new area of physics. These discoveries provide exciting opportunities to exploit topological quasi-particles for applications in spintronics and quantum computation. Major efforts around the globe are combining various aspects of theory, materials growth, spectroscopy, and device fabrication to understand and exploit new materials properties based on topology.

#### **2.3.1 Decadal Objectives**

We expect continued progress in the classification and discovery of new types of topological excitations, and the materials that harbor them. This includes various flavors of Dirac, Majorana,



Weyl, Dirac Loop, Fermi arc, Dirac lines, drumheads, band-touchings of many flavors, including linear, quadratic, and beyond. Spin-orbit interactions, time-reversal and crystal symmetries are key elements to produce topologically protected electronic properties while the key scientific methods include density functional theory, exploratory materials synthesis, and advanced spectroscopies.[41] As an example, figure 2.3.1 shows ARPES data for  $\text{Bi}_2\text{Te}_3$ , that provides experimental evidence for topologically protected surface states [42].



**Figure 2.3.1.** ARPES measurement of the topological insulator  $\text{Bi}_2\text{Te}_3$  [42].

The majority of the work in this area in the present decade has involved systems with minimal or only weak electronic correlations, and this enables use of density functional theory in the search for new phenomena and materials. It is believed that strong electronic correlations will bring an entirely new dimension to the field of topology, though theoretical tools to handle the combined effects of correlations and topology are in their infancy. It is envisioned that understanding the interplay between strong electronic correlations and topology will be a major challenge in condensed matter physics.

A critical aspect will be moving from the identification/classification of topological properties and excitations towards device structures that exploit the unique functionalities that topology can enable. This will likely dovetail with a concept that will become more and more important – that of controlling the ground state wavefunctions and the associated topological excitations. The existence of topological states of matter is a direct consequence of the quantum mechanical phase of a system of particles undergoing a twist. Can we learn to more directly image and then control this phase?

At present, most topological insulators are not actually insulating in the bulk because they are unintentionally doped. This is unfortunate because the bulk impurity states tend to short out and obscure the spin-polarized topological surface states. Can we synthesize topological insulators that are truly and robustly insulating so that we can isolate the spin-polarized surface states for potential applications?

We expect many efforts aimed at the development of methods and materials for isolating and controlling non-abelian anyonic quasiparticles. These are topological quasiparticles that are neither bosons nor fermions, so that the many body wave function picks up a non-trivial quantum mechanical phase under their exchange. Such particles are of great fundamental scientific importance and could potentially be the basis for (topological) quantum computation.

### 2.3.2 Impacts

Our appreciation of the topological nature of electronic physics has already completely altered our

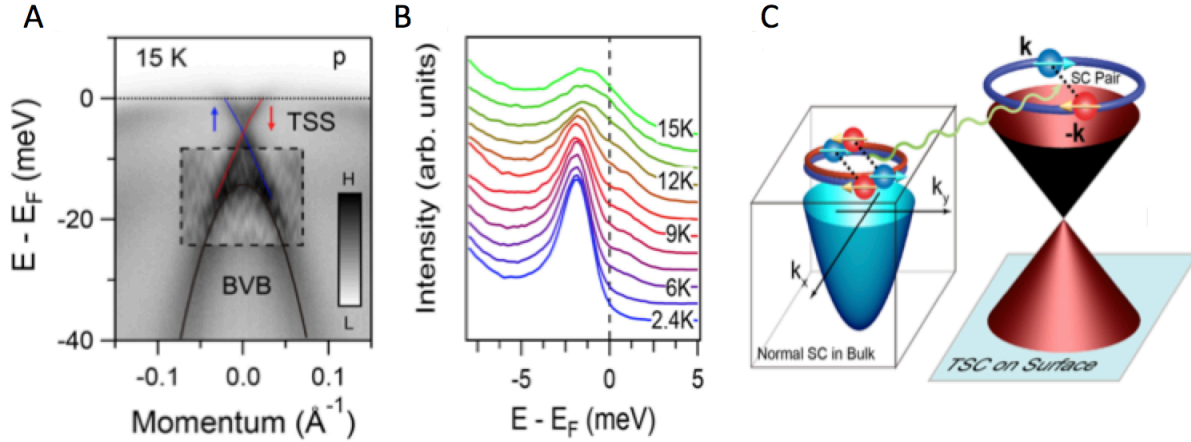
understanding of electronic materials, and led to the realization of quantum transport near room temperature and to the discovery of new types of topological excitations in solids. As we learn to predict and realize topologically protected electronic properties of bulk materials and devices we anticipate major technological impacts in crucial technological areas including energy, sensing, and quantum information processing.

### 2.3.3 Challenges

The community needs to continue developing advanced tools that can help us understand and control the topological character of materials. Especially with the push to study such things as topological superconductivity which has its most important signatures at very low temperatures and very low energy scales, the demands for more selective spectroscopies including higher energy resolution, lower sample temperatures, and higher fields will be much more severe, and new instrumentation will need to be developed. Though theory led the way in the initial phases of the topological revolution in materials science, continued progress particularly towards applications will be defined by advances in our ability to probe, visualize, and control topological states of matter.

ARPES and spin-ARPES have played a defining role in the discovery and elucidation of topological materials. The community was fortunate that much of this instrumentation was already available, largely built in the past two decades at synchrotron user facilities to probe high  $T_c$  superconductors. The scientific agenda that is pushing the field now demands significantly higher energy resolution, momentum resolution, spin-resolution (almost fully ignored in the US) and lower sample temperatures.

An example need for this improved resolution and sensitivity is the recent ultra-resolution ARPES work from Japan, which has uncovered a topological superconducting surface state in  $\text{FeTe}_{0.55}\text{Se}_{0.45}$ [43] (Fig 2.3.2), at present making this material the highest-temperature confirmed topological superconductor and a prime candidate for creating Majorana excitations that could be used for future quantum computing applications. The energy scale of the topological superconducting states is of order 2 meV. Probing or manipulating these states thus requires energy resolution of this order or better. For comparison 10 meV energy resolution is considered a high energy resolution ARPES experiment at one of the US synchrotrons while the very highest possible resolution there is 4 meV. Laser-ARPES enables higher energy resolution[44] that can even be better than 1 meV[45]. Such facilities are however, not available to many users and the  $k$ -space that can be accessed is severely limited. It is envisioned that an ultra-resolution ( $< 1$  meV) ultra-low temperature ( $< 1$  K) flexible ARPES instrument at a synchrotron with a wide range of photon energies, perhaps coupled with an STM and sample growth capabilities, would give ground-breaking new capabilities for the discovery and development of new topological superconductors – the critical ingredient for certain classes of quantum computation.



**Fig 2.3.2** Ultra-resolution ARPES utilized to determine the topological superconducting state in  $\text{FeTe}_{0.55}\text{Se}_{0.45}$  [43]. No user facility in the US has the capability to do ultra-resolution ARPES.

High magnetic fields are another critical probe of the band structure of topological materials and can identify Weyl and Dirac materials via Landau-level spectroscopy.[46-50] While lower fields probe the inherent band-structure, higher magnetic fields act as a tuning parameter that can reach beyond the quantum limit of topological insulators, e.g. Fig 3.1.7.[51-56] Finally, it is the coupling of the electronic spin and orbital degrees of freedom that enables topological band-structures to emerge in a majority of topological materials. Thus, applied magnetic fields, which inherently couple to both spin and charge, play a fundamental role in probing and manipulating the underlying topology of materials. Fields beyond 100 T such as the proposed 135 T millisecond magnet are needed to reach beyond the quantum limit of a large number of topological materials, even those with small Fermi surfaces.

One of the major challenges in general is to interrogate and control the quantum mechanical phase of a system, as opposed to just the amplitudes that most spectroscopies are sensitive to. New developments along these lines are strongly encouraged, for example phase-sensitive Josephson Junction-based STM experiments[57], or THz optical experiments where the electric fields (and not just amplitudes) are explicitly constrained. There may also be ways in which neutron interferometry can be used to directly detect solid state entanglement.

Ultrafast time resolved experiments, allowing for the creation of new types of non-equilibrium (Floquet) states and probing the associated short-lived atomic scale correlations and electronic properties help us move towards controlling electrons at the atomic scale. Such experiments can make important connections to applications of advanced electronic materials.

### 3. Instrumentation to enable the Quantum Leap

We have organized this central part of the report into three sections that emphasize distinct components of the experimental program. First and foremost, we must form novel quantum materials and novel quantum states of matter and this area is described in section 3.1. This area not only encompasses bulk and thin film synthesis but we include methods to perturb materials at low temperatures including high pressure and high magnetic fields because these may be considered as means of forming new states of matter within a given solid. Section 3.2 covers enhanced methods to probe quantum materials over many length and time scales and with a variety of different contrasts and sensitivities. Finally, section 3.3 discusses a specific application of quantum materials; quantum computation. Other applications are discussed in other areas of the report but quantum computation takes center stage as it relies on the quantum nature of electronic states to realize a new and potentially transformative technology.

#### 3.1 Forming Quantum Matter

##### 3.1.1 Synthesis

Contemporary areas of fundamental physics, such as high energy and astroparticle physics, are propelled by the advancement of sensitive (and usually large) ‘telescopes’ or ‘detectors’ that open new windows into hidden aspects of the universe that are only manifest beyond the reach of quotidian reality, whether at the galactic or subatomic scale. Quantum materials provide a realization of similar science on a benchtop, serving both as artificial ‘mini-universes’ that host emergent quantum phenomena, while at the same time serving as ‘detectors’ of these very phenomena. The excitement generated by the study of quantum materials arises from unique opportunities to *design, create, measure, and exploit new quantum mechanical behavior* that has *never before existed* in the universe. The construction of these benchtop mini-universes and detectors, in other words the synthesis of quantum materials, has played, and will continue to play, a pivotal role in this research enterprise. Without synthesis, major condensed matter physics discoveries, such as quantum Hall insulators, high temperature superconductors, topological materials, and quantum spin liquids, would not have occurred. Whether the underlying theory for these discoveries came first or later, the relevant emergent quantum phenomena could not have been realized without a synthesized material to measure. Apart from having a direct impact on curiosity-driven discovery, the synthesis of quantum materials has also been the driver for critical advances in information technology, ranging from the transistor, magnetic memories, and solid-state light emitters to the rapidly advancing frontiers of contemporary quantum information technology.

Since synthesis plays such a central role in the exploration of quantum materials, investments in this field necessarily take a different form than in many other fields: Not only is there not ‘one technique’ that dominates how quantum materials are made, but we understand so little about the fundamentals of how materials form, that we do not even know all the techniques that are possible. This makes for a very exciting environment, one wherein individuals or small teams can make contributions as effectively (if not more so) as large collaborations. This naturally leads to the “let

a hundred flowers bloom” funding model. But it also helps define an area for significant targeted future investments that could fundamentally transform the field of quantum materials. We envision three parallel tracks in this context: the ‘science of synthesis’ aimed at elucidating how materials are made and why they form; the development of low cost crystal growth techniques aimed at the rational control over nanoscale modulation and heterogeneity in macroscopic crystals; and the development of a ‘cloud’ approach to sophisticated epitaxial growth via a nationally coordinated network of epitaxial growth nodes.

### ***3.1.1.1 Searchlights for Quantum Materials Discovery***

Almost by definition, the field of exploratory materials synthesis is all about the crafting of new materials – whether by design or not – and, in particular, doing so in a regime (i.e. parameter space) where existing theories do not make robust predictions. That is, just as the materials genome initiative is focused on expanding the range of situations in which computational methods and big data analytics make concrete, valid predictions, exploratory materials synthesis focuses on expanding the range of possible synthetic conditions, and opening synthetic territory that was not previously accessible.

Specific to quantum materials, there have been remarkable advances in the last 20 years and especially in the last few years of theoretical predictions, going from vague concepts to making concrete, specific materials predictions. In the context of non-interacting phenomena, such as topological band structures, these predictions have been particularly well validated. We are now at the point where in certain cases the ability of theory to make concrete predictions has far exceeded our ability to actually create the proposed materials. Thus, a central challenge that needs to be resolved is to develop synthetic routes for comprehensive exploration of predicted materials.

This is a difficult enterprise because, fundamentally, we do not understand the chemistry of solid-state synthesis sufficiently well to *a priori* design a synthesis protocol for an arbitrary new quantum material (except in very limited circumstances). To design a synthesis route, we need to know many things: what are the individual mechanistic steps, i.e., the individual bond making and breaking steps that govern how a synthesis proceeds? How do external perturbations affect these reaction pathways? What is the unifying organizational framework for these reactions, and how does that gain us the ability to rationally design syntheses for arbitrary target materials? When we consider heterogeneous thin films such as epitaxial heterostructures and multilayers, these questions become even more complex since they are affected by other factors such as strain and far-from-equilibrium crystal growth conditions. Answers to these questions in the realm of solid state materials, particularly quantum ones, are mostly unknown. The discovery of revolutionary quantum materials clearly requires significant investments to advance solid state materials synthesis. For quantum materials, this has not occurred over the last 30 years. This stands in contrast to porous and nanoscale materials synthesis, both of which have seen significant investment in the chemistry community.

### 3.1.1.1.1 Scientific Requirements

#### Superconductivity

As we continue to struggle with the challenging mysteries presented by high temperature superconductors, a variety of new materials keep emerging with the tantalizing promise of alternate routes to unconventional high temperature superconductivity. A primary goal that emerges in this context is to develop routes for enhancing the relevant correlations between electrons in crystals with simpler, more isotropic structure. While bulk crystal growth will continue to be a mainstay in this exploration, the emergent properties enabled at heterogeneous interfaces will likely play an increasingly important role in this endeavor. A key example is provided by the exciting discovery of high temperature superconductivity at the interface between SrTiO<sub>3</sub> and FeSe[58]. The development of thin film synthesis capabilities with rapid *in situ* characterization of electronic structure, crystal structure, and transport will be a powerful tool to advance this frontier.

#### Quantum magnetism

As outlined earlier, a central goal in research on quantum materials is the large-scale quantum entanglement of wave functions and excitations. Quantum spin liquids provide a particularly exciting frontier in this context, one that has almost exclusively been driven by traditional synthesis routes such as bulk crystal growth and organic chemistry. Developing materials at the nexus of topology, interactions, and quantum magnetism could vastly expand this space.

#### Topological materials

Rapid advances have been made over the past 8 years in using bulk crystal growth and thin film deposition to realize a variety of predicted ‘topological materials,’ including topological insulators, topological crystalline insulators, topological superconductors, Weyl semimetals, and Dirac semimetals. Yet to be verified theoretical predictions still abound, suggesting that many more possibilities for topological materials exist in the vast database of materials that can in principle be synthesized. For instance, a recent theory of ‘topological quantum chemistry’ identifies non-trivial topological band structures for all possible 230 crystal symmetry groups[59]. Even this endeavor leaves out the relatively unexplored emergent phase space that is created by the effects of interactions on topological states. Finally, ‘emergent broken symmetry surface states’[60] arise when we systematically control some fundamental symmetry in a topological material. Examples include the breaking of gauge symmetry (Majorana modes) and time reversal symmetry (quantum anomalous Hall insulators). All these ideas provide clear scientific requirements that will help guide the development of instrumentation for the synthesis of quantum materials: rapid exploration of potentially interesting topological materials identified by topological quantum chemistry, rational design of topological materials with enhanced interaction effects, and the systematic engineering of topological materials with some broken symmetry.

### 3.1.1.1.2 Midscale Instrumentation

Retrosynthetic analysis, or the ability to design a series of reaction steps to produce a target

molecule, is the backbone of modern molecular and biochemistry: virtually every advance relies on the ability to routinely and selectively make specific compounds. It was enabled by 150+ years of exploration and analysis of molecular reaction mechanisms to create a versatile and well-understood toolbox of selective molecular reactions. There are many isolated examples of selective chemical reactions for solid state materials, but a lack of the mechanistic details severely limits their universality (i.e. scope). It also results in significant irreproducibility between different synthetic groups, or even between instruments within a group – e.g. a problem recently discussed at length for CVD synthesis of transition metal dichalcogenides. A platform for rigorously and systematically exploring the mechanisms of growth in solid state materials would be transformative, allowing for true materials synthesis by design. In terms of precise instrumentation, this requires the standup of converged synthesis-probe equipment to probe structure and dynamics – from the atomic to the macroscale and from the picosecond to hour timescale – in real time and under representative synthetic conditions, e.g. by combining x-ray, neutron, photon, and mass spectrometry probes in operando synthetic equipment. In addition to providing the capability, the mechanism of access must allow for extended studies, on a range of materials systems.

In addition to understanding mechanisms of reactions in existing synthetic approaches, it is equally important to expand the range of available synthetic capabilities. Recent advances have been made in the area of high pressure – the capability to carry out syntheses in supercritical fluids up to 300 bar at the “Platform for the Accelerated Realization, Analysis, & Discovery of Interface Materials (PARADIM; an NSF Materials Innovation Platform) is an example – the variety and availability of such unique capabilities should be expanded. On a longer timescale, it is exciting to envision entirely new synthetic approaches that might be possible. For example: what if, by harnessing the super-/sub-melt saturation composition oscillations that occur in crystal growth, one could controllably build in arbitrary layering, on an atomic layer by atomic layer basis, at the growth front ... i.e. MBE in a “crystal”? But it will also not be easy – in addition to knowledge of the kinetics and thermodynamics parameters (see above) – this requires the ability to, via the control of external variables, dynamically and systematically control fluid flows in a melt. This will require a concerted effort – combining state of the art tools for fluid dynamics modeling and parameter optimization, with new synthetic tools that allow for perturbations of the solid-liquid interface in real time. Such crystal MBE synthesis would be transformative – allowing for the full weight of available characterization tools to be applied to systems previously only available in fragile, small volume form. Crystal MBE would open up the use of complex heterostructures in new technologies.

Molecular beam epitaxy (and other ultrahigh vacuum deposition techniques) have already proven capable of creating a variety of quantum materials, resulting in major discoveries such as the quantum Hall effects (in the integer, fractional, spin, and anomalous realizations) and emergent phenomena at heterogeneous interfaces (e.g., ferromagnetism and superconductivity in oxide heterostructures). Some of these phenomena are the result of synthesizing thin film materials of exceptional perfection (such as GaAs/(Ga,Al)As heterostructures), while others have exploited the realization of novel materials (such as magnetically-doped topological insulators) and novel

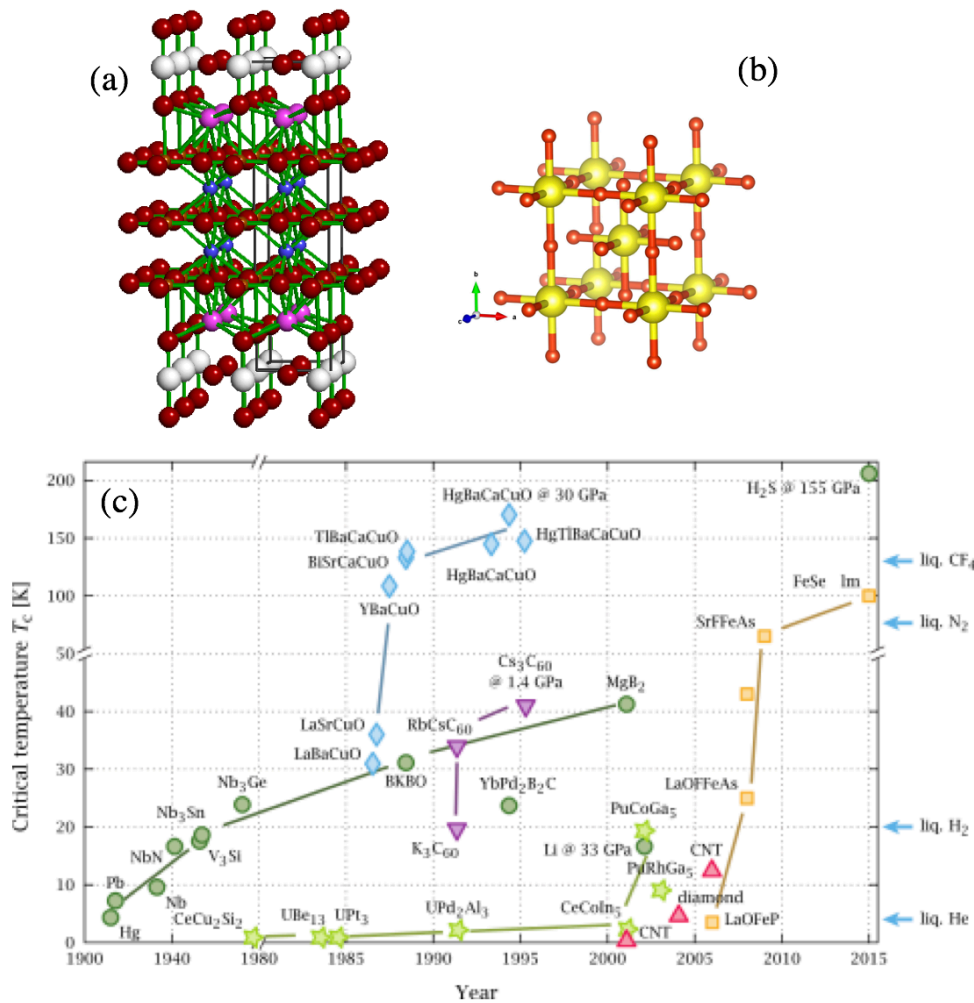
heterointerfaces (such as  $\text{LaAlO}_3/\text{SrTiO}_3$  and  $\text{FeSe}/\text{SrTiO}_3$ ). But the path towards such discoveries has been slow because these thin film deposition techniques are inherently expensive and limited in the scope of materials that can be rapidly explored. For example, if a crystal growth lab is focusing on the growth of topological insulators derived from the tetradymites (Bi- and Sb-chalcogenides), its operations cannot quickly shift toward the exploration of some other newly predicted material without expensive investments in time (cleaning, followed by bake out) and equipment (new effusion cells). If a crystal growth laboratory trains students and postdocs in MBE, given the high cost (about \$1M) of setting up individual MBE systems with limited materials scope, the alumni of such a group have limited opportunities to readily start their own high impact MBE programs as beginning assistant professors. A funding model that focuses on increasing support for the purchase or construction of individual MBE systems by single PIs will simply be ‘more of the same’ and may not provide the accelerated scale of discovery required for revolutionary advances in quantum materials. An alternate model might involve the funding of a coordinated nationwide network of MBE systems that are both locally and ‘remotely’ accessible to users. The coordination of materials synthesized at different nodes of this network and the transparent sharing of relevant data (growth parameters and resulting sample characteristics) would be crucial to its success. We note in this context that even when MBE growers report parameters such as beam fluxes in published work, these are often not easily translated into useful parameters for reproducing the growth in another chamber since the chamber characteristics (such as the source flange geometry and the source-substrate distance) are not typically reported. An essential aspect of a ‘cloud’ approach to MBE will be the wider adoption of automated robotic sample growth, an approach already in place in many industrial MBE operations and in a few academic laboratories. Another important aspect will be the more widespread adoption of *in situ* real time diagnostics of thin film growth and more standardized growth parameters: while reflection high energy electron diffraction is a standard real-time tool in MBE, *in situ* optical probes such as ellipsometry or x-ray techniques are less commonly used and would greatly advance the understanding of the growth process.



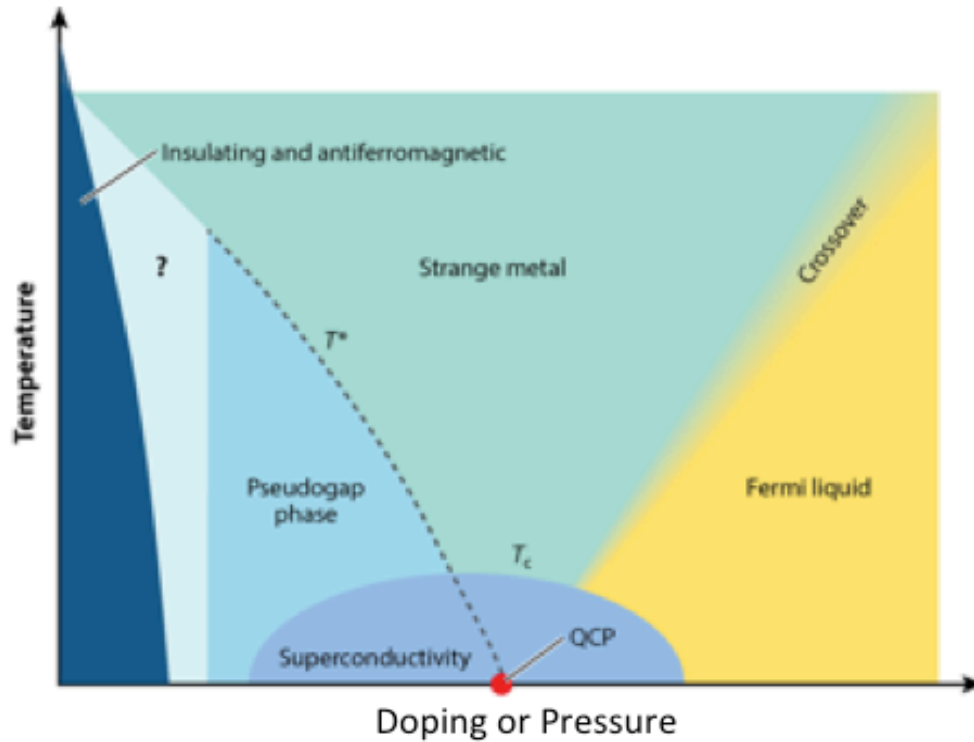
### 3.1.2 Extreme Conditions

#### 3.1.2.1 Pressure

Recent advances in the generation of extreme pressures under static and dynamic conditions in the laboratory are creating new opportunities for quantum materials. Static pressures up to 5 Mbar (500 GPa) can be reached in the laboratory achieving volumetric compression of many solids by an order of magnitude or more and providing exquisite control of interatomic distances that impact quantum properties. These advances have led to the discovery of altogether new phases and phenomena, and the prospect of synthesis and recovery of novel quantum materials for applications. With new dynamic compression techniques, still higher pressures at modest temperatures are possible, enabling experiments in previously inaccessible  $P$ - $T$  regimes of condensed matter where altogether new quantum behaviors are predicted to occur. Developments at major x-ray, infrared, neutron, and laser facilities, as well as improvements in dynamic compression methods, are enabling these advances in materials research at extreme conditions.



**Figure 3.1.1.** (c) Timeline of superconductivity (<https://en.wikipedia.org/wiki/Superconductivity>), showing the highest critical temperatures are those measured under pressure for (a) HgBa<sub>2</sub>Ca<sub>2</sub>Cu<sub>3</sub>O<sub>8.6</sub>[3,4] and (b) 'H<sub>3</sub>S'[5].



**Figure 3.1.2.** “Universal’ quantum phase diagram unconventional superconductivity near the antiferromagnetic quantum critical point. [61]

For all of these areas, mid-scale instrumentation is essential for the next quantum leap in understanding.

#### 3.1.2.1.1 Scientific Requirements

**Superconductivity:** There are exciting implications for superconductivity. To date, some 23 elements have been converted from non-metallic and/or non-superconducting states into new superconductors under pressure, including some with high critical temperatures.[62] The highest temperature superconductivity has been observed in materials under pressure, starting with the cuprates ( $T_c = 165$  K at 30 GPa)[63, 64] and now with the recent discovery of superconductivity in simple hydrides ( $T_c = 203$  K for  $H_2S$  above 150 GPa; Fig. 3.1.1).[65] High-pressure studies can expose mechanisms of superconductivity and lead to new superconductors that function above room temperature.

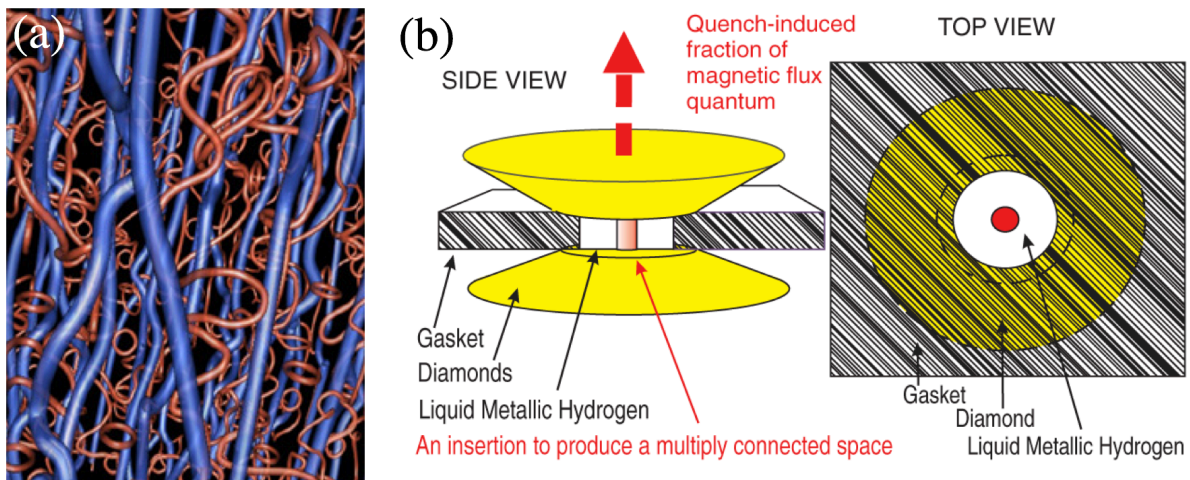
**New ferroelectric, multiferroic, and magnetic materials:** Compressing materials produces remarkable changes in electronic and magnetic properties and entirely new phases that in some cases can be recovered to ambient conditions. Where conventional Neel magnetism is suppressed, new forms of quantum matter can emerge including. Examples include unconventional superconductivity (Fig. 3.1.2), colossal ferroelectricity, multiferroics, spin liquids, and other strongly correlated electron systems.[66-68] As access to more extreme conditions of pressure and temperature is expanded so are opportunities to understand universal features of the phase diagram for strongly correlated electrons.

**Topological and 2D Materials:** Pressure provides a powerful means for exploring the behavior of topological and 2D materials. High-pressure transitions in known topological insulators[69] and 2D semiconductors and semimetals produce new phases, including topological insulating phases from ambient pressure metals with topologically protected surface states.[70] To understand these phenomena, new high-pressure tools need to be developed, including probes of surface states of materials under pressure.

**New phenomena in molecular systems:** There are also potential applications for the study of highly compressible molecular systems. Oxygen,  $\text{H}_2\text{O}$ , and other simple molecular materials transform to new structures with altered chemical bonding at megabar pressures[71-74] and provide the prospect of uncovering even more exotic states of systems at much more extreme conditions, with implications for our understanding of quantum materials and planetary science.

**Driving metals into insulators:** Not only can pressure turn insulators into metals, it can turn metals into insulators. These remarkable quantum phases, now known as high-pressure electrides, are characterized by interstitial electron localization.[75, 76] These materials can form from simple metals at pressures below 200 GPa.[77] Understanding the nature of these new quantum materials requires the development of new *in situ* techniques as well as reaching still higher pressures.

**Novel phenomena in dense hydrogen:** Hydrogen is a fully quantum mechanical system over the entire range of conditions. How its quantum properties evolve as a function of density is not well known though it is important both for condensed matter physics and astrophysics.[80, 81] Solid At sufficiently high pressures, hydrogen is predicted to be a high temperature superconductor well above room temperature,[82] and to have a fluid ground state that is both superconducting and superfluid – an altogether unique state of matter (Fig. 3.1.3).[78, 83] The search of these novel phenomena requires new classes of experiments using multiple extreme environments of pressure above 400 GPa, temperature, and magnetic fields.

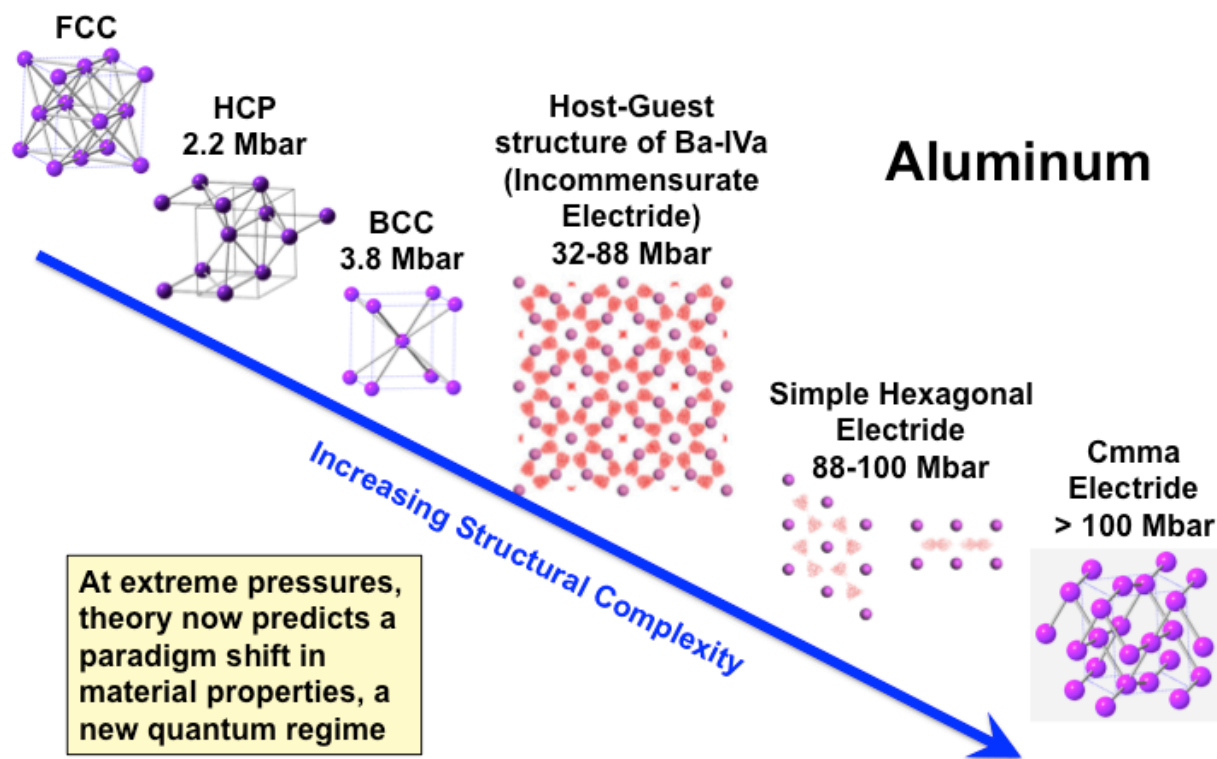


**Figure 3.1.3.** (a) Vortices in superconducting superfluid hydrogen predicted at high pressures above 400 GPa.[78] (b) Proposed experiment as a function  $P$ - $T$ - $H$  to test the prediction.[79]

**Unexplored regimes of cold dense matter:** Theory predicts remarkable structures of even simple elements when they exist under much more extreme conditions of pressure. Examples include Al and C, which may form complex electride-like structures up to multi-TPa pressures (Fig. 3.1.4).[79, 84] The pressures of these predicted exotic structures can now be explored with new dynamic measurement techniques but new probes are needed. New diagnostics, including measurements of temperature, on dynamic compression need to be developed in order to test these new predictions.

**Coupling theory and experiment:** Close interaction between theory and experiment has been an important element of high-pressure work to date. An example is the successful prediction of new superconducting hydrides and electrides,[76] but in other cases experiments have defied theoretical predictions. There is a need to develop a general theory of kinetic stabilization of novel high-density phases. We also point out that high-pressure science is a natural platform to learn and comprehend quantum mechanics. Greater synergy between theory and experiment is necessary to understand and guide the creation of new quantum materials.

**Materials synthesis:** There are important needs and opportunities for creating new quantum



**Figure 3.1.4.** Emerging structural complexity predicted in aluminum at extreme pressures.[79] All structures near 30 TPa are far from close-packed. Related behavior is observed experimentally in Li and Na.

materials discovered at high pressures for recovery to ambient conditions. In addition to direct high pressure synthesis, alternative synthetic routes such as CVD synthesis of metastable high-pressure phases by chemical kinetic stabilization may be employed (Fig. 3.1.5).[85] New quantum materials discovered at high pressures can be synthesized by various routes for study and use under ambient pressures.

### 3.1.2.1.2 Midscale Instrumentation

***New tools for probing materials at static pressure extremes:*** Despite recent progress, the sample containment devices used in high-pressure experiments impose limits on what can be measured. A new generation of instrumentation is necessary, including sensors embedded in diamond anvils for transport and magnetic measurements, nanofabrication of smart devices, fast spectroscopies (*e.g.*, 2D IR), and magnetic resonance methods.[86] This new suite of tools includes probes of 2D properties of interfaces, grain boundaries, and mesoscopic structures.

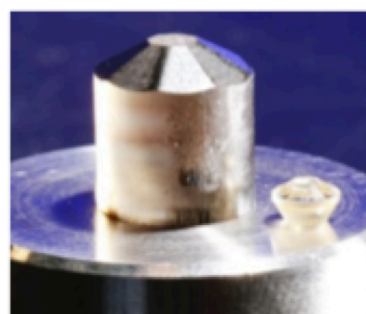
***X-ray and neutron facilities for pressure-induced quantum matter:*** Beamlines at national x-ray and neutron scattering facilities need to be upgraded, optimized, and equipped – and in some cases built – for a detailed view of pressure induced quantum matter. Synchrotron x-ray facilities will need to take advantage of new sources optimized to produce coherent x-rays, while new neutron instrumentation will need to be optimized to provide greater access with multiple probes. New diagnostics are needed for optical lasers and XFELs to direct the measurement of quantum properties.

***Reaching more extreme pressures:*** Reaching still higher pressures with both static and dynamic compression methods is required to address a number of the above questions. For static compression, this development includes further optimization of cell designs to reach TPa pressures.[87] For dynamic compression, the challenge is reaching and measuring lower temperatures at these and much higher pressures[88] to explore the new domain of ‘cold’ dense

(a)



(b)



**15 carat CVD  
diamond anvil**

**Figure 3.1.5.** (a) high-pressure apparatus. (b) 15 carat diamond anvil produced by CVD compared to a conventional 0.3 carat natural diamond anvil (courtesy of R. Boehler, Oak Ridge National Laboratory).



quantum matter where there is the potential for discovery new physical phenomena.[83]

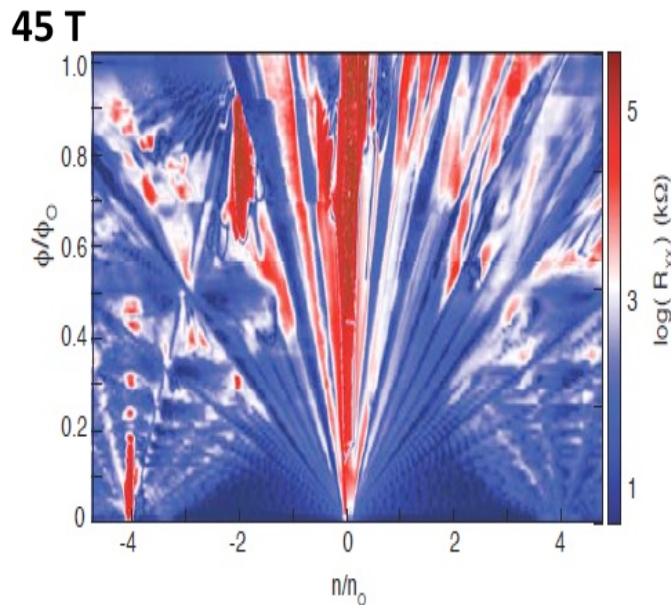
**Multiple extreme environments:** For many of the questions addressed above, new probes for measuring properties of materials under multiple extreme environments need to be developed. This effort includes extending measurements of Fermi surfaces under combined  $P$ - $T$ - $H$  conditions. In addition, there is the prospect of uncovering new phenomena by combining ultrahigh pressures with ultrahigh magnetic, electric, and radiation fields.[74, 78]

**New diagnostics on dynamic compression facilities:** A new class of quantum matter exploration can be launched to exploit existing dynamic compression facilities combined with sub-ps, sub-Å microscopes. Laser wake field acceleration of e- and THz energy enable  $\sim 10$  fs, sub-Å resolution electron microscopy/diffraction at  $>100$  MeV to explore many of the phenomena described above. With proper investments, new laser-driven plasma and electron sources with Compton scattering or B-modulation will enable the interrogation of new phases.[89, 90]

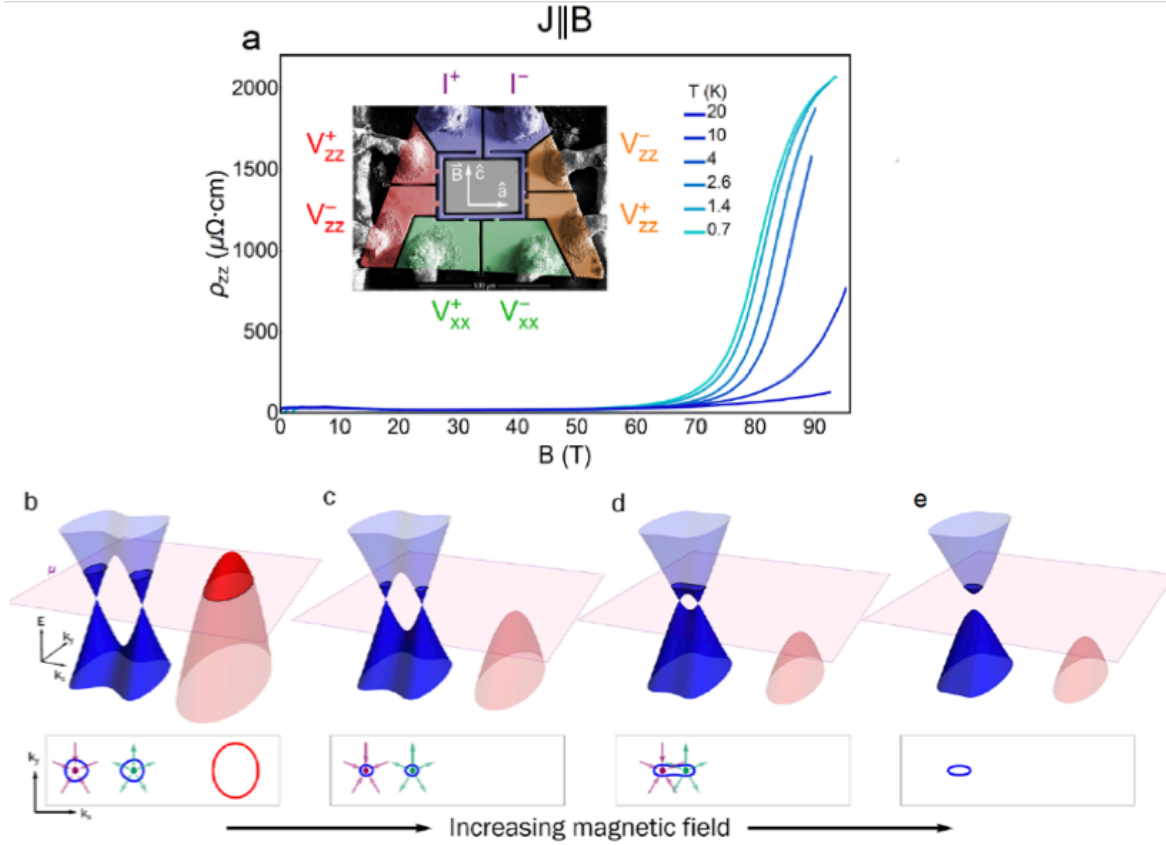
**Large volume synthesis of new quantum materials:** Unlike other countries, the US lacks the capabilities for large volume synthesis and recovery of high-pressure materials, including newly discovered quantum materials. New synthesis facilities should include capabilities for metastable low-pressure growth such as CVD that can be used to synthesize high-pressure phases. Expanded capabilities for synthesis under high pressure extremes is needed to exploit this promising area.

### 3.1.2.2 High Fields

The magnetic field is a unique quantum mechanical probe of materials. It couples to spins, electron orbits, and it can directly control the quantum-mechanical phase of electronic wave functions. Not only can an applied magnetic field induce new quantum phases, but it is as an exquisitely controllable perturbation that is the basis for powerful spectroscopic probes of quantum materials. Magnetic fields induce the quantum Hall effect, which is a property of the first known quantum topological state and generally play an important role in research on quantum and topological materials as reflected in the 2016 Nobel Prize[33-35]. Higher magnetic fields are poised



**Figure 3.1.6.** Two different quantization conditions - imposed by magnetic field and by a super lattice - interfere to create a fractal energy landscape in graphene. Resistance data are plotted on a color map. The left axis corresponds to magnetic flux quanta (magnetic fields up to 45 T) and the horizontal axis to electron density (gate bias)[1].



**Figure 3.1.7.** Magnetoresistance and sound speed measurements in microstructured samples of the Weyl semimetal TaAs up to 95 tesla, deep within its quantum limit, show a phase transition likely corresponding to Weyl node annihilation[51].

to transform our knowledge of multiple outstanding problems associated with the NSF Quantum Leap[91].

### 3.1.2.2.1 Scientific Requirements

**Two-dimensional systems:** Fig. 3.1.6 shows a graphene superlattice where two quantization conditions – the superlattice and the magnetic field – interfere to create a complex pattern of the *phase and amplitude* of the wave function[1, 92]. Both fractional and fractal Hall effects can be observed. The recent discovery of many 2D materials beyond graphene (e.g. phosphene, silicene and transition metal dichalcogenides like MoS<sub>2</sub>) provide a rich landscape for future high-field studies. In many of these materials, the carrier masses and exciton binding energies are much larger than in bulk semiconductors such that 50-200 tesla instead of 5-10 tesla is required to achieve a basic understanding of the semiconducting and optoelectronic properties[93-95].

**Topology in condensed matter physics:** The impact of topology in condensed matter in this century may become as profound as the concept of broken symmetry in the previous century[33-35]. High fields are required to identify and probe[46-50] topological states such as the spin Hall effect in graphene[46]. High-field landau level spectroscopy and Fermi surface studies can uncover

the topological nature of the band structure in Dirac or Weyl materials such as  $\text{Bi}_2\text{Te}_3$  [50]. Higher magnetic fields will allow us to reach beyond the quantum limit in more materials to probe new emergent strongly-correlated states of matter[51-56]. A new thermodynamic phase transition in TaAs was recently discovered in fields up to 95 tesla, beyond its quantum limit (Fig. 3.1.7)[51]. The quantum limit exceeds 100 tesla for the majority of interesting topological materials, even those with small Fermi surfaces.

***Spin-orbit coupling in insulating oxides:*** A knowledge frontier exists within materials where spin-orbit coupling is comparable in strength to electronic and magnetic interactions. The confluence of energy scales can lead to new states of matter and new functionalities, for example magnetism driven by Kitaev interactions and potential Kitaev spin liquids in  $4d$  and  $5d$  oxides [96, 97]. To understand the underlying physics, high fields are needed to separate out competing energy scales, and to identify the strength of interactions. The field-dependences of physical properties over the extended T-H phase diagram is a particularly valuable probe of theoretical models[98, 99].

***Quantum magnetism:*** Magnetic fields can induce states of matter in quantum magnets such as fractal series of magnetic states in frustrated dimer systems[100], spin ice[101], spin liquids[102-105], Bose-Einstein condensates[106, 107], Luttinger liquids[108] and possibly even supersolids[109, 110]. Quantum magnetism underlies the more complex phenomena of strongly-correlated metals and unconventional superconductors. High fields are needed to access new phases. In particular very sensitive measurements of e.g. heat capacity, neutron diffraction and electron spin resonance at higher DC fields are needed to definitively identify the scaling relations and symmetries of states like Bose glasses and supersolids[107, 109].

***Superconductivity:*** Although three decades have passed since the discovery of high- $T_c$  superconductivity, the mechanism remains unknown. Data up to 100 tesla show that magnetic fields of 130 – 150 tesla are required to access the quantum critical point and probe the mysterious normal state, for important high  $T_c$  superconductors such as  $\text{YBa}_2\text{Cu}_3\text{O}_7$ [111] (see Fig. 2.1.2). High fields are also a unique probe of the Fermi surface and the upper critical fields of superconductors. Fields exceeding 80 T have driven many recent advances in our understanding of high- $T_c$  superconductors. Higher fields and higher DC fields for key techniques like STM and neutron diffraction have the potential to finally solve the riddle of high-temperature superconductivity[111-116].

### 3.1.2.2.2 Midscale Instrumentation

***135 tesla Pulsed fields:*** Magnetic fields with timescales longer than milliseconds are necessary to access a wide range of physics and measurement techniques. Exceeding 100 tesla on millisecond timescales is necessary to explore many Weyl and Dirac materials beyond their quantum limit, to understand the excitons that govern optoelectronic properties of 2D superconductors, and to reach the quantum critical point at the heart of high- $T_c$  superconductivity. Higher fields allow us to study materials with functionality near room temperature for technological applications.



**60 tesla DC magnetic fields:** Clean DC magnetic fields access the definitive equilibrium ground states of materials. Higher DC fields are critical to achieve fundamental understanding and uncover new physics including topological states, 2D materials, and unconventional superconductivity. DC fields allow us to perform measurements requiring time scales of minutes to days, such as many optical techniques, thermodynamic probes, or ultra-sensitive measurements as in Fig. 3.1.6. DC fields also enable low temperatures down to hundreds of micro-Kelvin, and higher pressures so as to access an extended  $T$ - $H$ - $P$  phase diagram. Finally, scanned probes such as STM can be implemented in high-field superconducting DC magnets.

**40 tesla SC magnetic fields:** A 60 T DC magnet will require high temperature superconducting materials (HTS) on a much larger scale than has been demonstrated to date (~50 MJ required compared with <0.5 MJ to date). In addition, quench management for HTS materials is in its infancy compared to that for low temperature superconductors (LTS). Consequently, prior to building a 60 T DC magnet, it would be prudent to develop, test, and operate a 40 T superconducting magnet. This would allow the novel magnet technologies required for 60 T DC to be tested at an appropriate scale as well as meet the challenge of the 2013 MagSci committee that stated “A 40 T all-superconducting magnet should be designed and constructed building on recent advances in high-temperature superconducting magnet technology.”[91] Such a magnet would enable micro-Kelvin measurements, spatially-resolved atomic-resolution measurements, pump-probe optical techniques, among others. It would also dramatically lower power consumption and allow long hold times at high fields compared to all resistive and resistive-superconducting hybrid magnets.

**Magnets at x-ray, neutron, and other scattering facilities:** Combining both pulsed and DC magnetic fields with x-ray and neutron facilities is a severely underdeveloped capability in the US. x-rays at new high-flux coherent beamlines are an increasingly powerful probe of magnetism and thus these beamlines should have magnets. Higher magnetic fields can be achieved when a beamline is dedicated to magnetic field measurements and a scattering instrument is designed and built around the magnet. The importance of progress in our ability to conduct neutron and x-ray scattering experiments at high magnetic fields was emphasized in the two most recent National Academy Reports on high magnetic fields.[91, 117]

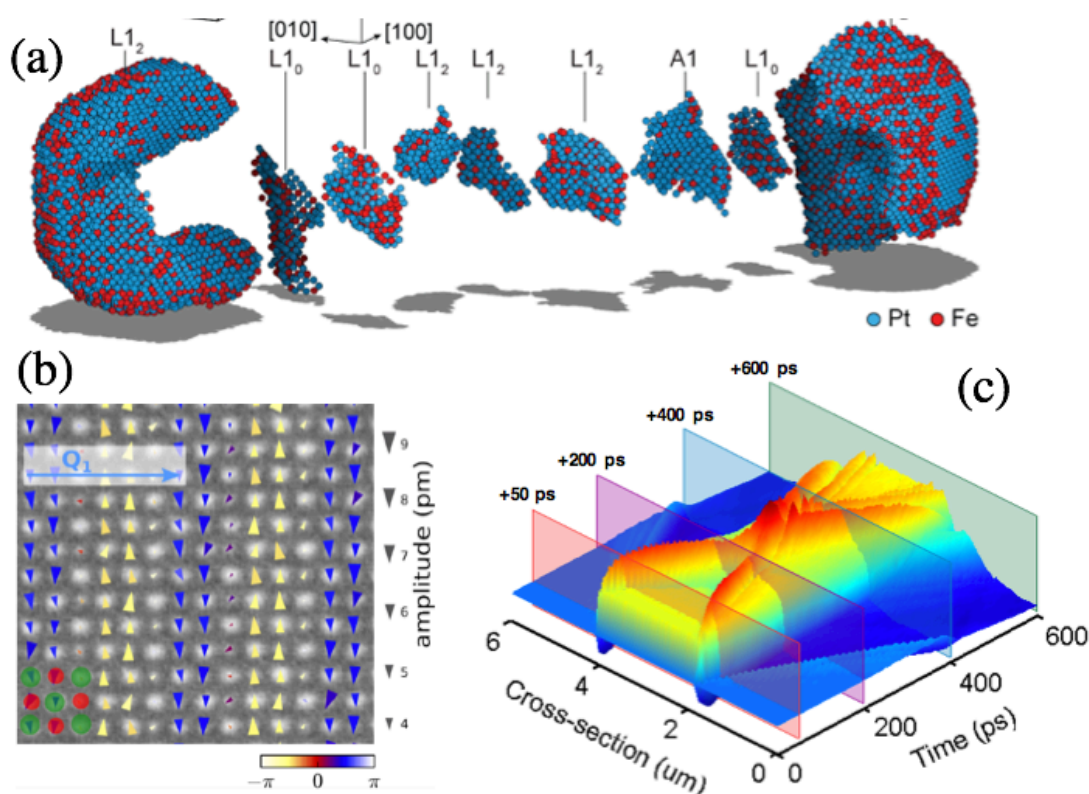
### 3.2 Probing Quantum Matter

Quantum materials have attractive properties such as unusually rich phase diagrams that support useful crystallographic, electronic, and magnetic phases that can potentially be rapidly and efficiently modulated for a host of applications. However, decades after their discovery, their complexity presents fundamental challenges for understanding and exploiting them. The inherent heterogeneity and multiple phases present in quantum materials demand a new and powerful set of hyper-spectral and multifunctional microscopies and spectroscopies. Indeed, this may be the only way to achieve a nuanced understanding of the underlying complex physics of quantum materials. Many nanostructured and complex materials will require a combination of different techniques (visible, THz, x-ray, electron, neutron etc.) to probe their coupled responses over

multiple length and time scales ( $\text{\AA}$  and attoseconds on up). Such experiments may also uncover new routes for accessing and manipulating previously unknown states of quantum matter.

### 3.2.1 Microscopy

The prospects for dramatic advances in understanding and exploiting quantum materials are very strong. An important factor has been revolutionary advances in photon, electron and neutron sources, in new computational imaging techniques, as well as in advanced spectroscopies that can probe interactions and correlations in real time.[118-126] Coherent (laser-like) light sources now span from the x-ray to the THz region of the spectrum, with exquisite control over the spectrum, waveform, pulse duration and polarization,[127-129] allowing us to go beyond static probes for rich new understanding of how materials function, and how we might very rapidly manipulate their properties. New computational imaging techniques harnessing powerful algorithms have pushed electron imaging to near-atomic resolution in 3D, have enabled sub-wavelength spatial



**Fig. 3.2.1. Recent advances in imaging quantum materials.** (a) Advances in electron microscopy now make it possible to determine the 3D coordinates of EVERY atom in an FePt nanoparticle (all 6,569 iron and 16,627 platinum atoms), to correlate 3D atomic arrangements and chemical order/disorder with material properties at the single-atom level. These measured atomic coordinates can be used as direct input for quantum calculations to predict the material behavior and identify what structure is best for next-generation battery materials. (b) Picometer periodic lattice modulation in charge density wave materials using scanning tunneling electron microscopy. These materials show promise for next-generation electronics. (c) Stroboscopic imaging of transport in nanostructures using coherent short wavelength light, where sub-wavelength spatial resolution imaging is now possible. Extending these capabilities into the soft x-ray region will make 3D functional imaging of quantum or designer nanostructured materials possible, to directly visualize how they work.

resolution imaging at short wavelengths for the first time, as well as 3D imaging of visibly-opaque samples. New scan probe tools have been developed that can directly interrogate complex phases such as magnetism and superconductivity with unprecedented spatial and energy resolution[130]. Mid-scale instrumentation to help address the following questions would have broad impacts:

***What materials and devices will be used in the next generation electronics and computers?*** We are in the middle of a materials revolution, with exciting opportunities being realized by synthesis of 2D and nanostructured 3D materials. Currently, a combination of synthesis and theory are driving discovery, and microscopy and scan probe tools are used to understand the results of synthesis. There is a significant opportunity to develop tools with high throughput and accessible while directly measuring the quantum properties of interest. The development of such tools will be essential to navigate the rapidly evolving materials landscape of the 21<sup>st</sup> century, by providing quick and accurate feedback to synthesis.

***How do these materials and devices work, and how can we improve their performance?*** The role of microscopy and scan probe tools is to provide fundamental insight into the quantum mechanical processes at work in new materials. Continuous advances in multimodal imaging techniques over wide ranges of time, length, and energy scales, together with direct measurements of equilibrium and dynamic quantum properties are needed to advance these goals.

***Where will the next breakthrough be?*** Several areas show great promise for dynamically manipulating materials and device properties using light or other external stimuli. However, we must sort through the diversity of ideas to find what approaches work best. Access to the formation and decay of quantum correlations and interactions, the interactions between atoms, spins, and charges in equilibrium, during phase transitions, or while externally-driven, would enable rapid feedback between understanding, theory, growth, characterization and application.

### **3.2.1.1 Scientific Requirements**

Looking to the future, there are exciting opportunities for advanced instrumentation to dramatically enhance progress in quantum materials, to help uncover how quantum many body phenomena emerge from atomic and nanoscale heterogeneity. Moreover, these same techniques can have a wide impact beyond quantum materials, by also addressing instrumentation needs for advanced nanotechnology development and even manufacturing as these techniques mature. Exciting new instrumentation capabilities that will be required to advance quantum materials include:

***Real-time imaging*** across broad length and time scales, with multiple contrast mechanisms, to probe the growth, properties (electronic, magnetic, structural), and function of quantum materials in real space and time. This might include new hybrid photon and electron imaging – from atomic to nano to macro, ultrafast to static, hyperspectral from THz to x-ray, attosecond to static, Å to full field.

***Functional imaging*** in combination with synthesis and hetero-structuring, gives us the ability to design and engineer low-dimensional quantum materials with unique properties that go beyond those possible in bulk quantum materials. However, nanostructured materials are still extremely

challenging to model, and new theories need to be validated through comparison with experiment. Functional imaging of such materials is needed to capture and understand how the nanoscale structures influence the functional properties of a material, whether for next-generation energy-efficient electronics, wearable devices, computing, data storage, or thermoelectrics.

***Quantum manipulation*** allows us to manipulate materials properties using light, external fields, pressure and other external parameters. Better tools are needed to understand how these materials are changing as we control their properties to optimize function. Since quantum materials are intrinsically susceptible to competing or intertwined orders, multiple views of the same material with varying contrasts and resolution are needed.

***Harnessing topology*** requires techniques that manipulate, control, or directly reveal the topological and/or entangled nature of quantum materials.

***Correlations and fluctuations*** inherently span multiple spatial and temporal scales, and new techniques are needed to probe these on their natural length and time scales, as well as the influence of interfaces. We need to capture the formation, spatio-temporal evolution and decay, dephasing, correlations, fluctuations of atoms-spins-charges-photons, in equilibrium, during phase transitions, and when driven far from equilibrium. New multi-dimensional spectroscopies from THz to multi-keV can be harnessed, as well new tools to probe solid state quantum entanglement.

***Faster, accessible, agile, adaptive and interactive*** probes are needed to accelerate advances in quantum materials. A portfolio of tabletop mid-scale instrumentation that can be optimized and duplicated for broad impact, as well as facility-scale instruments, are needed to accelerate the development pathways and enable iterative designs for quantum and indeed a wide range of nanostructured and inhomogeneous materials.

### ***3.2.1.2 Midscale Instrumentation***

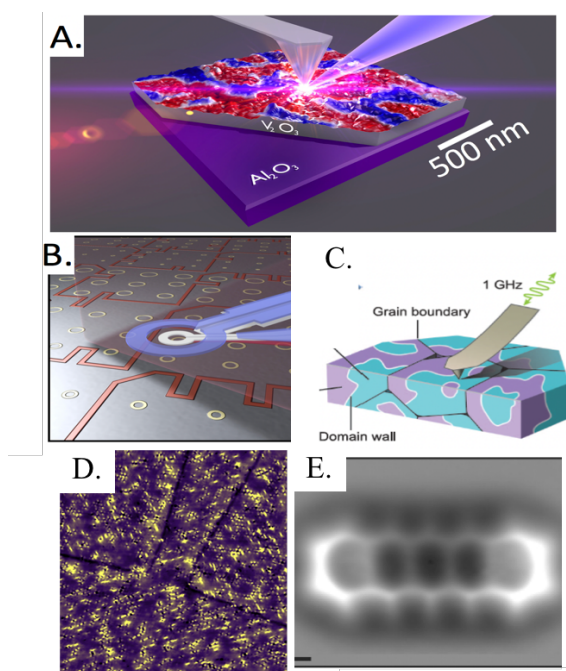
***Real-time 3d imaging of functional materials with multiple contrast mechanisms:*** Advances in ultrafast lasers over the past 10 years now make it possible for a single ultrafast laser to simultaneously deliver ultrafast coherent beams of light (i.e. directed and laser-like) spanning from the THz, to the infrared/visible/ultraviolet and soft x-ray regions. Moreover, this light emerges as a well-synchronized waveform that can capture and control material dynamics from attosecond time scales on up. The same laser can also generate ultrafast electron bursts from nano-tips. New mid-scale materials instrumentation based on ultrafast lasers would provide unique microscopes for imaging functioning materials systems in 3D – a capability that is not possible using current imaging techniques. Although most areas of imaging science are undergoing revolutions – as evidenced by the 2017 Nobel Prizes in Chemistry for cryo-electron imaging,[122] yet they still represent slow, static imaging of surfaces or very thin samples.

New laser-based mid-scale instrumentation could probe and image materials using photon/electron sources that are agile in energy, wavelength, repetition rate, power, pulse duration and spectral bandwidth. This instrument could deliver multiple, synchronized probes, to the same sample - combining the advantages of imaging and advanced spectroscopies to capture the dynamic

electronic, magnetic, order, and structural properties of a material. These include time-, spin- and angle-resolved photoemission spectroscopy (ARPES), x-ray absorption spectroscopies (XAS, EXAFS), as well as a variety of magnetic spectroscopies. Advances in light sources for ARPES, for example, will enable high time- and energy-resolution, to make it possible to explore the role of fluctuations in superconducting systems. In addition, full field, dynamic imaging is now possible, to capture the fastest charge (scattering and screening) and spin (e.g. spin currents and skyrmions) dynamics in thick (visibly opaque) inhomogeneous materials using soft x-ray imaging, and in the future, atomic-level resolution using ultrafast electron imaging. This would enable dynamic probing of the emergent static and dynamic properties and couplings in quantum materials with a variety of contrast mechanisms (e.g. electronic, structural, magnetic), on all timescales relevant to function. It would also be possible to coherently manipulate the electronic, spin, and lattice degrees of freedom, and observe how the material properties change, and also to probe coherence and correlations.

Finally, another very significant of such a mid-scale instrument is that it can greatly accelerate iterative and smart design of a wide range of nanostructured and inhomogeneous materials. A current bottleneck to progress in materials science is that access to advanced imaging techniques at large scale facilities may not be possible at the relevant project time scales, which can inhibit efficient interactions between design, synthesis, characterization, and theory.

***Opportunities in scanning probe instruments:*** Scanning probes give us direct information on quantum phenomena that occur over an extended energy range from sub meV-1eV[135]. Looking



**Figure 3.2.2.** A: nano-infrared image of a phase separated electronic state in  $V_2O_5$  (blue: insulating, red: metallic) amidst the insulator-to-metal transition. Adapted from [123]. B: Scanning SQUID microscope that can measure a spatially varying magnetic fields with high sensitivity and spatial resolution on the order of  $1\ \mu\text{m}$ . Courtesy of Moler group, Stanford University [131]. C: Illustration of microwave impedance microscopy (MIM) setup [132]. D: Atomic-resolution scanning tunneling spectroscopy visualization of electron nematicity [133]. E: Non-contact Atomic Force Microscopy visualization of individual bonds in pentacene [134].

to the future, there are opportunities for exploring some of the most enigmatic properties of quantum materials. These include well-known problems such as the Mott transition and strongly correlated phases in its vicinity, as well as newly observed quantum phenomena such as transient photo-induced superconductivity and “hidden” phases in the vicinity of phase transitions. Many topologically non-trivial states of matter imply the existence of so-called edge effects literally occurring at the physical boundaries of the specimens. The study of edge current in quantum Hall systems is a text book example. Scanning electrical, optical and microwave probes will allow one to visualize and thoroughly investigate electronic, photonic and polaritronic edge phenomena. Furthermore, nano-optical probes are well suited to explore and exploit propagating polaritonic modes of electronic, excitonic and magnetic origin[136]. These hybrid light-matter modes enable extraordinarily strong electric fields that pave the way for the exploration of non-linear phenomena [137]. Finally, scanning tunneling and non-contact atomic force probes are intrinsically capable of resolving the quantum electronic and chemical properties with sub-atomic spatial resolution and thermally limited energy resolution simultaneously[138]. New tools that utilize the existing strengths of these probes but add new functional and dynamic capabilities will play an important role in harnessing the properties of quantum materials.

***Integrating scan probes into the quantum factory:*** The idea of the “quantum factory” was put forward as a place where quantum materials are synthesized and quantum devices are fabricated. Given that scan probes have the capability to directly address local quantum properties, they would play an important role in the characterization of these materials and devices. The instrumentation needs here are to take existing scan probe technologies and make them agile, easy to use and high throughput. A key driver of cost is the facilities required to properly integrate high-resolution scan probes into the quantum factory environment. Near-field probes that have atomic resolution require a controlled mechanical and acoustic environment to achieve peak performance[139]. Quantum coherent properties of solid state materials are also usually observed in cryogenic environments. The current generation of high-resolution scan probes operate in a liquid helium based traditional cryogenic environment. They are consequently low throughput and labor intensive to maintain and operate, and integrated cryogenics development needs to be undertaken to make them suitable for a quantum factory environment. Upon proper integration into the quantum factory, the new scan probe tools would essentially play a role equivalent to transmission electron microscopes (TEMs) in determining structure of new materials, but instead provide direct imaging of the low-energy quantum properties. Examples of such tools that require significant investment in facilities include scanning tunneling microscopy and near-field optical probes (for electronic structure), scanning SQUID microscopes (for magnetism) and non-contact atomic force microscopes (for chemical bonding).

***Making hybrid scan probe tools:*** Nano-optical probes are good examples of what can be achieved here. By marrying two techniques (infrared optics and atomic force microscopy (AFM)), new functionality is achieved, while beating the optical diffraction limit. With the advent of new lasers for example, there are opportunities to develop hybrid instruments that can attack new problems. As an example, consider the possibility of non-equilibrium superconductivity that might be

possible to induce through strong field excitation[126]. If one could indeed measure the superconducting gap and other quasiparticle properties under illumination, this would enable new science. The problem here is making the very different timescales of typical scan probes (ns or longer) and dynamical electronic phenomena (fs-ps) compatible with each other. By proper mixing/lock-in techniques, this should be possible, but it is a significant development challenge.

***Scan probes in new environments:*** Traditional tools like electron transport and thermodynamic measurements can be applied to quantum materials under a wide variety of external conditions such as high magnetic fields, varying temperature, uniaxial and hydrostatic pressure, current flow, etc. In the past few years, it has become possible to apply high-resolution scan probe techniques to study quantum materials in some of these environments[140, 141], but cost is often a prohibitive factor in the development of these capabilities. An example of a potential new investment is to achieve scanning tunneling and non-contact atomic force microscopy under high magnetic fields (>20 T).

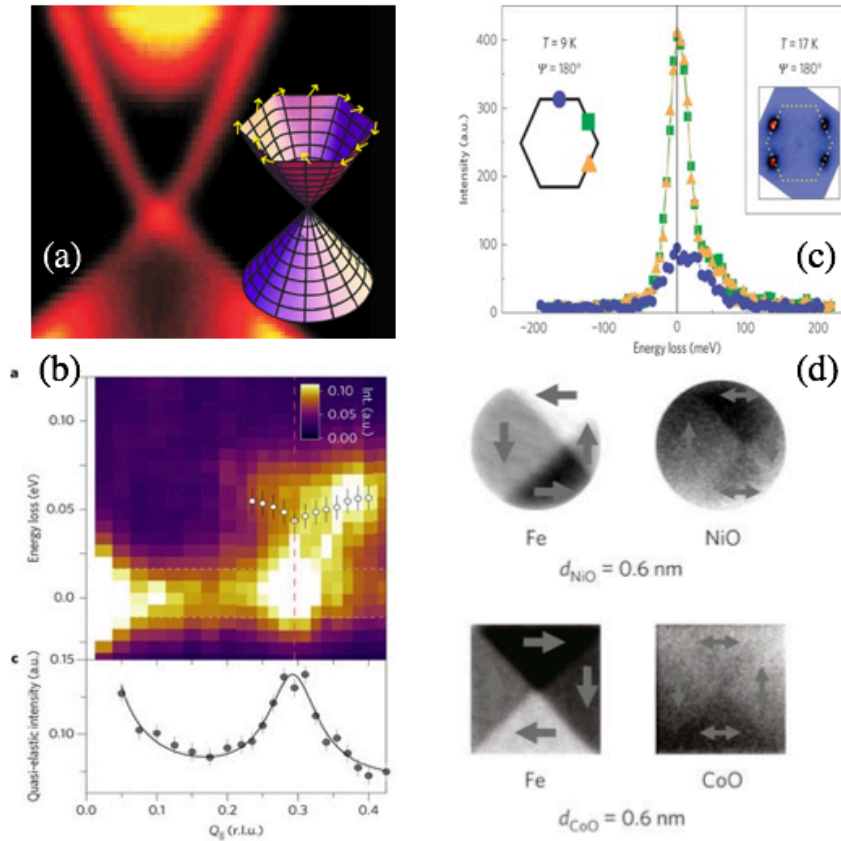
***New functionality in scan probes:*** Understanding and manipulating new/intertwined phases of matter requires tools that go beyond spectroscopy and can instead directly measure the phase of interest with high spatial resolution. Recent achievements in this area include probes that can directly measure superconductivity[57] and magnetic structures[125]. Targets for new functionality include the ability to measure local spin with atomic precision (magnetic exchange force microscopy) and probes that directly measure fractional charge in topological matter.

### **3.2.2 Scattering**

Neutron and x-ray-based scattering, diffraction, and spectroscopies play a major role in understanding the unusual properties of quantum materials. Many of these exhibit remarkable emergent properties that are proposed for applications in future information processing, notably those based on spintronic and quantum information technologies. In these materials, quantum-mechanical phenomena can persist far beyond the atomic. These long coherence lengths allow quantum phenomena like matter wave interference and entanglement to be observed and controlled, and thereby provide enormous potential for transformative applications including new information processing and storage schemes.

Superconductors are exemplary of quantum matter since the condensate wave function is macroscopically coherent. This allows, for example, the design of superconducting Q-bits for quantum information processing but operating for now only at low temperature. By contrast, the length scale over which spin wave functions are coherent and quantum spintronic devices might be designed can approach 1  $\mu\text{m}$ . Similarly, spin-momentum locking in topological insulators enhances carrier mobility, but the relevant coherence length remains finite and will determine the range over which quantum entanglement can occur. Achieving new quantum information technologies using these materials will require developing submicron structures that are optimized to minimize decoherence while also processing quantum information and translating the result to the macroscopic world.





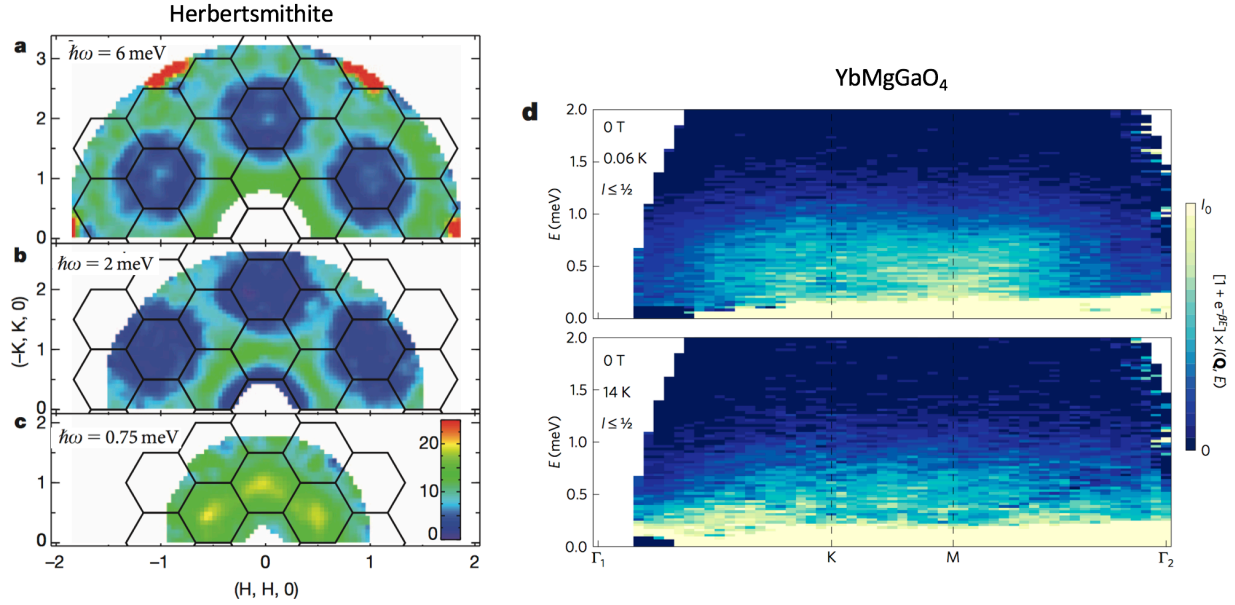
**Figure 3.2.1.** X-ray spectroscopies and scattering tools with spectroscopic contrast have become indispensable to understand these emergent phases of quantum matter. Examples include a) ARPES studies of topological insulators[40], b) RIXS studies of quantum materials like cuprate superconductors[142], c) REXS studies in of frustrated quantum spin liquids[143], and d) XMLD studies of antiferromagnetic spin vortices[144]. Moreover, these tools are increasingly being used as contrast mechanisms in x-ray nanoprobe so that quantum matter can be probed at a scale below the intrinsic quantum coherence lengths and, therefore, in functional submicron devices.

### 3.2.2.1 Scientific Requirements

The sensitivity of neutron and x-rays to relevant interactions in quantum materials have made them essential for understanding their emergent properties. x-ray techniques have provided the direct demonstration and discovery of new electronic phases of topological matter with angle-resolved photoemission spectroscopy (ARPES), key contributions to the ongoing effort to understand novel forms of superconductivity with resonant inelastic x-ray scattering (RIXS) and ARPES, evidence for Kitaev interaction induced magnetic order in  $\text{Na}_2\text{IrO}_3$  with resonant elastic x-ray scattering (REXS), and the characterization and understanding of functional spin textures with x-ray circular and linear dichroism (XMCD and XMLD) (Fig. 3.2.1).

Neutrons provide quantitative access to correlations of electronic spin and orbital angular momentum in space and time from the atomic scale to the device scale. Neutron diffraction provides unique access to magnetic ordering at the atomic scale in anomalous superconducting materials and their parent compounds, to magnetic structure on the mesoscale in the mixed phase of type II superconductors, and to magnetism within engineered hetero-structures on length scales extending up to microns. Neutrons are also important for the determination of chemical structure where their unique sensitivity to light atoms and to different isotopes and nuclear spin





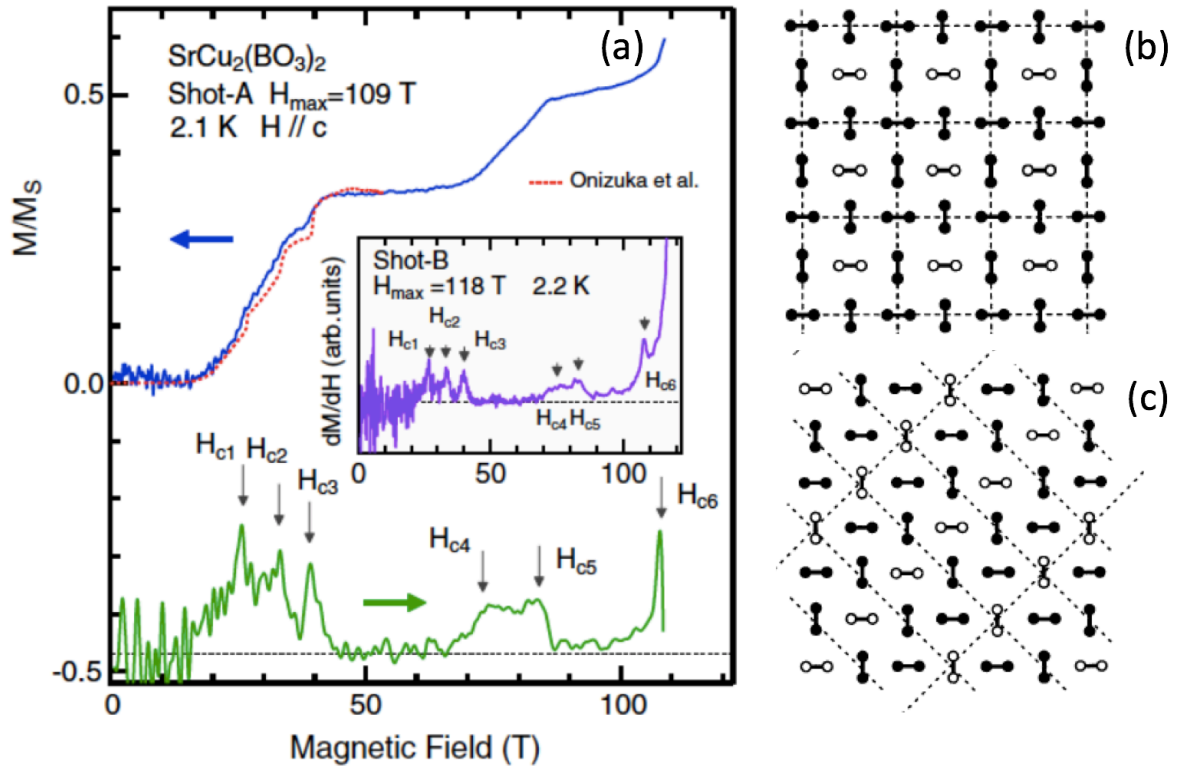
**Figure 3.2.2.** Inelastic neutron scattering data from (a-c) Herbertsmithite[21] and (d) YbMgGaO<sub>4</sub>,[22] wherein spin degrees of freedom on kagome and triangular lattices respectively fail to develop long range magnetic order in the low temperature limit. For a conventional ordered magnet, such data would contain sharp lines of high intensity regions associated with single magnon-creation from which the magnon dispersion relation and life time can be extracted. The broad structures seen for spin-liquid candidates indicates neutrons scatter through multiparticle processes or that there are no well-defined quasi-particles.

complements x-ray techniques. Inelastic neutron scattering provides access to dynamic correlations and a detailed view of collective quasi-particles including magnons, spinons, and phonons with sufficient resolution and sensitivity to probe energy transfers from neV to eV with optimized instrumentation. Recent examples include evidence for spin resonances in anomalous superconductors,[145-147] fractionalized excitations in one-[148-150] and two-dimensional quantum spin liquids,[21, 22] emergent magnetic monopoles in spin ice,[151] and determination of the chemical and magnetic structure of a wide range of quantum materials including copper oxide[152] and iron pnictide [153] superconductors.

Despite steady advances in x-ray and neutron scattering instrumentation, the fundamental challenges and technological opportunities associated with quantum materials calls for a quantum leap. Rather than broken symmetries, quantum matter is characterized by entanglement and yet we do not have experimental tools to probe solid state entanglement.

Neutron scattering played a central role in establishing the concept of collective quasi-particles such as magnons and phonons in solids. Conventional ordered magnets have sharp dispersion relations from which detailed information about magnetic interactions and the structure and life-times of quasi-particles can be inferred. Fig. 3.2.2 shows examples of the excitation spectra measured for quantum spin liquid candidate materials at low temperatures.[21, 22]

Acquisition and analysis of electronic scattering continua with high resolution and dynamic range is needed as well as new methods to infer relevant information about the underlying quantum state



**Figure 3.2.3.** (a) Magnetization curve of the quantum magnet material  $\text{SrCu}_2(\text{BO}_3)_2$ . [154] The anomalies are thought to be associated with distinct arrangements of magnetized and non-magnetized spin pairs as indicated in (b) and (c) for the  $\frac{1}{3}$  and  $\frac{1}{2}$  magnetization plateaus respectively [155]. Instrumentation for neutron scattering in this high field range is needed to explore these novel states of matter, their excitations, and the nature of the phase transitions between them.

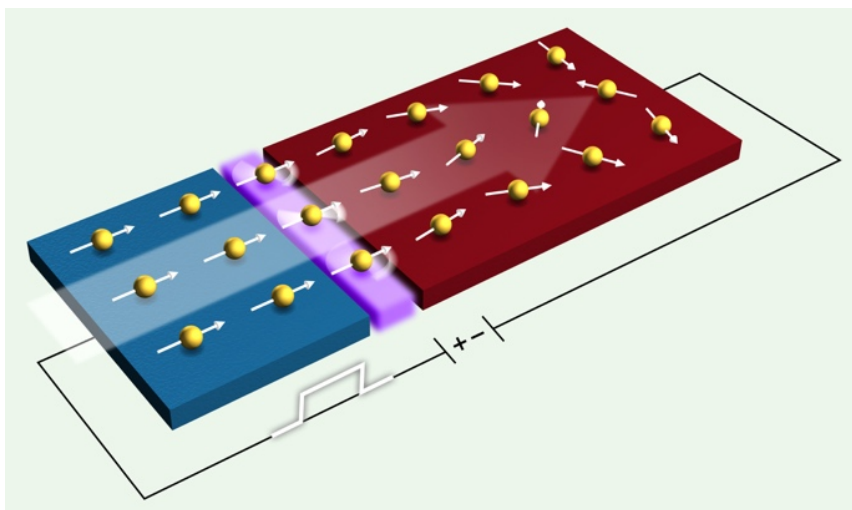
in a model independent fashion. The exploration of novel quantum states of matter will require advanced neutron and x-ray scattering techniques that can be applied to small ultra-high-purity single crystalline samples and engineered heterostructures under extreme conditions of magnetic field and pressure. Fig. 3.2.3 shows a sequence of magnetization plateaus in  $\text{SrCu}_2(\text{BO}_3)_2$ . [154] It is thought that these are associated with distinct orderings of singlet and triplet states of dimers on the Shastry-Sutherland lattice. [155] Exploration of these unique states of quantum matter, their excitations, and the phase transitions between them awaits the development of instrumentation to conduct comprehensive high resolution inelastic neutron scattering experiments in fields beyond 30 tesla.

An important role is envisaged for x-ray beams in the development of quantum information technologies. As presently applied, x-ray tools often lack the spatial resolution to probe quantum matter on the length and time scales over which quantum phenomena are coherent. But x-ray wavelengths are ideally matched to both the energies of the relevant electronic states as well as the important length scales, so their application as nanoscale probes is feasible, relevant, and imminent (Fig. 3.2.4).

The combined spectral, spatial, and temporal sensitivity enabled by emerging high brightness x-ray sources will accelerate this transformation of the powerful x-ray tools illustrated in Fig. 3.2.1 into spectroscopic nanoprobes. These nanoprobes will be able to measure the decoherence of wave functions near defects and interfaces, the influence of device morphology on emergent quantum phenomena, and the motion of quantum information at the heart of emerging quantum technologies. These experiments will investigate not only the spatial and temporal heterogeneity observed in many forms of idealized, pure quantum matter, but also their manifestation in candidate device structures. This includes thermal fluctuations, which tend to cause quantum decoherence, as well as quantum fluctuations, which will need to be an integral part of quantum information technologies based on quantum matter.

It is important to consider how x-ray nanoprobe tools might be deployed for maximum impact in the coming decades. One of the most important issues will be uncovering the influence of device morphology and composition on the intrinsic spatial fluctuations present in quantum matter. This will naturally require a mutual convergence of x-ray and other nanoprobes with sample preparation. This will allow us to evolve the technique from statistical approaches that average over spatially inhomogeneous states, to a sample-centric one where correlations between structure and electronic responses can be directly uncovered within the relevant length scales.

The present trend to add *in situ* growth capabilities to ARPES end stations is expected to continue in the future; growth, analysis, and modeling will be even more fully integrated and broadly coupled to a range of techniques. In such a sample-centric approach, it will be possible to carefully engineer sequences of samples to expose the influence of composition and morphology on quantum states. Such an approach would be prohibitive in the present state of affairs where samples must be prepared and preserved under strictly identical conditions at various technique-centered facilities. In an integrated sample-preparation environment, advanced techniques such as machine-learning guided searches for new materials and properties can then be shared among multi-modal x-ray (and non-x-ray) nanoprobes, allowing much faster development of useful



**Figure 3.2.4.** The decay of the polarization of spin currents injected from a magnetic material into a nonmagnetic material was recently detected using a high sensitivity soft x-ray scanning transmission microscope. Such currents can be deployed at submicron scales for both classical and quantum information storage and processing. [156]

emergent properties in a dense parameter space. The complementarity of these tools applied to the very same regions of these samples will transform our ability to understand, control, and exploit heterogeneity in quantum materials.

### **3.2.2.2 Midscale Instrumentation**

The following are some examples of new experimental modalities that would accelerate our understanding of quantum materials and their technological deployment.

X-ray tools are intrinsically multi-modal. Different atomic species and different electronic and spin configurations can be probed with a given technique simply by tuning the wavelength, detected particle (electrons vs. photons) or polarization. These give sensitivity to chemical species, electronic structure, spin, and orbital characters. Also, soft x-ray tools, which are ideal for probing electronic and spin textures, can be beneficially coupled to hard x-rays tools, which provide a detailed study of underlying lattice structure, mesoscale organization, and associated strain in thin films and nanostructures. Coupling soft and hard x-ray tools, particularly nanoprobe, is challenging due to distinct optical designs in these two regimes, but progress is being made along these lines.

Integration of x-ray techniques with non-x-ray techniques is also very important and will become closer and more common in the future. For example, ARPES measures the single-particle spectral function, which describes the coupling between fermionic motion to bosonic excitations, while RIXS measures the two-particle dynamic structure factor of those same bosonic excitations, which are dressed by the same fermionic fields. It would be advantageous to apply these tools to the same sample, with the same contrast mechanisms, to provide a more complete understanding of the motion and lifetime of quasiparticles and their relationship to emergent properties. Even more powerful would be the application of both as nanoprobe spectroscopies. NanoARPES is presently being developed at a few facilities around the world, but nanoRIXS is a much more challenging technique that will require innovative spectrograph designs particularly to integrate it with nanoARPES.

Scanned probe microscopies are obvious candidates for coupling to x-ray tools. This is happening at x-ray facilities around the world, and the development of nanoprobe x-ray techniques at high brightness x-ray facilities will clearly accelerate this process. In addition to scanning tunneling microscopy and spectroscopy, an interesting possibility is scanning near-field infrared and scanning microwave microscopies. This combination will connect the conductivity tensor near interfaces, defects, and grain boundaries to the underlying electronic phase behavior, lattice strain, etc.

Spin polarized inelastic neutron scattering is needed to understand low energy collective phenomena in bulk quantum materials, especially when spin, lattice, and orbital degrees of freedom are entwined and when coherent modes do not dominate the spectrum. While existing instruments do not offer an adequate information rate or dynamic range, focusing, multiplexing, and broadband polarization techniques developed in the last decade point to a new generation of

instrumentation that will be two orders of magnitude more efficient than current instrumentation. A new generation of ultra-efficient neutron spectrometers to chart collective electronic dynamics in quantum materials from 0.05 meV to 200 meV would have major impacts.

High magnetic fields open a new dimension in the exploration of quantum materials. While thermo-magnetic and transport measurements reveal the presence of novel quantum correlated states of matter as ultra-high magnetic fields, the ability to probe the underlying electronic correlations at the atomic scale is critical to understand such novel quantum states of matter. High field neutron scattering is ideal, but at the moment only available in fields up to 14.5 tesla. A dedicated and versatile instrument for neutron scattering in fields beyond 25 tesla is however, technically feasible and would be a major resource to understand quantum matter. High Tc superconducting tapes and a paradigm shift towards non-insulated superconducting magnet design may make it feasible to build a split-coil magnet for scattering and spectroscopy in fields approaching 40 tesla.[157] By combining time of flight and crystal spectroscopy it is possible to build a neutron spectrometer that employs a broad band of neutrons both before and after the sample while continuously covering a wide range of scattering angles. Such a spectrometer would make optimal use of the solid angle available in a split coil magnet and provide unprecedented capabilities for experimental studies of field-driven quantum matter.

Similarly, light-scattering capabilities at high magnetic fields, from the far-infrared to the x-ray range and including THz experiments do not exist within the US. These experiments elucidate spin texture and control of quantum matter at high fields; including the nature of the quantum phase transition under the superconducting dome in high-temperature superconductors. Furthermore, extreme magnetic fields pave a road towards discovery of quantum materials as they uniquely tackle strong correlations from single particles (QHE and beyond), probing the coupling of high fields to spin and charge degrees of freedom – when these degrees of freedom are strongly coupled and when strong fields are required to disentangle. Building a high-field split coil at a synchrotron would be an important mid-scale direction. Also, more recently, light-source technologies have been developed in the mid-scale range of cost (including far infrared free-electron lasers, inverse Compton sources, high-order harmonic generation (HHG) sources) that can be built at a dedicated high-field split-coil magnet.

There is presently no experimental tool to probe the defining characteristic of quantum entanglement in solids. A versatile experimental probe of entanglement would have major impacts on the field, with an even greater possible impact if spatial resolution on the device scale were possible. It seems likely that entangled beams of neutrons or x-rays will be important but there is not yet a proven concept so this remains an exciting challenge to the community.

### **3.3 Controlling and integrating Quantum Matter**

#### **3.3.1. Materials Issues in Quantum Information**

Quantum systems feature unique properties that do not have classical analog. These can be utilized to represent and process information that is fundamentally more powerful than what is classically

possible. Superposition of available quantum states and quantum entanglement among multiple quantum particles provides additional resources that can be utilized for information processing. One such application is quantum computation, where access to a vast Hilbert space and carefully designed quantum algorithms, composed of individual quantum logic gate operations that evolve quantum states in such Hilbert space, can lead to substantial advantage for certain computational tasks compared to classical computers. These quantum resources tend to be highly fragile, as interaction of the quantum degrees of freedom with the environment lead to the loss of quantum coherence. In many implementations of quantum bits (qubits) today, the longevity of the qubits and the accuracy of the quantum logic operations are limited by issues in the underlying materials, such as isotopic purity of the host material, unintended coupling to environmental degrees of freedom (*e.g.*, lattice vibrations and impurities), and drifts in the coherent control signals arising from the optical/microwave devices that generate them. Analogous to the advances in materials that enabled the performance and complexity of today's silicon complementary metal-oxide-semiconductor (CMOS) devices at the core of classical computing, the scientific progress towards the fundamental understanding of the materials issues are expected to propel the progress of quantum computing technology in the near future. However, unlike the CMOS technology, which is an irrefutable technology of choice for classical information processing, the physical implementation of quantum computing is still in its early stages and multiple candidate systems, from atomic qubits to various forms of solid-state qubits, are being explored. Thus the range of opportunities for mid-scale instrumentation to study the material properties is quite broad.

### **3.3.1.1. Scientific Requirements**

There are several possibilities for studying the fundamental materials issues through midscale instrumentation, and each comes with a specific set of scientific requirements. One major topic is the issue of surface imperfections in various implementations of the qubit. Unlike in the bulk of the material, the surface of a device used to define or confine a qubit can have significant amounts of defects, adsorbates and contaminants that can affect the quality of the qubit or quantum gate operations on them. Examples include the loss or decoherence mechanisms on superconducting circuits and microwave cavities, anomalous heating of the motional degrees of freedom in surface ion traps, and electric field fluctuations arising from the surface contaminants that affect the qubit coherence in gate-defined quantum dots or defects in crystals such as the nitrogen-vacancy centers in diamond. In most of these qubit implementations, the critical performance characteristics, such as the coherence time (over which the qubits' quantum properties are maintained) or the fidelities of the quantum logic gates, are limited by the properties of the materials with which the qubits are realized. Since surface defects and sub-monolayers of surface adsorbates or contaminants can dominate these noise properties, it is imperative that a careful exploration of these surface properties be performed in an ultra-high vacuum (UHV) environment. Furthermore, as the restructuring of the surface can happen instantaneously through contact with the atmosphere (oxidation, etc.), integrating the surface characterization methods with surface preparation capabilities (such as material deposition, etching and surface treatment) to enable *in situ* monitoring of surface properties right after preparation is absolutely critical. Experimental

infrastructure to support such surface studies requires major development of dedicated instrumentation.

#### **3.3.1.2. Midscale Instrumentation**

One possible solution to enable thorough studies on the impact of surface defects on the qubit performance is to construct dedicated instrumentation for multi-modality *in situ* materials analysis relevant for qubit technologies. The system should have a modular design with a controlled environment throughout the modules, such as UHV, electromagnetic fields, and temperature. Various modular chambers should be connected by transfer mechanisms supporting full UHV conditions. Possible modules integrated in the system could include, as a baseline.

- (1) various surface characterization capabilities, such as reflection high energy electron diffraction (RHEED) tools, low energy electron diffraction tools, x-ray photoelectron spectroscopy (XPS) tools and Auger spectroscopy tools, to name a few
- (2) integration capability to soft x-ray sources (such as in synchrotrons) for further surface characterization
- (3) materials processing capability, such as etching and ion milling, cutting techniques (e.g., focused ion beam, etc.) and surface treatment options (high temperature annealing, etc.)
- (4) provide access to variable temperature environment for characterization (10mK – 300K+)
- (5) scanning probe tools for surface characterization, such as near-field scanning optical microscope (NSOM), scanning tunneling microscope (STM) and atomic force microscope (AFM)
- (6) thin-film deposition and material growth capabilities (such as atomic layer deposition techniques, sputtering and evaporation, and epitaxial growth capabilities)
- (7) flexible user ports to add optional customized processing and characterization capability for a specific qubit technology of choice.

To support various qubit technologies, one might require incorporation of tools, such as high numerical optical access across a wide temperature range (for atomic and other optically-addressed qubits like NV centers and quantum dots), high frequency and microwave electronics, ultra-low temperature environment, and specific measurement capabilities including the coherence ( $T_1$  and  $T_2$  measurements, for example). Open questions for such a facility remain including how to provide adequate user support and manage the various user demands and the extended experimental durations typically necessary for qubit characterization experiments.

#### **3.3.2. Hybrid Quantum Photonic Integrated Circuit (PIC) Facility**

Many technology platforms for quantum information processing involve photons, or optical addressing of qubits using photonics technology. These qubit platforms utilize lasers, photon detectors and waveguides for propagating and routing photons within the system. Recent progress in the photonic integrated circuit (PIC) technology in the research community have been

remarkable and has led to rapid adoption by leading groups in the photon-based qubit technology[158, 159] and optically-addressed solid-state qubits that benefit from strong qubit-photon coupling such as quantum dots[160] and NV centers in diamond[161]. However, the technical expertise and intense fabrication infrastructure necessary for realizing PICs has kept it less accessible to the research groups that do not have access to major fabrication infrastructure or communities with less experience with micro-fabrication (such as the atomic physics community). Providing a lower-barrier access for such fabrication-intensive technologies to a broader community of researchers in quantum information science can have a transformative impact on many qubit platforms that extensively utilize light, such as atomic ions, neutral atoms, defects and spins in solid state systems. In these cases, a foundry-like model to provide standardized processes and to train non-expert users can expand the accessibility of high-impact technologies like PIC to a broader user base.

### ***3.3.2.1. Scientific Requirements***

A typical silicon-based complimentary metal-oxide-semiconductor (CMOS) foundry today offers mature fabrication processes targeted for rapid ramp up to production-scale operation. This type of approach is less amenable to developing new devices, fabrication and integration processes required for research and development purposes. Recent availability of silicon photonics foundry is also designed for high-throughput volume applications, and operate at a price point of  $\sim \$10\text{k}/\text{mm}^2$ , where the dominant cost stems from making high-resolution optical masks. A more adequate model for a shared instrumentation facility would be designed with a focus on lower-throughput, flexible process development procedures targeted to enable new quantum technologies. In such a facility, one would offer a suite of fabrication processes and corresponding design rules that are currently mature, with an operating model that enables integration of new processes over time as they mature. To achieve this goal, the facility should be designed to provide a diverse set of enabling technologies, including

- (1) Photonic light wave circuits (PLCs) to realize optical waveguides for flexible routing of photons on a chip
- (2) High-performance single photon detectors (SPDs) and photodiodes to monitor photons in the system
- (3) Silicon optical bench integrate both active and free-space optical components on the chip as necessary
- (4) Electro-optic (such as lithium niobate components) and piezo-electric (such as aluminum nitride devices) thin films for additional photonic functionality
- (5) Wafer-scale material integration techniques like wafer bonding, and flip-chip bonding
- (6) Packaging capability such as precision pick-and-place and high-density wire bonding.

The technical goal of the facility is to support heterogeneous integration of various photonics components such as laser sources, optical detectors, modulators, waveguides and diffractive



optical elements on the chip and support post-processing steps and packaging procedures.

#### ***3.3.2.2. Midscale Instrumentation***

A potential midscale instrumentation facility for this purpose would be a research-grade foundry providing hybrid quantum photonic integrated circuit technology to a broad set of users in quantum information processing research that do not have access to this transformative capability. The facility would be either housed at or provided in partnership with a currently established national lab or university-based user facility, as it would require significant investment in the instrumentation as well as the staff support that will be required to develop the fabrication and integration processes, maintain the facility, train the users at various levels and offer the necessary services to the larger scientific community. Successful examples of similar operating model include the Sandia National Lab's facility that provides both micro-electromechanical systems (MEMS) technology and ion trap chips to a wider user community, Center for Nanoscale Science and Technology (CNST) at the National Institute for Standards Technology (NIST) that provides academia, industry and other government agencies with nanotechnology ranging from discovery to production, and the California NanoSystems Institute (CNSI) housed at the University of California at Los Angeles, jointly operated with UC Berkeley and UC Santa Barbara, supporting researchers across the University of California system in a range of nanotechnology research, education, and commercialization efforts. For this type of a facility to have a lasting impact, professional staff operating and maintaining the facility is critical to its success. Therefore, in addition to establishing the facility, creating a sustaining operating model with user engagement and cost recovery is crucial.

## **Appendix A: Workshop participants by panel**

The workshop was organized into two different sets of panels so that each participant appears twice below. On the first day of the workshop (December 5, 2016) the panels were arranged by technical/instrumentation disciplines. On the second day of the workshop (December 6, 2016) the participants were organized by scientific panels. Each panel had two panel leads that organized the discussions and were responsible for the panel report.

### **December 5: Instrumentation panels:**

- **Panel 1: Beamline instrumentation (da Vinci)**
  - **Steve Kevan, Lawrence Berkeley National Laboratory (Panel lead)**
  - **Alan Tennant, Oak Ridge National Laboratory (Panel lead)**
  - Collin Broholm, Johns Hopkins University
  - Dan Dessau, University of Colorado Boulder
  - Alex Grutter, National Institute of Standards and Technology
  - John Hill, Brookhaven National Laboratory
  - Peter Johnson, Brookhaven National Laboratory
  - Jonathan Lang, Argonne National Laboratory
  - Mark Lumsden, Oak Ridge National Laboratory
  - Chuck Majkrzak, National Institute of Standards and Technology
  - Ray Osborn, Argonne National Laboratory
  - Jacob Ruff, Cornell University
  - John Tranquada, Brookhaven National Laboratory
  - Igor Zaliznyak, Brookhaven National Laboratory
  
- **Panel 2: Microscopy, spectroscopy, and scan probe (Gallery III)**
  - **Margaret Murnane, University of Colorado Boulder (Panel lead)**
  - **Abhay Pasupathy, Columbia University (Panel lead and plenary speaker)**
  - Dmitri Basov, Columbia University (plenary speaker)
  - William Graves, Arizona State University
  - Mohammad Hamidian, University of California, Davis
  - Pinshane Huang, University of Illinois, Urbana-Champaign
  - Ania Jayich, University of California, Santa Barbara
  - Lena Kourkoutis, Cornell University
  - Kathryn Moler, Stanford University
  - Jiwoong Park, University of Chicago
  - Richard Sandberg, Los Alamos National Laboratory
  - Arvinder Sandhu, University of Arizona
  - Tom Silva, National Institute of Standards and Technology

- **Panel 3: Extreme Sample environments (Gallery II)**
  - **Russell Hemley, Georg Washington University (panel lead and plenary speaker)**
  - **Vivien Zapf, Los Alamos National laboratory (panel lead)**
  - Mark Bird, National High Magnetic Field Laboratory
  - Nick Butch, National Institute of Standards and Technology
  - Gilbert Collins, University of Rochester
  - Shanti Demyad, University of Utah
  - Laura Greene, National High Magnetic Field Laboratory
  - Neil Harrison, Los Alamos National Laboratory
  - Zahir Islam, Argonne National Laboratory
  - David Larbalestier, National High Magnetic Field Laboratory
  - Ross McDonald, Los Alamos National Laboratory
  - Doan N. Nguyen, Los Alamos National Laboratory
  - Viktor Struzhkin, Carnegie Institution for Science
  - Stan Tozer, National High Magnetic Field Laboratory
  
- **Panel 4: Synthesis and materials discovery (Picasso)**
  - **Tyrel McQueen, Johns Hopkins University (panel lead)**
  - **Nitin Samarth, Penn State University (panel lead)**
  - Charles Ahn, Yale University
  - Danna Freedman, Northwestern University
  - Supratik Guha, Argonne National Laboratory
  - Zhiqiang Mao, Tulane University
  - Darrell Schlom, Cornell University (plenary speaker)
  - Steve Wilson, University of California, Santa Barbara
  - Haidong Zhou, University of Tennessee, Knoxville
  - Jiwoong Park, University of Chicago
  
- **Panel 5: Quantum Structures and Devices (Gallery I)**
  - **Oskar Painter, California Institute of Technology (panel lead)**
  - **Jungsang Kim, Duke University (panel lead)**
  - David Awschalom, University of Chicago
  - Nathalie de Leon, Princeton University
  - Mark Eriksson, University of Wisconsin-Madison
  - Kai Mei Fu, University of Washington
  - Greg Fuchs, Cornell University

- Evelyn Hu, Harvard University
- Richard Miren, National Institute of Standards and Technology
- Shayan Mookherjea, University of California, San Diego
- Mark Saffman, University of Wisconsin-Madison
- Matthias Steffen, IBM (plenary speaker)
- Hong Tang, Yale University
- David Weld, University of California, Santa Barbara
- Chee-Wie Wong, University of California Los Angeles

#### **December 6: Scientific panels:**

- **Panel A: Superconductivity (Gallery III)**
  - **Peter Abbamonte, University of Illinois, Urbana Champaign (panel lead)**
  - **Peter Johnson, Brookhaven National Laboratory (panel lead)**
  - Mark Bird, National High Magnetic Field Laboratory
  - Nick Butch, National Institute of Standards and Technology
  - Shanti Demyad, University of Utah
  - William Graves, Arizona State University
  - Laura Greene, National High Magnetic Field Laboratory (co-chair)
  - Neil Harrison, Los Alamos National Laboratory
  - Jonathan Lang, Argonne National Laboratory
  - David Larbalestier, National High Magnetic Field Laboratory
  - Zhiqiang Mao, Tulane University
  - Ray Osborn, Argonne National Laboratory
  - Viktor Struzhkin, Carnegie Institution for Science
  - Hong Tang, Yale University
  - John Tranquada, Brookhaven National Laboratory
  
- **Panel B: Quantum Magnetism (Picasso)**
  - **Steve Wilson, University of California, Santa Barbara (Panel lead)**
  - **Igor Zaliznyak, Brookhaven National Laboratory (Panel lead)**
  - Gilbert Collins, University of Rochester
  - Greg Fuchs, Cornell University
  - Russell Hemley, Georg Washington University (Plenary speaker)
  - Mark Lumsden, Oak Ridge National Laboratory
  - Chuck Majkrzak, National Institute of Standards and Technology
  - Martin Mourigal, Georgia Institute of Technology
  - Jacob Ruff, Cornell University

- Richard Sandberg, Los Alamos National Laboratory
- Alan Tennant, Oak Ridge National Laboratory
- Stan Tozer, National High Magnetic Field Laboratory
- Vivien Zapf, Los Alamos National laboratory
- Haidong Zhou, University of Tennessee, Knoxville
  
- **Panel C: Topological insulators, semi-metals, and superconductors (da Vinci)**
  - **Collin Broholm, Johns Hopkins University (Panel lead)**
  - **Daniel Dessau, University of Colorado, Boulder (Panel lead)**
  - Charles Ahn, Yale University
  - Mohammad Hamidian, University of California, Davis
  - Ania Jayich, University of California, Santa Barbara
  - Lena Kourkoutis, Cornell University
  - Ross McDonald, Los Alamos National Laboratory
  - Kathryn Moler, Stanford University
  - Abhay Pasupathy, Columbia University (Plenary speaker)
  - Nitin Samarth, Penn State University
  - Darrell Schlom, Cornell University (plenary speaker)
  - Steve Kevan, Lawrence Berkeley National Laboratory
  
- **Panel D: Applied quantum materials (Gallery I&II)**
  - **Evelyn Hu, Harvard University (panel lead)**
  - **Jiwoong Park, University of Chicago (Panel lead)**
  - David Awschalom, University of Chicago
  - Nathalie de Leon, Princeton University
  - Mark Eriksson, University of Wisconsin-Madison
  - Danna Freedman, Northwestern University
  - Kai Mei Fu, University of Washington
  - Alex Grutter, National Institute of Standards and Technology
  - Supratik Guha, Argonne National Laboratory
  - Pinshane Huang, University of Illinois, Urbana-Champaign
  - Jungsang Kim, Duke University
  - Richard Miren, National Institute of Standards and Technology
  - Shayan Mookherjea, University of California, San Diego
  - Margaret Murnane, University of Colorado, Boulder
  - Doan N. Nguyen, Los Alamos National Laboratory
  - Oskar Painter, California Institute of Technology

- Jiwoong Park, University of Chicago
- Mark Saffman, University of Wisconsin-Madison
- Arvinder Sandhu, University of Arizona
- Tom Silva, National Institute of Standards and Technology
- Matthias Steffen, IBM (plenary speaker)
- David Weld, University of California, Santa Barbara
- Chee-Wie Wong, University of California Los Angeles

## Appendix B: Workshop Agenda

The workshop took place at the Hilton Hotel, 950 North Stafford Street, Arlington, VA 22203 (12/5-6/2016) and at the National Science Foundation, Room 970, Stafford Place 1 Arlington, VA 22203 (12/7/2016). The agenda is below.

Time	Title	Speaker or panel leads
Monday December 5, 2016 (Arlington Hilton, Gallery I&II)		
7:30	Continental breakfast	
8:30	Welcome to NSF	Linda Sapochak
8:35	Quantum leap	Tomasz Durakiewicz
8:55	NSF midscale instrumentation	Guebre X. Tessema
9:15	Workshop goals and methods	Collin Broholm
9:25	Scan probe optical imaging	Dmitri Basov
10:00	Coffee break	
10:15	Breaking synthesis rules to cultivate quantum materials	Darrell Schlom
10:50	To the extremes of pressure	Russell Hemley
11:25	Instrumentation for quantum computing	Matthias Steffen
12:00	Working Lunch	
1:15	Parallel panel sessions: Instrumentation	
Present, discuss, write.	P1: Beamline instrumentation (da Vinci)	Stephen Kevan and Alan Tennant
	P2: Microscopy, spectroscopy, scan probe (Gallery III)	Margaret Murnane & Abhay Pasupathy
	P3: Extreme sample environments (Gallery II)	Russell Hemley and Vivien Zapf
	P4: Synthesis and materials discovery (Picasso)	Tyrel McQueen and Nitin Sammarth
	P5: Quantum structures and devices (Gallery I)	Oskar Painter and Jungsang Kim
5:30	Refreshments followed by Working Dinner	
Tuesday December 6, 2016 (Arlington Hilton, Gallery I&II)		
7:30	Continental Breakfast	
8:30	Topology in strongly correlated systems	Piers Coleman
9:05	P1: Summary of discussions	Stephen Kevan and Alan Tennant
9:25	P2: Summary of discussions	Margaret Murnane & Abhay Pasupathy
9:45	P3: Summary of discussions	Russell Hemley and Vivien Zapf
10:05	Coffee	
10:20	P4: Summary of discussions	Tyrel McQueen and Nitin Sammarth
10:40	P5: Summary of discussions	Oskar Painter and Jungsang Kim
11:00	Advances in microscopy for quantum materials	Abhay Pasupathy
11:35	Fundamental and applied quantum materials	Supratik Guha
12:10	Working Lunch	
1:30	Parallel panel sessions: Fundamental Science and Applications	
Present, discuss, write.	PA: Superconductivity (Gallery III)	Peter Abbamonte and Peter Johnson
	PB: Quantum Magnetism (Picasso)	Igor Zalianyak and Stephen Wilson
	PC: Topological Materials (da Vinci)	Collin Broholm and Dan Dessau

	PD: Applied Quantum Materials (Gallery I & II)	Evelyn Hu and Jiwoong Park
<b>4:15</b>	<b>Coffee</b>	
4:30	PA: Summary of discussions	Peter Abbamonte and Peter Johnson
4:45	PB: Summary of discussions	Igor Zalianyak and Stephen Wilson
5:00	PC: Summary of discussions	Collin Broholm and Dan Dessau
5:15	PD: Summary of discussions	Evelyn Hu and Jiwoong Park
<b>5:30</b>	<b>Main meeting Adjourns</b>	
	<b>Wednesday December 7, 2016 (NSF Stafford Place 1; Room 970)</b>	
<b>7:30</b>	<b>Continental Breakfast</b>	
8:30	Report writing	Leads: P1-P5 & PA-PD
11:30	Meet NSF staff	Co-chairs
<b>Noon</b>	<b>Working Lunch</b>	
<b>1:30</b>	<b>Adjourn</b>	



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