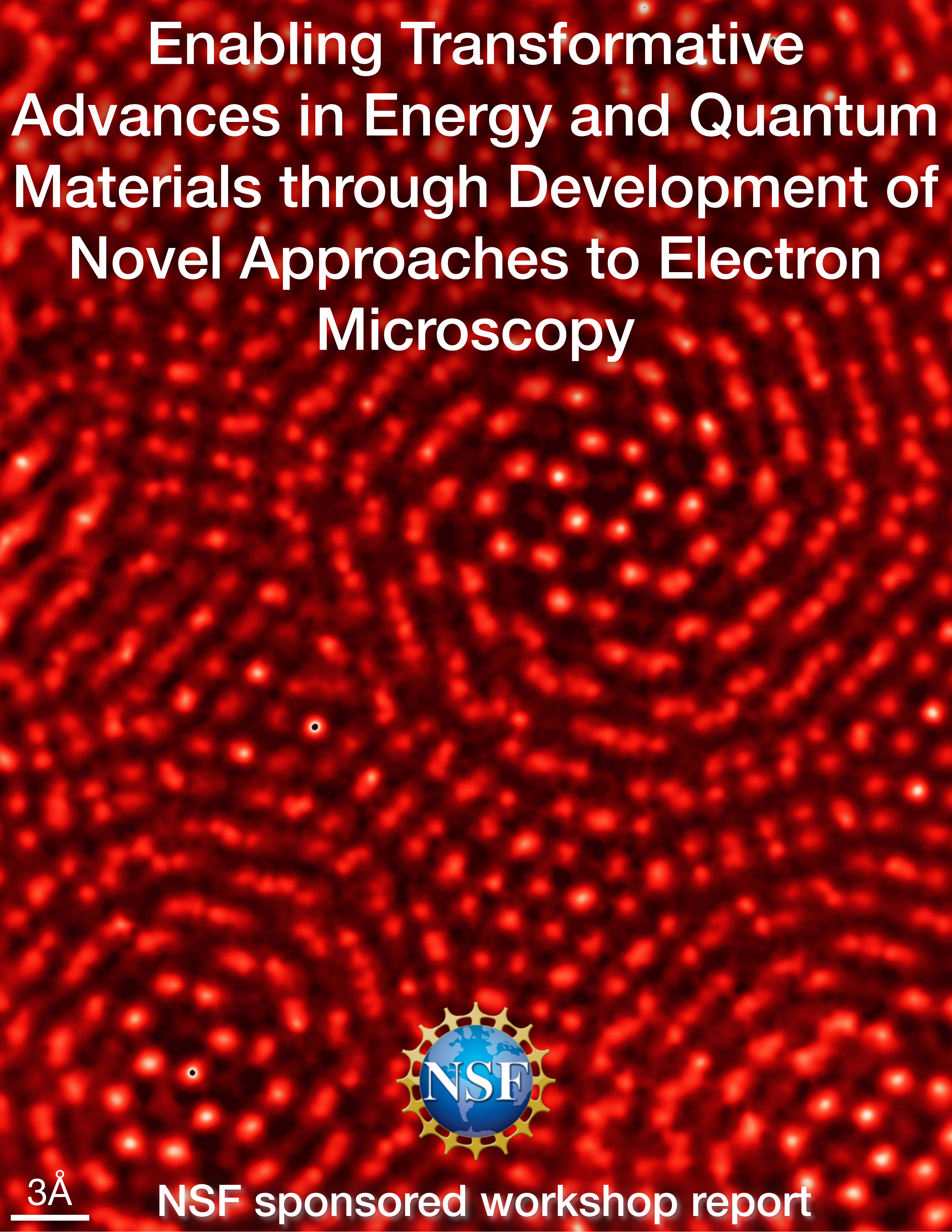
Enabling Transformative Advances in Energy and Quantum Materials through Development of Novel Approaches to Electron Microscopy



NSF sponsored workshop report

Workshop Report on Enabling Transformative Advances in Energy and Quantum Materials through Development of Novel Approaches to Electron Microscopy

Virtual Workshop: September 4 and 6, 2020.

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Cover image: Electron ptychography of a twisted bilayer of MoS_2 demonstrating deep sub-Ångström resolution at 80 keV. Image adapted from reference (Jiang, Chen et al. 2018).

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Executive Summary

Advances in electron microscopy over the past decade have opened new windows into how materials and devices function at the atomic scale. Today, microscopy provides unprecedented insights into physical, chemical and biological processes and structures. With rapid developments of data-based tools and approaches for electron microscopy, we are now at the start of a new era of atomic-scale characterization of *complex* and *heterogeneous* materials systems and problems. This workshop report describes a future road map for electron microscopy with potential to transform virtually all areas of physical, chemical, biological and materials science and engineering. In this report, we focus specifically on two key areas, Energy and Quantum Materials. We define grand challenges and identify infrastructure needs to address these challenges. Building such infrastructures and making it widely available will have broad impacts beyond energy and quantum science and engineering.

Modern society thrives on a constant supply of energy and information which impacts human health, food and water, transportation, production, security and financial systems. Environmental challenges and information security, drive the need for improvements in energy and information technologies. Breakthroughs in sustainable energy and quantum information systems will be transformative by providing access to abundant clean energy as well as rapid and secure information processing. Advanced materials play a key role in these areas and developing a fundamental understanding of the relationship linking atomic structure and functionalities is essential to creating new technologies. Electron microscopy is a primary tool for probing atomic-level structure and properties in materials, but current systems are not able to provide the information required to understand many of the key complex material behaviors associated with function. Currently available hardware is often insufficient to detect required signals and the increasing complexity of instrumentation yields large datasets which routinely outstrip existing processing capabilities. Completely new approaches to hardware/software integration along with re-engineering of human operator-microscope interfaces and interactions are required.

To address these limitations, a virtual workshop was held in 2020 attended by leading scientific experts representing areas of sustainable energy, quantum materials, data science and electron microscopy. The primary goal of the workshop was to define an electron microscopy road map and infrastructure needs to accelerate scientific discovery and progress in energy and quantum materials. To guide infrastructure development, six grand challenges were identified to enable transformative progress.

Grand challenges for sustainable energy:

- How might we image catalytic reaction pathways taking place on particle surfaces?
- How might we determine the structure, composition and evolution of solid-liquid interfaces under conditions relevant to electrochemical devices?
- How might we observe transport pathways through materials and interfaces?

Grand challenges for quantum science and technology:

- How might we understand quantum decoherence in materials and devices?
- How might we map electron correlations and the emergence of new phases?
- How might we measure magnetic moments of individual atoms?

Infrastructure needs were identified focusing on six key areas covering hardware, data science, theory/simulations, open-source software, training and community engagements. The most important hardware need included more stable cryogenic microscopes, faster and more sensitive electron detectors and improved *in situ* capabilities with the ability to synchronize with

a wide range of applied stimuli. Full integration of data science and simulation methods into instrument design is essential to maximize the impact of next generation microscopes. Such an approach requires co-design of microscope systems with edge computing hardware and software together with direct integration with large-scale, high performance computing resources.

The immense opportunities for scientific and societal impact through electron microscopy discussed in this report call for new investments in electron microscopy infrastructure as well as hardware, software and methods development programs. Such investments will be critical in retaining US leadership in electron microscopy instrumentation and innovation for accelerated scientific discovery and development of new technologies.

1. Introduction

Progress in science and engineering research has a major impact on society affecting almost every sphere of human activity including construction, manufacturing, transportation, energy, the environment, agriculture, and human health (NAE 2017). Advances in each of these areas has a large impact in economic development contributing directly to improvements in quality of life. Technological advances often flow from development of devices and processes that rely on properties of the materials employed in their manufacture. Indeed, the properties and functionality of materials and materials systems are critical in the performance of many engineering applications such as catalysis, buildings/structures, computing, communication, energy conversion/storage, and biomedical devices. Transformative advances may lead to completely new technologies with potential that can only be imagined at the present time. For example, the new area of *quantum science and devices promises to revolutionize computing, sensing and encryption in ways that may be difficult to anticipate*. Similar arguments can be given for the impact of materials advances in many fields and in some areas relatively small improvements can make a large impact on society. For instance, it is estimated that *90% of all manufactured products involve catalytic processes somewhere in their production chain*, and such products have considerable impact in energy, healthcare (pharmaceuticals), materials synthesis, transport, and the environment contributing 30–40% of global GDP (Bravo-Suárez, Chaudhari et al. 2013). The ability of engineering to build an environmentally sustainable society will be critically dependent on creating new materials and materials systems with functionalities that enable the development of clean, efficient technologies.

While materials are ubiquitous in engineering, there are many properties of technologically relevant materials systems that remain unknown or poorly understood. For example, various manifestations of interfacial processes (solid-solid, solid-liquid and solid-gas interfaces) play a key role in catalysis, separations, energy storage/conversion, environment, optoelectronic devices communication, and quantum systems. However, our knowledge of interfacial phenomena that occurs when systems interact across varying length scales remains limited. Extreme conditions, weak interactions or rapid dynamics may make interfacial structures inaccessible with current *in situ* characterization technology. For example, many electrocatalytic systems – such as the hydrogen evolution reaction for water splitting – only operate in strong caustic or acidic electrolytes and often at very high current densities, conditions which are incompatible with many *in situ* characterization tools (Osterloh 2013, Pinaud, Benck et al. 2013). Interfacial structures often catalyze chemical conversion processes that are critical to fields such as pharmaceutics, agriculture, energy storage and conversion, yet we still have only a rudimentary knowledge of the processes taking place and how interface structure and functionality can be manipulated and controlled. Space charge effects may drastically affect transport processes and fundamentally regulate the properties of surfaces and interfaces (Feng, Lugg et al. 2017, Vikrant, Chueh et al. 2018). Similarly, the promise of devices based on quantum materials is vast, but we struggle to locally sense quantum phenomena in materials systems. The complexity and weakness of the interactions make it challenging to link materials structure and chemistry with quantum phenomena like topology, entanglement and transduction.

Many of the performance improvements for applications may not be possible with existing materials and will rely on the discovery of new materials which are tailored for specific engineering applications. The vision of the Materials Genome Initiative (MGI) is to integrate modern tools for computation, experimental measurement, and digital data in order to dramatically reduce the time and cost associated with materials discovery, design, and commercialization. However, in many engineering applications, microstructural features such as interfaces, defects, and surface structures are critical for regulating functions such as transport and chemical transformation. Accurate theoretical predictions of the functional properties in micro- or nano-structured systems is often not yet possible.

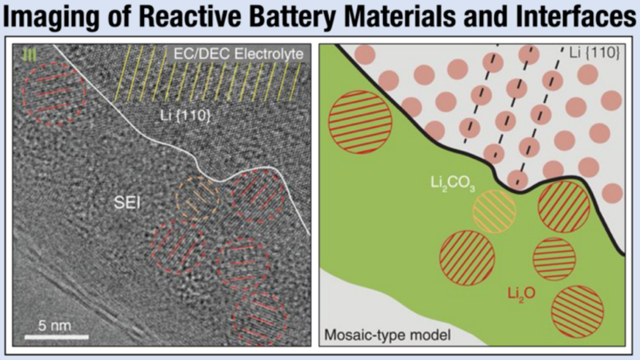
Fortunately, fundamental knowledge in energy and quantum materials may be addressed through advanced nanoscale and atomic resolution probing of materials. Electron microscopy (EM) imaging, spectroscopy and diffraction is a primary tool for probing materials structure, chemistry and properties at the atomic level. Tremendous advances have taken place over the last ten years in the areas of aberration correction, monochromation, detectors, and *in situ/operando* probing along with theoretical advances in our understanding of image and spectral signal generation. These advances in microscopy have resulted in an enormous increase in our understanding of materials impacting many areas of engineering, biology and physical sciences. The enormous impact of microscopy to science and engineering was recently recognized with the award of the 2017 Nobel Prize in Chemistry to Dubochet, Frank, and Henderson for cryo-electron microscopy and the more recent award of the Kavli Prize in 2020 to Haider, Krivanek, Rose and Urban for the development of aberration correctors.

1.1 Role of Materials Development in Energy Conversion Process

Materials are ubiquitous in current and future energy technologies. Structural materials play a major role in energy conservation, e.g., light weight for aerospace application, higher temperature stability to improve the efficiency of Rankine cycles in power plants. The definition of energy materials can be very broad but the workshop focused primarily on fundamental questions that must be addressed to develop future ***sustainable technologies*** associated with "free" intermittent sources such as solar and wind along with required storage solutions. More specifically, the primary focus was on technologies based on thermal, thermo-chemical, photo-chemical, and electrochemical processes.

At present in the US, nearly 80% of our energy is derived from fossil sources (EIA 2020). Many of the chemical conversion processes employed in manipulating fossil sources operate far from the maximum efficiency. Due to convenience, high energy density and existing energy infrastructure, it will take time to fully transition away from strong dependence of fossil fuels. To quote Richard Heinberg "Fossils fuels are the equivalent of a huge inheritance……other energy sources are more analogous to wages: we will have to work for what we get and our spending will be restricted to our immediate income" (Heinberg 2005). Indeed, projections out to 2050 from the US Energy Information Administration suggest that fossil fuel utilization may remain close to 70% both domestically and internationally. This reality makes it clear that in addition to developing fossil-free energy sources, it is also critical to develop far more efficient technologies to utilize fossil fuels (especially natural gas). These ***transitional technologies*** must push to the thermodynamic limits for chemical conversion processes for hydrocarbons. Much of the

Imaging of Reactive Battery Materials and Interfaces



Cryogenic cooling in TEM allows reactive materials such as lithium metal to be imaged at atomic resolution. The structure of the solid-electrolyte interface (SEI) layer that forms on the surface of the Li electrode during battery cycling is resolved (Li et al., 2017).

fundamental knowledge in thermal, catalytic and electrochemical processes required to develop transitional technologies, will also be applicable to sustainable technologies.

Many of these transitional and sustainable technologies rely on materials functionalities and properties related to light harvesting, transport, reactivity and stability. Catalysis plays a key role in many of the chemical transformations taking place in energy conversion systems. Catalysts serve as kinetic switches to accelerate desirable reactions while shutting down undesirable reactions. In heterogeneous catalysis, enzymes and materials surfaces orchestrate and control the kinetic pathways. The chemical reaction is controlled by selective charge transport taking place on the surface at so-called active sites. Understanding the relationship between catalyst structure and reactivity is essential to manipulating and controlling catalysis to increase the efficiency of an energy conversion processes. Transport in many forms is also critical to developing new and improved energy conversion and storage processes. In a battery or fuel cell, electronic and ionic transport is essential to device function. How does a material's atomic structure, nanostructure, defects, composition and bonding regulate such processes and how can we manipulate the material to enhance conductivity? How can we enhance the ability of a material to not only store ions but also enable the system to easily and reversibly charge and discharge thousands of times with no capacity fade? Phonon transport regulates heat transfer in thermal processes and is also key in technologies such as thermo-electrics. Phonon coupling with photons and electrons giving rise to polaritons and polarons which may be exploited in both solar and thermally driven devices.

For all of these functionalities, the atomic level structure, composition and bonding of the materials systems will regulate and control the fundamental behaviors required for energy conversion systems. Developing a fundamental understanding of these structure-function relationships will be impossible without a detailed dynamic atomic level view of the materials. *In situ* probing of static and dynamics material structure under will not be sufficient. Future electron microscopes must provide atomic level characterization of technologically relevant functionalities such as transport and reactivity not only in perfect crystals but at defects, interfaces and surfaces.

1.2 Role of Materials Development in Quantum Science

The development of materials and their integration in devices has shaped modern society through technical achievements such as the invention of the transistor, the laser, and the integrated circuit, which today form the backbone of modern computers, telecommunications, and the internet. New quantum technologies including quantum computing, quantum information processing, quantum networking and quantum sensing are expected to have similarly transformative impacts on modern society and will likewise depend on the development of next-generation materials. The potential of a *Quantum Leap* and the research that is necessary to make such a quantum revolution a reality is recognized as one of NSF's *Ten Big Ideas*.

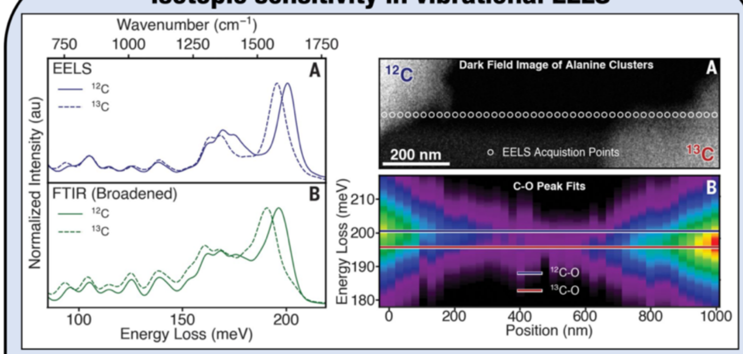
A large number of materials systems are currently being explored as building blocks for various quantum technologies including superconducting qubits, semiconductor quantum dots, defect centers in solids such as diamond, silicon carbide and aluminum nitride, and quantum materials in which correlations give rise to new quantum states. In all cases, materials have to be optimized in order to maintain and control their quantum properties. Being able to harness these properties in practical quantum technologies requires integration of materials into device structures which presents additional challenges especially because of the increased sensitivity to even low levels of disorder or fluctuations.

The goal in designing new quantum materials is to enhance and maximize desired quantum properties as well as to realize new ways to control and manipulate quantum states. The ability to rationally design such materials rather than discovering them accidentally requires advancing our microscopic understanding of how new properties or quasi-particles emerge on both the atomic and mesoscopic length scales. In addition to providing a platform for developing next-generation quantum technologies, quantum materials research is also of fundamental scientific interest. Quantum materials can exhibit exotic physical properties, including topological protection, colossal magnetoresistance, high temperature superconductivity, multiferroicity, and nanoscale electron self-organization. The ability to tune and control such functionalities in new materials will lead to transformative applications in energy, sensing, and information technologies. The interactions between charge, spins, and the atomic lattice are so strong that most phenomena occur at atomic length scales. For these reasons, new capabilities in probing materials structure and functional characteristics down to the atomic scale will underpin the realization of quantum materials, devices, and systems.

1.3 Problems and Opportunities for Electron Microscopy

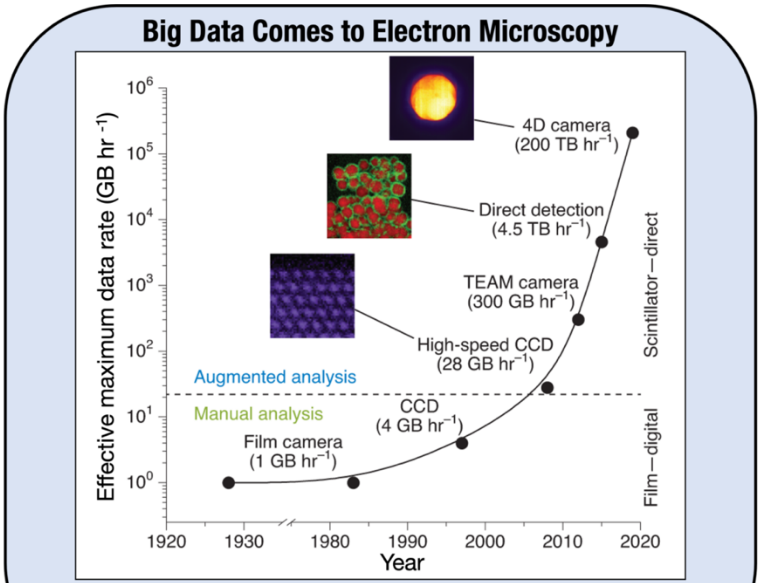
As remarkable as advances in TEM have been over the last 10 years, they have not yet been effectively integrated, combined, and implemented in a way to optimize the solution of complex materials problems. In some areas, current technology is insufficient to provide the necessary information for solving critical materials problems. For example, while modern TEM-based electron energy-loss spectroscopy can now provide energy resolutions of better than 5 meV, the energy scale of many quantum phenomena is measured in small fractions of a meV. Subtle changes in the nanoscale distribution and character of electrons may fundamentally alter the properties of materials, yet there are no good methods to reliably detect small changes or correlations over length scales relevant to quantum systems. Cooling to cryogenic temperatures without compromising resolution in imaging, scattering and spectroscopy experiments will be required for a range of quantum and energy systems but is not currently available (Minor, Denes et al. 2019). New detectors with high detective quantum efficiency and high readout rates (Hart, Lang et al. 2017, Faruqi and McMullan 2018, Ercius, Johnson et al. 2020, MacLaren, Macgregor et al. 2020, Plotkin-Swing, Corbin et al. 2020) offering temporal resolutions better than 10^{-3} s can in principle allow the observation molecular intermediates on surfaces and at interfaces critical for catalysis, energy storage, and conversion. In practice, radiation damage at high electron dose and poor signal-to-noise (SNR) restrictions at low electron dose severely limit the information on atomic level dynamic processes. Potential strategies to mitigate this issue depending on the situation and include cooling/heating of the sample, machine learning approaches for noise reduction, and even developing a so-called quantum microscope (Kruit, Hobbs et al. 2016, Juffmann, Koppell et al. 2017) to enhance SNR through quantum interference effects.

Isotopic sensitivity in vibrational EELS



Vibrational spectroscopy with monochromated EELS performed in a STEM resolves carbon-site–specific isotopic labels in real space with nanoscale spatial resolution (Hachtel et al., 2019).

Progress in TEM-based characterization has driven developments in computational approaches such as *ab initio* methods, signal simulation, and inverse/reconstruction algorithms



Effective maximum microscope data-production rates by year showing the rapid increase associated with better detector technologies (Spurgeon et al., 2021).

that assist in extracting relevant information from complex experiments. To date, however, the immense volume and variety of data collection has vastly outpaced downstream analysis approaches. In turn, a vast majority of collected data is under-analyzed, leaving crucial physical insights unexplored (Spurgeon, Ophus et al. 2021). Given the increasing complexity of instrumentation and the enormous datasets routinely generated from new detectors, data processing is now a significant limitation. For some experiments the mere task of rapidly saving and moving the data is a formidable computing challenge.

It is important to re-engineer human operator-microscope interfaces and interactions. Engineering designs centered on high-throughput automation and data intensive workflows could provide materials scientists and engineers with rapid experimental feedback on a more statistically significant volume of samples, rather than a limited subset. Additionally, experiments requiring significant input and control from the human operator can risk introducing observational bias and hampering data interpretation. Such limitations may be overcome by co-designing data acquisition with scientific computing systems. Data-based approaches may also provide a path to correlate multimodal signals spanning varying length and time scales to understand functionalities of quantum and energy materials. Contemporary materials modeling and simulation will not only impact data analysis and interpretation but also inform improved experimental design.

1.4 Workshop Goals

There are many potential opportunities to make progress on the questions raised above and engaging the community in a workshop to identify the strategically most important new directions was necessary. The workshop brought together experts in materials, microscopy, and data science to identify the most important problems in energy and quantum materials and to articulate new microscopy infrastructures that should be developed. ***The primary goal of the workshop was to define a future road map for electron microscopy and to identify infrastructure needs to accelerate scientific discovery and progress in the fields of energy and quantum materials.***

Due to the COVID 19 health crisis, the workshop was virtual, and we worked with Knowinnovation, who have extensive experience in facilitating creative scientific workshops. In addition to engaging approximately 40 leading microscopists in the US, a dozen international experts in either energy or quantum were invited to articulate and initially lead discussions of the most important outstanding questions in their respective fields. Data scientist were also invited to inform the community of new developments in data science and the need to engage data scientist at all levels in the design process.

One motivation for the workshop was to discuss possible opportunities to seek support for microscopy infrastructure development through programs like the NSF Midscale Infrastructure Program. During the workshop there was also a strong desire by the workshop participants to build a strong community that would facilitate rapid development of microscopy hardware, software and education. This is reflected in the six infrastructure solutions deemed necessary by the workshop participants to drive the electron microscopy community which, in addition to hardware and software development, also proposed solutions in, outreach to scientist and engineers, education and more active engagement of the general public to recognize the role of microscopy in society.

The output from the workshop is given in the following three chapters. Chapter 2 and 3 explore problems and opportunities for advances in energy and quantum materials respectively. Chapter 4 describes solutions that the community should explore to strengthen and build microscopy infrastructure with the US. Various appendices list workshop participants, agendas and more detailed data from some of the workshop topics.

2. Energy Materials Science Drivers, Critical Questions and Solutions

The workshop focused primarily on fundamental questions that must be addressed to develop future sustainable technologies associated with "free" intermittent sources such as solar and wind along with required storage solutions. Such an infrastructure must develop inexpensive and environmentally benign approaches to harvest, store and release the energy to match demand. A big success in this space is the photovoltaic panel. Starting in the 1950's with the development of the modern semiconductor solar cell based on p-n junctions, and intensive research motivated in part by the needs of the space program, solar panels are now widely available. Commercially available single crystal and polycrystalline solar cells have efficiencies ~70% of the Shockley-Queisser limit and continued improvement in manufacturing has reduced cost to the point where they have become a commodity. A similar story can be told for wind turbines which are operating close to the Betz limit especially for offshore installations. While the cost per watt of these energy sources continues to fall, the intermittent character of solar and wind means large-scale penetration can only move forward if the energy storage problem is addressed. The most promising storage options rely on thermal, thermochemical, photochemical and electrochemical processes. Technologies based on these approaches have the greatest potential to shift society to a zero-carbon emission energy infrastructure in the long term. Moreover, finding solutions to the many challenges associated with exploiting solar/wind resources would also find application in development of transitional technologies.

Initially, workshop participants identified major problems in the sustainable energy space which served as science and engineering drivers to motivate the infrastructure developments in microscopy. It is impossible to consider the problem of energy storage without discussing batteries. Batteries permit direct storage and release of electricity generated by solar and wind sources. It is the dominant storage mode for small devices such as cell phones and computers and is making significant penetration into the personal transportation market with the growth of electric car sales. It is not clear whether batteries will offer a viable solution for grid level storage of electricity from solar and wind generation. There remain significant problems in their limited energy density, charging speed and durability. Battery functionality relies on reversible intercalation and charge transport process taking place in the anodes, cathodes and electrolytes and for improved performance these processes must be understood across multiple length scales. The phase changes and transport mechanisms that allow electrons and ions to rapidly move in and out of the electrode materials in a reversible manner are poorly understood. Many aspects of the ion and electron transport taking place across the electrode/electrolyte interfaces are unknown. Understanding and controlling transport and phase transformations in battery materials will not only enable large scale storage solutions for stationary sources but will also have a dramatic impact on all areas of the transportation sector.

Another approach to large-scale energy storage is through fuel generation such as hydrogen. In water electrolysis, for example, electrolyser efficiencies are in the 70 – 80% range and this technology is commercially available today. For solar fuels, there is considerable interest in developing photocatalytic, electrophotocatalytic and thermo-chemical technologies to directly generate hydrogen from photons to reduce overall system costs. Carbonaceous fuels can also be generated via artificial photosynthetic systems where CO_2 reduction can be achieved, for example, using solar hydrogen. Extensive research has been conducted over many decades but to date no viable commercial technologies have been developed for direct solar-to-fuel conversion.

The materials challenges that must be solved to develop a technology with these chemical processes are formidable, but the potential rewards are enormous. Photon generated electron-hole pairs must be continuously separated and transferred to surface catalytic sites (typical a co-catalyst) where catalytic reaction occur. This process is plagued with recombination hindering efficiency. The most efficient catalytic systems are not stable and deactiviate due to the harsh reaction conditions. For example, in many materials systems, it is thermodynamically more favorable for the material to decompose rather than the H_2O or CO_2 reactants leading to severe photocorrosion of the catalyst. Developing an atomic level understanding of the reaction pathways is essential to control the selectivity to disable undesirable reactions which poison or destroy the catalyst. Thermal catalysis is central to sustainable and transitional energy conversion and storage systems, but our understanding of the interplay between nanoparticles surfaces, phonons and activation of reactants is missing. The problem of inefficient activation of reactant permeates all energy systems based on chemical conversion, and can only be solved by mapping out the fundamental atomic and chemical processes taking place on catalyst surfaces under reaction conditions.

In addition to making fuels from photons, it is also necessary to improve methods of making electricity from fuels. In a fuel cell for example, achieving the thermodynamic conversion efficiency is limited in part by poor performance and lack of understanding of the catalytic processing taking place on the electrode materials particularly the oxidation/reduction reactions on the cathode. There are considerable gaps in our understanding of the oxygen exchange reaction and how it is influenced by surface structure. To address this problem, it is not enough to perform *in situ* observations. We must be able to visualize not only the catalyst surface structure, but also the molecular intermediates so that we can link local chemical conversion and kinetics to local surface structure.

Ionic transport processes are also critical to fuel cell and battery operation. Charge transport also plays a key role in other energy conversion processes such as photocatalysis. At present the role of atomic level defects, interfaces and surfaces on regulation of ion, electron and hole transport is poorly understood. First principles calculations of transport at complex interfaces are often beyond current capabilities and macroscopic experimental transport measurements typically average over many heterogenous structural components. To deepen our fundamental understanding of charge transport we must be able to make direct, atomic-level observations of transport taking place at defects, interfaces and surfaces. Corrosion is associated with ion transport between a solid and a liquid so observing the atomic structure of the solid-liquid interfaces may provide solution to address durability issues.

Thermal science plays a critical role in energy conversion and phonons transport controls how heat is moved within the system. To fully master and manipulate thermal processes, we must understand local phonon transport especially across interfaces and near defects. How does local material structure and composition regulate phonon modes and transport? How can we observe these processes at high spatial and temporal resolution in order to develop and match materials to desirable phononic properties?

To solve many of the problems discussed above, the workshop identified three grand challenges which if addressed would lead to transformative progress in sustainable and transitional energy technologies. These challenges are:

QE1: How might we image catalytic reaction pathways taking place on particle surfaces (gas-solid reactions triggered by heat,light or electric field)?

QE2: How might we determine the structure, composition and evolution of solid-liquid interfaces under conditions relevant to electrochemical devices?

QE3: How might we observe energy, charge and mass transport pathways through materials and interfaces with atomic resolution?

Below, we discuss the solution of these grand challenges through the advancement of electron microscopy infrastructure. Antecedent questions to address these grand challenges are discussed along with anticipated necessary observations including:

What measurements are required?

What precision, sensitivity, spatial resolution, and temporal resolution is needed?

What new data science and modelling approaches are required to extract the scientific information from large noisy datasets?

Many of the hardware and software needs to solve one grand challenge may also impact other grand challenges. To avoid excessive repetition, we present a detailed discussion of approaches for QE1 with the recognition that many of the same strategies will also benefit QE2 and QE3.

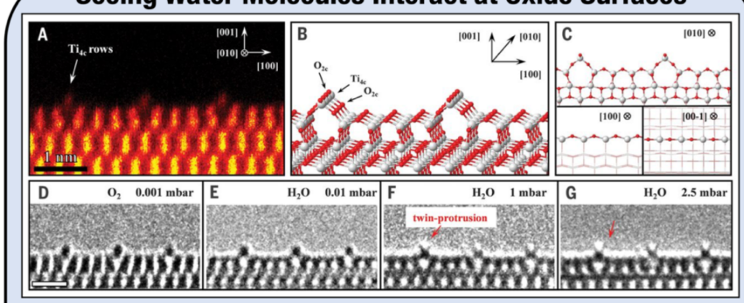
2.1 QE1: How might we image catalytic reaction pathways taking place on particle surfaces (gas-solid reactions triggered by heat, light or electric field)?

It is now relatively common to perform atomic resolution imaging of the surface of a catalyst under reaction conditions. For gas phase reactions, MEMs based holders and differentially pumped TEM columns allow nanoparticles to be imaged in the presence of reactant gases and at elevated temperatures (Yokosawa, Alan et al. 2012, Tao and Crozier 2016). Different approaches to *in situ* and *operando* measurement have been performed to correlate the observed surface structures directly to catalytic processes and chemical kinetics (Vendelbo, Elkjær et al. 2014, Vincent, Vance et al. 2020). There has been some success in developing an understanding of the chemical engineering aspects of *in situ* electron microscopes so that averaged quantitative chemical kinetic measurements can be performed. However, such an approach does not provide information on local intermediate reactions taking place on the surface. To observe reaction pathways on the catalysts, it is necessary to observe the chemical processes taking place directly on the nanoparticle surface at specific locations such as step edges, terraces or interfacial sites. Several strategies to accomplish this goal were discussed in the workshop.

2.1.1 Finding Adsorbates and Intermediates

In energy conversion applications, the reactants and products are usually light element molecules (e.g., H_2, CO, CH_4, O_2 etc.…) and direct observation of such adsorbates on catalytic nanoparticle surfaces is extremely challenging due to weak contrast and radiation damage considerations. However, there have been two reports in which the authors claim to have made direct observation of observing CO, OH and H_2O adsorbates on nanoparticles surfaces in the electron microscope (Yoshida, Kuwauchi et al. 2012, Yuan, Zhu et al. 2020). These observations, though noisy, suggest that it is possible to observe a *static* adsorbate. To observe the reaction pathway, it is necessary to image the evolution and transformation of the adsorbate species on the surface with high spatial and temporal resolution. While this is an extremely challenging, with significant improvements in sensitivity, experimental design and data analysis, it should be feasible. At the more active sites, the atoms making

Seeing Water Molecules Interact at Oxide Surfaces



The TiO_2 (001) surface shows a (1 x 4) surface reconstruction in vacuum. In a water vapor environment two water molecules adsorb at the protruding Ti row (labelled in A) and give rise to the twin protrusions (labelled in F) (Yuan et al., 2020).

up the surface of the nanoparticle may undergo significant displacement due to the significant bonding and debonding associated with converting reactants into products. For example, during oxygen exchange on non-stoichiometric oxide surfaces, cation displacements can be on the order of tens of picometers. Observing dynamic fluctuations on the surface of the catalyst under *operando* conditions would indicate the likely locations of the most active sites. It is not clear if phase contrast TEM or STEM approaches are best for imaging molecular dynamics on a nanoparticle surface. 4D STEM and pytchography provides signals with greater post-processing flexibility offering the potential to explore the sensitivity of different parts of the electron angular scattering distribution for molecular imaging. STEM techniques also enable locally probing of chemistry using spectroscopy providing critical information of the character of the intermediates. However, a current limitation of the 4D STEM approach is that the image frame rates are too slow to track structural dynamics at millisecond temporal resolutions or faster.

2.1.2 Turnover Frequencies, Cyclic Behavior and Temporal Considerations

Catalytic turnover frequencies at an active site can range anywhere from 1 to 100 conversions per second so temporal resolution on the order of 0.001s or faster are desirable but ultrafast approaches may not be required. A catalytic reaction typically moves through a series of metastable states separated by transition states associated with energy barriers. The lifetime of the metastable states may be measured in thousandth to tenths of seconds depending on the thermodynamics and kinetics of the local energy landscapes. The transition states are very short lived and will not be directly observable with the techniques described here. However, the duration of the metastable states and transition probabilities will provide information on the thermodynamics and kinetics as the system evolves. Information on these intermediates will provide valuable input for first principles modelling. The information will be local and thus the kinetics and reaction pathways associated with different nanoparticle facets as well as edge, corner and defects sites will be determined. This will not only provide a fundamental understanding of how atomic structure on surfaces regulate and control chemical reactions but will also point the way forward for designing surfaces with desired chemical reactivity.

The catalytic turnover process is cyclic but not periodic in time. In the conventional picture, adsorbates arrive at an active site, undergo dissociation, molecular components eventually re-assemble into product molecules, product molecules desorb, and the entire process begins again. For thermochemical reactions, the energy to activate reactants and overcome kinetic barriers comes from thermal fluctuations which excite the intermediates into transition states. The fact that the process is cyclic offers opportunities to address signal-to-noise and radiation damage limitations by average signals recorded from the same metastable state. Thus, it will be necessary to develop data science methods to recognize the spatio-temporal fingerprints of the cyclic turnover processes. Approaches such as change point analysis and detection of rare events in noisy temporal data stream is essential (Matteson and James 2014).

It may be possible to make the turnover processes periodic by using external triggers. For example, for photocatalytic process this can be accomplish using a visible light pulse and for a thermochemical reaction by an IR pulse. Electrochemical reactions could be triggered with electric field pulses. These approaches would allow much more control in experimental design and would increase the power of data science methods to explore the role of heterogeneity on the evolution of metastable intermediates. Such experiments will require the development of electron detector systems that can be precisely synchronized at the 10^{-3} – 10^{-9}s level with the application of a wide range of externally stimuli.

2.1.3 Electron Beam Effects

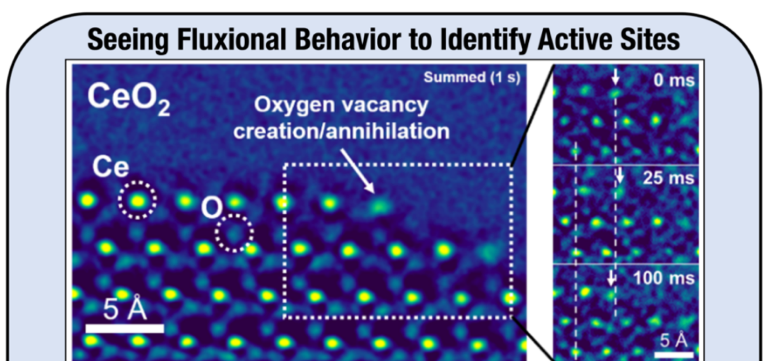
For all electron microscopy work, it is necessary to understand and manage electron beam effects. The electron beam may cause the molecules to desorb from the surface or it may drive different chemical transformations than those of interest. Radiation damage often involves radiolytic bond scission follow by diffusion of fragments leading to irreversible structural change. The ability to control the effect of radiation damage on the observation may be greatly enhanced using a pulsed electron source. If a short duration pulse is employed, it may be possible for bonds to reform before the next pulse arrives. If desorption takes place, then the remaining vacant site will be rapidly repopulated with adsorption of another reactant. Flexible pulsed electron sources are critical to increase our understanding of localized chemical reaction pathways and kinetics.

2.1.4 Efficient Hypothesis Testing with Modelling and Simulation

With predictions and simulations, it should be possible to generate a series of hypotheses for what the experimental image contrast should look like as the chemical conversion process moves from reactants through intermediates to products. These hypotheses would be employed to design workflows to maximize the information content of and experiment. There may be substantial prior information from techniques like IR, STM, etc... which can guide the development of reasonable structural hypothesis. A series of initial estimates of likely configurations determined by say, DFT or structure libraries, could be employed to simulate images and diffraction patterns for immediate comparison during an experiment. With such an approach, we may be able to observe the location of intermediate species at a single site before the product rapidly desorbs from the surface. Complete integration of computational materials science, data bases and surface chemistry will be critical for success. There is need to develop rapid algorithms which can guide the experimental data interpretation under near on-the-fly conditions. This does not necessarily imply inversion of experimental data but rather developing efficient feedback mechanisms to vary the theoretical models so that iterative forward simulations converge rapidly to the experiment using data-driven approximators and generative models. This integration of experiment and near on-the-fly simulation will be essential for creating efficient workflows to maximize the efficiency of extracting information from large dynamic datasets. A special role here will be played by Bayesian methods (Gelman, Carlin et al. 2013) from simple inferential models to deep probabilistic programming, naturally comporting to the structure of prior knowledge and largely known causal links in modern physical sciences.

2.1.5 Electron Microscopy to Image Catalytic Reaction Pathways

The advances in electron microscopy required to accomplish the scientific goals described above are now explicitly described. These needs are presented in terms of advances in hardware, data science and simulation. However, it is important to emphasize the integration of these three components directly into a novel electron microscope platform is essential for transformative advances. The new science that will be available from the enormous data



Cation column fluctuations associated with the creation/annihilation of oxygen vacancies at surface site. A sequence of 25 ms exposures resolves dynamic fluctuations in the Ce column position (right, white arrow) (Lawrence et al., 2020).

sets generated by the proposed new detectors will be inaccessible unless directly coupled with automated analysis.

2.1.5a Hardware

Commercially available direct electron cameras currently provide 1000 fps (3 – 4 times faster on small readout areas). It is desirable to sample statistically large sample areas (100 nm wide at 0.5 Å resolution) at high spatial and at high temporal resolution to provide flexibility and variability in averaging techniques. A camera to perform parallel-beam TEM imaging would be able to readout images at least 10k x 10k pixels in size at 10,000 frames per second with detective quantum efficiencies near the theoretical limit (McMullan, Chen et al. 2009, Ruskin, Yu et al. 2013, Faruqi and McMullan 2018). *In situ* 4D STEM and pytchography provides signals with greater post-processing flexibility than broad beam TEM imaging, and ptychography in particular makes it possible to trade real-space for reciprocal space pixels, trading frame rate for data rate (Chen, Odstrcil et al. 2020). Recent developments of 4D STEM have demonstrated cameras that can generate almost 100,000 fps albeit at 1 bit depth, i.e. a data rate of 20 Gbits/second (Ercius, Johnson et al. 2020). However, the current generation of detectors is not fast enough to enable 4D STEM imaging at millisecond time resolution. For example, to achieve temporal image resolutions of one millisecond for a 1000 x 1000 pixel image, a diffraction pattern readout rate of 10^9 pixels/second is required, which is four orders of magnitude above current technologies. Even a 256 x 256 pixel image requires a readout rates on the order of 10^7. So, to enable millisecond resolution 4D STEM, a major goal for future electron detectors is to increase the readout rate to approximately 10^7 fps and many bits deep while maintaining a near-ideal DQE and MTF. It will also be necessary to develop scanning systems that can perform 1000 real-space STEM images per second on areas on the order of 100 nm in width with no distortions or flyback errors.

Many of the structure changes associated with locally observing a surface reaction may be associated with subtle changes in EM signals. It is essentially to integrate the application of *in situ* stimuli (e.g., heat, light, electric field) with synchronized detector readout. This would enable pump-probe strategies to be developed in which the detector readout is synchronize or output frames tagged with the arrival of say an IR pulse at a catalyst. The ability to precisely identify images, diffractions or spectral frames recorded at different points in a rapid cyclic application of external stimuli will be essential to map out structural dynamics associated with localized surface chemistry.

Similarly, electron sources should be developed that enable variation in pulse duration, frequency and intensity to synchronize with applied stimuli. To provide flexibility the pulse rate should be at least several orders of magnitude higher than the maximum turnover frequency of a catalysts so pulse rates of around 10 – 100 kHz would be desirable. Pulses should be long enough to avoid the Boersch effect to ensure a spatial resolution of 1 Å or better. Such sources may also play a critical role in managing electron beam damage effects.

2.1.5b Data Science, Modelling and Simulation

As the readout rate increases, on-the-fly edge computing methods become more important and may involve simple neural networks or machine learning codes (e.g., for denoising of raw data coming in at fast rates). As the volume of data generated increases with high readout rates, most of the intensity in the detector pixels will be zero and it should be possible to employ lossless compression techniques (Datta, Ng et al. 2021). Careful consideration should be given to on-the-fly processing if compression occurs via edge computing at the detector. The term "on-the-fly" is used throughout this report to mean "…something that is being changed while the process that the change affects is ongoing." If the process is raw data being generated by a detector at high rates, then edge computing will be required for on-the-fly processing in real-time. If the process is running an experiment on a microscope that takes hours or days, then on-the-fly may mean high level processing that is performed in seconds or minutes.

In the presence of high shot noise, a wide range of data manipulation techniques must be developed to extract scientific relevant information about the catalyst surface. Adsorbates species may remain static while the system is in a metastable state and so frame averaging could be performed to increase the signal-to-noise. One could envisage a 100,000-frame movie being binned into say 5 unequal time segments corresponding to 5 different metastable structures. Knowing which set of frames belong to a particular intermediate species would require the development of data analysis techniques that can detect the likely location of the frames where a transition occurs. This is an example of detection of a rare event. This analysis should ideally be performed on-the-fly so substantial computations power must be readily available at the microscope. On-the-fly denoising techniques based on convolutional neural networks should be developed so that structural information can be more easily visualized during an experiment. These approaches must include confidence estimates on the reliability of the output from the neural networks. Data science techniques such as change point analysis or hidden Markov models should be developed to refine the analysis in near real time so that it can be used to adapt conditions during an experiment. This integration of experiment, simulation and first principles modelling will be an essential part of the process of designing, performing and analysis data on-the-fly.

2.2 QE2: How might we determine the structure, composition and evolution of solid-liquid interfaces under conditions relevant to electrochemical devices?

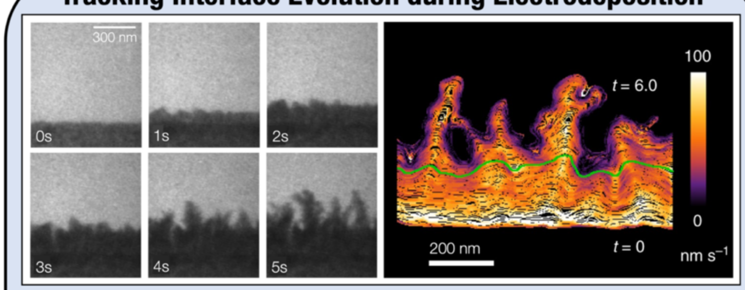
Many electrochemical energy conversion devices such as batteries, fuel cells, electrolyzers and photoelectochemical water splitting involve processes at solid-liquid interfaces. The liquids may be strongly alkaline or acid, aqueous or organic, and may also react with air. The solid-liquid interface represents one of the most challenging interfaces to investigate at atomic resolution using microscopy approaches. While there has been significant progress in liquid cell microscopy over the last 10 years, visualizing the interface under *operando* conditions is very challenging. For the simplest systems, the structure of the crystalline components at the interface may be determined using established microscopy technique and conventional surface science descriptors. The liquid part of the interface is much more challenging. Even in pure water, evidence suggests that some form of partial order may occur under some conditions. It is not clear how such structures should be described. In the presence of multiple solute ions, the structure of the charge double layers may be very complex requiring detailed probing with an array of imaging, diffraction and spectroscopy approaches. Under reaction conditions, these liquid "thin films" are likely to undergo dynamic structural changes which will certainly strongly impact the ionic and electronic transport that is critical for electrochemical functionality. Most liquid environments are very susceptible to electron beam damage making the characterization problem much more challenging.

There are several possible approaches to observe the liquid-solid interface. In many cases, even being able to characterize relatively static interfaces under equilibrium conditions would represent an important step forward. Recent and exciting progress has been made in this area by using cryogenic techniques (Li, Li et al. 2017, Wang, Li et al. 2018, Zachman, Tu et al. 2018). This involves rapidly freezing the sample in a cryogen to fix the liquid-solid interface. The sample is then observed at cryogenic temperatures which also dramatically reduces electron beam damage making atomic resolution imaging and spectroscopy feasible. Methods for reliable and reproducible sample preparation are critical to ensure that interface structures are well preserved.

Electrochemical samples and devices have to be frozen fast enough so that cryo-S/TEM measurements are performed on unaltered cryo-immobilized solid-liquid interfaces. Interfaces in operational electrochemical systems evolve over time. Understanding how the structure and chemistry of these interfaces changes during operation will require controlled freezing on the millisecond time scale to create snap shots of the evolution over minutes to hours. The freezing process itself is especially important for extended systems which may involve thick liquid layers such as battery coin cells or similar devices often used for electrochemical measurements. Even when properly frozen, such large samples have to be thinned under cryogenic conditions prior to high-resolution characterization by S/TEM. Recent advances in cryo-FIB based sample thinning are promising, however, new developments in instrumentation and workflow for cryogenic sample preparation of solid-liquid interfaces are urgently needed to improve success rate and throughput. This also includes the multiple steps currently needed to transfer the sample into the cryo-S/TEM. A platform that allows for storage and direct transfer of frozen thinned samples into the microscope without breaking vacuum would overcome problems due to ice contamination. A key limitation for atomic-scale spectroscopy and 4D STEM measurements at cryogenic temperature is the lack of appropriate cryo-S/TEMs. The stability of available cryogenic stages which also allow for 5-axis sample control is poor which limits atomic-scale measurements especially when the signal is low, e.g., spectroscopy (Goodge, Bianco et al. 2020). Improvements in cryo-stage stability of 10x or more, i.e., drift rates of 5-10 pm/sec, are needed to enable experiments for high-resolution characterize of these complex solid-liquid interfaces.

To observe the fluxional behavior that may occur at the solid-liquid interface it is necessary to make *in situ* or *operando* observations. Fundamental questions that must be addressed with *operando* observations include local characterization of the electric double layer, tracking ion transport within the electrolyte, structural changes in the interfacial layer with applied voltage and changes in pH. Electron beam damage is particularly severe for such measurements and many of the techniques described in the previous section on gas-solid interfaces can also be adapted for the liquid-solid interface. Low fluence rates are essential but generates signals which are extremely noisy. Denoising techniques that can reliably handle interfaces which are periodic and disordered must be developed to enable the extraction and rapid analysis of structural information. The ability to control the effect of radiation damage on the observation may be greatly enhanced using a pulsed electron source. If a short duration pulse is employed, it may be possible for the electronic structure to rapidly recover before the next pulse arrives. Very small diameter beams must be employed for TEM approaches so that if damage to the liquid occurs during atomic level observation, fresh liquid will continually flow in to restore the original liquid. An alternative strategy would be to follow the "diffract and destroy" approach where short intense pulses would allow structural information to be acquired quickly before the molecular species diffuse away. Electron sources should be developed that enable variation in pulse duration, frequency and

Tracking Interface Evolution during Electrodeposition



Liquid cell TEM enables quantification of transient growth front dynamics during electrodeposition of copper. The growth undergoes a transition at ~ 3s after which the roughness of the growth front increases more rapidly. (Schneider et al., 2017).

intensity to match the liquid damage rates and flow characteristics of the liquid cell. To provide flexibility the pulse rate should be highly variable to match system response.

In electrochemical devices there is a strong desire to develop techniques that allow atomic resolution analysis to be performed over extended length scales so that the response of anode/electrolyte/cathode interfaces can be studies simultaneously. The ability to image a 100 μm length battery stack, and yet get near-atomic resolution information and maintain a low enough dose from a cell "*in operando*" would be an aspirational goal. This requires the development of much larger detection systems and the ability to control and correct aberrations across such a large field of view. The enormous data sets that will be generated will also require the development of sophisticated automated analysis techniques which will include phase identification, interface characterization and classification of temporal fluctuations.

2.3 QE3: How might we observe energy, charge and mass transport pathways through materials and interfaces with atomic resolution?

Transport through solid-solid, solid-liquid and solid-gas interfaces are ubiquitous in energy systems and understanding the processes at the atomic level is required so that we can manipulate, control and optimize materials design parameters. Many of the *in situ* and *operando* techniques can now apply stimuli to materials systems which can drive transport process but typically phonon, electron, hole and ion transport is usually measure macroscopical. It would be highly desirable to be able to directly sense the transport process and pathways with atomic resolution using some form of imaging, diffraction or spectroscopy. The strategies, problems and opportunities to locally characterize transport will be different depending on the phenomena of interest. It is useful to discuss each transport process separately and consider opportunities for each to be measured.

Ion transport is central to electrochemical technologies and in many energy conversion devices an important situation is ionic transport through ceramics. In these applications, the ions or interest are typically derived from light atoms and included H, Li, Ca, Na and O species. The transport mechanism usually occurs via a vacancy hoping (i.e., oxygen in fluorite structures) or interstitially (i.e. Li in layered electrode materials). Device functionality relies on reversible charge transport process taking place in the anode, cathode and electrolyte control and these processes must be understood across multiple length scales. Many aspects of ion transport across the electrode/electrolyte interfaces remain unknown. Developing strategies to visualize ion transport will in part be influenced by the kinetics of the process i.e., slow or fast transport. The rate of transport can be regulated by temperature to take into account detector speeds and SNR requirements to reliable sense the perturbation caused by the dynamic change. The process of ionic transport is associated with the motion of point defects moving through the crystal lattice. An antecedent question for transport detection is can we visualize light element point defects in ceramics? The defect may lead to local changes in composition, bonding and lattice distortion or a combination of all three so point defect detection should be possible at least in principle using imaging, diffraction and spectroscopy tools. For example, an oxygen vacancy can lead to strain and distortions in the surrounding cation sublattice of over 10 pm which should certainly be detectable with imaging and diffraction techniques where the precision of detecting atomic column positions is less than 1 pm. Such distortions have recently been employed to map oxygen vacancy diffusion and it should be possible to extend such measurements to atomic resolution (Ding, Choi et al. 2020). For interstitial species such as Li or protons, the lattice distortion may be considerably less so other approaches may need to be found which may include spectroscopy or local measurement of the phase shift of the electron beam using ptychographic or holographic approaches. Isotopic tracer species may also be exploited using atomic resolution vibrational EELS mapping. A major challenge with all these approaches is that in 3D materials, the perturbation in a projected signal will be small. The binding energy of a defect that readily undergoes transport will also be small and so the local atomic environment around the defect may be significantly perturbed by the electron beam necessitating the use of low dose approaches.

Even if identification of individual point defects is challenging, mapping high ionic current pathways may would also lead to new advances for electrochemical devices. For example, existing biasing and heating holders should be able to drive ion currents though interfaces and grain boundaries so that high conductivity pathways can be identified. However, some sort of dynamic stimuli may be required to facilitate identification the subtle changes created in the structure when transport is taking place.

Electron transport through metals will be difficult due to rapid screening by conduction band electrons so there will be no lattice distortion or significant change in bonding. In electroceramics, electron transport of often associated with localized polaron hoping in which the electron jumps between cations in ionically bonded materials. The changes in cation states can be detected with EELS although the change in the spectroscopic signal associated with a single electron jump will be extremely small. Polaron hopping should also give rise to local lattice distortion due to local changes in ionic bonding making the detection problem similar to the ion transport problem. Detecting the transport of electrons and holes from dissociated excitons (e.g. electrons and holes in silicon) will require a different approach. The small change in occupancy in the delocalized valence and conduction bands may not be detectable with local probes. Electron transport may be associated with very small induced electric and magnetic fields. Whether the influence of the fields on the fast electron gives rise to a detectable phase shift is not known at present. Scientifically the only reason to perform such experiments in an electron microscope is to characterize transport in the vicinity of defects. Recent work from electron holography has shown that static space charge layers can be detected (Xu, Liu et al. 2020). Defects may serve as charge traps and charge accumulation which may be easier to detect. Dynamic *in situ* stimuli may be critical to detect the changes associated with electron accumulation and transport.

Phonon transport is also associated with lattice distortion but since they travel at the speed of sound high temporal resolution is required. Considerable progress has been made in using ultrafast microscopy to visualize the lattice distortions associated with phonon transport. Further developments and improvements of ultrafast approaches will be critical to understand transport in phonons. Vibrational EELS is now able to locally probe phonon modes with atomic resolution at interfaces and surfaces. The ability to gain information on the local phonon density of states will contribute to understanding phonon behavior near structural heterogeneities.

2.3.1 Electron Microscopy to Study Transport

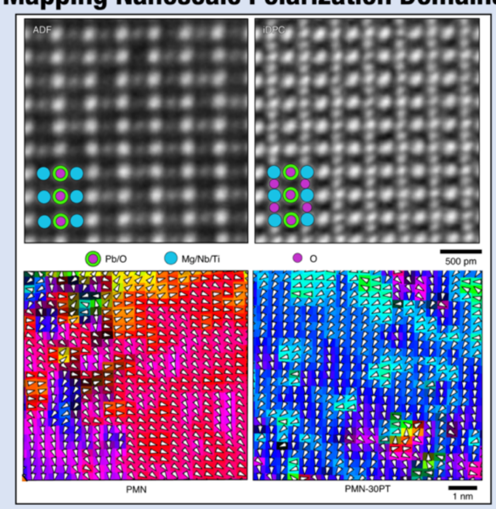
Many of the electron microscopy needs to investigate transport parallel those already discussed in the previous sections. For example, application of periodic electric fields coupled with synchronized detector readout would enable summing of frames recorded at equivalents temporal points in the periodic cycle revealing subtle changes and transients associated with transport. The key will be the ability to apply the thermal and/or bias rapidly without introducing any image drift or disruptions to the electron optics. The development of detectors where the exposure and readout is synchronized with applied stimuli may be critical to detect the subtle changes associated with transport. Data processing combined with simulation must be developed to enable recognition and track point defect motion associated with transport. Near on-the-fly matching of the difference signals may be essential to identify and differentiate the signal fingerprint defects associated with transport. For processes which cannot be locally triggered, identification of the spatial and temporal location of rare events is a major challenge that must be solved to observe the fundamental atomic level processes taking place. Even with detector speeds of 1000 fps and frame sizes of 5000 x 5000 pixels, successful identification of the location and time of, say, a single ion jump by casual inspection will be unlikely. Regardless of the signal of interest (image, diffraction or spectroscopic), it essential to develop near on-the-fly methods for detection of rare events.

3. Quantum Science Drivers, Critical Questions and Solutions

New technologies enabled by quantum materials and devices hold the promise of transformative impact across many sectors including health, energy, information, and national security. Quantum materials provide opportunities for realizing functionalities not available in conventional materials such as multiferroicity, colossal magnetoresistance, and high temperature superconductivity (Keimer and Moore 2017, Tokura, Kawasaki et al. 2017, Feliciano, Manuel et al. 2020). In this broad class of materials, quantum phenomena often emerge at atomic length scales due to strong interactions between charge, spins, and the lattice. These interactions also afford various tuning knobs such as light, strain, and external fields which may be used to further control and manipulate such properties for applications (Basov, Averitt et al. 2017). Understanding the nature of these interactions, the relevant length scales at which quantum phenomena arise, and the role of disorder requires advanced spatially resolved probes (Moler 2017).

Sensitive real-space probes are also critical for progress in quantum devices using more conventional materials. Solid-state qubits based on Josephson junctions, semiconductor quantum dots and defects centers in materials such as diamond, silicon carbide and silicon have rapidly developed over the past few years (Maurer, Kucsko et al. 2012, Christle, Falk et al. 2015, Miao, Blanton et al. 2020). Fabrication of such devices that harness the quantum effects necessary for new technologies present significant challenges given their high sensitivity to the defects, imperfections, and fluctuations. One of the key problems that must be addressed is how a quantum state can be maintained over sufficiently long time scales. Understanding sources of quantum decoherence in materials and solid-state devices is therefore critical.

Mapping Nanoscale Polarization Domains



Simultaneous ADF and iDPC STEM imaging allows for quantification of nanoscale heterogeneities and their connection to local polarization in a relaxor ferroelectric (Kumar et al., 2021).

Whether designing novel quantum materials or engineering quantum behavior in more conventional materials, the ability to directly probe effects at the atomic scale will inform fundamental scientific understanding and enable the realization of new functional materials and devices.

The last decade of electron microscopy has opened an era of atomic resolution imaging and spectroscopy of materials and devices. Recent developments in novel imaging and detector modes, electron spectroscopy, and *in situ* sample control have enabled functional properties of materials to be probed at high spatial resolution. They have allowed direct visualization and quantification of functional structural order parameters with picometer precision, directly mapping such phenomena as ferroelectric polarization, unconventional ferroelectric behavior in superlattices, and charge-ordered phases. Despite these promising advances in the field of electron microscopy, major limitations in instrumentation and methodology for collecting and extracting the relevant information from quantum materials and devices remain. Overcoming these limitations in materials characterization will underpin the development of new quantum devices and systems.

The workshop identified three grand challenges in the area of quantum science and technology which can be uniquely addressed with new midscale electron microscopy-based instrumentation. These challenges are:

QQ1: How might we understand mechanisms for quantum decoherence in materials and devices?
QQ2: How might we map electron correlations and the emergence of new phases in quantum materials?
QQ3: How might we measure magnetic moments of individual atoms?

Progress in each of these areas will have a transformative impact on quantum science and technology. The three grand challenges provide the priorities which should guide the development of electron microscopy infrastructure over the next 10 years. Below, we present what this workshop has identified as new opportunities in these areas as well as the advances in electron microscopy infrastructure that are needed to address the three questions.

3.1 QQ1: How might we understand mechanisms for quantum decoherence in materials and devices?

The key challenge in quantum technologies is understanding and maintaining quantum coherence in the presence of real operating environments. In different applications, however, this manifests with different levels of complexity. *Quantum sensing*, for example, may not require many interacting qubits, but the coherence of a qubit needs to be preserved within the target sensor. In solid-state sensors, the material has to maintain the coherence of the solid-state qubits inside. An important challenge is understanding how the material's atomic-scale environment contributes to decoherence and how materials and devices can be improved to maintain coherence. Furthermore, the effect of the target on the coherence of the probing qubit must also be considered: How does the sensor maintain its coherence in the presence of the real target, i.e., an environment which may not be fully controlled? How can quantum coherence be protected in a noisy environment? Similar questions are also relevant for known applications in *quantum computing*, *quantum simulation* and *quantum networking*, where maintaining coherence is critical. Compared to sensing, these situations are often even more delicate because many interacting qubits are required. The ability to measure sources of decoherence and ultimately control the coherence time in these materials and devices will be paramount for significant advances in quantum technologies.

3.1.1 Probing Sources of Decoherence in Solid-State Qubits

Solid-state qubits range from superconducting qubits to defect centers in materials such as diamond, silicon carbide, silicon etc. Decoherence of the quantum state can be induced by uncontrolled fluctuations in magnetic, strain or electric fields within the environment. These fluctuations can arise from a variety of different sources in a material. A crystalline defect can, for example, introduce a fluctuating magnetic field, or uncontrolled or inhomogeneous strain. Electron microscopy can play an important role in understanding how that atomic-scale environment can lead to fluctuations. Microscopic insights into the underlying mechanisms that limit current devices will enable the development of rational approaches for future improvements to quantum systems. This becomes particularly important when qubits are integrated into a larger system.

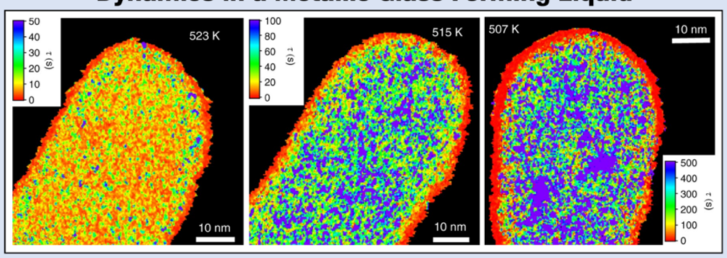
3.1.2 Controlled Positioning of Defects for Solid-State Qubits

In addition to probing the fluctuations that can cause decoherence, electron microscopy may also be used as a tool to precisely position and monitor individual defect centers in materials. Key questions that drive this field forwards are: How can single vacancy centers in diamond be produced? How can a single divacancy in silicon carbide be placed in exactly the right spot? Being able to controllably position such defect centers with nanometer precision would allow tunable coupling between multiple qubits or between a single qubit and a photonic or phononic cavity mode. Such coupling will be important for scaling quantum systems. Again, the problem is not only one of materials synthesis or device manufacture, but also inherently one of careful measurement and observation: An instrument which can create single defects in a material with nanometer precision will require not only a way to controllable introduce detects into a material but also a framework to monitor where and when the defect is formed.

3.1.3 Conditions for Relevant Measurements

Despite many efforts to probe individual qubits through a number of techniques, it is not yet fully understood which measurements are most relevant for understanding the origin for decoherence. While electron microscopy techniques are traditionally well-suited to correlated measurements, which may disentangle competing order parameters, one major limitation thus far has been accessing the necessary experimental conditions. The most advanced EM materials characterization requires very stable conditions, which have thus far precluded its extension to the kinds of environments, e.g., low temperatures, which are relevant for the specific quantum systems.

Dynamics in a Metallic Glass Forming Liquid



Direct visualization of spatially heterogeneous fluctuations of atomic arrangements in the supercooled liquid state of a Pt-based metallic glass enabled by electron correlation microscopy with sub-nanometer resolution (Zhang et al., 2018).

3.1.4 Electrically Gated Quantum Dot Qubits in Silicon

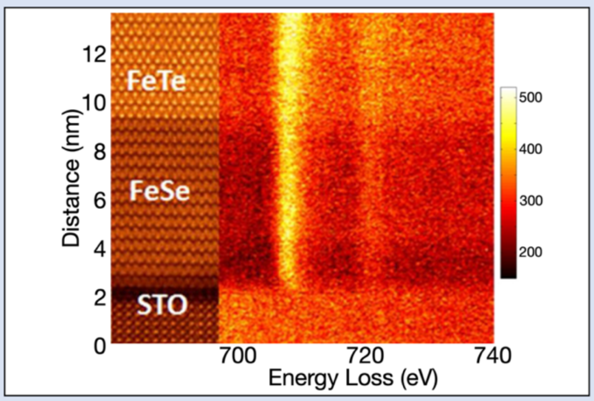
One solid-state alternative for qubits which is closest to the current technology for classical computers are electrically gated silicon quantum dot qubits. Although silicon-based quantum computers would operate fundamentally differently than classical microelectrons (even at cryogenic temperatures), a key motivation for silicon-based qubits is leveraging the extensive existing knowledge of semiconductor device fabrication. To go beyond classical computing will require large systems which integrate millions of qubits especially when taking into account error correction. The successful construction and operation of such systems requires the behavior of each individual qubit to be predictable and controllable. A major limitation to progress in this field is device yield. The failure rate is high and limited largely by materials issues. There is a critical need for an approach to extract atomistic information about the complex interface in devices. These techniques must also be able to account for the non-trivial topology of these interfaces. For silicon-germanium qubits, for example, the interface structure plays an important role in lifting the valley degeneracy in silicon and for determining the precise energy of this splitting, which is a key parameter for functional qubits. Key advances in silicon-based qubit technology can be made by focusing on the complex, buried interfaces in these devices at the relevant cryogenic temperatures.

3.1.5 Electron Microscopy to Advance Understanding of Quantum Decoherence

Electron microscopy has rapidly advanced over the last decade. The critical next step for developing quantum technologies will be leveraging these advances to identify impurities, defects and fluctuations associated with decoherence in materials and devices? Analyzing a billion atoms at a time at high spatial resolution over time scales of a few minutes will be a leap forward in the realization of functioning quantum systems.

Rapid identification of areas of interest may be accomplished using low magnification ptychography guided by ML or by new data driven methods enabling rare event detection by multimodal probes. Given the dose sensitivity of individual defects, targeted efforts should focus on developing low-dose approaches. Once coarse identification is completed, detailed autonomous or semi-autonomous experiments can be performed to probe the structure and functional properties of local defects and impurities. This may or may not require some depth resolution to avoid defects on the surface caused by sample preparation of electron transparent TEM samples. These experiments will require improvements by 10x or more in sample stability and positioning, especially at cryogenic temperatures relevant for quantum systems. As a benchmark target, instruments operating at temperatures of a few Kelvin or lower will enable quantum decoherence studies in a range of systems. In addition to the much-needed improvements in sample stability and positioning, rapid STEM measurements at ~1 ns/pixel of areas on the order of 100 nm in width will require fast detectors and scanning systems that overcome current limitations in distortions and flyback errors. Such rapid STEM will enable atomic-resolution STEM over large areas necessary to identify low concentrations of active defects in quantum devices while minimizing effects from mechanical instabilities. Coupled with recent advances in electron sources which allow high currents of ~1 nA to be focused into the ~1 Å probe, such rapid measurements will also improve time resolution of STEM. Acquisitions with ~1 ns/pixel will enable 1024x1024 pixel STEM imaging with 1,000 frames per second and measurements of fluctuations on time scales up to GHz using stationary beams.

Interface Electronic Structure Measured at 10 K



Cryo-STEM-EELS across an FeSe/$SrTiO_3$ interface shows a blue-shift of the Fe-L_3 edge within 2 unit cells of the interface when the sample is cooled to 10 K (Zhao et al., 2018).

Compared to devices for classical computing, solid-state qubits are more sensitive to non-uniform strain. New methods must therefore be developed to map strain smaller than 10^{-4} in such devices at temperatures below 4 K. Furthermore, fluctuations in strain fields and in magnetic fields can give rise to decoherence (discussed above). Direct measurement of such fluctuations due to the presence of impurities and defects are needed and may be enabled by 4D STEM using fast detectors with the high dynamic range necessary for such measurements. For micron scale devices, advances in sample preparation are also needed to make large areas accessible to high resolution electron microscopy.

Detecting coherent states by interference between multiple beams might be possible, but the small signal of such measurements will require very high dynamic range detectors and possibly a more coherent electron source.

3.2 QQ2: How might we map electron correlations and the emergence of new phases in quantum materials?

Electrons in quantum materials can interact strongly with each other or with the atomic lattice, giving rise to novel electronic states with properties not achievable in conventional materials. Correlations between electrons, for example, give rise to high-temperature superconductivity in materials with complex and highly anisotropic crystal structure. These unconventional superconducting states are often in competition with other correlated states of spin, charge and the atomic lattice. One example is charge ordering, in which electrons as well as the atomic lattice form periodic patterns that lift the symmetries of the crystal. Charge ordered phases are not only intertwined with superconductivity, but also underlie other exotic electronic phenomena such as metal-insulator transitions and colossal magnetoresistance. Mapping out different and often intertwined phases within the complex phase diagrams of quantum materials is important to understand the underlying physics of these systems and control them. Ultimately, this may even allow for predictive design of entirely new materials and functionalities.

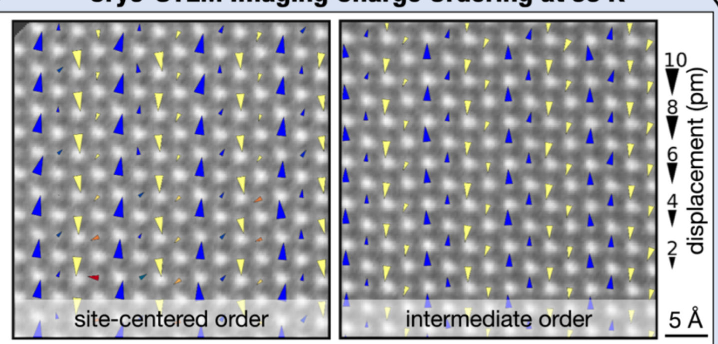
Being able to probe where electrons reside and how they couple to the atomic lattice will advance our fundamental understanding of quantum phases, their origins and their interactions with other electronic states. X-ray and neutron scattering are powerful techniques for investigating quantum materials, and structural refinements have provided important information about the crystal symmetries in these systems. Such techniques, however, do not provide the spatial resolution often needed to map local variations of lattice or electronic degrees of freedom within quantum materials or to isolate certain sample geometries. The recently discovered family of superconducting infinite layer nickelates, for example, has so far only been stabilized in epitaxial thin films which preclude the use of bulk probes. Local probes are also required for the direct mapping of spatial inhomogeneities, and the presence of competing and intertwined states. While scanning probe techniques have enabled exquisite measurements of structure and electronic states, they are predominantly limited to surfaces.

With the tremendous advances in hardware and software over the last decade, electron microscopy has become an indispensable tool for atomic-scale measurements of materials and their interfaces. Local symmetries can be measured using scanning diffraction experiments with sub-nanometer beams, picometer scale displacements of atomic columns can be quantified by direct atom tracking, and spectroscopy provides information about local electronic structure, charge distribution at interfaces, and collective modes such as phonons. Key limitations for mapping electron correlations and the emergence of new phases in quantum materials and the need for new electron microscopy infrastructure will be discussed next.

3.2.1 Electron Microscopy to Map Correlations in Quantum Phases

Despite the progress in electron microscopy, measurements of many quantum phases and correlations are still out of reach due to the lack of suitable instruments that operate at the relevant cryogenic temperatures where these states emerge (Minor, Denes et al. 2019). Ultra-stable cryogenic instruments must be developed to probe quantum materials. The required temperature range is set by the quantum materials of interest: while ambitious temperatures below 1K will be required to probe some quantum phenomena, even the development of ultra-stable instruments operating at more moderate cryogenic temperatures between tens of Kelvin to room

Cryo-STEM Imaging Charge Ordering at 93 K



Cryo-STEM resolves how atoms displace when a manganite crystal is cooled into its charge-ordered phase. Real-space mapping enables identification of two distinct ordered states which coexist within the same material (El Baggari et al., 2021).

temperature will enable measurements of a large range of quantum materials, including high-temperature superconductors, 2D magnets, topological insulators etc. Approaches in cryogenic cooling have so far focused on improving the design of side-entry cryogenic holder that also allow for 5-axis sample control and *in situ* temperature control. The stability of such traditional side-entry holders is, however, limited. New ultra-stable cryogenic electron microscopes need to be developed to enable high-resolution experiments, especially those that rely on weaker signals such as spectroscopic mapping by STEM-EELS or imaging of fields by 4D STEM. Such stable cryogenic instruments would open a new frontier for discovery and development of quantum materials.

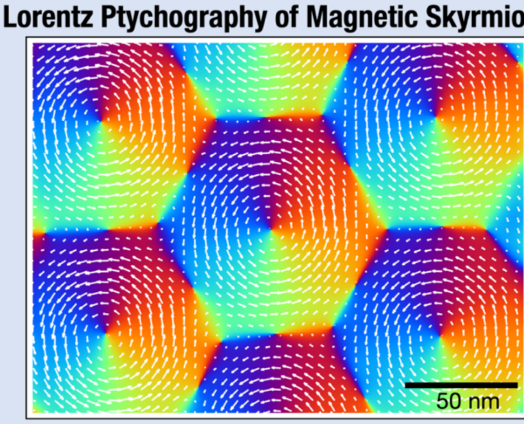
Hardware advances are also required to spatially probe currently inaccessible spectroscopic signatures of many exotic states. State-of-the-art STEM instruments have demonstrated energy resolutions of about 4-5 meV and can access excitations above ~25 meV, but the energy scales of many quantum states are on the order of ~1-2 meV. Achieving micro-eV electron spectroscopy will require improvements in electron sources, monochromators, spectrometers, and system electronics. Together with ultra-stable cryogenic operation, such instruments would provide ground-breaking experimental capabilities for local spectroscopic measurements of quantum materials such as mapping of superconducting gaps. Scanning nanobeam electron diffraction may provide an alternative approach to mapping subtle changes in the distribution and character of electrons, which can fundamentally alter the behavior of a material. The signal-to-background-ratio problem inherent to the fact that most of the electrons in the material are not involved in the states of interest has to be overcome, however.

Quantum phases can be created or tuned through appropriate drive systems such as light, magnetic and electric fields. Time-resolved measurements with high brightness sources, fast detectors with readouts that can be synchronized and labeled in time with application of external stimuli and dynamic control will enable new studies of emergent phases in correlated electron systems. In some materials, ultrashort light pulses can also induce metastable non-equilibrium states some of which may be sufficiently long-lived to be probed at high spatial resolution.

3.3 QQ3: How might we measure magnetic moments of individual atoms?

Being able to resolve the magnetic moments of individual atoms will enable direct mapping of spin textures in materials and devices, order and disorder in quantum magnets and spin liquids, and more broadly the emergence of new phases in quantum materials. Quantum magnets, for example, have spin structures that are dense in real space. While sensitive probes exist that measure very small magnetic moments, in a quantum magnet, the states of interest are often antiferromagnetic or frustrated so that no net external magnetic field is produced. The ability to measure individual spins within a solid would allow mapping of their arrangement

Lorentz Ptychography of Magnetic Skyrmions



Magnetic Skyrmions in FeGe at 94 K measured by Lorentz Ptychography using a 4D-STEM. The white arrows are the induction field vectors with lengths representing the magnetic field magnitude. The high signal-to-noise ratio makes it possible to see topological changes as the amplitude tends to zero (Z. Chen et al., 2019).

in such quantum magnets including the ordered ground states as well as inhomogeneities and defects that often play a critical role in quantum systems. Understanding fundamental interactions and correlations will further accelerate the development of quantum technologies.

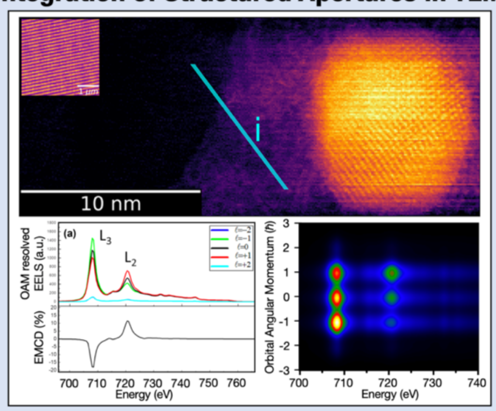
3.3.1 Electron Microscopy to Measure Magnetic Moments of Individual Atoms

Spin detection by holography or 4D-STEM depends on the detection of small phase changes or deflection of the electron beam. The detection sensitivity in turn depends on the electron dose used. The ability to measure small deflections therefore require larger dynamic range detectors as well as brighter electron sources. When considering a magnetic moment of a single atom, the deflection of an electron beam due to the Lorentz force should be measurable in a few minutes using 1 nA of beam current focused into a 1Å probe. This, however, also requires a high dynamic range detector which can measure a few billion electrons.

An alternative approach is to couple spectroscopy with structured electron beams. The magnetic moments could be measured by EELS/EMCD, where two atomic-size probes with opposite orbital angular momentum are used to extract the magnetic signal. This also involves detecting very small signals, and is particularly sensitive to multiple scattering in the sample that changes the beam profile, but may be very valuable in 2D materials where these effects are smaller.

Along with these hardware improvements, there is an additional need to develop new, efficient algorithms capable of extracting the relevant information from complex, multidimensional dataset. For all of the methods discussed above, stage stability improvements will be key, especially at low temperature where mechanical drift rates are an order of magnitude higher than at room temperature. Weak signals require long acquisition times to build up a sufficient signal-to-noise ratio suitable for extracting the relevant information. Stages need to be stable over the length of the experiment (drift rates less than 10 pm per image, or 10 pm/minute). Currently even the best room temperature stages show a stability of 500 pm/min.

Integration of Structured Apertures in TEM



Structured apertures enable (top) phase measurements by interferometry to simultaneously measure high- and low-scattering materials (Yasin et al., 2018) and (bottom) magnetic measurement when combined with EELS (Rotunno et al., 2019).

Ultra-stable low temperature instruments are needed to access many of the important magnetic phases, including 2D quantum magnets such as CrI_3, and $Cr_2Ge_2Te_6$ which may prove a promising platform for electrically switchable spintronic devices, spin currents and spin valves. Coupling such cryogenic operation with *in situ* capabilities including biasing, heating, photoexcitation will allow for tuning of such quantum states. New lens designs are also required to allow the magnetic field at the sample to be tuned without sacrificing the imaging resolution. This is critical for magnetic structures that can be altered or even destroyed in the high magnetic fields of the objective lens typically used in aberration corrected instruments. Recent advances in Japan have pushed towards lattice resolution (non-magnetic) imaging in variable magnetic fields, a first step in this direction. Significant continuing developments are needed to improve resolution and enable single magnetic moment detection.

4. Infrastructure Needs and Solutions

The primary goal of the workshop was to define a future road map for electron microscopy and to identify infrastructure needs to accelerate scientific discovery and progress in the fields of energy and quantum materials. As such the workshop participants focused on six key areas essential to the future success of the field in the context of the science drivers discussed above.

(1) Hardware development
(2) Integration of data science approaches
(3) Integration of real-time simulations and computation
(4) Development and distribution of open-source software
(5) Training of current and future electron microscopy users
(6) Community engagement

Major opportunities in taking a community focused approach to address these infrastructure needs were identified and are presented next. Specific needs for energy and quantum materials are described and motivated in detail in chapters 2 and 3. There was significant overlap in the infrastructure needs for both areas. Moreover, many of the hardware and software components may be developed in modular form for implementation in a wide range of instruments across the US. In the following six infrastructure solutions (IS) sections, we provide succinct lists of critical developments identified as necessary by the workshop participants.

For easier navigation and rapid connection of proposed infrastructure with grand challenge problems, the technical IS sections (hardware, software, theory) indicate how critical developments impacts the previous energy and quantum sections through back references to previous section numbers and a few keyword identifiers. Many of the infrastructure solutions have potential impact beyond energy and quantum materials, touching many areas of science and technology where electron microscopy makes a significant contribution.

4.1 IS1: Infrastructure for hardware development - How do we as a community enable, accomplish, and accelerate hardware development?

Development of hardware for electron microscopy is broadly recognized by the community as critical for advancing our ability to probe materials and devices. In addition to the specific hardware needs identified in the previous two sections, there are also new opportunities at the intersection of hardware, software and data analytics. To retain US leadership in electron microscopy instrumentation and innovation, we must invest in infrastructure that enables and accelerates hardware development for electron microscopy in the US.

Goal: Development of unique electron microscopy instrumentation to address science drivers.

Current Approach and Limitations: The key limitation in enabling new electron microscopy capabilities for energy and quantum science and technology is that the US trails Europe and Japan in technical expertise for the development of electron microscopy instrumentation. Much of the knowledge and expertise in hardware development currently resides in industry. Industry, however, is limited in freedom to explore, develop and share new and potentially transformative ideas. At universities and academic institutions, on the other hand, higher-risk research is possible but long-term engineering expertise is often lacking. As hardware development has moved to industry, the broader EM community has lost the expertise required to build innovative new instruments. Significant investment is required to train the next generation of instrument builders in the US.

Solution: National-scale facilities or platforms with mechanisms to retain long-term engineering expertise are needed to design and build prototype machines, each with different unique capabilities to enable new scientific and engineering breakthroughs. These prototype machines may or may not be developed from existing commercial instruments. Such national-scale facilities should support extended visits by researchers from academic institutions in the US including faculty, postdoctoral and graduate student researchers. These visits, including sabbaticals, may also provide a path to implement parallel instrumentation developments in their campus labs, for which funding should also be made available through such a national infrastructure. This approach provides an avenue to take full advantage of the higher-risk research possible at universities and leverages the expertise of the broader microscopy community in order to make new capabilities available to all facility users. The proposed infrastructure model makes full use of the professional engineering and continuity of technical support available at national laboratories to design and implement new electron microscopy capabilities. This program differs, however, from current national lab electron microscopy facilities in that a wide variety of specialized expertise at US universities is included to develop new instrument capabilities. To nucleate such national-scale implementation efforts, significant investment is required in building up instrumentation development expertise by focusing on design and construction of components critical for addressing the scientific questions identified in the previous sections. Collaborations and coordination between individual groups based on broad scientific goals will allow for efficient parallel developments towards new measurement capabilities.

Immediate Needs*:* There is significant overlap in the key required hardware needs for problems in both energy and quantum materials. Many components may be developed in modular form for implementation across a wide range of instruments. Important hardware needs include:

- Ultra-stable cryogenic stages: 10x improvements or more in sample stability at a broad range of cryogenic temperatures relevant for quantum and/or energy systems.
  Impact: High-spatial resolution structural and chemical mapping of processes at solid-liquid interfaces in electrochemical systems (Section 2.2). Access to electronic and magnetic phases in quantum materials and devices including solid-state qubit systems, superconductors, topological insulators and quantum magnets (Sections 3.1-3.3).
- Electron detectors:
  - Increased readout rate of 10^7 Hz for low dynamic range STEM detectors while maintaining near-ideal DQE and MTF for *in situ* 4D STEM.
    Impact: Direct imaging of catalytic reaction pathways by tracking structural dynamics on catalyst nanoparticle surfaces with site specificity (Section 2.1.5a).
  - 10-100x improvements in readout rates of high dynamic range STEM detectors for functional imaging by 4D STEM.
    Impact: Detection of magnetic moments of individual atoms for direct mapping of spin textures in materials and devices including quantum magnets and spin liquids (Section 3.3) and measurements of emergent field properties in engineered quantum materials (Section 3.2).
  - Fast electron detector systems with readouts that can be synchronized and labeled in time with application of cyclic stimuli (heat, electric field, light etc.).
    Impact: Tracking the dynamic evolution of cyclic processes such as catalytic surface reactions at active sites (Section 2.1.5a) or site-specific facile ion transport across interfaces (Section 2.3.1).

- Adapting and expanding current electron sources to enable variation in pulse duration, frequency and intensity with synchronization with cyclic stimuli (heat, electric field, light etc.).

  Impact: Dose efficient tracking of reaction pathways in photocatalytic, thermochemical and electrochemical processes (Sections 2.1.3 and 2.1.5a), and imaging of ion transport across solid-liquid interfaces (Section 2.3.1).
- Scanning systems for rapid STEM measurements at 100-1,000 fps on areas on the order of 100 nm in width with no distortions and flyback errors.
  Impact: Atomic-resolution imaging over large fields of view to capture heterogeneity in complex interface systems (Section 2.2) and to identify low concentrations of active defects in quantum devices (Section 3.1.5).
- Hardware (monochromators, spectrometers, electronics, temperature control) for micro-eV spectroscopy.
  Impact: Map spectroscopic signatures of exotic quantum states such as variations in the energy gap in superconducting or topological systems (Section 3.2).
- +

Impacts: Several key opportunities for scientific and technological advances in energy and quantum systems with new hardware components are highlighted above. Furthermore, a national-scale electron microscopy infrastructure will have long-term impacts through (i) the development, design and implementation of instruments with unique capabilities that enable experiments in quantum and energy science not currently possible through commercial solutions and (ii) strengthening the instrumentation development expertise and knowledge among students, faculty and postdocs in the US for sustained advances in electron microscopy.

4.2 IS2: Data Science - How do we as a community enable, accomplish, and accelerate the integration of data science (and data storage) into next generation TEM?

The community recognizes that a computational imaging approach with full integration of data science into instrument design, data acquisition and on-the-fly processing is essential to maximize the impact of next generation microscopy on energy and quantum materials.

Goal: Development of infrastructure solutions at the intersection of data science and hardware that enables efficient extraction of relevant materials information.

Current Approach and Limitations: Data-based tools (AI, ML, simulations) have the potential to accelerate acquisition, extraction and interpretation of relevant information from electron microscopy signals. Many such tools are, however, not integrated into currently available electron microscopy platforms and are only available for post processing of data. Codes are often developed in 'bubbles' and not easily available for wide use. Furthermore, ML methods are often sensitive to the out-of-distribution artefacts, meaning that slight changes of imaging parameters may lead to catastrophic failure of a ML model. Existing instrument platforms provide limited accessibility for users to install open-source code for control, acquisition and analysis. Manufacturers and researchers are concerned about intellectual property and mechanisms for recognition of credit for contributions. The development of data tools by the broader community also relies on availability of appropriate data. There is, however, a community concern regarding lack of data validation, inadequate metadata etc. In general, data and data-based tools in microscopy fail to meet the FAIR standards for open science (Wilkinson, Dumontier et al. 2016).

Solutions: A unified cyber infrastructure designed for materials' electron microscopy must be created to accelerate integration of data science approaches. The impact of individual group efforts could be dramatically expanded by building a standard open-source framework and infrastructure for sharing and reuse of data and models. To enable optimized workflows as well as ML-guided autonomous experiments for high-throughput, high-resolution characterization of materials, integration of hardware and software has to occur at the design stage.

Widely accepted benchmark datasets for electron microscopy relevant problems need to be developed to enable testing and training of new data approaches. This will help build acceptance of new models and tools, since they will have reliable benchmarking. It is necessary to create systems for image classification / segmentation / identification from simulated electron scattering signals (images, diffraction patterns, spectra, etc.). These systems may benefit from new ML models / methods that build on known physics of electron microscopy problems (e.g., crystal symmetries, reciprocal vs. real space, etc.). Physics-informed ML models may also enable fast, approximate data inversion and analysis with rapid feedback during the experimental sessions, e.g., fast approximate ptychographic reconstruction, tomographic reconstruction, etc. These approximate inversion models can be used together with iterative forward simulations to develop and refine 3D structures from experimental data.

Immediate Needs*:* Many of the data needs were common to problems in both energy and quantum materials and can also be developed in modular form to directly impact many TEM labs around the country.

- Microscope control systems to optimize data acquisition for high-throughput and hypothesis testing.
- Edge computing hardware and software to co-locate sufficient computational power with microscopes to enable rapid, in some cases real-time, analysis and feedback to the operator.
- Regardless of the signal of interest (image, diffraction or spectroscopic), it is essential to develop on-the-fly methods for detection of rare events in space and time.
- Develop data science methods to recognize the spatio-temporal fingerprints of metastable states that may repeat in noisy data streams.
- Large dataset size will require the development of sophisticated automated analysis techniques to test specific hypotheses including phase identification, interface characterization and classification of spatial and temporal fluctuations.
- Systems for creating image classification / segmentation / identification from simulated electron scattering signals (images, diffraction patterns, spectra, etc.).
- On-the-fly compression techniques will become increasingly important as datasets increase in size.
- On-the-fly denoising techniques should be developed so that approximate structural information can be more easily visualized.

Impacts: Integration of data-based tools into instrument design, data acquisition and on-the-fly processing will maximize the impact of electron microscopy across multiple fields by accelerating acquisition, extraction and interpretation of relevant information from electron microscopy experiments. For example, optimized data acquisition through microscopy control and on-the-fly detection of rare events will enable identification and characterization of active sites in catalytic and electrochemical processes at complex interface (Sections 2.1.5a, 2.3.1) as well as active defects in quantum devices (Section 3.1.5). New *in situ* and *operando* approaches for energy systems will benefit from on-the-fly compression and denoising techniques (Section 2.1.5b), and rapid image classification and segmentation will provide users the feedback needed to identify key areas of interests for detailed analysis. More generally data-tools for electron microscopy, especially when integrated at the instrument design stage, will increase the speed at which materials and devices can be characterized and will make reliable atomic-resolution analysis of materials broadly accessible.

4.3 IS3: Theory - How do we as a community enable, accomplish, and accelerate real-time integration of simulations and computations into next-generation TEM?

Developing a more efficient hypothesis driven approach to experiments requires implementation of on-the-fly hypothesis tests which in turn requires on-the-fly simulations of TEM image, diffraction and spectroscopy signals. Creating an automated loop of experiment→analysis→model refinement would not only dramatically decrease the time to solve critical materials problems but is also essential for high-throughput, potentially autonomous analysis. Such an approach would also dramatically enhance the accessibility of advanced electron microscopy to non-specialists.

Goal: Enable, accomplish and accelerate real-time integration of simulations and computations into next-generation TEM.

Current Approach and Limitations: Currently, there is almost no real-time simulation performed at the microscope during the experiment. Signal simulation is entirely performed either prior to the experiment or as a post processing exercise. This is true even for approaches that are currently fairly reliable such as multislice simulations of images and diffraction patterns. However, many of the currently available methods (e.g., multislice approaches) do not provide an easy way to incorporate mesoscopic data from phase-field and finite-element models, for example. There is a need to develop new methods and new algorithms to deal with the complexity of mesoscopic microstructures. The computational power on many current microscopes is very limited and with relatively modest investment (in comparison to the cost of the EM platform) it could easily be increased by one or more orders of magnitude. There is no currently available workflow platforms that can perform quantitative comparison between theory and experiment, test hypotheses from the operator and suggest changes to experimental conditions and procedures to increase the rigor of the theory-experiment comparison. Most S/TEMs currently cannot implement any kind of live "decision processes".

Solutions: Develop integrated software which enables a theory-simulation-experiment loop that allows for physics-guided data reduction. This will enable hypothesis validation/modification and experimental control adjustments. The ability to immediately exclude/reject hypotheses "instantly" from observations is a much easier task than solving for a ground truth. Workflows for various types of analysis with system control to rapidly evaluate hypotheses need to be established. Simulation and theory can also generate microscope parameters on-the-fly which may allow for real-time correction critical for accurate execution of workflow procedures.

Immediate Needs: Significant design modification to existing TEM columns must include direct integration of high-performance computational capability along with more sophisticated system control to enable high-level workflow and decision-making capability.

- Development of control to enable complex data acquisition sequences based on high level workflows.
- Integration of microscope edge computing with large-scale high-performance computing resources at the institutional and national facility scale.
- Establish and curate standards, protocols for data/method validation and for tests of new analytical methods.
- Establish large databases of simulated images, diffraction patterns and spectra which can be matched with experiment for quick screening.
- Integration of TEM signal simulation software with suitable cost functions and feedback loops to enable automated quantitative interpretation of image, diffraction and spectral signals during an experiment.
- Integrate multiscale simulation tools (DFT, phase field, finite element, etc.) with large scale electron diffraction and image simulations, to enable immediate local determination of materials properties going beyond atomic coordinates.

Impacts: Integration of simulations and computations into next-generation TEM will enable on-the-fly hypothesis testing and decision making for high-throughput, potentially autonomous experiments. Such an approach would decrease the time to solve critical materials problems and would also make advanced electron microscopy more accessible to non-specialists.

4.4 IS4: Software - How do we as a community enable, accomplish, and accelerate the development and distribution of open-source software for electron microscopy?

The role of software tools for data analysis in electron microscopy has dramatically increased over the past several years. Advances in instrumentation have led to large data streams requiring sophisticated analysis tools to make most of the available data. Making these tools user friendly and accessible will empower the broader scientific community to make use of advanced electron microscopy techniques.

Goal: Create a cyberinfrastructure for electron microscopy with incentives for ongoing community contribution and development of advanced software tools for data analysis.

Current Approach and Limitations*:* There are enormous variations in workflows/analysis approaches across different groups and it is difficult to converge on one solution or set of solutions which can be supported by a large number of developers and widely adopted by the field. This problem is exacerbated by proprietary formats and limited access to metadata. There are not strong incentives for the community to collectively engage since it is hard to publish or get "high impact" credit for developing important, user-friendly software. It is also difficult to get reviewers and funding agencies to see software as a critical part of infrastructure.

Solutions: We must build strong community support for open software development. This must be supported by funding agencies, vendors, scientific journals, and adoption of science enabling software tools. Following the NSF HDR strategy, we need to generate a cyberinfrastructure for open-source software development for electron microscopy. The needs listed below impact all areas.

Immediate Needs*:*

- Establish opportunities for communication and collaboration between open-source EM software developers to facilitate coordination and avoid duplication.

- Establish and curate standards, protocols, and validation to certify the quality of open-source software.
- Establish a centralized location for certified open-source software.
- Establish a community wide approach for long-term stability and updates of open-source software including funding for maintenance costs.
- Incorporate open-source software into TEM platforms to support on-the-fly data analysis.

Impact*:* Building infrastructure for sustained development and maintenance of data analysis tools that are reliable, reproducible and accessible will empower all electron microscopy users, experts and novices, to make most of the rich data streams generated with modern electron microscopes.

4.5 IS5: Training - How do we as a community educate current and future TEM users?

Effective education and training of current and future TEM users in the capabilities of and new opportunities with modern electron microscopes is key to expanding the impact of electron microscopy.

Goal: Develop a new model for graduate education that trains students in diverse fields about capabilities of modern electron microscopy.

Current Approach and Limitations*:* A coordinated, community supported model for graduate training in electron microscopy is currently lacking in the US. Courses on electron microscopy techniques are taught at individual academic programs especially at institutions with strong electron microscopy expertise. More broadly accessible are summer schools and workshops at select programs including at ASU, Lehigh and PARADIM, however, each is limited in the number of participants they can support. In addition, rapid developments of new techniques and areas that play an increasing role in electron microscopy such as data science present further complexities in education and training. Furthermore, there currently does not exist a centralized training platform for complex experiments.

Solutions: Building a virtual infrastructure for practical electron microscopy training and workforce development will democratize electron microscopy education. In addition to learning opportunities and instruction on the fundamentals of electron microscopy (hardware, optics, techniques), such infrastructure should include integrated, seamless remote operation of modern instruments for hands-on training as well as practical training in data analysis which is increasingly becoming the bottleneck in the field. Leveraging the knowledge of experts in the field to educate about novel developments will make this infrastructure effective and avoid duplicated efforts at individual institutions. It will also allow students to learn what can be accomplished with a technique or instrument available nationally and not only at their home institutions. Focus should be on new capabilities. A modular structure may allow timely integration of high-quality content covering new topics. Materials developed for this education infrastructure should be revised frequently to ensure that they are up to date. To increase opportunities for practical training in the operation of electron microscopes, construction of full microscope simulators may also be beneficial. Training a future workforce on advanced microscopy techniques impacts all areas.

Immediate Needs*:*

- Develop online teaching materials.
- Create and fund online classes.
- Pursue cross-credit for different institutions for online EM classes.

- Expand opportunities for in-person summer schools with hands-on components for practical training in instrument operation, maybe facilitated by remote access.
- Broaden training opportunities in data analysis tools through interactive, virtual short courses or summer schools.

Impact: Infrastructure that leverages the knowledge of experts in the field to educate and train current and future electron microscopy users will expand the impact of electron microscopy and drive scientific and technological advances with these tools.

4.6 IS6: Community Engagement - How do we communicate to the science and engineering communities, the public, and policy makers the value of TEM for their research needs?

It is very important to communicate the transformative impact of advanced electron microscopy to different stakeholders. The science and engineering community must be made aware of cutting-edge capabilities to enable them to take advantage of the resources to advance their research objectives. The general public and policymakers to ensure that they understand how investments in microscopy help to drive progress in a wide range of areas including energy, communication, human health and the environment.

Goal: Strengthen and enhance community engagement to highlight the value and impact of electron microscopy for advances in science and technology.

Current Approach and Limitations: Universities and national labs have extensive outreach programs to reach the science and engineering communities. At national user facilities there are benchmarks to measure the effectiveness of these programs to drive constant improvement. Many universities and national labs also have industrial affiliate programs to communicate the power of microscopy to industry. On the education side there are numerous outreach activities for schools run by universities and professional societies but there is very little coordination, integration or sharing of resources. Collectively, the electron microscopy community is not doing much at the national level to reach the general public and policymakers. The Microscopy Society of America and the Microanalysis Society do not engage in policy outreach perhaps due to budget constraints. Furthermore, there seems to be a dearth of participation by the academic microscopy community in lobbying and policy making. There is no nationally coordinated communication plan to share the excitement and impact of microscopy with the general public.

Solutions: We need to establish stronger connections to the general public, possibly with coordination through the microscopy professional societies (to address policy makers). (It is not necessary for societies to hire lobbyists, but rather to have resources to train the community to "lobby" or serve as strong advocates for microscopy. Also linking and coordinating these activities with larger societies like MRS, APS and ACS (through possible partnerships) would enable efficient leveraging of existing infrastructure. We need to develop coordinated programs to communicate to the public the impact electron microscopy has had on their lives. This should go beyond simply capturing their imagination, lasting impact will come from demonstrating the practical importance and relevance to daily lives. First and foremost, we must educate people on what an electron microscope does and why it is important. The many pictures of the COVID-19 virus have been seen by most of the general public, for example, but very few will know about the critical role electron microscopy plays in generating such images. This ties into the need for microscopy to be seen as relevant to society. We also need to clearly communicate how future technology development will absolutely need advances in electron microscopy technology. This message must constantly be communicated to public policy makers to strengthen the link between investment in electron microscopy and economic development.

Immediate Needs*:* It is necessary to build a network of community members that have a strong interest in leading lobbying and outreach to the general public. Some immediate activities suggested by workshop participants include:

- The national societies should establish focused interest groups or subcommittees that will constantly drive national coordination and implementation of these activities related to lobbying and public outreach.
- Create training programs or workshops focused on developing effective communication skills for electron microscopists at all levels.
- Develop communication plans which provide a roadmap for the community to engage in effective lobbying and education.
- Develop collections of stories that clearly illustrate the links between fundamental electron microscopy discoveries and products that are crucial to quality of life.

Impact: Effective communication of the value and impact of electron microscopy to the broader scientific community and the public will drive progress in science and engineering. In the short term, it will raise awareness of the unique capabilities enabled by advances in electron microscopy and how these capabilities can accelerate scientific discovery in a diverse range of fields. In the longer term, it will serve a critical role in educating the community of the importance of sustained development and innovation in electron microscopy.

Workshop Agenda

Friday, September 4th, 2020

1030 - 1700 EST

Workshop Day One (Overview)

Identify, explore and formulate key scientific drivers in the fields of Energy and Quantum science and technology.

1030 - 1200 EST

Session One: Discuss and articulate interesting questions in Energy and Quantum science.

Session Two: Continue to discuss and articulate interesting questions in Energy and Quantum science in new groups.

Breakouts

1200 - 1230 EST

Break - Watercooler

1230 - 1400 EST

Session Three: Final opportunity to discuss and articulate interesting questions in Energy and Quantum science in new groups.

Session Four: Explore interesting directions in <u>Data</u> science that could enhance electron microscopy for energy and quantum materials in new groups.

Breakouts

1400 - 1500 EST

Break - Watercooler

1500 - 1630 EST

Session Five: Converge on the most interesting questions in energy and quantum (looked at through the lens of measurement and data)

Breakouts

Sunday, September 6th, 2020

1030 - 1700 EST

Workshop Day Two

Develop potential approaches to the questions explored on day one with a view to creating a roadmap for electron microscopy.

1030 - 1200 EST

Session One: What are the antecedent questions - the new experiments/measurements must be performed to address the scientific drivers?

Breakouts

1200 - 1230 EST

Break - Watercooler

1230 - 1400 EST

Session Two: Thinking about the EM Community - What is the ideal infrastructure landscape in the next 10 to 15 years? Writing session to capture ideas.

Breakouts

1400 - 1500 EST

Break - Watercooler

1500 - 1630 EST

Session Three: Present your outline infrastructure proposals and get peer feedback.

Workshop Participants

Co-Chairs:
Peter Crozier, *Arizona State University*
Lena Kourkoutis, *Cornell University*

Organizing Committee:
Joshua Agar, *Lehigh University*
Peter Ercius, *Lawrence Berkeley National Laboratory*
Stephen House, *University of Pittsburgh*
Sergei Kalinin, *Oak Ridge National Laboratory*
David McComb, *Ohio State University*
Andrew Minor, *University of California, Berkeley and Lawrence Berkeley National Laboratory*
Seda Ogrenci-Memik, *Northwestern University*
Frances Ross, *Massachusetts Institute of Technology*
Mary Scott, *University of California, Berkeley and Lawrence Berkeley National Laboratory*
Toby Scott, *Knowinnovation*
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Participants:
Nasim Alem, *Penn State University*
Harry Atwater, *California Institute of Technology*
Robert Cava, *Princeton University*
Maria Chan, *Argonne National Laboratory*
Gang Chen, *Massachusetts Institute of Technology*
Susan Coppersmith, *UNSW Sydney*
Yi Cui, *Stanford University*
Abhaya Datye, *University of New Mexico*
Peter Denes, *Lawrence Berkeley National Laboratory*
Vinayak Dravid, *Northwestern University*
Jennifer Dionne, *Stanford University*
David Flannigan, *University of Minnesota*
Andrew Herzing, *NIST*
Robert Hovden, *University of Michigan*
Juan Carlos Idrobo, *Oak Ridge National Laboratory*
James LeBeau, *Massachusetts Institute of Technology*
Suljo Linic, *University of Michigan*
Ania Jayich, *University of California, Santa Barbara*
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