Center for Nanophase Materials Sciences Strategic Plan 2019–2023

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Executive Summary

The Center for Nanophase Materials Sciences (CNMS) is a Nanoscale Science Research Center (NSRC) established as part of the Department of Energy (DOE) Office of Science contribution to the US government's National Nanotechnology Initiative. Located at Oak Ridge National Laboratory (ORNL), CNMS provides a diverse user community with access to forefront nanoscience research capabilities, expertise, and equipment, and these resources are offered free of charge to users who intend to publish the results in the open literature. The center emphasizes a highly collaborative environment where users often take advantage of multiple resources for studies that include making, characterizing, and understanding nanomaterials. CNMS users include experienced research professionals as well as postdocs and students, coming from academic, industrial, or government-funded institutions from across the U.S. and abroad.

The resources at CNMS are also applied to execute a cutting-edge in-house science program focusing on *understanding the effects of dimensional and spatial confinement on formation and function, in order to enable the design of responsive nanomaterials that efficiently capture, transport, and/or convert energy*. This research effort not only contributes to our understanding of nanoscale mechanisms, but also leads to the development of advanced capabilities in synthesis, characterization, imaging, and theory/modeling/simulation that will benefit future users. This vigorous, DOE reviewed in-house research effort is organized into four research themes that catalyze new nanoscience capabilities and knowledge for energy generation, storage and use, and create the integrated environment needed for CNMS to be a forefront user facility where staff expertise enables user science.

CNMS is unique among user facilities in providing specific approaches and methods not accessible to the user community elsewhere: a broad spectrum of imaging capabilities that include scanning transmission electron microscopy emphasizing electron energy loss spectroscopy, scanning probe microscopies, He-ion microscopy, atom probe tomography, and mass spectrometry based chemical imaging; nanofabrication efforts that have created opportunities for 3D direct-write nanofabrication and positioning of individual dopants; selective deuteration and precise synthesis of macromolecular materials; all in combination with a comprehensive suite of functional characterization and a fully integrated effort of theory, modeling, and simulation that explicitly underpins much of the work.

This Draft Version of the Strategic Plan (subject to discussions at the on-site meeting with the CNMS Scientific Advisory Committee, May 2018) outlines the scientific agenda that will drive the scientific progress of CNMS in the coming five years. It reflects our plans to maintain a vibrant environment for users from the nanoscience community in the broadest sense, implementing modern materials science methods based on a close integration of high performance computing, data analytics, and artificial intelligence. The center emphasizes energy related topics such as batteries, photovoltaics, catalysis, etc., and delivers new advances in soft matter, 2D materials, and multiscale integration of nanomaterials. In addition, CNMS specifically seeks to address additional emerging opportunities in quantum information science, neuromorphic computing, and those at the interface between biology and materials research.

CNMS benefits from an intrinsically strong interaction with ORNL signature strengths in multiple areas and takes advantage of the distinctive capabilities of other DOE user facilities at ORNL, including the Oak Ridge Leadership Computing Facility, the Spallation Neutron Source, and the High Flux Isotope Reactor. CNMS thus acts as a gateway for *neutron sciences*, providing an environment for researchers to integrate neutron studies into nanoscience efforts, and for *computational nanoscience*, enabling the community to take advantage of ORNL's powerful computational resources.

With the strong connection to core ORNL strengths and a clear focus on addressing the needs of future users, CNMS will continue to thrive as a critical center for advancing nanoscience throughout the scientific community in partnership with users from national laboratories, universities, and industry.

1. Introduction: Nanoscience and CNMS

Nanoscience, conventionally defined as the study of structures, materials, and phenomena on the scale of nanometers (~1-100 nm), has led to groundbreaking technological advances and applications in areas spanning from batteries to photovoltaics, from catalysis to drug delivery, from stronger composites to hydrophobic surfaces, and many more. Yet, despite the unquestionable success and societal benefits of nanotechnology, there remain significant scientific and technological challenges to fully understand the nature and consequence of nanoscale confinement and to precisely create the nanomaterials and nanostructures that are needed for future applications. Fortunately, significant advances have been made in methods to manipulate matter at the scale of individual atoms, to image nanostructures with atomic resolution, and to use modeling and simulation of increasingly larger and more complex nanoscale assemblies. In combination, these advances are poised to lead us to the point where we can create – with atomic precision – the desired structures for which advantageous properties are being predicted through theory, modeling, and simulation (TM&S), and for which functional imaging reveals the behavior of each atom. Overcoming the challenges in integrating prediction, formation, and characterization will lead to applications of nanotechnology in new areas, enabling new ways of harvesting and converting energy, and spanning from Quantum Information Science (QIS) to research at the interface between biology and materials science.

Research in the area of nanoscience is fundamentally multi-disciplinary, not only because chemical and quantum mechanical behaviors become intrinsically intertwined even in the simplest cases of nanomaterials, but also because the application of nanoscience affects such a broad spectrum of disciplines: for example, experts in quantum computing and specialists in medicine may both need to address fundamental nanoscience challenges that are outside of their own expertise. It is in this context that the Center for Nanophase Materials Sciences (CNMS) operates as a user facility that allocates resources to users solely based on the scientific merit of the proposed research, as determined by peer review.

CNMS is one of five Nanoscale Science Research Center (NSRCs)[1] established as part of the Department of Energy (DOE) Office of Science contribution to the US government's National Nanotechnology Initiative and thus contributes to the goal of working toward "a future in which the ability to understand and control matter at the nanoscale leads to a revolution in technology and industry that benefits society." Co-located at Oak Ridge National Laboratory (ORNL) with the Spallation Neutron Source (SNS), the High Flux Isotope Reactor (HFIR), and the Oak Ridge Leadership Computing Facility (OLCF), CNMS benefits from ORNL's expertise in materials research and computational sciences and provides, in particular, a connection between researchers in nanoscience and neutron sciences.

[1] The five NSRCs are the Center for Functional Nanomaterials, CFN (at Brookhaven National Laboratory), CINT (jointly at Los Alamos National Laboratory and Sandia National Laboratories), the Center for Nanoscale Materials, CNMS (at Argonne National Laboratory), the Center for Integrated Nanotechnologies, the Molecular Foundry (at Lawrence Berkeley National Laboratory), and CNMS.

Various BES documents highlight the importance of nanoscience in a broad range of fields. The report from the Basic Energy Sciences Advisory Committee (BESAC) on "Challenges at the Frontiers of Matter and Energy: Transformative Opportunities for Discovery Science" (November 2015) lists five such opportunities that have the potential to further transform key technologies involving matter and energy:

1. Mastering hierarchical architectures and beyond-equilibrium matter
2. Beyond ideal materials and systems: understanding the critical roles of heterogeneity, interfaces, and disorder
3. Harnessing coherence in light and matter
4. Revolutionary advances in models, mathematics, algorithms, data, and computing, and
5. Exploiting transformative advances in imaging capabilities across multiple scales.

Opportunities #1 (hierarchical architectures) and #2 (heterogeneities, interfaces, and disorder) are inherently nanoscience topics, and #5 underpins our work in imaging by various complementary approaches. An increased awareness of the importance of artificial intelligence (AI) in experimental research and of making such approaches accessible to the user community, the need for advanced data analytics, and a close and deliberate integration of TM&S into all aspects of CNMS research, directly align with opportunity #4, while the connections to QIS illustrate the relevance of harnessing coherence (#3).

Subsequent BES Basic Research Needs (BRN) Workshops on Quantum Materials (February 2016), Synthesis Science (May 2016), Innovation and Discovery of Transformative Experimental Tools (June 2016), Next Generation Electrical Energy Storage (March 2017), and Catalysis Science (May 2017) all include discussion of nanoscience topics in these energy related areas. Additionally, two areas related to information technology were addressed by Roundtable discussions: Neuromorphic Computing (October 2015) and Quantum Systems / Quantum Computing (October/November 2017), both of which provide strategic guidance to CNMS in planning future activities and illustrate the future needs of users.

A user facility for the nanoscience community is not only relevant in the context of the multi-disciplinary nature of nanoscience research: the model also addresses the importance of shared resources at a time where the cost of scientific equipment is increasing more rapidly that funding in many areas. However, making commercial equipment available to a broader audience is a minor mission of CNMS. Our main interest lies in developing the capabilities for the next-generation nanoscience researcher, which can only be accomplished by combining technique development with cutting-edge research. This is enabled through a two-pronged approach in which CNMS researchers are often deeply involved in the research efforts of the users (i.e., highly collaborative studies leading to joint publications) and where CNMS researchers pursue their own scientific careers. Our in-house research effort, organized in four scientific Themes as described below, is the primary vehicle for this scientific activity. In addition, CNMS researchers actively participate in research projects funded by other agencies to which they submit proposals and often participate in large, multi-institution teams. ORNL-internal funding opportunities to develop new concepts, namely the Seed projects (typically one-year efforts with a proof-of-concept mission) and the Laboratory Directed Research and Development (LDRD) program (typically 2-year projects), continue to play an important role in maintaining scientific diversity of thought, providing leadership opportunities to junior researchers, and maintaining a vibrant communities consisting of a balance of students, postdocs, and permanent staff.

As we describe below, modern materials research is increasingly relying on a very direct integration not only of TM&S, but also of data analytics and AI approaches. Many of these techniques are still in their infancy, and CNMS sees it as one of its core missions to contribute to the development of 21st century research approaches that take full advantage of tools that have just recently been introduced to the community and that are often not yet integrated into the efforts of our users.

DOE fosters an environment in which the five NSRCs are collaborating and coordinating activities, while at the same time critically evaluating each of the NSRCs in terms of well-defined metrics (primarily number of users and number of publications). Each year CNMS supports approximately 650 unique users from more than 100 different institutions spanning from academia to industry, and from around the world. The user community is diverse, ranging from students who work closely with CNMS staff—learning unique skills from experts and gaining access to cutting-edge instrumentation as they advance their research—to "partner users" who collaborate with staff to develop new capabilities and instruments that are then made available to the broad CNMS user community. About 10% of all users are theory users who work with the staff of the CNMS Nanomaterials Theory Institute (NTI) to gain access to expertise, specific computational software, and computational resources. About 40% perform synthesis and/or nanofabrication and typically use a broad range of characterization tools to verify the quality of the synthesized materials or to investigate their novel properties. The remaining 50% of the users come to CNMS specifically for characterization, using the broad range of tools including microscopy (electron, He-ion, scanning probe), atom probe tomography (APT), X-ray diffraction, optical spectroscopies, and other techniques. Collectively, CNMS staff and users publish more than 400 scientific articles in peer-reviewed journals per year, with more than a third appearing in journals with an impact factor > 7. In addition, CNMS researchers co-authored 19 patents in FY2017. Reflecting the impact of CNMS research, we note that more than 43,000 papers have cited CNMS publications since its inception.

2. The CNMS Research Groups and Key Capabilities

CNMS is organized into six research groups, whose primary responsibility is to maintain and operate the research laboratories and capabilities that are used for the user effort and the in-house research, but also to provide staff with a productive collaborative environment with an emphasis on mentoring that includes students, postdocs, and staff.

Providing a broad palette of research capabilities requires a highly diversified staff, with researchers from different disciplines working closely together. To this end, it is often desirable to rely on researchers from other divisions to support users in specific areas, and funding from DOE-SUFD is then used to support these specialized researchers.

Conversely, as mentioned above, CNMS researchers actively seek financial support from other funding sources to maintain a vibrant and broadly diversified research environment. Funding from DOE-BES Materials Science and Engineering Division (MSED) and Chemical Sciences, Geoscience, and Biosciences (CSGB) Divisions, other DOE programs (BER, EERE), form internal sources (LDRD), corporations (through Strategic Partnership Projects), etc., collectively contribute to about one fifth of all of the available research funds.

All researchers at CNMS are responsible both for working with users and for performing their own in-house research within the framework of the DOE-reviewed research project. Research staff are expected to split their SUFD-funded effort evenly between in-house research and user support; technicians and technical staff generally play a larger role in the user effort through their direct involvement with instrumentation, which is dedicated 80% of the time to the user effort. In contrast, postdocs do not have an obligation to support users, but typically choose to collaborate with users in areas related to their expertise.

Research Groups

The following is a brief description of each of the groups within CNMS:

- **Scanning Probe Microscopy.** The Scanning Probe Microscopy Group is a leader in imaging of material functionality at the nanoscale dimensions where properties are established and where quantum mechanics is manifest. The group develops and applies scanning probe microscopies to map local physical and electronic structure, electronic and ionic transport, spin, electromechanics, quasiparticle behavior, and other responses in materials of reduced dimensionality, including wires, surfaces, defects, and films. The group maintains ten atomic force microscopes with a variety of capabilities ranging from piezoresponse to surface potential and dielectric response in an array of controlled environments including liquid, humidity controlled, and ultrahigh vacuum. The group also operates nine variable or low temperature scanning tunneling microscopes in ultrahigh vacuum including unique instruments designed for *in situ* studies of oxides, high magnetic field response, or with multiple probes for transport. Users are provided access to staff with considerable expertise in the capabilities of these instruments including CNMS patented imaging modes and advanced statistical analysis. This expertise, when added to access to high quality microscopes, is an attraction for many users.

- **Electron and Atom Probe Microscopy**. The Electron and Atom Probe Microscopy Group comprises the former ShaRE User Program and specializes in the development and application of advanced analytical electron microscopy and atom probe tomography techniques towards understanding materials structure and chemistry at the atomic level. The extensive array of specialized scanning transmission electron microscopes (STEMs) available in the facility have enabled new studies across a broad range of nanomaterials, e.g., 2D chalcogenides, catalysts, and thin film heterostructures, which are specifically aimed at understanding the physical phenomena responsible for controlling materials performance.

The group maintains laboratories/facilities located on ORNL's central campus, including in the Advanced Microscopy Laboratory (AML), a dedicated, low-noise facility for performing high-resolution STEM imaging and electron energy loss spectroscopy (EELS), as well as laboratories in the High Temperature Materials Laboratory for conducting APT to elucidate microstructure-processing-property relationships that define materials behavior. These laboratories contribute to CNMS in-house science by providing a critical understanding of the role of Å-level structures (e.g., defects, interfaces, junctions, phase interactions, etc.), chemistry, and physical properties that provide insight into materials behavior, functionality, and degradation, Staff are developing novel imaging, spectroscopy, and operando techniques necessary to enable new scientific discoveries, e.g., novel *in situ* sample holders and more recently, 4D STEM and ptychography methods.

The group is responsible for providing users with access to state-of-the-art analytical STEM and APT capabilities (as well as staff expertise) that can be coupled directly with synthesis and performance studies conducted at CNMS, or to perform stand-alone experiments to characterize novel materials prepared at their home institutions.

- **Macromolecular Nanomaterials.** The Macromolecular Nanomaterials Group specializes in the synthesis and characterization of well-defined polymers and molecular building blocks with various architectures by a wide range of methodologies, including "living" anionic polymerizations, controlled radical and ring-opening polymerizations, controlled cross-coupling reactions, and most recently, polymerization under confinement (custom-designed flow reactors). The group's work on isotope-labeling (especially deuterium labeling) of soft matter is crucial to neutron scattering studies of structure and dynamics. The group maintains five synthesis laboratories equipped with four custom-built high vacuum lines and glass blowing stations. The group also has a broad spectrum of polymer characterization instruments, including chromatography (gel permeation chromatography (GPC) in various mobile phases, temperature gradient interaction chromatography (TGIC)), light scattering (dynamic and static), spectroscopies (FTIR, Nuclear Magnetic Resonance Spectroscopy (NMR), Matrix Assisted Laser Desorption Ionization-Time of Flight Mass Spectrometry (MALDI-TOF-MS), broadband dielectric spectroscopy), mechanical (rheometer, dynamic mechanical analysis), thermal (differential scanning calorimetry (DSC) and thermogravimetric analysis (TGA)), and thin film evaluation (ellipsometry, including phase modulated).

  These laboratories provide precise synthesis and rigorous characterization of the polymers and molecular building blocks that are essential to address the scientific questions in the CNMS in-house and user science. The group is responsible for providing users with well-defined soft matter, especially isotopically labeled polymers and molecular building blocks for their neutron studies, which accounts for about half of the user proposals for synthesis.

- **Functional Hybrid Nanomaterials.** The Functional Hybrid Nanomaterials Group specializes in research involving the controlled synthesis of nanostructures and thin films, exploration of their fundamental optical and electronic properties, and use of these properties as *in situ* and *operando* diagnostics of evolving structure and function. The group is recognized for the use of time-resolved, *in situ*, laser-based diagnostics to develop growth models for the controlled synthesis of nanomaterials, especially for carbon nanomaterials (including nanotubes, nanohorns and graphene) and thin film oxide heterostructures, and more recently two-dimensional (2D) metal chalcogenide crystals. In addition, the group develops new spectroscopic techniques to explore the heterogeneity and photoresponsivity of nanomaterials in different environments, especially photocatalysis, photovoltaics, and multimodal sensing. The group maintains six laboratories that are responsible for providing optical excitation for synthesis and characterization, and corresponding electronic, magnetic, and optoelectronic functionality. High-power nanosecond-pulsed lasers are employed in the growth of new thin films and nanostructures by pulsed laser deposition and gas-phase spectroscopy. Ultrafast (picosecond and femtosecond) pulsed lasers are employed for tunable-Raman spectroscopy and ultrafast white-light pump-probe spectroscopy characterization. The group also maintains laboratories for x-ray diffraction and for fundamental measurements of electronic, optoelectronic, and magnetic properties, as well as processing equipment for the assembly of soft and hard materials and electrodes. These laboratories contribute to the CNMS in-house research by providing well-defined and characterized nanomaterials that are central to the development of atomistic models correlating nanostructure and properties. For structural information, the group has worked closely with the staff at the SNS to develop neutron reflectivity and other scattering techniques for thin-film organic electronic materials. More generally, they are developing multimodal quartz-crystal microbalance platforms and computational approaches to measure and correlate structural, optical, and electronic properties changes of nanostructures with interrelated environmental effects such as humidity, temperature, and gas exposure. The group offers all of these capabilities and interrelationships to users.

- **Nanofabrication Research Laboratory**. The Nanofabrication Research Laboratory Group specializes in research in direct-write nanofabrication and micro- and nano-fluidics. Direct-write nanofabrication focuses on the fundamental properties of beam (electron, ion, photon) induced deposition and etching on substrates at the nanoscale, and incorporates a variety of imaging, characterization, analytical, and computational approaches. The micro- and nano-fluidics work focuses on understanding chemical reaction-diffusion systems in crowded and confined spaces, and incorporates a variety of fabrication, characterization, analytical, and computational approaches. The group maintains a 10,000 square foot class 100/1000 cleanroom facility housing a comprehensive suite of micro- and nanofabrication capabilities; optical, electron, and scanning probe characterization tools; and has an affiliated nanobiosciences laboratory that allows for the characterization of wet, soft, or biological materials. These laboratories contribute to the CNMS in-house research by allowing for the controlled fabrication, synthesis, and characterization of a wide variety of materials over length scales that vary from nanometers to centimeters.

Operating within the Nanofabrication Research Laboratory Group is the **Chemical Imaging Team,** a leader in high-resolution multimodal chemical imaging at the nanoscale. The team develops and utilizes complementary characterization techniques based on scanning probe microscopy, ion microscopy, optical spectroscopy and mass spectrometry in order to visualize surfaces, defects, interfaces, and devices to explore and correlate their physical and chemical properties. In addition to the core CNMS instrumentation, this Team also provides support for ORNL's AFM/ToF-SIMS operated through the Materials Characterization Core (an ORNL cost-recovery center), making it possible to offer a combination of scanning probes and SIMS-imaging to the users as it is being developed, just as CNMS's AFM-based MS-techniques and the integration of SIMS on the He-ion microscope (HIM) are forefront capabilities under active development.

- **Nanomaterials Theory Institute.** CNMS incorporates a substantial theory, modeling and simulation effort in the form of the Nanomaterials Theory Institute (NTI) that is cross-matrixed with the Computational Sciences and Engineering Division at ORNL. The NTI works primarily on atomistic simulations at the level of electronic structure and molecular dynamics, but also have developments and capabilities for coarse-grained and multi-scale modeling. Overall, the calculations support and challenge experimental work in polymer synthesis, functional oxides, layered materials, quantum materials, carbon nano-architectures, electrochemistry, catalysis, photovoltaics, optoelectronics, and electronic device performance. This effort also includes modeling for the interpretation of neutron scattering experiments and hence provides a cross synergy with the needs of SNS/HFIR in this domain.

  The NTI capabilities, research and development are a key component to all areas of the CNMS in-house research. The NTI also maintains allocations of computational time for its staff and CNMS users at the National Energy Research Scientific Computer Center (NERSC) and the Oak Ridge Leadership Computing (OLCF) facilities through competitive proposal processes (INCITE, ALCC, DD). The NTI also has dedicated in-house computational resources that are provided through a mid-sized compute cluster, software stack and data environment.

In addition to these research groups, CNMS interacts closely with and contributes strongly to the **Institute for Functional Imaging of Materials (IFIM).** ORNL established IFIM to play a coordinating role between imaging activities across ORNL. IFIM quickly identified a lack of accessible data analytics tools and approaches as a common theme and is working across

Directorates to provide the necessary infrastructure and methods. IFIM's Director, Sergei Kalinin, is a member of the Scanning Probe Microscopy Group of CNMS.

Operations and Support. CNMS management is responsible for all aspects of the environmental, safety, and health (ESH) program and its communication and implementation with users, staff, and visitors via the divisional operations program. CNMS policies and procedures, utilized in conjunction with all applicable ORNL and DOE requirements, ensure that CNMS is operated in a safe, compliant, and effective manner. The CNMS program follows ORNL ESH&Q program requirements as managed in the Standards Based Management System (SBMS) and incorporates state-of-the-art ESH assessment and control methods specifically tailored to the nuances of a leading edge engineered nanomaterial research user facility including equipment, materials, and processes that define CNMS operations. The ESH&Q programs minimize negative impacts on the research mission by ensuring that users, staff, and visitors are provided adequate resources and support to create the most positive research environment possible and reduce time spent on non-research-based activities.

Key Capabilities

The following paragraphs summarize the key capabilities are offered to the user community at CNMS. More details can be found at cnms.ornl.gov:

- **Imaging, Microscopy, and Nanoscale Characterization**: Scanning probe microscopy for imaging and dynamics in nanostructures including ionic and electronic transport, electromechanics, energetics, magnetic properties, chemical reactions, and electronic, structural and spin phases and transitions. The CNMS has developed microscopy techniques for resonance imaging, high data acquisition rates, and spin-dependent transport. A complementary suite of aberration-corrected STEMs for atomic-level imaging and spectroscopy, with an emphasis on ultrafast image acquisition, *in situ*/*operando* microscopy, and low-voltage/low-dose capabilities for beam-sensitive materials. CNMS is the only NSRC to offer APT to the user community, and these research efforts are continuously focused on application of the technique to nanoscale materials systems. Special emphasis on multimodal and chemical imaging (based on mass spectrometry and on optical spectroscopy) and He-ion microscopy (HIM). Development and implementation of deep data analytic methods.

- **Theory, modeling, and simulation**: Multiscale modeling, nanomaterials design, virtual synthesis, and characterization using high performance computing capabilities to establish and enhance links with experiments, including neutron nanoscience, and to aid understanding, prediction, and exploration.

- **Synthesis and fabrication:** Controlled synthesis and directed assembly of nanomaterials in a Class 1000 cleanroom environment ("Nanofabrication Research Laboratory"); chemical and biological functionalization of nanoscale materials, with a special emphasis on the directed assembly of 3-D structures. Synthesis of 2D materials, hybrid structures, and epitaxial oxide layers.

- **Soft matter synthesis and characterization**: Synthesis and molecular level characterization of small molecule building blocks, polymers, and polymer-modified interfaces, including biologically inspired systems, site-specific deuteration of molecules and polymers for neutron scattering studies.
- **Functional characterization of nanomaterials**: Optical characterization and laser spectroscopy. Electrical and optoelectronic characterization. Magnetometry and magnetotransport. X-ray diffraction, including small-angle X-ray scattering.

3. The CNMS In-House Research Effort: Four Scientific Themes

The CNMS in-house research effort aims to *understand the effects of dimensional and spatial confinement on formation and function of nanomaterials and nanostructures. This will enable the design of responsive nanomaterials that efficiently capture, transport, and/or convert energy as well as structures for sensing and information transmission, manipulation, or storage*. Reflecting the importance of combining activities in synthesis and in understanding of nanomaterials, the in-house research is structured into four themes. Two of these themes (Hierarchical Assembly and Directed Nanoscale Transformations) focus primarily on the formation of materials and an understanding of synthesis and assembly mechanisms. The other two themes (Electro-Ionic Interactions and Heterogeneities in Quantum Materials) emphasize the study and understanding of nanomaterials functionality.

Within the themes focusing on the formation of nanomaterials, **Hierarchical Assembly** seeks to understand the impact of material building block structure and environment or interface architecture on the formation of functional materials, and thus emphasizes topics of active matter and self-assembly. In contrast, **Directed Nanoscale Transformations** seeks to illuminate the basic scientific questions of how matter can be transformed locally and will thus enable novel nanofabrication approaches. In combination, these two themes tackle nanomaterials assembly both from the bottom-up (self-assembly) and top-down (fabrication) perspective, as we realize that the two approaches will need to merge for the formation of complex materials exploiting phenomena at multiple length scales. The unique CNMS expertise in using focused beams and local probes to measure and manipulate materials, the abilities to precisely synthesize molecules and functionalize surfaces, theoretical approaches to help guide and understand assembly and transformation, and the use of *in situ* neutron and microscopy approaches to observe nanomaterials formation, will contribute to the success of these themes.

Within the themes focusing on the function of nanomaterials, the effort on **Electro-Ionic Interactions** investigates nanoscale ionic transport and its consequence on lattice deformations and electron transport. In contrast, **Heterogeneities in Quantum Materials** studies the effects of compositional and structural heterogeneities on quantum behaviors of fundamental energy carriers and seeks to reveal quantum interactions at the heterogeneities. In combination, the two themes emphasize the most relevant electronic and ionic functionalities of nanomaterials, which are often coupled, and rely on materials and structures that are created within the efforts on Hierarchical Assembly and Directed Nanoscale Transformation. At the same time, the understanding of these functional behaviors then plays a role in understanding many of the mechanisms of nanomaterials formation through its investigation of energy and mass transport at the nanoscale.

Neutron scattering plays a role in all four of these themes, most directly so in Hierarchical Assembly and Electro-Ionic Interactions, while Directed Nanoscale Transformation and Heterogeneities in Quantum Materials have a much stronger reliance on spatially resolved measurements provided by scanning probes, HIM, and new opportunities presented by ORNL's purchase of the monochromated, aberration-corrected STEM (MAC-STEM) and a low-temperature, high-resolution four-probe Scanning Tunneling Microscopy (STM).

While the majority of the work at CNMS is motivated with specific energy applications in mind, including energy storage (batteries, supercapacitors, etc.), energy conversion (photovoltaics, thermoelectrics, solid state lighting), and catalysis, new "beyond-Moore" computing approaches also strongly benefit from the work in these research themes. QIS and quantum computing are most closely represented in Directed Nanoscale Transformations and Heterogeneities in Quantum Materials, while topics related to neuromorphic computing are part of the research in Electro-Ionic Interactions and Hierarchical Assembly.

Hierarchical Assembly

The overarching goal of the Hierarchical Assembly theme is *to understand the impact of material building block structure and environment or interface architecture on the formation, structure, and response of functional materials*. Understanding the role of these factors in driving key steps in material formation is essential to the predictive design of new classes of nanoscale and responsive materials. In parallel to, and intertwined with, the development of direct write techniques or conventional nanofabrication processes that actively shape or form materials into specific geometries, we recognize the need to develop strategies that can be used to push materials into desired states over extended length scales. These strategies will center on tuning local enthalpic and entropic interactions in combination with non-equilibrium processes as means of achieving desired structural and morphological properties that ultimately translate into targeted material function and dynamic responsive behavior.

This theme addresses the specific aims of

1) Active Matter and Molecular Self-Organization: understanding the dynamic assembly of molecular and polymeric materials under constant excitation, to understand how the coordinated motion of individual building blocks can be used to synthesize materials capable of responding to external stimulus.
2) Assembly of Active Interfaces: understanding the link between structure and dynamic transitions in hierarchical and hybrid material systems in response to changing environments or stimulus.

3) Evolving Materials: understand how 'fluid,' loosely coupled, macromaterials (i.e., lipid bilayers and copolymer membrane mimics) can be developed to undergo *dynamic* reorganization and shifts in their steady state structure as they are driven repeatedly away from equilibrium.

Directed Nanoscale Transformations

This theme seeks to understand and control the dynamic changes of chemical and structural states that materials undergo in confined and non-equilibrium conditions by focusing on three specific aims:

1) Energy Transfer: How is energy transferred from a beam or field to initiate a transformation? Can we optimize and control this transfer?
2) Energy Landscape: What is the influence of the environment on a transformation? Can we assess and tune the underlying energy landscape to select and guide transformations?
3) Energy Flow: How do the results of previous transformations affect subsequent ones? Can we use intelligent feedback to guide a cascade of transformations?

The starting point for this work is the observation that experimental platforms to visualize structure and function, such as electron, ion, and scanning probe microscopes and associated spectroscopies, can also be used to induce transformation, thus enabling the study and control of the processes of highly localized structural, chemical, and electrochemical changes. The understanding gained here will enable us to direct matter with atomic precision to create 3D nanoscale structures with desired form and function.

The expertise in controlling scanning probes, electron beams, and ion beams is crucial to the success of this work. CNMS has attained strong leadership in this area through its work in functional imaging of materials and through LDRD efforts in atomic-scale fabrication described below. The ability to manipulate and study matter in a tight research loop at the atomic- and nano- scales will be of key interest to a broad user community, including those from the QIS field.

Electro-Ionic Interactions

The goal of this theme is to understand and modify bias-induced material functions, which rely on the coupling of ion and electron transport through the bulk and along/across interfaces. The work in this theme is based on the foundation laid over the past 3 years, which was focused on electromechanical phenomena based on bias-induced mass transport. We will expand the concepts explored in that theme and include pure ionic transport as well as electron transport. This is critical since electronic and ionic currents are coupled to maintain charge neutrality. To achieve the overarching goal, three specific aims will guide the research:

1) Ion-Induced Electromechanical Phenomena
2) Ionic Transport at Interfaces
3) Coupled Electron-Ion Transport under High Electric Fields

The theme will continue working on previous focus areas of ion-conducting polymers and ion conducting ferroelectrics (aim 1). Here we want to explore how we can understand and enhance electromechanical responses and explore the size limit of electromechanical systems. This will include quasi-2D materials in the few layer regime, closely connecting to the Heterogeneities in Quantum Materials theme.

In the area of solid-solid interfaces (aim 2) we will answer how ionic transport is facilitated across interfaces through internal fields. This will be achieved by new STEM techniques coupled with complimentary SPM-based techniques, which will allow us to separate effects of chemistry, strain, and fields. In addition, the challenging but important field of soft-hard interfaces will be included where the ion conduction pathways are not clear. This research area has a strong connection to the Hierarchical Assembly theme.

Finally, we will explore systems with strongly coupled electron-ion transport in the regime of high electric fields (aim 3) to answer the question of how long-range ionic motion modifies electron transport in oxide materials. We will utilize unique capabilities such as microwave microscopy and APT to provide insight on processes relevant for information storage.

Heterogeneities in Quantum Materials

The overarching goal of this theme is to understand the effects of compositional and structural heterogeneities on quantum behaviors of fundamental energy carriers and reveal quantum interactions at the heterogeneities. This is achieved by studying the effects of heterogeneities, including defects, interfaces, and disorder on electronic, magnetic, and vibrational behaviors and their interactions in low-dimensional quantum materials, focusing on an atomistic-level understanding of quantum states and interactions in materials with physical dimensions that are comparable to the quantum phase coherence length of the electrons. The understanding will feedback to the design of new quantum materials that address DOE BES needs and impact energy transport, quantum computing, and low power computing. The effects of heterogeneities are treated in three aims:

1) Quantum Structures: What do heterogeneities look like structurally, electronically, and magnetically?
2) Quantum Behaviors: How do heterogeneities affect the quantum behaviors of fundamental energy carriers?
3) Quantum Interactions: What role do heterogeneities play in the quantum interactions and how can this be used to enable energy conversions?

This research theme builds directly on expertise in atomic-level control and understanding of materials including heterogeneities, through innovative development and utilization of scanning probe and STEM imaging and spectroscopy, combined with first-principles calculations. It takes advantage of capabilities developed in Directed Nanoscale Transformations and Hierarchical Assembly Themes that focus on formation of nanomaterials with precisely controlled defects and interfaces and provides information on electronic behaviors to aid the understanding of Electro-Ionic Interactions where heterogeneities, particularly interfaces, often play a key role. The gained knowledge will push beyond our current understanding of "ideal" materials and provide the critical knowledge needed to control the couplings of electrons and spins in materials and create the designer quantum states that will contribute to the foundations of QIS. Users at CNMS will be able to take advantage of the developed experimental techniques to correlate atomic-scale structural information such as electronic and magnetic structures of defects obtained from quantitative microscopic studies with functionalities from optical and electrical spectroscopy characterization.

4. Cross-Cutting Focus Areas and New Directions

The four scientific Themes described above provide the general framework for the in-house research at CNMS. In addition, and often cross-cutting these Themes, we have identified a number of focus areas and new directions into which we plan to invest, both through equipment and staffing decisions, and which we continuously re-evaluate in the context of the needs of the user community and of new opportunities arising in the various areas of nanoscience. Input from the CNMS Scientific Advisory Committee, the User Executive Committee (UEC), feedback received at the DOE Triennial Reviews, as well as direct interactions with the other NSRCs, all contribute to the planning of future directions.

Modernizing Materials Science

In is clear that interest in deep data analytics, AI and machine learning (ML) for science and engineering problems/applications, continues to rapidly increase. This is driven in part by the quantity and quality of data that can be collected and stored, as well as the computational power available to process this data, together with the successes that have been achieved in applying AI to numerous applications in science and engineering. At CNMS, we are proactively involved in integrating theory-based computational approaches, deep data analytics including ML, and AI approaches to address problems in nanomaterials science, and to provide such emerging capabilities to the user community.

Although theory, modeling, and simulation have been reasonably successful in predicting single materials functionalities, modern materials development requires understanding, designing, and predicting materials with *competing functionalities* that often give rise to inhomogeneous ground states and chemical disorder as well as properties that emerge from reduced dimensions or strong quantum confinement. From occupancy in equivalent or weakly nonequivalent positions, to chemical phase separation in physically inhomogeneous systems (e.g., manganites, high temperature superconductors), we must be able to treat these mixed degrees of freedom. Being able to do so can provide a route for rapid screening by first-principles methods of realistic materials structures incorporating the complexities of real materials, such as extended defects, disorder and impurities, as well as the complexities due to strong electron correlation (common in quantum materials) and proximities to phase transitions. However, achieving this requires a foundation rooted in quantum approaches that is capable of effectively utilizing high performance computing (HPC), a framework for high-throughput calculations of multiple configurations and structures, and capabilities for extensive data assimilation, validation, and uncertainty quantification.

Theory, modeling, simulation, and data analytics in the materials design/optimization loop: In order to move beyond traditional theory-based avenues boasting improved capabilities in accuracies and scales, we are utilizing the spectacular progress over the last 10 years in imaging, X-ray, and neutron scattering, which together provide quantitative structural and functional information from the atomic scale to the relevant mesoscales where real world functionality often emerges. However, this frequently involves dealing with complex multi-dimensional data that require statistical approaches, decorrelation, clustering, and visualization techniques. For example, STEM allows the real-time visualization of solid-state transformations in materials, including those induced by an electron beam and temperature, with atomic resolution. However, despite the ever-expanding capabilities for high-resolution data acquisition, the standard manual and *ex-situ* analysis of the collected volumes of data becomes nearly impossible due to sample dynamics. To circumvent this problem, we are developing a deep learning framework for dynamic STEM imaging that is trained to find the structures (defects) that break a crystal lattice periodicity. This approach will ultimately need to be extended to interactive knowledge discovery to deliver direct manipulation and design of materials (as is the focus of the Directed Nanoscale Transformation theme).

In general, data from state-of-the-art imaging, spectroscopy, and scattering approaches can be integrated with theory for significantly improving the quality and rate of theoretical predictions and for accelerating the design and discovery of functional materials. To this end, we are establishing workflows for data and imaging analysis that are made broadly available through Jupyter notebooks, for example, to provide the relevant atomic and magnetic configurations as well as response behaviors of materials. Ultimately, this can be used as direct input into the first principles simulations, and subsequent refinement of theoretical parameters via iterative feedback. Additionally, through IFIM, efforts have established an open-source set of utilities for image processing and scientific analysis of imaging modalities including multi-frequency scanning probe microscopy, scanning tunneling microscopy, X-ray diffraction microscopy, and STEM, called *pyCroscopy*.

Opportunities in Quantum Information Science

Quantum Information Science (QIS) is generally described as the confluence of two of the great scientific and technological revolutions of the 20th century: quantum mechanics and information theory (see, e.g., “A Federal Vision for Quantum Information Science,” Subcommittee on Quantum Information Science, National Science and Technology Council, January 2009). QIS builds on uniquely quantum phenomena, such as superposition, entanglement, and squeezing to obtain and process information in ways that cannot be achieved based on classical behavior (“Advancing Quantum Information Science: National Challenges and Opportunities,” Interagency Working Group on QIS, National Science and Technology Council, July 2016). BES convened two roundtable discussions on QIS – one focused on quantum computing

(“Opportunities for Quantum Computing in Chemical and Materials Sciences,” Oct. 31-Nov. 1, 2017) and one on quantum systems (“Opportunities for Basic Research for Next-Generation Quantum Systems,” Oct. 30-31, 2017). As the latter of these reports points out, such quantum systems are expected to lead to enhanced resolution in imaging, sensors, and detectors; advanced cryptography for more secure communication; and significantly larger computational capabilities at speeds far greater than those possible in classical computing. But realizing these advances requires breakthrough advances in understanding how such quantum systems behave and how they can be controlled and utilized. Creating and controlling quantum states within molecules and materials involves significant hurdles within materials science and chemistry that are fundamental nanoscience challenges. As was also pointed out in the above-referenced July 2016 report, “Much of the research in QIS has been conducted within existing institutional boundaries. ... The next critical steps in QIS R&D will require increased collaborations across these boundaries. Teams with a diverse range of skills will be needed.” This is precisely the type of an environment that CNMS provides its user community – a collaborative center where researchers from different disciplines and backgrounds work hand-in-hand on cutting-edge instrumentation and in a setting that emphasizes integration of TM&S into all steps of the studies.

It is thus not surprising that QIS is playing a rapidly increasing role at CNMS. In addition to the unique capabilities developed through our in-house research and interactions with users, CNMS benefits from the broad quantum materials effort being pursued at ORNL’s neutron sources and using ORNL’s computational resources, and the very direct interactions with ORNL’s Quantum Information Sciences group (within the Computational Sciences Directorate): In fact, interactions with this group have helped in shaping the CNMS direction in QIS, and allowed CNMS to build the bridge between the materials manipulation and imaging side, which is clearly within the scope of nanoscience, and the information science aspect of QIS, which remains outside the scope of CNMS research.

Two of our in-house research themes, Directed Nanoscale Transformation and Heterogeneities in Quantum Materials, have a direct link to QIS. In fact, some of the most spectacular achievements within the Directed Nanoscale Transformation effort, namely the direct placement and manipulation of individual dopants within a 2D lattice, have been prominently featured in the above-mentioned Roundtable report. Combining such exquisite control over individual dopant placement with simultaneous progress in nanoscale 3D direct-write fabrication will lead to entirely new opportunities to create, and interface with, artificial quantum systems. CNMS is actively planning investments in equipment that will enable a more direct integration between such dissimilar approaches, using combinations of electron and ion beams with *in situ* monitoring and operando quantum observations. At the same time, CNMS nanofabrication techniques are already being used to create structures that enable the integration of quantum emitters with plasmonic or nanophotonic structures, or cathodoluminescence observations in a STEM. 4-probe SPM approaches will similarly make it possible to manipulate dopants and observe the consequence on quantum behaviors *in situ*. Developments of post-reconstruction data processes has enhanced the capabilities of APT to now quantitatively determine the interface widths in semiconductor quantum well structures, and STEM techniques are now capable of observing every individual dopant in transition metal dichalcogenides.

Finally, CNMS is also actively exploring ways to integrate quantum sensing into advanced measurement and imaging approaches. For example, CNMS is teaming with the ORNL Quantum Information Science group on a Seed project to apply quantum noise reduction (squeezed light) to scanning probe microscopy.

Polymers and Soft Matter

Polymer and soft matter research, rather than being a separate research direction or stand-alone capability, is seen as cross-cutting all of our science themes, and the Center's established capabilities in synthesis, site-specific deuteration, and broad functional characterization, strongly support these activities. For example, the effects of isotopic substitutions on electron transport is studied in the Heterogeneities in Quantum Materials theme; Electro-Ionic Interactions includes the study of electroactive polymers, Hierarchical Assembly places a special emphasis on bottlebrush block copolymers; and Directed Nanoscale Transformations addresses the conversion of polymers into nanoribbons. These activities closely tie to other soft-matter efforts at ORNL (including within the Neutron Sciences Directorate, within the BES-MSED portfolio and the applied sciences efforts at the Carbon Fiber Technology Center and the Manufacturing Demonstration Facility) and are benefitting from recent LDRD-funded activities.

Polymers are a versatile class of materials that often provide key advantages over other materials for energy applications: (1) *Light weight* (built from the lightest atoms such as H, C, O, N); (2) *Unique viscoelastic* properties (only polymers exhibit rubber elasticity and unmatched ductility); (3) *Extremely broad range of tunable properties* (e.g., modulus from kPa to GPa, conductivity from insulators to conductors, selective permeability, etc.); and (4) *Wide variety of scalable synthesis and processing methods*. These unique features endow polymers as a material of choice for many current, and especially for future technologies, which are directly relevant to DOE missions, including energy generation (photovoltaics), conversion (fuel cells), storage (batteries, capacitors), energy efficiency (lightweight materials, lubrication), novel computing (neuromorphic computing), and separation technologies (natural gas purification, water purification/desalination). Since polymer-based materials have essentially an unlimited range of tunable parameters (e.g., chemical composition, structure, and architecture, ionic groups, copolymers and blends, and hybrid/composite materials), rational design of functional polymeric materials requires a deep fundamental understanding of the interplay between structure, morphology, and dynamics on multiple length and time scales. To this end, we seek *to understand how the chemical structure, molecular architecture and composition, nanoscale organization, and dynamics of macromolecular and hybrid materials influence response (stimuli responsive and adaptive) and properties in order to design the next generation of functional materials for energy technologies.*

To implement this ambitious vision, we use our expertise in atom precise monomer and polymer synthesis including deuteration, exquisite characterization of structure, morphology and dynamics by a variety of state-of-the-art methods, including an array of microscopies (note specifically CNMS's recent acquisition of the low-voltage/low-dose cryo-capable JEOL NEOARM), computational modeling from quantum to coarse-grained and continuum levels working on high-performance computing platforms (pre-exascale, moving quickly toward exascale), and an array of neutron scattering techniques through direct integration with SNS and HFIR. For example, for understanding interfaces within polymers (soft-soft) or between polymers (soft) and "hard" materials (oxides, metals, ceramics), our integration with expertise in materials imaging via scanning probe and electron microscopies alongside strong connection to theory/simulation, opens unique opportunities. In particular, advances of multiparametric and multifunctional characterization with high resolution, exceptionally fast detectors/cameras and acquisition capabilities, and the capacity to operate under various media, SPM and (S)TEM are now well positioned to tackle the challenges of understanding the structure, dynamics, transport (ions, molecules), and properties of increasingly more complex polymers and their composites. Moreover, the combination of two or more SPM-based modes to characterize multiple aspects of polymers is very promising way to characterize polymers and their interfaces with exceptional spatial resolution: for example, combining AFM and scanning electrochemical microscopy (AFM-SCEM) can provide spatially correlated electrochemical and nanomechanical information paired with high-resolution topographical data of soft electronic devices. When paired with precise synthesis and detailed modeling/theory, polymer imaging will provide a transformative step for realizing our ambitious overarching goal.

Precise synthesis of macromolecules (as well as molecular building blocks and isotopically labelled compounds) not only affords the opportunity to create a virtually unlimited array of tailored molecular architectures that display different properties and functions, but also supports the critical needs of CNMS theme science and user projects (including users of neutron sources). While the synthesis effort in the Macromolecular Nanomaterials laboratories establishes a unique position among the NSRCs in soft matter research, the synthesis of target materials within a reasonable time frame continues to be a challenge. Although there was significant progress in the synthesis of macromolecules with precisely controlled architecture and functionalities in the last decade, the need to synthesize macromolecules with precise primary structural parameters (such as sequence, composition, tacticity, branching, topology, dimensionality, functionality) efficiently, polymerize pre-organized monomers or nano-objects to create architectures encoded with designed functions, and to develop new macromolecular materials with theoretical screening combined with new chemistry from readily available monomers, remains imperative. Our vision for macromolecular synthesis research at CNMS is to be recognized internationally as the place for precise polymer synthesis, rigorous characterization, and selective isotopic labelling.

To this end, we use an integrated approach with CNMS's strong scientific programs and the broad spectrum of core facilities and expertise at ORNL (especially supercomputing and neutron science). This enables us to provide both nanoscience and neutron users the opportunity to address their pressing scientific questions and technological challenges in a timely manner. Here we strive to:

i. Provide expertise and capabilities for innovative polymer synthesis by enhancing our signature expertise in anionic polymerizations and directly utilizing the advantages of flow chemistry. The addition of flow chemistry, which uses channels or tubing to conduct a reaction in a continuous stream rather than in a flask, can provide chemists with unique opportunities to increase yield, enhance reactivity/selectivity, or in some cases enable new reactions.

ii. Provide leading efforts for precision isotopically labelled materials synthesis, especially deuteration, enabled by exploring and understanding their influence on the physio-chemical properties.

iii. Develop *in situ* soft matter characterization techniques and harness the power of predictive theory and modeling in synthesis.

As an integral part of the ORNL's research community, our macromolecular research is well-positioned to achieve the above-discussed goals. We have dedicated staff, engaged users, state-of-the-art in-house instrumentation, and we are adjacent to core facilities (supercomputing and neutron sources). More importantly, the vibrant CNMS theme science programs directly integrate macromolecular science. In particular, the flow chemistry platforms we are currently developing will enable us to carry out controlled reactions not possible using bench approaches. This will also accelerate polymer discovery by flow reactions in tandem with external control (such as light mediation).

We must keep pace with emerging trends in polymer science. For example, the application of polymers in the neurosciences and other biologically related areas are exciting new areas that will be reflected in future user demands. Additionally, conjugate materials (molecules built from synthetic components and bio-components) that mimic specific aspects of biological complexity and active matter (non-equilibrium) are intriguing areas with a large growth potential.

Neutron Nanoscience

The direct proximity of CNMS to ORNL's two neutron sources, HFIR and SNS, provides a unique opportunity to use neutrons as probes to answer nanoscience questions. In fact, 18% of all CNMS users are also users of either SNS or HFIR or both. We are actively taking steps to increase the interactions between the neutron and nanoscience efforts, in both CNMS staff science and user projects. For example, ORNL established a Soft Matter Council of members from both the nanoscience and neutron science areas within ORNL that has the mandate to identify areas of common interest and opportunities. Additionally, a polymer focus group between SNS/HFIR-CNMS was established to coordinate outreach in terms of neutron and nanoscience, and a polymer working group spanning ORNL was established to coordinate a strategy to launch a new initiative in soft matter at ORNL. Finally, administrative steps, such as a simplified access route for CNMS users to SNS and HFIR and vice versa have been implemented. Neutron–nano interactions are currently strongest in the area of soft matter research, in which CNMS deuteration capabilities are a key resource for the neutron scattering programs, and the sensitivity of neutrons to light elements allows the investigation of assembly, structure, and dynamics in complex macromolecular systems. The demand for the selective deuteration capabilities will continue to increase as the number of soft-matter users at both facilities increases. In parallel, CNMS is investigating the impact of deuterium substitution on structural and functional properties, e.g., via quantum zero-point energy and electron-phonon coupling.

CNMS continues to partner with SNS/HFIR to develop new capabilities for neutron studies of materials, including specialized *sample environments* and new characterization techniques that can be used in concert with neutrons to provide multi-dimensional information on material structure and function (e.g., vibrational spectroscopy of bulk materials at SNS and at the atomic-level using MAC-STEM). Similarly, CNMS provides a key component to future neutron studies of soft matter and bio-inspired materials via the fabrication of *nanostructured templates* having lateral dimensions sufficient for neutron experiments and the development of computational models and simulations that probe the same length and time scales. As more instruments become integrated into the neutron user program, and as the Proton Power Upgrade at SNS proceeds, vibrant interactions between CNMS and the Neutron Sciences Directorate continue grow.

As an illustrative example, neutron reflectometry is used in critical experiments to provide depth information for materials. This is particularly the case in the study of assembly and self-organization of macromolecular and polymeric materials (where the specialized sample environments provide simultaneous optical and spectroscopic information), or in investigating electronic and magnetic reconstruction at atomically abrupt interfaces within an oxide heterostructure. Complementary techniques, such as scanning-probe-based 3D force imaging to study ionic layering in liquids, or real-space cross-sectional STEM for epitaxial structures, will benefit from (and contribute to) these neutron capabilities. Also, at this crossroad, we have been involved in developing integrated experimental-theoretical efforts to describe the structure and dynamics of inhomogeneous polymeric systems, paying special attention to the local charge regulation and electric polarization in an external electric field, and the resulting microphase separated morphologies. Neutron spin-echo, broadband dielectric spectroscopy and neutron reflectivity in the presence of an applied electric field are ongoing for verifying predictions of the models for the ion transport and the chain dynamics. A specific Neutron reflectivity set-up in the presence of applied electric field was developed at the Spallation Neutron Source (SNS) as a part of an active laboratory directed research between SNS and CNMS.

CNMS research in energy storage and conversion is another specific example of the benefits of proximity to the ORNL neutron sources. Issues of ionic transport in inorganic solids, including lithium ions and protons, are investigated using the SNS instruments NOMAD instrument (Nanoscale-Ordered Materials Diffractometer) and POWGEN (powder diffractometer) for structure resolution. Dynamic ionic transport studies within the Electro-Ionic Interactions theme and in user work take advantage of quasi-elastic scattering on the SNS BASIS (Backscattering Spectrometer). Additionally, the SNS VULCAN (Engineering Materials Diffractometer) instrument has provided the capability for operando charge and discharge studies of whole battery cells to understand structure evolution within a cell, and an *in situ* high-temperature environment for real-time characterization of structure and property evolution for energy-related materials.

The SNS vibrational neutron spectrometer (VISION) uses neutrons to probe molecular vibrations while simultaneously collecting diffraction data and provides an opportunity for the study of *in situ* catalysis and study of active sites in materials difficult to probe using other spectroscopic techniques. CNMS scientists are in a unique position to help SNS staff develop environmental capabilities that will attract users studying catalytic and energy-related processes to both facilities. CNMS provides unparalleled local capabilities for materials synthesis, and CNMS researchers work closely with SNS scientists to develop capabilities of interest to users of both facilities. For example, we are collaborating (LDRD) on the development of hyperspectral compressive imaging with neutrons. Specifically, by using a reconfigurable spatial modulator for neutrons and by exploiting VISION's already existing neutron detectors, VISION can be outfitted with the world's first "single pixel" neutron camera. The end-result will be the only instrument of its kind in the world: a neutron-imaging vibrational spectrometer and diffractometer.

Closely related to the interactions with the neutron community and taking advantage of the sensitivity of neutrons to spins, is our effort at CNMS to strengthen and broaden research on *magnetism and magnetic materials as well as the class of quantum materials*. This is particularly relevant in the context of increased interactions with the QIS community. On the synthesis side, pulsed laser deposition (PLD) of oxide layers and an evaluation of opportunities to expand PLD to non-oxide materials (such as halides) will play an important role. In combination with capabilities in molecular-beam epitaxy of metallic materials, these synthesis efforts strategically position us to take advantage of CNMS characterization strengths in measuring magnetism and magneto-transport using new methods in spin-polarized scanning tunneling spectroscopy and 4-probe STM along with cutting-edge approaches for extracting magnetic information from EELS data (discussed in the imaging strategy) that will play a significant role in connecting neutron results with spatially resolved information. Work on the MAC-STEM will also enable a visualization of phonons at a local scale – providing a unique synergy with inelastic neutron scattering. CNMS is leading the development of these spatially resolved approaches, making them accessible to a broad range of users, including those who access SNS/HFIR. Theoretical contributions, including work on multiple orbital models of transition metal oxides with realistic interactions, help researchers understand and develop experiments by studying not only static properties at zero temperature but also dynamical properties at finite temperature.

CNMS also maintains and further develops a host of complementary techniques that includes: NMR, MALDI-TOF-MS, Raman spectroscopy, light scattering and rheometry, X-ray diffraction (XRD), and magnetic (QuantumDesign MPMS SQUID magnetometer), as well as magneto-transport (QuantumDesign PPMS) characterization. These provide different levels of valuable information (composition, spatiotemporal properties and structure, magnetic and physical properties) that are not easily accessed from neutron scattering and thus provide another valuable experimental modality for neutron sciences users and staff.

Finally, we note that CNMS continues to contribute to the operation of SNS through the fabrication of the diamond stripper foils, development of sensors to measure stresses on the spallation targets, and development and nanofabrication of zone plates.

Imaging Strategy

A broad range of imaging approaches is needed for success in nanoscience, both the staff science pursued in the scientific themes and the science carried out by the user community. CNMS combines key strengths in SPM, analytical STEM, HIM, and APT. Within SPM, low-temperature and variable-temperature STMs are designed and built fully in-house. Based on our expertise in 4-probe STM, a scanning tunneling potentiometry method has been developed, which has been extended to spin-polarized probes. Similarly, band excitation and multidimensional spectroscopic modes for probing bias- and temperature-induced phase transformations and electrochemical processes have created new opportunities for exploration in areas as diversified as ferroelectrics, multiferroics, and energy storage/conversion, and are expanded to "G-mode" methods in which the entire tip trajectory is being recorded, increasing data speeds and eliminating the need of curve fitting and a-priori assumptions. These capabilities have propelled CNMS to the forefronts of these fields and engendered a vibrant user program. A new USM microscope from Unisoku operating at temperatures down to 2K and magnetic fields up to 9T opens opportunities for atomically resolved structure and excitations, including novel quantum quasiparticles. Similarly, ORNL's acquisition of a true low temperature Scienta-Omicron four-probe STM will enable both electron and spin transport measurements in nanomaterials.

CNMS continues to be recognized as a world-leader in using STEM-based EELS to study materials behavior, which is being further advanced by CNMS's recent investment in the JEOL NEOARM aberration-corrected TEM/STEM, which is capable of operation at 30kV and low-electron-dose (for beam-sensitive materials such as soft matter) and is equipped with significantly advanced capabilities for simultaneous EELS and energy dispersive X-ray spectroscopy (EDS) data acquisition, and an ultrafast pixelated detector for ptychography, and also by ORNL's recent investment in the Nion MAC-STEM and aberration-corrected EEL spectrometer, which will push the limit of achievable energy resolution to the sub-10nm regime. APT will continue to focus on expanding methodologies toward the 3D atom-by-atom tomography analysis of new nanoscale materials, an area which has undergone significant growth as we aim to study non-conductive materials using APT. These recent advancements in STEM and APT, which are being coupled with CNMS priorities in data analytics, AI/ML, and TM&S, are truly novel amongst the NSRCs, and will enable new scientific studies focused on materials functionality and behavior.

Across many of these imaging platforms, materials are not simply imaged in the sense of determining atom locations but rather are interrogated to reveal spatially resolved maps of physical properties and functionalities, which increasingly can be connected to physical models through the use of AI approaches. This will eventually complete the transition from "seeing where atoms are" to "seeing what atoms do" and finally to "understanding why they do what they do." Local stimuli (e.g., electric fields, strains, electron beams) can reversibly probe a material's response or produce localized modifications, chemical reactions, or nanostructure assembly. Mapping of physical properties results in multi-dimensional data sets for which the historically successful analysis approaches of fitting and parameter extraction have become inadequate, especially in cases where a phenomenological parameterization at all locations in a material cannot be performed without loss of information. This points to the need for "deep data" approaches, an area in which the close link between theory, computation, and imaging strategically positions CNMS to provide the required breakthroughs.

The CNMS vision to provide staff scientists and users with forefront tools, expertise, and approaches is supported by our plans to

- extend our development of novel SPM techniques, particularly by pushing our G-mode acquisition to higher speeds to examine the dynamics of ionic and electronic motion.
- continue to develop and implement state-of-the-art techniques for probing phonons and optical excitations, magnetic and electrical responses, and lattice vibrations through the use of ORNL's MAC-STEM. These studies will serve to strengthen the interaction and collaboration with SNS and HFIR and enable advances in QIS, and they will be facilitated by incorporating the ultrahigh-energy resolution, aberration-corrected EELS on the MAC-STEM with world-leading energy resolution.
- fully implement the capabilities afforded by the new JEOL NEOARM TEM/STEM, including capabilities for low temperature (cryo-EM), low voltage, and low dose imaging and analysis for soft matter, biological, and beam-sensitive materials; highly advanced capabilities for the simultaneous acquisition of atomic-level EELS and EDS data; provide new platform to exploit ultrafast data acquisition capabilities enabled by pixelated detector and simultaneously develop algorithms, AI, ML, and data analytics to facilitate ptychography and 4D STEM methodologies (and make methods accessible across the entire suite of CNMS STEM and APT instrumentation/facilities)
- expand expertise in quantum quasiparticle excitations seen with low-temperature, high field, scanning tunneling spectroscopy for novel applications in QIS
- advance the scientific basis of HIM through the recently implemented, one-of-a-kind mass spectrometry approach into HIM so as to provide spatially resolved and surface-sensitive chemical information
- develop of chemical imaging using mass-spectrometry approaches, as mentioned above for the case of HIM, MALDI-TOF, and TOF-SIMS, but also increasing spatial resolution using a rapidly heated tip in scanning probe microscopy, to provide a seamless platform of chemical imaging across multiple length scales and probing different materials
- emphasize continued improvements for *in situ* and operando functional imaging and spectroscopy of materials processes at high spatial and temporal resolution; such studies are being facilitated and enhanced by CNMS's greater focus on ultrafast data acquisition, data reconstructions through advanced data analytics, and continuous specialized holder development

- implement CNMS advances on ORNL's new low-temperature Scienta-Omicron 4-probe SPM instrument to map electronic current and voltage drops through potentiometry and to visualize magnetic properties and spin transport using spin-selective tunneling.

Linking many of these approaches are efforts to combine data streams (e.g., APT data with STEM chemical and lattice imaging and spectroscopy data) to provide unprecedented 3D, multi-modal insights into nanomaterials structure and behavior.

Coordination between the different imaging efforts at ORNL will continue to be emphasized, with core strengths being identified an exploited. This includes working together on advancing materials research supported by the Materials Sciences and Engineering Division at BES through two ORNL FWPs, through enhancing research capabilities on the Nion MAC-STEM, and through the Scienta-Omicron 4-Probe STM. These two instruments are ORNL resources that benefit multiple research programs including the CNMS. IFIM efforts ensure that CNMS imaging science works in concert with other imaging efforts across ORNL (particularly chemical imaging and STEM-based approaches) and data analytics capabilities. Therefore, IFIM provides a platform for enhanced information exchange, efficient resource utilization, and identification of common needs across the various imaging platforms.

3D Nanofabrication

Being able to create arbitrary three-dimensional nanostructures has long been a dream of researchers. Two-photon polymerization, as implemented in the Nanoscribe tool at CNMS, provides an attractive route to the creation of such structures with feature sizes approaching 100 nm and is now made available to the user community. Future work will emphasize the development of novel photoactive precursors for the formation of 3D structures built with different materials and in combination with bottom-up and top-down pre- and post-processing strategies. Integration of two-photon polymerization into already established fabrication sequences will create novel opportunities for the user community in utilizing this commercial tool.

High-resolution 3D metallic structures are also being fabricated via electron or ion beam–induced deposition (EBID or IBID), in which a volatile organometallic precursor decomposes in a controlled manner under electron or ion impingement, thereby depositing materials in the area local to the well-defined electron or ion probe. Research at CNMS, in collaboration with users, has pushed this nanoscale analog of 3D printing (or additive manufacturing) to the point where structures with exquisite geometries and precise dimensions can be created, including structures consisting of high-purity metal or functionalized to act as chemical sensors.

A closely related technique using He^+ (which has the potential of achieving a purer product and a more efficient conversion of the precursor into the desired material) has been demonstrated and implemented on the HIM. Both electrons and He-ions can also be used to create structures from a liquid precursor, using a Protochips liquid cell and the electrolytic transformation of an aqueous precursor.

Nanomaterials Synthesis

As described in the BRN reports for "Synthesis Science" and "Quantum Materials" (mentioned earlier), accelerating the discovery of quantum materials with new approaches integrating emerging theoretical, computational, and *in situ* characterization tools to achieve directed synthesis with real-time adaptive control is a key BES priority. Atomically-thin, two-dimensional materials have emerged as a platform for the theme science at CNMS because they are computationally tractable and because each atom can be imaged individually. The explosion of interest in atomically-thin two-dimensional crystals worldwide has been addressed at CNMS through the development of several custom-built, in-house tools for the vapor transport growth of the most popular emerging new materials, especially the transition metal dichalcogenides (TMDs), including GaSe, $MoSe_2$, MoS_2, WSe_2, and WS_2 as well as doped variants, alloys, and heterostructures. CNMS is very closely collaborating with the program "Growth Mechanisms and Controlled Synthesis of Nanomaterials" within the Synthesis and Processing Sciences area of MSED, which has developed alternative synthesis strategies. This FWP addresses very fundamental aspects of understanding the formation of nanomaterials from their basic building blocks – an understanding that leads to capabilities to synthesis the materials needed for this work and for the users. These interactions are highly synergistic and allow each program (MSED and SUFD) to benefit from the understanding and instrumentation that is being developed separately.

Processing setups for stamping layers exfoliated from bulk crystals have also been developed including novel noble metal dichalcogenides such as pentagonal $PdSe_2$. Associated transfer, lithography, and methods have been developed to characterize, manipulate, and induce point defects or phase transformations by electron/ion-beam in these novel two-dimensional crystals.

In order to meet the needs of users for the rapid exploration of atomically-thin materials, non-equilibrium laser-based synthesis and processing of TMDs developed at CNMS in the past few years will be expanded and emphasized. PLD is especially well suited to the rapid exploration of atomically-thin layers. Established CNMS facilities for the PLD of novel metal oxides and heteroepitaxial superlattices will be expanded and modified to address the many new families of atomically-thin layers, such as chalcogenides and chlorides, that are envisioned for emerging quantum materials. This requires upgrading the PLD laboratory for reactive/toxic gas handling, integrating load-locked analysis chambers to rapidly characterize multiple samples for composition and thickness *in situ*, and new multi-target ablation capabilities for rapid exploration of non-equilibrium synthesis conditions with stoichiometry control.

Synthesis strategies that utilize the variety of pulsed laser sources and tunable wavelengths at CNMS will be developed for rapid exploration of materials with *in situ* optical diagnostics designed to provide rapid feedback of materials functionality and properties on samples compatible with the atomic-resolution electron microscopy capabilities at CNMS.

Optical Spectroscopy

A comprehensive combination of linear and nonlinear optical spectroscopic tools has been developed at CNMS, including high- and low-frequency Raman scattering, photoluminescence (PL) and PL lifetime measurements, micro-absorption, second harmonic generation, ultrafast pump-probe spectroscopy, and others to characterize low-dimensional materials. These capabilities have attracted many users, especially in the field of 2D materials where the optoelectronic properties of atomically-thin specimens evaluated at the sub-micron scale can be correlated with the atomic-resolution electron microscopy and STM characterization of defects, dopants, and stacking configurations. Bridging length scales to macroscale response, *operando* optical spectroscopic characterization techniques have been developed to evaluate the interface-directed assembly and functionality of ensembles of nanomaterials as part of a multimodal characterization suite. Optical spectroscopy is integrated with one or more property-measurement techniques (e.g., AC impedance, QCM-based adsorption, stiffness, photocatalysis) to evaluate the changing optoelectronic properties of nanomaterials correlated to their performance as sensors, catalysts, and other emerging applications of interest to users.

An important research direction is the emerging field of quantum materials, which requires exploration of a wide variety of time, length, and energy scales for their characterization. Currently, single quantum emitters including defect-trapped excitons in 2D-materials (e.g., in transition metal dichalcogenides and color centers in hBN and GaN) have provided tantalizing new frameworks for integrated quantum repeaters, quantum communication, and quantum sensing. However, improved understanding and control of the nanoscale emitter dynamics is critical to developing quantum information technologies.

Fully characterizing and controlling ultrafast quasiparticle interaction dynamics with individual color centers requires a high-speed, hyperspectral platform capable of describing interactions with sub-ps timing resolution, few nanometer spatial resolution, meV spectral resolution, and control over spin and orbital angular momentum at low temperatures. There is no commercially available tool capable of describing all these degrees of freedom simultaneously. To achieve high spatial and time resolution we are planning to combine commercial low temperature cathodoluminescence (CL) SEM microscopy with existing CNMS femtosecond lasers by pumping an SEM electron gun with fs-laser pulses to create fs-electron beams for CL. This will enable the complete characterization and understanding of coherent sub-ps dynamics in novel quantum emitter platforms.

5. User Needs and Outreach

Anticipating and Accommodating User Growth

As user facility, we strive to increase the number of users, their satisfaction, and the number of measurable outcomes such as publications, patents, etc. With the merger between CNMS and the former Shared Research Equipment (ShaRE) User Program in 2013, the total number of users for both user programs initially decreased, as a deliberate effort was made to align the former ShaRE resources with CNMS and nanoscience goals (eliminating some high-capacity microscopes and investing the resources into more specialized efforts). Despite the initial loss of “capacity” users, the combined program quickly re-established growth in user numbers and productivity: Over the time interval from FY2015 to FY2017, the user numbers grew at an annualized rate of ~7.5% to over 660 users in FY2017; the number of peer-reviewed scientific publications in regular journals increased at an annualized rate of ~10.5% to 435, while the operating budget increased at an annualized rate of 3.5% during the same time period (not adjusted for inflation). Under the same boundary conditions, such dramatic growth will be difficult to maintain in the long run as it is only possible through a concerted effort to provide users with the best possible support. Therefore, a significant effort in increasing efficiency in our user operation will be needed to maintain and grow our user numbers. Increased automation, streamlined training, workshops to provide users with the skills to best prepare for user visits and then perform the data analysis independently, and increased reliance on tools such as Jupyter notebooks (an open-source web application that allows researchers to create and share documents that contain live code, equations, visualizations and narrative text) all contribute to this effort.

At the same time, it is important to consider today what resources the users will need in the coming years. In consultation with the CNMS User Executive Committee, feedback received at the annual user meeting, and evaluation of our strategies with our Scientific Advisory Committee, we identified key areas in which we anticipate future growth. These areas include

- Quantum information systems and quantum computing are seen as key growth areas, as mentioned above, but there are also emerging opportunities in neuromorphic computing enabled by our work on soft matter, electro-ionic coupling, and membranes as artificial synapses.
- In energy *applications*, we will continue to bring fundamental science to applied projects, and support applied and industrial researchers with the relevant expertise and instrumentation.
- Biology, bio-medical research, and bio-imaging are areas where CNMS currently plays a somewhat limited role, supporting the community through our bio-affiliate laboratories operated jointly with the Biosciences Division at ORNL. Particular areas of continued and increased relevance include plant-on-chip and organisms-on-chip approaches, where nanofabrication capabilities enable specific biological studies, and bio-imaging, where our expertise in mass spectrometry-based imaging (MALDI-TOF-SIMS, TD-MS AFM, and HIM-SIMS) all are expected to significantly contribute.
- Data analytics and AI approaches, as discussed above, are expected to become increasingly important in all areas of nanoscience. We anticipate that a user community will grow to take advantage specifically of the tools and methods developed at CNMS in this rapidly evolving arena.

CNMS also continues to improve its outreach to neutron scattering users and seeks to develop greater visibility within that growing community. For several years, CNMS has offered its users an opportunity to request neutron scattering time as part of a CNMS user proposal; more recently, an equivalent mechanism has been implemented to provide streamlined CNMS access to the neutron user community as part of the neutron proposals. CNMS thus continues to support neutron scattering user projects either through its Rapid Access proposal process or as regular proposals, in both cases accepting the peer reviews from the neutron scattering proposals. Extended outreach in this area also includes a SNS/CNMS effort via a Polymer Focus Group that regularly travels ("a neutron and nanoscience road show") to different institutions to present overviews targeting formation of direct connections/collaborations.

Finally, we need to point out that despite the significantly increased demands on CNMS staff as a consequence of a user number that is growing faster than our budget, our user satisfaction, as measured through its annual user satisfaction survey, remains very high (with only around 1% of respondents to the CNMS user survey indicating either dissatisfied or very dissatisfied). We are confident that the alignment around key future areas, in combination with a vigorous in-house research effort and a highly motivated and skilled workforce, will enable us to continue to serve a highly satisfied user community.

User outreach

CNMS can only fulfill its mission as national and international user facility if researchers from across the world and from different types of institutions are aware of the benefits of CNMS to their research. While the CNMS website (cnms.ornl.gov) has recently been revised to present information about the user effort in a more accessible way, and additional improvements are being implemented in FY2018 (more detailed descriptions of the various capabilities), and while the capabilities of all NSRCs are compared and searchable on the NSRC-common portal website (nsrcportal.sandia.gov), it is clear that a web presence alone is not sufficient for informing a broader community.

The most significant and effective user outreach is through peer-to-peer communication between scientists. Key to such communication is a community of users who refer to work performed at CNMS (especially in conference talks and in scientific publications), and a steady flow of high-visibility scientific publications, press releases, DOE Web Highlights [science.energy.gov/bes/highlights/], etc. In this context, all CNMS staff who present at conferences are required to indicate on the title slide of their presentation that CNMS is a scientific user facility, where all resources are available free of charge to users who intend to publish their results.

CNMS will continue to partner with the other NSRCs to hold specific events (such as the upcoming 1-day Presidential Symposium at the ACS conference, featuring NSRC user work in the area of ionic transport) and feature booths at related exhibitions. Such booths, staffed by NSRC researchers who attend the conference, provide a forum to discuss the NSRC model with other conference attendees who may not be familiar with these facilities.

The user meeting, organized annually by the UEC and at times held jointly with the SNS/HFIR user meeting, fosters a sense of community between the users, enables neutron users to learn about CNMS capabilities, and includes workshops that educate the community about CNMS capabilities. In addition, CNMS is actively reaching out to various institutions through targeted visits. Examples include events such as a mini-workshop at Georgia Tech (2017) focusing on chemical imaging, travel to universities in conjunction with ORNL-wide partnering efforts (e.g., Virginia Tech, April 2018), and a series of university visits organized jointly with SNS/HFIR targeting specifically the soft-matter and polymer community. Active engagement of CNMS staff in DOE efforts (such as the above-mentioned Roundtable on Next-Generation Quantum Systems, co-chaired by the CNMS Director and including a CNMS staff as co-author of the report), participation on panel reviews, conference and conference symposium organization, etc., also play a role in raising awareness of the CNMS capabilities.

Finally, CNMS is host to numerous tours of visitors coming to ORNL. Many of these tours target delegations from other universities, trade organizations, research teams, etc. While at times a somewhat disruptive and time-consuming activity, we see such visits as an opportunity to increase broad awareness of CNMS across multiple organizations.

Industry interactions

CNMS has been successful engaging with industry through multiple pathways. While the number of industrial researchers is small (typically below 5% of all on-site users), the industry interactions take many forms. For example, CNMS hosts many academic researchers who are funded by industry to perform their work (e.g., a student funded by a company performing work as CNMS user). Even CNMS staff on many occasions are funded by industrial partners and then perform this work as users at CNMS).

CNMS researchers continuously develop licensable intellectual property, as evidenced by 19 patents issued in FY2017 and several fully licensed technologies. The development of new intellectual property is consistent with the CNMS's position as a world-class research institution.

CNMS also has several industry sponsored research projects and industrial collaborations. A few examples include:

- Proctor & Gamble (P&G) has worked with CNMS for 7 years on a range of projects, starting from developing computational protocols and new methods to evaluate complex product formulations, to how specific formulations wet and interact with surfaces to how they degrade over time, to optimization of polymer bottles that house the formulations. These fundamental research projects capitalized on unique CNMS capabilities and staff as well as integrating to SNS/HFIR and HPC.

- HRL Laboratories and CNMS have worked on understanding the factors which control the atomic-scale interface between silicon and silicon/germanium layers. This fundamental research project capitalized on unique CNMS capabilities and staff and has a direct impact on an applied problem: improving electronic device manufacturing.

- Lockheed Martin Space Systems and CNMS developed a deeper understanding of the mechanism of graphene ion milling using unique CNMS resources. The project was fundamental in nature – the basic research involved CNMS instrumentation and staff that resulted in highly publishable results; those published results also improved the understanding of proprietary manufacturing processes for the user.

Despite the successes, CNMS leadership is determined to improve the accessibility of CNMS resources for industrial users because industrial recruiting has been and remains a challenge. Industrial users often have different goals and timelines for their research than academic users, and the CNMS review process historically did not accommodate those differences. The CNMS is developing strategies to include more industrial scientists in the reviewer pool, and the CNMS Outreach Coordinator (and other CNMS staff) work with potential industrial users to help them to better understand and navigate the review process.

Potential industrial users often perceive a conflict between the need to maintain trade secrets and the need to publish results for non-cost recovered (e.g. free) access to government user facilities. The numerous examples of successful interactions with industry, both at CNMS and other DOE user facilities, indicate that these concerns can be addressed, but a bigger effort needs to be made to educate industry researchers on these topics.

To address these issues, CNMS is reemphasizing outreach strategies for industrial users. The message that the need for publication of results is not detrimental for industrial research is central for in-person visits and meetings, for example at recruitment booths at industry-heavy conferences (e.g., TechConnect). ORNL has robust IP protection mechanisms including Materials Transfer Agreements and Non-Disclosure Agreements, and CNMS management is working to ensure that CNMS researchers are aware of the mechanisms and requirements for information protection. CNMS will be deploying a webpage specific for industrial users, to spell out in clear terms the benefits and obligations of a user agreement, how new IP is handled, how information is protected, and what information is appropriate for publication. We believe that the key to improving industrial outreach is clear communication to industrial users to mitigate concerns specific to them.

Finally, CNMS has set aside travel funds for researchers to specifically target industry interactions, both at specialized conferences and through site visits.

Unique strengths for Specific User Areas

In addition to the strategic areas discussed in Sect. 4 above, the broad range of capabilities and resources at CNMS attracts users from many areas of research that are not (or not any more) at the heart of our research activities. A final version of this Strategic Plan will include examples of such activities as sidebars. Particularly worth mentioning are breakthroughs in catalysis (both experimentally and computationally), the characterization of semiconductor devices, user efforts investigating membranes, for example for desalination, a large effort to support the metallurgy and radiation-effects communities, and numerous examples of users from biology.

6. Instrumentation and Space Utilization

The previous sections, especially the description of strategic enhancements to the user and in-house research environment, clearly indicate a need for continued investments in future instrumentation. This includes in particular *major investments in small-angle x-ray scattering, lithography, and dedicated APT sample preparation facilities (focused ion beam(FIB) instrument).*

Additional and significant investments are needed to maintain the existing suite of instrumentation and continuously replace equipment when maintenance becomes impossible or the instruments become obsolete, to provide users at a minimum with current-generation capabilities. These include the purchase of computing resources (managed through the Oak Ridge Compute and Data Environment for Science (CADES)), the numerous highly used pieces of equipment for characterization and fabrication, and routine laboratory equipment. As mandated by SUFD since 2015, but implemented at CNMS already before, we continue to invest approximately 10% of our total funds into recapitalization.

With the cost of liquid helium and the reliability of supply being significant issues, the purchase of helium recovery units is being considered. In particular, a free-standing unit to supply, for example, the SQUID magnetometer has been evaluated as an alternative to a more expensive building installation that would recover helium from all instruments, including the nuclear magnetic resonance instrument. Recovering helium has become much easier because modern re-liquefiers require very little maintenance and staff attention (unlike when CNMS first began operation). Possible use of Laboratory overhead funds for a site-wide He-recovery system is also being considered.

Given the recent acquisition of the JEOL NEOARM TEM/STEM, CNMS will no longer provide access to the Zeiss Libra TEM nor the previously shared Hitachi HF3300 TEM/STEM. This will result in quiet space being made available for the new instrument, and in reduced maintenance and hourly costs.

Finally, access to instruments provided at ORNL through the cost-recovery method of the Materials Characterization Core (MCC) service center has become a unique opportunity for CNMS to save up-front equipment costs, in exchange for providing staffing and expertise to operate high-end equipment that is also used by other programs at ORNL. These instruments, i.e., the MAC-STEM, the TOF-SIMS, the Hitachi NB-5000 FIB for STEM sample preparation, and the low-temperature 4-probe SPM, are available to users in projects that directly involve CNMS staff, and are critical for the development of new areas, including those in QIS, soft matter, and MS-based chemical imaging.

7. Staffing Plans and Career Development

Meeting CNMS goals requires that the facility employ and retain outstanding scientists, engineers, and support/operations staff. Researchers hired into CNMS become part of a highly collaborative and productive research environment with outstanding facilities and the opportunity to focus on important research areas. They also interact directly with leading research groups through the CNMS user program and are encouraged to participate in major national and international conferences. It is therefore not surprising that staff members from CNMS at all levels are being recruited to other institutions, and that CNMS needs to make a constant effort to retain and to attract the leading researchers needed to operate an attractive user facility.

CNMS strives to assist with the career development of research staff into new or expanding areas of nanoscience that directly impact our mission and have high potential to engage an expanding user community. Part of the future success of CNMS will be linked to expanding opportunities for outstanding researchers within the program. CNMS also strongly encourages and fosters its researchers to take advantage of LDRD and other opportunities to develop their own ideas if they are aligned with the mission of the CNMS but outside the scope of the peer-reviewed theme science. Such venues encourage creativity and contribute to staff retention and at the same time provide a testing ground for novel ideas that may eventually be rolled up into the user program or the theme science if they prove successful. To increase the rate of success, CNMS fosters a team environment where individual researchers' proposals are being thoroughly and constructively evaluated by peers and line management.

CNMS also makes a strong effort to nominate members for awards from societies, leading to several Fellow awards from major societies (APS, MRS, AVS, AAAS, etc.) every year. CNMS researchers are often recipients of UT-Battelle Awards, and CNMS has its own internal annual awards (distinguished paper, most notable user project, distinguished patent, outstanding technical contribution, postdoctoral award). To encourage visibility of CNMS researchers within ORNL, staff members are actively encouraged to serve on ORNL committees, which often provides a very valuable career development opportunity. Similarly, CNMS has promoted early and mid-career staff to Theme Leader positions.

As CNMS has transitioned to an environment where only a limited number of postdoctoral fellows can be funded through SUFD monies, the roles of technicians and technical professionals are being re-evaluated and strengthened. Over time, this will lead to a research environment with enhanced and streamlined user support and increased institutional memory (i.e., less susceptible to information loss with the departure of postdocs).

In parallel, however, CNMS is strengthening the integration of students into our research efforts, with the goal of reaching a better balance between students, postdocs, technical professionals, and research scientists. CNMS has been a particularly active participant in two graduate research and education programs at ORNL: 1) The Bredesen Center for Interdisciplinary Research and Graduate Education in collaboration with the University of Tennessee, Knoxville; and 2) The GO! Graduate program in collaboration with the UT-Battelle core universities. In these two programs, CNMS has served as the research home for nearly 30 Ph.D. candidates in a variety of disciplines associated with nanoscience. The four that have graduated as of today all immediately found jobs: one as a postdoc, three in permanent positions (Whirlpool, KPMG Data Analytics, Jet Propulsion Laboratory). Eighteen CNMS staff members are serving as either major professors or research mentors for these students. The net result has been enhanced educational opportunities for the students and professional growth for the CNMS staff. Supervision of postdocs and students is a key element in the career development of researchers, allowing staff at all levels to develop and test their management and organizational skills. CNMS will simultaneously take advantage of a re-structured internal training program at ORNL that emphasizes the “soft skills” (communication, conflict resolution, delegation, etc.) that graduate school and postdoctoral experience often fail to adequately address.

8. Outlook

Nanoscience has seen tremendous achievements in the last decade, with fascinating advances in the creation, manipulation, and observation of nanostructures and nanomaterials now becoming broadly integrated into numerous areas of research. We are approaching the point where researchers can control the formation of nanomaterials at the atomic scale, observe the behavior of each individual atom that constitute a nanostructure, and use theoretical approaches and computational tools that capture the behaviors of exactly those structures that are observed experimentally. These advances now allow us to tackle the significant challenges of understanding the pathways by which such structures are formed and modified by localized stimuli, how matter can be programmed to arrange itself into desirable, hierarchical assemblies, how the coupling of electronic and ionic phenomena leads to novel functionalities, and how individual defects and heterogeneities affect the quantum behavior of nanostructures.

These challenges, collectively addressed in the in-house research effort of CNMS, are fundamentally interdisciplinary, and require approaches that combine elements of AI, deep data analytics, close integration of TM&S, and the most advanced tools for imaging, functional measurements, and spectroscopy. CNMS will play a key role in defining the approaches and methods that will be used by the next generation of scientists, and in making them available to a broad spectrum of users.

The most important resource that we have to offer our users are our researchers who lead the development of these approaches. As user facility, we strive to go far beyond making equipment available for use – our main mission is to provide a comprehensive research environment and solutions to complex problems. The users who come to CNMS are leaders in their fields. Many of them help define and develop our approaches. Others work on areas that are complementary to our strengths, and the interactions between users and staff lead to a vibrant environment that encourages and enables the exploration of new ideas.

With an ever-increasing urgency to provide solutions to energy sustainability and security, with new opportunities related to QIS, quantum materials, and neuromorphic computing, and with key strengths and leadership in soft matter, neutron nanoscience, and imaging at multiple length scales, this is an exciting time for CNMS. We will continue to invest into our equipment and our people, and we will continue to grow as a resource for the national and international user community.

9. List of Abbreviations

2D	Two-Dimensional
AC	alternative current
AFM	Atomic Force Microscopy
AI	Artificial Intelligence
ALCC	ASCR Leadership Computing Challenge
APT	Atom Probe Tomography
ASCR	Advanced Scientific Computing Research
BASIS	Backscattering Spectrometer (SNS)
BER	Biological and Environmental Research (DOE)
BES	Basic Energy Sciences
BESAC	Basic Energy Sciences Advisory Committee
BRN	Basic Research Needs (Workshop)
CADES	Oak Ridge Compute and Data Environment for Science
CFN	Center for Functional Nanomaterials
CINT	Center for Integrated Nanotechnologies
CL	Cathodoluminescence
CNM	Center for Nanoscale Materials
CNMS	Center for Nanophase Materials Sciences
CSGB	Chemical Sciences, Geosciences, and Biosciences (BES)
DOE	Department of Energy
DSC	Differential Scanning Calorimetry
EBID	Electron Beam Induced Deposition
EDS	Energy Dispersive X-ray Spectroscopy
EELS	Electron Energy Loss Spectroscopy
EERE	Energy Efficiency and Renewable Energy (DOE)
ESH	Environmental, Safety, and Health
ESH&Q	Environmental, Safety, Health, and Quality
FTIR	Fourier-Transform Infrared Spectroscopy
GPC	Gel Permeation Chromatography
HFIR	High Flux Isotope Reactor
HIM	Helium Ion Microscopy
IFIM	Institute for Functional Imaging of Materials
INCITE	Innovative and Novel Computational Impact on Theory and Experiment
LDRD	Laboratory Directed Research and Development
MAC-STEM	Monochromated, Aberration-Corrected STEM
MALDI	Matrix Assisted Laser Desorption/Ionization
MCC	Materials Characterization Core
ML	Machine Learning
MPMS	Materials Properties Measurement System (QuantumDesign)
MS	Mass Spectrometry
MSED	Materials Sciences and Engineering Division (BES)
NERSC	National Energy Research Scientific Computer enter
NMR	Nuclear Magnetic Resonance
NOMAD	Nanoscale-Ordered Materials Diffractometer (SNS)
NSRC	Nanoscale Science Research Center
NTI	Nanomaterials Theory Institute
OLCF	Oak Ridge Leadership Computing Facility
ORNL	Oak Ridge National Laboratory
PL	Photoluminescence
PLD	Pulsed Laser Deposition
POWGEN	Power Diffractometer (SNS)
QCM	Quartz Crystal Microbalance
QIS	Quantum Information Science
SBMS	Standards Based Management System
SCEM	Scanning Electrochemical Microscopy
SEM	Scanning Electron Microscopy
ShaRE	Shared Research Equipment User Program
SIMS	Secondary Ion Mass Spectrometry
SNS	Spallation Neutron Source
SPM	Scanning Probe Microscopy
SQUID	Superconducting Quantum Interference Device
STEM	Scanning Transmission Electron Microscopy
STM	Scanning Tunneling Microscopy
SUFD	Scientific User Facilities Division (BES)
TD-MS	Thermal-Desorption Mass Spectrometry
TEM	Transmission Electron Microscopy
TGA	ThermoGravimetric Analysis
TGIC	Temperature Gradient Interaction Chromatography
TM&S	Theory, Modeling, and Simulation
TMD	Transition Metal Dichalcogenide
TOF-SIMS	Time Of Flight - Secondary Ion Mass Spectrometry
UEC	User Executive Committee
VULCAN	Engineering Materials Diffractometer (SNS)
XRD	X-ray diffraction