Center for Nanophase Materials Sciences Strategic Plan 2021-2025

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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 U.S. Department of Energy (DOE) Office of Science contribution to the U.S. 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, staff expertise, and state-of-the-art instrumentation, 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 synthesizing, characterizing, and understanding nanomaterials. CNMS users include experienced research professionals as well as postdocs and students that come 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 focused on *understanding the effects of temporal and spatial confinement on the 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.* The CNMS 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, and simulation that will benefit future users. As CNMS and the broad research community continue to advance new research, novel capabilities must continually be created to address new challenges and exploit new opportunities. CNMS leadership, working with its User Executive Committee (UEC) and Science Advisory Committee (SAC), continues to identify and advance capabilities to serve the user community as well as its research themes to ensure CNMS continues to be a world-leading facility that is accessible by researchers so that they are able to perform high-impact nanoscience, not just today, but for future generations of researchers. CNMS' research efforts are organized into four research themes that catalyze new nanoscience capabilities and knowledge for energy generation, quantum information science, novel polymer synthesis, and nanoscale manipulation of matter, all of which create the integrated environment needed for CNMS to be a forefront nanoscience user facility where staff expertise and state-of-the-art facilities enable world-leading 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 innovative opportunities for 3D direct-write nanofabrication, positioning of individual dopant atoms, and integrating next-generation materials into functional architectures; precise synthesis of macromolecular materials and selective deuteration; all in combination with a comprehensive suite of functional characterization and a fully integrated theory, modeling, and simulation effort that explicitly underpins nearly all the work.

This Strategic Plan outlines the research agenda that will drive the scientific progress of CNMS over the next 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 close integration of high-performance computing, data analytics, and artificial intelligence. The center emphasizes energy related materials topics and delivers new advances in soft matter, two-dimensional (2D) materials, and multiscale integration of nanomaterials. In addition, CNMS specifically seeks to address and integrate emerging opportunities in quantum information science, artificial intelligence and machine learning, and those at the interface between biology and materials research.

CNMS contributes to and benefits from robust partnerships with ORNL signature strengths in multiple strategic areas and takes advantage of the distinctive capabilities of other DOE user facilities located at ORNL, including the Oak Ridge Leadership Computing Facility (OLCF), the Spallation Neutron Source (SNS), and the High Flux Isotope Reactor (HFIR). For example, CNMS maintains a strong link to *neutron sciences* through research efforts that tie nanoscience with neutron capabilities, thereby providing a direct platform for the broader research community to integrate neutron studies into nanoscience efforts. This is exemplified by CNMS' strong research efforts and knowledge in polymer synthesis and site-specific deuteration. CNMS also uses its expertise in materials, physics, chemistry, and computational sciences to incorporate and develop predictive materials sciences approaches. In this way, CNMS acts as a gateway for *neutron sciences* by providing an environment for researchers to integrate neutron studies into nanoscience efforts and for *computational nanoscience*, enabling the community to take advantage of CNMS expertise and ORNL's powerful computational resources.

At CNMS, our cutting-edge in-house theme science program and our vibrant user science portfolio work hand-in-hand to provide a unique environment that enables the science community to predict, design, produce, and characterize the (nano)materials needed to address the most important issues facing our nation. With our strong connections 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 across a broad spectrum of disciplines 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 strong composites to hydrophobic surfaces, and many more. 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 with, and complemented by, the ability to integrate these elements into complex material systems, 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 of integrating prediction, formation, and characterization will lead to the application 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 CNMS operates as a user facility that allocates resources to users solely based on the scientific merit of the proposed research as determined by external peer review.

CNMS is one of five Nanoscale Science Research Centers (NSRCs)[1] established as part of the U.S. Department of Energy (DOE) Office of Science (SC) contribution to the U.S. government's National Nanotechnology Initiative (NNI) 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 to benefit society." CNMS is located at Oak Ridge National Laboratory (ORNL), which is home to the nation's largest materials science program, the Spallation Neutron Source (SNS), the High Flux Isotope Reactor (HFIR), and the Oak Ridge Leadership Computing Facility (OLCF). This co-location of facilities allows CNMS users to benefit from ORNL's expertise in physical and computational sciences and provides a connection between researchers in nanoscience and neutron sciences. As such, CNMS is not only at the forefront in the context of the multi-disciplinary nature of nanoscience research but is also positioned to address the critical importance of providing shared resources. As a user facility, it is CNMS's primary goal to work

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

with the user community by providing world-leading capabilities and facilitating the advancement of nanoscience.

Various Basic Energy Sciences (BES) documents highlight the importance of nanoscience across a broad range of scientific fields. The report from the Basic Energy Sciences Advisory Committee (BESAC) "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.

Hierarchical architectures (Opportunity #1) and heterogeneities, interfaces, and disorder (Opportunity #2) are inherently nanoscience topics, and Opportunity #5 underpins our work in imaging by employing 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 (Opportunity #3).

BES Basic Research Needs (BRN) Workshops on Quantum Materials (February 2016), Synthesis Science for Energy Relevant Technology (May 2016), Innovation and Discovery of Transformative Experimental Tools (June 2016), Next Generation Electrical Energy Storage (March 2017), Catalysis Science (May 2017), Chemical Upcycling of Polymers (April 2019), Producing and Managing Large Scientific Data with Artificial Intelligence and Machine Learning (October 2019), and Transformative Manufacturing (March 2020) all include discussions of nanoscience topics in energy related areas. Additionally, areas related to information technology were addressed by other Roundtable discussions: Neuromorphic Computing (October 2015), Opportunities for Quantum Computing in Chemical and Materials Sciences, and Opportunities for Basic Research for Next-generation Quantum Systems (October/November 2017), which provide strategic guidance to CNMS for 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 when the cost of scientific equipment is increasing more rapidly than funding opportunities in many areas. Making commercial equipment available to a broader audience is a minor mission of CNMS; however, 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 and leadership opportunities. Our in-house research effort, organized in four scientific themes as described below, is the primary vehicle for pursuing scientific activities. CNMS researchers can also actively participate in research projects funded by other agencies to

which they submit proposals and often participate in large, multi-institution teams (e.g., Energy Frontier Research Centers or Energy Materials Network Consortia). ORNL-internal funding opportunities to develop new concepts, namely Seed projects (typically one-year efforts with a proof-of-concept mission) and the Laboratory Directed Research and Development (LDRD) program, continue to play an important role in maintaining scientific diversity, providing leadership opportunities to early- and mid-career researchers, and maintaining vibrant communities consisting of a balance of students, postdocs, and permanent staff.

As we describe below, modern materials science research increasingly relies 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 views such methods as integral to its core mission to contribute to the development of 21st century research approaches that take full advantage of tools that have 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 collaborate and coordinate activities, while at the same time critically evaluating each of the NSRCs in terms of well-defined metrics (primarily the 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 and faculty 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% of the users 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 users come to CNMS specifically for characterization, using the wide range of tools available including microscopy (electron, He-ion, scanning probe), atom probe tomography (APT), X-ray diffraction (XRD), optical spectroscopies, and other techniques. Collectively, CNMS staff and users publish more than 400 scientific articles in peer-reviewed journals per year, with more with more than one half of the publications appearing in journals with an impact factor > 5 and about one third of the publications in journals on the NSRC high-impact journal list. In addition, CNMS researchers co-authored 18 patents since FY2017. Reflecting the impact of CNMS research, we note that more than 48,000 papers have cited CNMS publications since its inception.

2. The CNMS Research Groups and Key Capabilities

CNMS is organized into nine research groups whose primary responsibility is to maintain and operate the research laboratories, oversee all the capabilities used for the user effort and the in-house research, and to provide staff with a productive and collaborative research environment with an emphasis on mentoring for 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 ORNL divisions to support users in specific areas; funding from the BES Scientific User Facilities Division (SUFD) is used to support these specialized researchers.

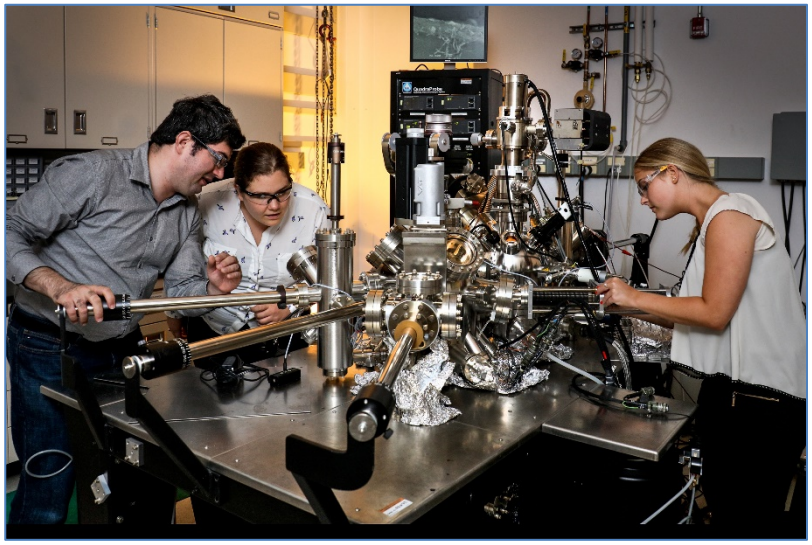
Conversely, as mentioned above, CNMS researchers can actively seek financial awards/support from other funding sources to maintain a vibrant and broadly diversified research environment. Funding from DOE-BES Materials Sciences and Engineering Division (MSED) and Chemical Sciences, Geoscience, and Biosciences (CSGB) Division, other DOE programs (Biological and Environmental Research (BER) and Energy Efficiency and Renewable Energy (EERE), for example), ORNL internal sources (Seed and LDRD), with industrial partners (through the ORNL Strategic Partnership Projects (SPP) Office), etc., collectively contribute to about 20-25% of all of the available research funds supporting CNMS staff research.

All researchers at CNMS are responsible for working with users and for performing in-house capability science within the framework of the DOE-reviewed CNMS program. 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 who do not have an obligation to support users typically choose to collaborate with some users in areas related to their expertise.

Staff at the CNMS discussing research and future directions in an open-format (Town Hall) setting.

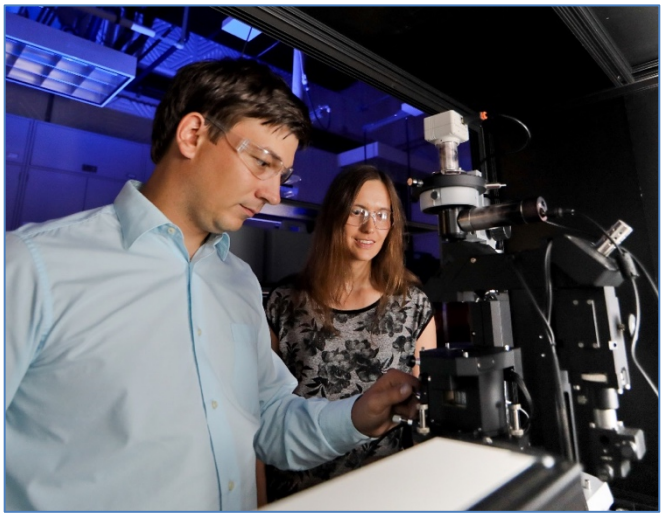
CNMS Research Groups

The following sections provide a brief description of each of the nine research groups within the CNMS.



Scanning Tunneling Microscopy (STM). The STM Group specializes in imaging and spectroscopy characterization of low-dimensional materials to establish structure-property relationships with atomic resolution. The group develops and applies both single-tip and multi-probe STM to map the local physical and electronic structure, charge and spin transport, quantum electronic behavior and quasiparticle state, and other fundamental behaviors of quantum and molecular systems including wires, surfaces, defects, films, and heterostructures. The group maintains seven variable or low-temperature STM systems in ultrahigh vacuum (UHV) including unique instruments designed for *in situ* studies of oxides, high magnetic field response, magnetic domains and spin textures, or with multiple probes for quantum transport and topological edge current measurements. Furthermore, the team is leading the development of atomically precise synthesis and manipulation via combining STM and on-surface chemical reactions for controlled synthesis and analysis of molecular nanostructures.

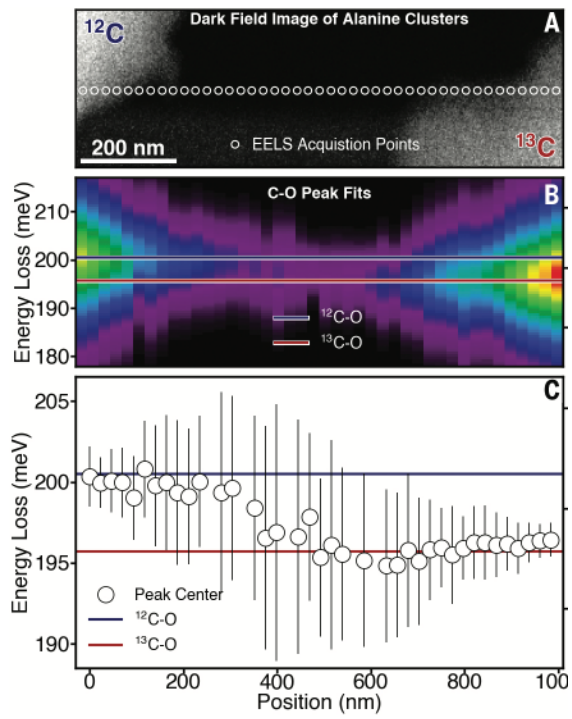
Functional Atomic Force Microscopy (F-AFM). The F-AFM Group is a leader in imaging a material's chemistry, functionality, and transformations at nanoscale dimensions to understand how properties and behaviors manifest at reduced length scales and dimensionality and how these translate to macroscale properties. The group develops and applies force feedback-based scanning probes to map the local physical and electronic structure, chemical, and molecular constituents, electronic and ionic transport, as well as bias, optically, and thermally driven phase transformations. The group maintains ten AFMs with a variety of capabilities ranging from advanced quantitative piezoresponse force microscopy (PFM) and spectroscopy, microwave microscopy, mechanical property mapping, time-resolved surface potential, and dielectric response in an array of controlled environments



including liquid, humidity control, and UHV and a suite of capabilities for chemical imaging including a time-of-flight secondary ion mass spectrometry (ToF-SIMS) system with AFM, an in-house developed ambient photo-thermal desorption AFM coupled to a mass spectrometer (AFM-MS), a NanoIR that enables nanoscale spectral imaging of material response to infrared (IR) optical excitation, and a coupled Raman-AFM system. Furthermore, the team is leading developments using AI and machine learning (ML) implemented on several edge-computation platforms including dedicated central processing units (CPUs), field-programmable gate arrays (FPGAs), and large-scale graphics processing unit (GPU) clusters for spectral-image analysis and for real-time control of reactions and transformations at the tip-surface junction.

Users are provided access to staff with considerable expertise in the many capabilities of these instruments including CNMS-patented imaging modes and advanced statistical analysis. This expertise, together with access to high quality microscopes, is a major attraction for many users.

Scanning Transmission Electron Microscopy (STEM) and **Materials MicroÅnalysis (MMÅ)** are two newly formed CNMS groups that specialize in the development and application of advanced analytical STEM and atom probe tomography (APT) techniques toward understanding and discovery of materials properties at the nanometer and atomic scales. Both microscopy groups complement each other and focus on maintaining an extensive array of specialized instruments (six aberration-corrected STEMs) to enable new science across a broad range of nanomaterials.

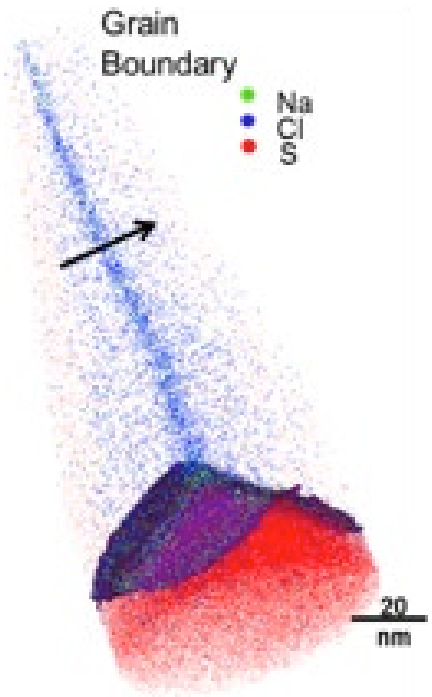


(A) Dark-field image of ^{12}C (left) and ^{13}C (right) Alanine clusters. EELS acquired along line profile between clusters (circles). (B) Fits of magnitude and position of C–O EELS peak, plotted for each point in line profile in (A). (C) Peak center and standard deviation for fits shown in (B), demonstrating high–spatial resolution identification of isotopic species.

The STEM Group primarily focuses on the direct development of novel electron spectroscopy and imaging techniques for the study of materials at the nanometer and atomic scale. In particular, the group aims to develop and exploit 4D STEM imaging, monochromation, electron energy loss spectroscopy (EELS), Cryo-EM, and is currently exploring imaging under variable-magnetic-field conditions that involves fully remote instrument operation, the development of scriptable automated procedures, and advanced data analysis using open-source python-based modules. For examples, the STEM Group aims to understand the electron optical conditions to measure temperature anharmonicity, phonon population, and even thermal transport, thermal conductivity, and heat capacity of materials at the nanoscale. Similarly, the group aims to extract charge density inhomogeneities of materials via 4D STEM atomic-scale imaging, as well as the

identification of magnetic properties of materials via spectroscopy and 4D STEM and the measurement of ion transport, phonon and plasmon dispersions, the detection of excitons, Moire potentials, ferroelectric properties of materials, and the detection of isotopic species in materials.

The MMÅ Group specializes in the development and utilization of state-of-the-art electron and atom probe microscopy methods to probe a material's nanoscale structure and chemistry through atom probe tomography (APT), atomic resolution imaging, analytical microscopy (EELS and EDS), electron diffraction, *in situ/operando* microscopy, cryo-EM, and advanced data analytics correlate function and structure with materials properties. The group develops novel methods for advanced and automated data processing of EM and APT data sets and quantitative microscopy through image analysis coupled with simulations. The group also focused on *in situ/operando* microscopy using unique *in situ* holder platforms for insight into materials behavior with full environmental control in the solid, liquid, or gas phase and with controlled electrochemical, electrical bias, and thermal exposure. Materials transformations, reaction mechanisms, and kinetics are quantitatively analyzed with time-resolved imaging/spectroscopy/diffraction data acquired with high-speed cameras and detectors.

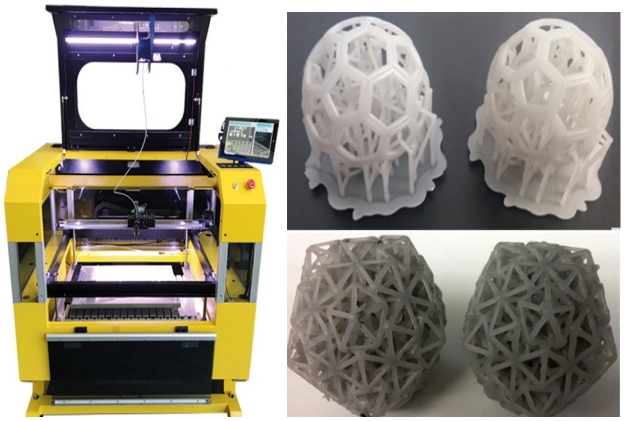


APT reconstruction of CdTe/CdS interface within CdTe-based solar cell. Na, Cl, and S atoms are green, blue, and red dots, respectively, along with a magenta S isoconcentration surface (22 at.%) to highlight CdTe/CdS interface.

The STEM and MMÅ groups maintain laboratories/facilities located on ORNL's main/central campus, including the Advanced Microscopy Laboratory (AML), a dedicated, low-noise facility for performing high-resolution STEM imaging and 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 the critical understanding of the role of Å-level structures (e.g., defects, interfaces, junctions, phase interactions, etc.), chemistry, and physical properties that provide knowledge of materials behavior, functionality, and degradation. Staff develop the 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 STEM and MMÅ groups are 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. Both groups work in the implementation of the developed techniques to the daily workflow of the CNMS' User Program.

Macromolecular Nanomaterials (Macro). The Macro Group specializes in the synthesis and characterization of well-defined polymers and molecular building blocks with various architectures by employing 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 site-specific isotope-labeling (especially deuterium labeling) of soft matter is crucial to neutron scattering studies of structure and dynamics. The importance of structure-composition-properties is emphasized as it relates to designing new macromolecules: from understanding fundamental phenomena related to chain dynamics and molecular conformations to applications as that require unique bulk material properties and response. The group maintains five synthesis laboratories equipped with 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), light scattering spectroscopies (dynamic and static), Fourier-transform infrared (FTIR) spectroscopy, 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).

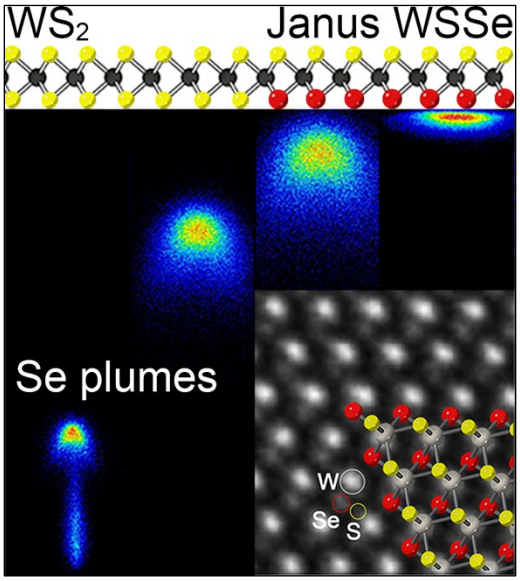


(Left) Hyrel Hydra 16A 3D printer recently acquired by CNMS. (Right) Images of 3D printed Silicone-Acrylate blend forming bro-icosahedron and Bucky ball shapes.

Laboratories supported by the Macro Group provide precise synthesis and rigorous characterization of polymers and the molecular building blocks that are essential to address the scientific questions in 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 neutron studies, which accounts for about half of the user proposals for synthesis. In addition, the newly established 3D printing facility will enable macromolecular materials studies under extrusion processing, photo-reactive conditions, and multi-materials fabrication methods. This includes fused deposition modeling (FDM), selective laser sintering (SLS), stereolithographic apparatus (SLA), and viscous solution printing (VSP). It is envisioned that fundamental phenomena studies and application properties from nanoscopic to macroscopic can be undertaken by the Macro Group and theme science (e.g., HCMC) within the unique environment of CNMS and its users.

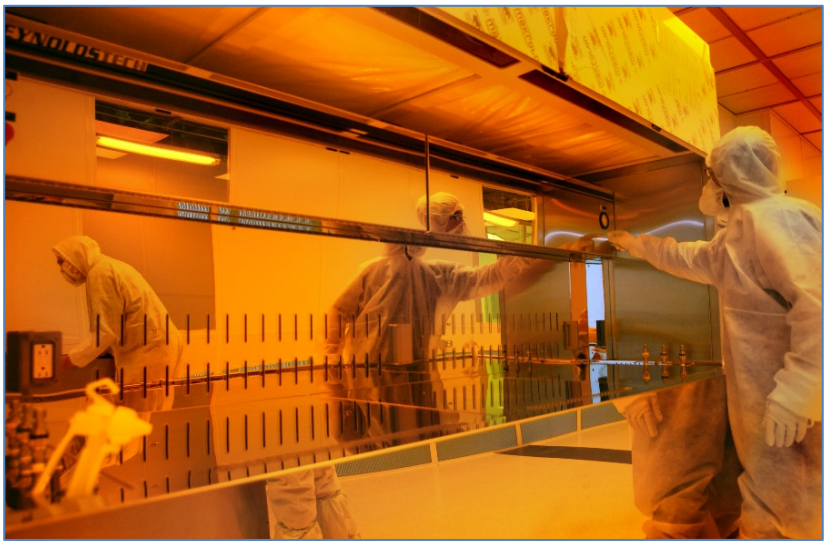
Functional Hybrid Nanomaterials (FHN). The FHN Group specializes in research involving the controlled synthesis of nanostructures and thin films, exploration of their fundamental optical and electronic properties, and use of these properties as *in situ* and *operando* diagnostics of evolving

structure and function. The group is recognized for using time-resolved, *in situ,* laser-based diagnostics to develop growth models for the controlled synthesis of nanomaterials, especially



carbon nanomaterials (e.g., nanotubes, nanohorns, and graphene), thin film oxide heterostructures, and more recently 2D metal chalcogenide crystals. The group also develops new spectroscopic techniques to explore the heterogeneity and photoresponsivity of nanomaterials in different environments, especially photocatalysis, photovoltaics, and multimodal sensing. The FHN Group maintains eleven laboratories at CNMS that are responsible for the synthesis of nanomaterials, their hybrid assembly in functional architectures, and the characterization of their electronic, electronic, catalytic, and quantum functionalities. High-power ns-pulsed lasers are employed in the growth of new thin films and nanostructures by pulsed laser deposition (PLD) using *in situ* spectroscopic and plasma diagnostics. Ultrafast (ps and fs) pulsed lasers are employed for tunable-Raman spectroscopy and ultrafast white-light pump-probe spectroscopy characterization of vibration modes and quasiparticle dynamics, respectively. The group maintains laboratories for conducting fundamental measurements of electronic, optoelectronic, catalytic, and quantum properties, as well as processing equipment for the assembly of organic, 2D materials, and hybrid perovskites for electronic and quantum devices. These laboratories contribute to CNMS theme science 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 works closely with the staff at the SNS to develop neutron reflectivity and other scattering techniques for thin film and bulk hybrid perovskites. To assess the functionality of nanomaterials, group members are developing multimodal quartz-crystal microbalance (QCM) platforms and ML approaches to measure and correlate structural, optical, and electronic property changes with interrelated environmental effects such as humidity, temperature, and gas exposure.

Nanofabrication Research Laboratory (NRL). NRL research addresses the properties of nanomaterials through their integration into systems with precisely defined functionality across multiple length scales. These are made to enable observation and measurement of properties for both individual nanoscale elements as well material ensembles under controlled conditions, thereby providing a practical link between nanomaterials and a macroscopic world. The NRL Group specializes in sculpting, connecting, and integrating a breadth of materials synthesized across the CNMS into practical, functional solutions targeting a variety of applications.

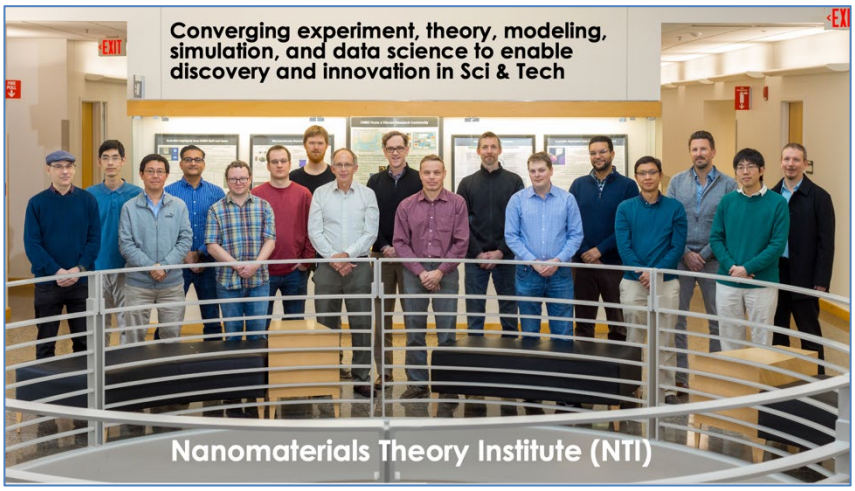


The NRL Group is focused, in part, on the development of direct-write nanofabrication processes across nano, micro, and millimeter scales. In conjunction with experiment, these efforts incorporate a variety of characterization, theory, and computational approaches. Specifically, the NRL wants to target the science behind the dynamic processes that are relevant in nanoscale (and beyond) direct-write technologies, as well as address the practical side of on-demand component creation for micro- and nano-fluidic systems. The interest in nanoscale direct-write is driven by applications that require precise control of nanoscale features – both physically and chemically – on a wide variety of surface topographies and materials. Micro- and nano-fluidics work focuses on the creation of interfaces to soft materials and polymer assemblies that respond and evolve to external stimuli in crowded and confined spaces. These efforts integrate a variety of fabrication, characterization, analytical, and computational approaches.

The group maintains a 10,000 sq. ft. class 100/1000 cleanroom facility that houses a comprehensive suite of micro- and nanofabrication capabilities; optical, electron, ion, and scanning probe characterization tools; as well as an affiliated nanoscale bioscience laboratory for the characterization of wet and soft biological materials. These laboratories contribute to the CNMS theme research by allowing controlled fabrication, synthesis, and characterization of a wide variety of materials over length scales that vary from nanometers to centimeters.

Nanomaterials Theory Institute (NTI). CNMS incorporates a substantial TM&S effort through the NTI. 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 and ML-based approaches. 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, all of which are integral to group, theme, and user science efforts.

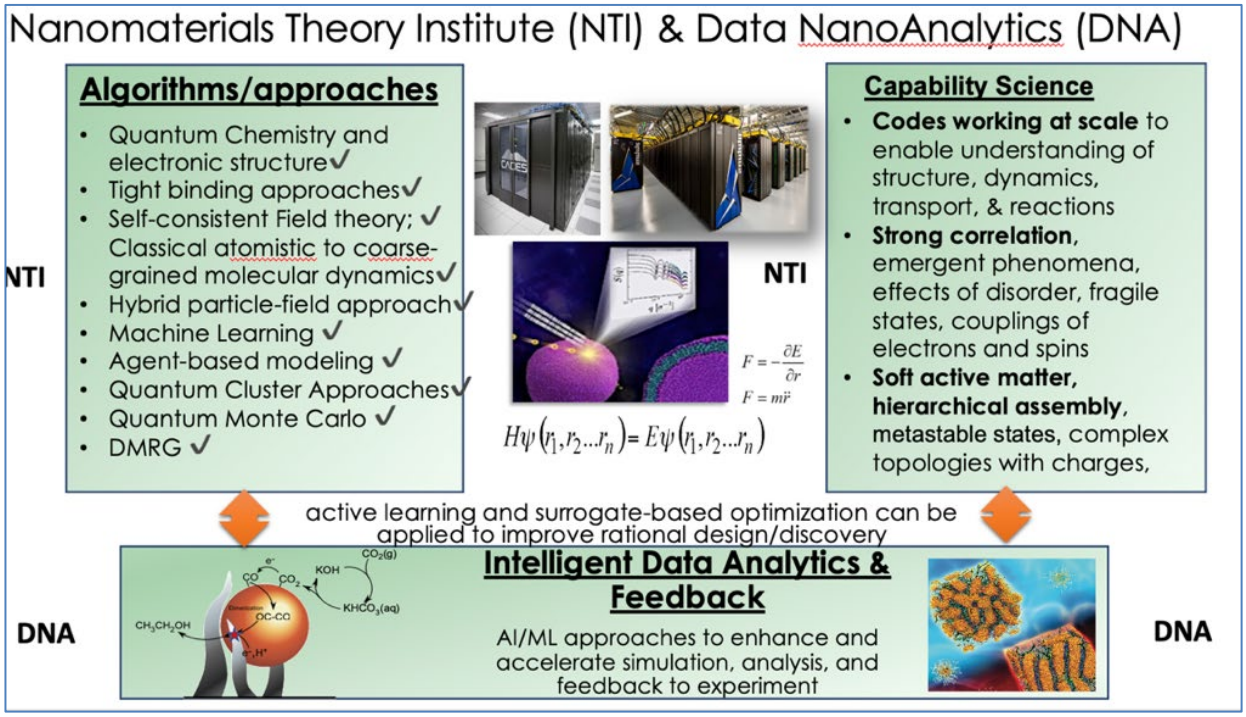
This work also includes modeling for the interpretation of neutron scattering experiments and hence provides a cross-synergy with the needs of SNS/HFIR in this domain.



NTI capabilities, research, and development are key components for all areas of the CNMS theme and user science. The NTI also maintains allocations of computational time for its staff and CNMS users at the OLCF facilities and the National Energy Research Scientific Computer Center (NERSC) through respective competitive proposal processes (INCITE, ALCC, DD), alongside a dedicated CNMS in-house computational resource (a mid-sized computer cluster, software stack, and data environment). The NTI works toward (see figure below):

1. *Generating reliable predictions from multiple levels of theory and experimental matching*: Predictions (with errors!) for structure, dynamics, transport, and other physical and chemical properties using different levels of TM&S, as well as computing a wide range of experimental observables measured at CNMS/SNS.
2. *Forefront nanoscience through high-performance computing:* Fundamental understating of correlated electron materials, non-equilibrium and metastability in nanomaterials, and active/adaptive processes.
3. *Using AI/ML to 'stitch' different levels of theory to achieve efficient multi-scale modeling*: ML/AI-steered model development to predict structure, properties, transport ,and dynamics across length/time-scales from pertinent data (via high-throughput computing) to understand hidden-variables that lead to material functionality at the mesoscale.

4. *Performing theory-experiment matching to discover 'new'/'missing' physics (in collaboration with DNA)*: iterative feedback between theory and experiment and matching in the latent-space (requires developing invertible representations in collaboration with the broader ORNL-AI community) to identify missing/new physics. We recognize this to be a Grand Challenge problem that will take beyond 'a few years' but we are working towards overcoming the various technical challenges.



NTI and DNA Groups: intimate integration between nanoscience computational codes/development and forefront science.

Data NanoAnalytics (DNA): A recent re-organization at CNMS established the DNA Group that strives to develop the bridges between the unique instrumental facilities available at the CNMS, advanced computational and theoretical capabilities at CNMS and at ORNL overall, and our in-house and user research, by developing instrument- and domain-specific ML tools and workflows. The specific targets of the DNA Group are:

1. Development of the analysis methods and workflows for experimental data from STEM, SPM, and chemical imaging and other characterization tools to derive materials- and system-specific insights into generative physical and chemical phenomena;
2. In collaboration with ORNL-wide AI efforts, integrate these methods in the sustainable data analytics and ML workflows that minimize the entry barriers for CNMS staff and users and allow reproducible data analyses;
3. Interact with the CNMS instrumentation staff/groups in the development of the ML tools and edge-specific algorithms and software for real-time data analytics, automated experimentation in electron, scanning probe, and chemical imaging, as well as robotic and automated synthesis.

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 environmental, safety, health, and quality (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, training, 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 that are offered to the user community at CNMS. More details can be found at cnms.ornl.gov:

- **Imaging, Microscopy, and Nanoscale Characterization**: STM for imaging, spectroscopy, and manipulation of the structural, electronic, magnetic, and transport properties in low-dimensional material systems. AFM for imaging and dynamics in nanostructures including ionic and electronic transport, electromechanics, chemical imaging, energetics, magnetic properties, chemical reactions, and electronic, structural, and spin phases and transitions. The CNMS has developed SPM microscopy techniques for resonance imaging, high data acquisition rates, microsecond time resolution, and spin-dependent transport. A complementary suite of aberration-corrected STEMs for atomic-level imaging and spectroscopy, with an emphasis on ultrafast image acquisition, ultrahigh energy resolution (phonons), *in situ*/*operando* microscopy, EELS, low-voltage/low-dose capabilities for beam-sensitive materials, and 4D STEM imaging techniques such as differential phase contrast and ptychography. CNMS is the only NSRC to offer APT to the user community and these research efforts continue to be focused on application of the technique to nanoscale and non-conventional (non-conductive) material systems. Special emphasis on multimodal and chemical imaging (based on mass spectrometry and optical spectroscopy) and He-ion microscopy (HIM). Development and implementation of deep data analytic methods.
- **Theory, Modeling, and Simulation**: Scale-spanning modeling/simulation, high-throughput screening, 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.
- **Science and user driven data analysis workflows**: The wealth of instrumentation available at CNMS yields large volumes of scientific data in the form of the multidimensional data sets, process and characterization histories, etc. CNMS team strives to develop and provide consistent and traceable workflows that allow end to end conversion of instrumental data to the physical and chemical descriptors and models and providing science-driven outcomes

- **Synthesis and Fabrication:** Controlled synthesis and directed assembly of nanomaterials in a Class 1000 cleanroom environment (the NRL). 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 by chemical vapor deposition (CVD) and PLD with *in situ* optical spectroscopic diagnostics. Controlled processing and assembly of organics, 2D crystals, and hybrid perovskites in glovebox inert environment. High-throughput synthesis platform for quantum materials by PLD utilizing AI/ML optimization.
- **Soft Matter Synthesis and Characterization**: Synthesis and molecular level characterization of small molecule building blocks, polymers, copolymers, and polymer-modified interfaces, including biologically inspired systems and site-specific deuteration of molecules and polymers for neutron scattering studies (deuteration).
- **High-throughput Synthesis**: Efforts on the use of continuous flow chemistry and polymerization enable control of reaction parameters (e.g., pressure, temperature, laminar flow) to enable more efficient yields and stereochemical control.
- **Rheological Behavior and Simulations**: The investigation of flow properties in matter enables a better approach for understanding the parameters that govern phase separation, interfacial interactions, and process control relevant to polymer applications. Simulation and theory are essential for predictive and empirical guidance and interpretation of results.
- **Thermomechanical and Dielectric Properties**: The understanding of properties in polymers (thermoplastics, elastomers, thermosets) can be elucidated through novel characterization methods giving a deeper understanding of structure-property relationships and dynamic macromolecular chain and reaction behavior.
- **3-D Printing**: Capabilities to utilize additive manufacturing as both a fabrication method and a tool for new materials investigation. In addition to supporting user needs for shaping polymers into unique and complex geometries, efforts around this this capability will look to expand techniques for operando characterization and the creation of new materials for evaluation.
- **Functional Characterization of Nanomaterials**: Characterization techniques include optical characterization and laser spectroscopy mapping (microRaman, photoluminescence (PL), second harmonic generation (SHG), ultraviolet-visible (UV-VIS) spectrophotometry), electrical and optoelectronic characterization, ultrafast optical pump-probe dynamics measurements, ultrafast nm-resolution cathodoluminescence (CL), quantum coincidence measurements, catalytic properties, magnetometry and magneto-transport, XRD including small-angle/wide-angle X-ray scattering (SAXS/WAXS), and QCM-based platforms tailored for gravimetric/optoelectronic and multimodal characterization.

3. CNMS In-house Capability Research Effort: Four Scientific Themes

The CNMS in-house science effort aims to *understand the effects of temporal 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 nanomaterials, CNMS

in-house research is structured into four themes. Two of these themes, Harnessing Complex Macromolecular Conformations (HCMC) and Directed Nanoscale Transformations (DNT), focus primarily on the formation of new nanomaterials and understanding synthesis and assembly mechanisms. The other two themes, Metastability in Driven Nanomaterials (MDN) and Heterogeneities in Quantum Materials (HQM) emphasize the study and understanding of nanomaterials functionality.

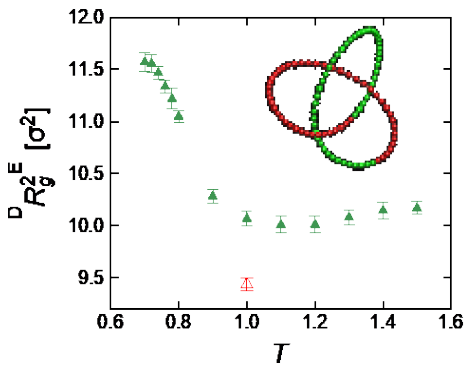
Within the themes that are focused on the formation of nanomaterials, HCMC seeks to understand the macroscopic response of chain conformations that form under non-equilibrium conditions and the interphase and interfacial effects that control these responses. In contrast, DNT seeks to illuminate the basic scientific questions of how matter can be transformed locally, thus enabling novel nanofabrication approaches toward atomic precision. In combination, these two themes tackle nanomaterials assembly both from the bottom-up (self-assembly) and top-down (fabrication) perspective, realizing 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 macromolecules 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 focused on understanding specific functionalities formed at the nanoscale, the effort on HQM seeks to understand the role(s) of heterogeneities in defining and affecting coherence and entanglement of quantum states to enable new materials for applications in energy and QIS. In contrast, MDN strives to understand and control the hysteretic response of nanomaterial systems under applied stimuli (electric, stress, or optical fields) to deterministically create long-lived metastable states that ultimately dictate energy transport in material. 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 HCMC and DNT. At the same time, these functional behaviors play a key role in understanding many of the mechanisms of nanomaterial's formation through investigation of energy and mass transport at the nanoscale.

Neutron scattering plays a role in all four themes, most directly so in the HCMC and MDN themes, while DNT and HQM 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 Nion Hermes UltraSTEM (MAC-STEM) and the low-temperature, high-resolution four-probe 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, etc.), 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 DNT and HQM themes, while topics related to neuromorphic computing are part of the research in the MDN theme.

Harnessing Complex Macromolecular Conformations (HCMC)



We simulated the effects of chain topology on lamellar-disordered transition temperature using coarse-grained molecular dynamics simulations. Simulations will be used to study topological effects in polymers and copolymers.

Control and characterization of equilibrium chain conformations in polymer systems is now feasible and significant advances have been made in connecting chain configurations and conformations to macroscopic properties. Moreover, advances in synthetic polymer chemistry and topology control have enabled improved physical models beyond scaling with chain length and interaction parameters in both solution and bulk form. Nonetheless, chain conformations under non-equilibrium conditions that result from either the presence of applied forces or processing protocols are less emphasized and not yet understood. Additionally, understanding interphase and interfacial effects that are essential for "real world" and industrial applications are lacking. Due to a limited number of studies probing chain conformations in non-equilibrium conditions, their connections with macroscopic responses remain unclear. *The overarching goal of this theme is to bridge this gap in our understanding by connecting non-equilibrium chain conformations to macroscopic responses using state-of-the-art synthesis, characterization, and modeling.* To achieve our overarching goal, three specific aims will be pursued:

1. *Understanding Structural and Polarization Dynamics in Polar Polymers:* Connections between non-equilibrium chain conformations and their responses can be investigated by designing experiments with carefully chosen polymers (non-polar and polar). Complementary simulations can unveil the role of entropy production by using electrostatics as a gauge for tailoring and measuring responses to applied fields, allowing us to explore the presence and tunability of these responses via small molecule and ion intercalation. The synthetic capabilities at CNMS related to polyelectrolytes and polyzwitterions coupled with small-angle/wide-angle X-ray scattering (SAXS/WAXS) and neutron reflectivity tools are vital to achieving this aim.
2. *Understanding Topological Effects in Polymers and Block Copolymers:* Advances in synthesizing topologically complex macromolecules (geometrical topology) and compositions such as graft copolymers and knotted polymers provide a library of materials whereby topological effects can be tuned in a facile manner by varying chain architecture and measuring responses to various non-equilibrium conditions. TM&S together with synthesis capabilities related to cyclic polymers will be used to achieve this aim.
3. *Understanding Processing Effects:* Effects such as solvent evaporation, thermal treatment, and mechanical deformation on polymer properties and responses will be established. Polymer non-equilibrium states in real-time will be studied to enable correlating macromolecular properties during a synthetic process, a key toward property optimization in technologies involving 3D printing.

These three aims will provide critical understanding of entropy-driven processes that are key toward designing processing pathways and macromolecular architectures for accessing metastable

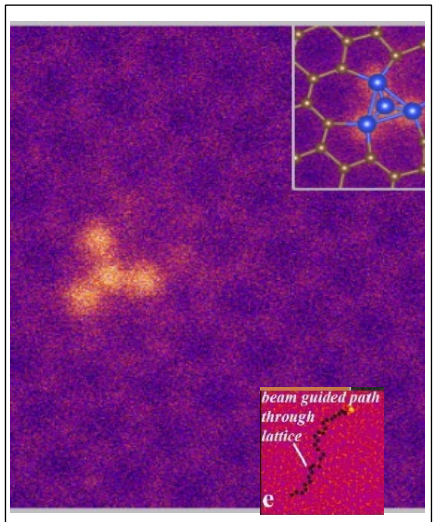
morphologies, e.g., a materials-by-design concept for functional soft materials that operate synchronously with processing conditions.

Directed Nanoscale Transformations (DNT)

The overarching goal of the DNT theme is *to dynamically control changes of chemical and structural states that materials undergo in confined and non-equilibrium conditions to structure new materials from atoms up.* The fundamental challenge in achieving this goal lies in understanding the dynamic processes that give rise to atomic transformation and building a framework to control it in order to achieve a directed nanoscale transformation. To overcome this challenge, two correlated research aims that are focused on understanding the interplay of energy and matter governing material transformations and feedback for direct control:

1. *Understanding Energy Transfer:* Determine the role of energy transfer from beams of energetic particles (electrons, ions, lasers) and external fields (scanning probes) into materials to enable control over highly localized transformations by altering bonds at the atomic and molecular level into excited metastable states.
2. *Understanding the Role of Energy Landscape:* Determine how energy landscape parameters can be locally and globally tuned to guide the directed fabrication and atomic manipulation of functional 1D-3D nanostructures on demand from the single atom level up.
3. *Guided Energy Flow for Controlling Transformations:* Develop ML methods for bottom-up descriptions of transformations and reinforced learning to predict and guide transformations.

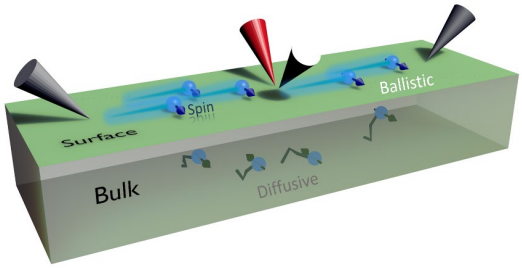
The starting point for this work is the observation that experimental platforms to visualize structure and function, such as electron, ion, and scanning probe microscopes and associated spectroscopies, can also be used to induce transformation, thus enabling the study and control of the processes guiding 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.



Directed transformation-assembly of silicon atoms into a graphene monolayer.

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. The ability to manipulate and study matter in a tight research loop at the atomic and nanoscale will be of key interest to a broad user community, including those from the QIS field.

Heterogeneities in Quantum Materials (HQM)



Spin-polarized four-probe STM measures spin-dependent electrical conductance on topological insulator Bi_2Te_2Se. By reducing the probe spacing at low temperature, most electrons are "pushed" to the topological surface states, showing a quasi-ballistic spin-polarized conductance.

Quantum materials are typically heterogeneous with structural defects, impurities, surfaces, edges, interfaces, and disorder. While heterogeneities are often viewed as unwanted liabilities in conventional systems, they offer a powerful platform to create and control coherence and entanglement in quantum materials. It is the electronic and magnetic structures of heterogeneities that often define and affect the system's quantum phenomena such as cherence, tunneling, interactions, entanglement, and topological effects. *The overarching goal of this theme is to understand the roles of heterogeneities in defining and affecting coherence and entanglement of quantum states to enable new materials for applications in energy and QIS.* This goal is achieved by revealing the atomistic-level understanding of structure-property relationships in quantum materials that occur at structural and compositional heterogeneities such as defects, interfaces, and disorder. This understanding includes the interactions between electronic, magnetic, photonic, and vibrational states of heterogeneities that render coherent and entangled quantum states necessary to enable the design of new generations of quantum materials, including topological matter (topological insulators, Weyl and Dirac semi-metals, and quantum spin liquids) and quantum light emitters based on heterogenous 2D materials.

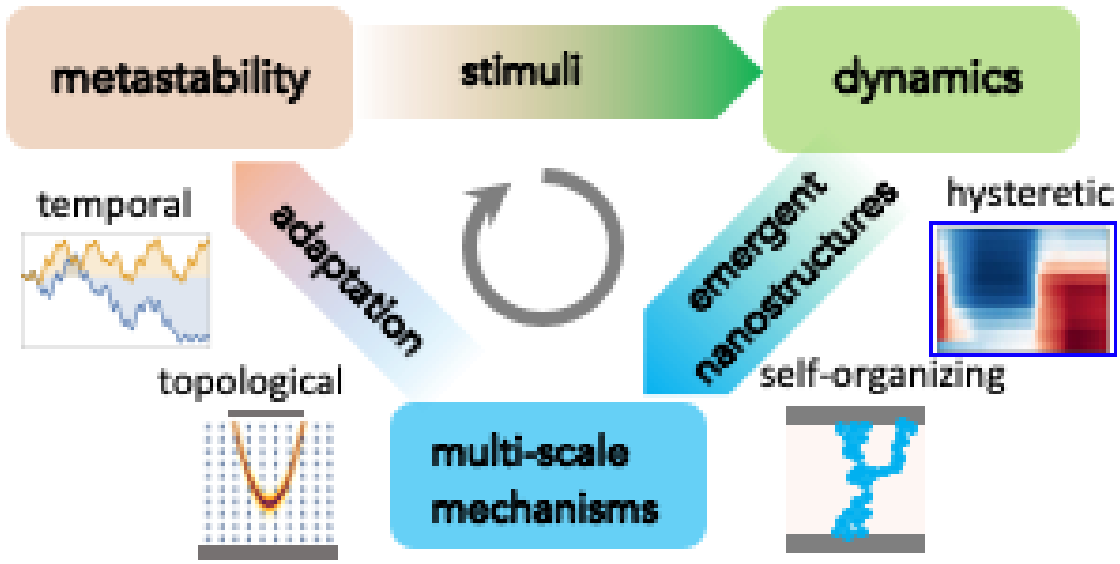
Research focused on effective ways of connecting the heterogeneity of structures to the quantum behaviors of real materials will allow us to develop unique capabilities for the scientific community. In this way heterogeneities can be rationally controlled to create desirable quantum states and interactions of various energy states and thus enable a quantum leap in materials research. Heterogeneities in quantum materials will be studied through the execution of three aims:

1. *Reveal the Structure of Heterogeneities*: new approaches to reveal the structural, electronic, magnetic, vibrational, and optical degrees of freedom of heterogeneities.
2. *Understand the Effect of Heterogeneities*: correlations of coherence, topology, and entanglement of quantum states with heterogeneities.
3. *Control Heterogeneities*: toward tailored interactions between energy carriers for energy conversion and QIS applications.

This research theme builds directly on expertise in atomic level control and understanding of materials, including the characterization of heterogeneities in low-dimensional (LD) materials through innovative development and utilization of scanning probe and electron microscopy and spectroscopy (such as spin-polarized STM, atomic-resolution 4-probe STM, MAC-STEM, low-frequency Raman, and ultra-fast laser spectroscopy) combined with first-principles calculations. These aims motivate key directions in other CNMS themes, e.g., DNT and HCMC, driving the development of materials and heterostructures with precisely controlled defects and interfaces in order to test theoretical predictions relating atomistic structure with function. These aims also provide information on electronic behavior to understand MDN, where defects and interfaces play a key role. With this assistance, the HQM theme will extend beyond our current understanding of

"ideal" materials and provide the critical knowledge needed to control the coupling of electron, spin, and photon interactions in materials to create the designer quantum states that will be the foundations of QIS.

Metastability in Driven Nanomaterials (MDN)



MDN seeks to unravel and control dynamics of driven nanomaterials, paving way to complex spatio-temporal functionalities such as self-organization, regulation, adaptation, and computation.

Predicting and measuring the non-equilibrium dynamics of the driven state is a fundamentally challenging task, generally involving complex spatio-temporal correlations and path-dependence, multiplicity of possible metastable states and the experimental and theoretical challenges associated with inclusion of stochasticity and energy dissipation in the description of material dynamics. However, time-dependent functionalities define the overwhelmingly complex, yet highly functional dynamics of non-equilibrium mesoscale and macroscale biological and natural systems including spontaneous symmetry breaking, self-organization, self-regulation, adaptation, and high degree of autonomy. Replicating such functionalities in nanoscale systems would necessarily set the stage for a new generation of applications while providing a new fundamental setting for further development of non-equilibrium thermodynamics. *The overarching goal of this theme is to reveal, understand, and control the dynamics of driven nanomaterials and nanoscale structures, with the aim of creating temporal, learning, and adaptive functionalities the drastically deviate from the equilibrium state.*

The theme will draw on the expertise at CNMS to address the specific challenges of driven nanomaterials. For example, the recent rapid proliferation of ferroelectric and ionic nanomaterials now enables us to predict their near-equilibrium properties with high accuracy. Yet, the observation of oscillatory, chaotic, adaptive, and self-organizing phenomena in such nanomaterials is rare. Meanwhile the operating principles of some material systems – such as resistive switches – continue to avoid understanding arguably because of the disconnect in available knowledge of the properties that determine time-evolution of such systems in the driven state. The work in this

theme will continue to grow and synergize CNMS strengths in hysteretic materials, SPM, nanofabrication, computation, and ML methodologies toward further probing and understanding of the time-domain and structural complexity of the metastable states. In addition to growing the user community, the theme seeks to develop the perspective of the function-structure paradigm, where the function, rather than thermodynamic stability, defines the structure of a nanomaterial, and the latter can therefore continuously adapt to the surroundings and operating environments in broad conceptual analogy to the fascinating functionalities of the biological machine. Three interconnected aims will guide the theme research:

1. *Scalable Modeling of Metastability:* A key effort in the theme is to apply the concepts of stochastic thermodynamics – that captures materials under the influence of external forces and/or noise – to known and emerging responsive nanomaterials prolific in metastable states. We will develop scalable theoretical and computational methodologies that seek a reliable balance between accuracy and scalability to predict underpin material response to external stimuli.
2. *Control of Cooperative Phenomena in Driven Nanomaterials:* The range of nanomaterials exhibiting distinct dynamics in the driven state can potentially be very large, not least because of the strong fields, energy densities, and fluctuations accessible in driven nanoscale systems. We seek deeper understanding of metastability underpinning such responses and new temporal dynamics in the driven state. To better understand the physics involved in generating metastable states in such systems, we will advance experimental approaches that enable precise control of driving stimuli and introduce new approaches to stimulate materials while analyzing real-time response of the system at the nanoscale.
3. *Advancing New Material Functions through 'Learning' and Adaptive Behavior:* Increased understanding of driven nanomaterials will introduce a function-structure paradigm in materials discovery by enabling the functions to be encoded into adaptive materials in a self-consistent cycle of stimulus-response-refinement. Informed by aims 1 and 2, we will seek the fundamental understanding of the material mechanisms that can perform the adaptation and pursue new adaptive material systems as well as "encoding" protocols that can reveal the full potential of this paradigm for present and future technology.

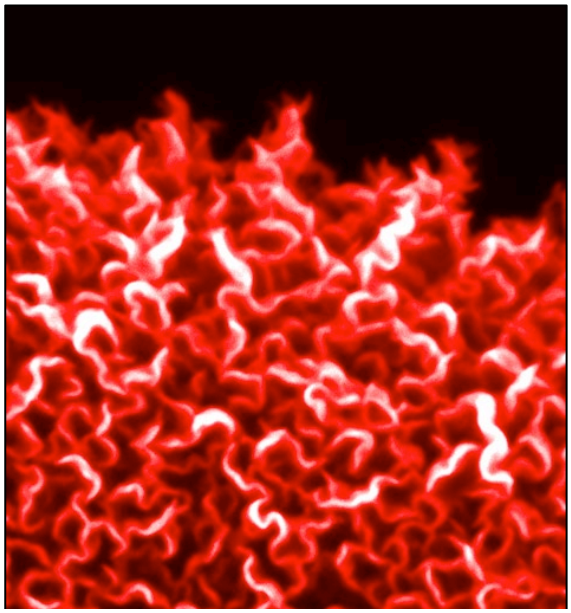
The MDN theme expands on the previous focus on responsive nanomaterials to include understanding of non-equilibrium response and control over metastability. The synergy of developing methods to stimulate and observe non-equilibrium response and the scalable theoretical methods that can support and eventually guide material dynamics is anticipated to create a new generation of experimental and theoretical user capabilities, and thus nucleate new user directions aimed at driven, adaptive, and active materials for the future of computing and energy functionalities.

4. Cross-cutting Focus Areas and New Directions

The four science/capability 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 that are continuously re-evaluated 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 (SAC), the User Executive Committee (UEC), feedback received following the DOE Triennial Reviews, as well as through direct interactions with the other NSRCs, all contribute to the planning our future directions.

Modernizing Materials Science

Experimental characterization methods and theoretical calculations are now the two central paradigms of modern physical and chemical sciences. However, it is well realized that both experiment and theory give only approximation of reality, due to incompatible and poorly controlled approximations. At CNMS, we are proactively involved in developing machine learning tools that aim to integrate the theorist- and experimentalist insights on the materials to build a comprehensive integrated picture, deploy these methods as a part of automated experimental workflows that accelerate materials understanding and discovery, and to make these tools broadly available to the user community.



New active functionalities emerge from materials when formed in a fashion to exemplify nanostructures. Shown is a surface coated with carbon nanospikes synthesized via plasma enhanced chemical vapor deposition (PECVD).

Although TM&S have been reasonably successful in predicting single material 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.

TM&S 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 and 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 datasets 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 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 DNT 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 an ecosystem of open-source packages (*pycroscopy*) and elucidated through Jupyter notebooks, to provide, for example, the relevant atomic and magnetic configurations as well as the 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. Indeed, the *pycroscopy* ecosystem contains an open-source set of utilities for image processing and scientific analysis of imaging modalities including multi-frequency SPM, STM, X-ray diffraction microscopy, and STEM and includes regular updates by both staff and users to the CNMS.

The second fundamental research direction is related to knowledge extraction from microscopy data. For scattering techniques such as X-ray and neutron scattering the approaches to extract materials related knowledge such as structure, presence of correlated disorder, or discovery of phase transitions and novel functionalities, are well established, as are links to existing physical models. In comparison, microscopy tends to yield large volumes of high veracity data containing information on structure, functionality, presence of novel physical phenomena, etc. Our ability to process and interpret this data is, at present, limited to direct comparisons with theory (i.e., conforming the feasibility of observed behaviors without making quantitative connections that will either refine the theoretical model or interpret observation) or human-based analysis. While adequate at times when a single data set (good image) required days of microscope set-up and tuning barely twenty years ago, this is grossly insufficient now when advanced microscopy systems are readily available. Ultimately, we aim to develop ML assisted pathways toward the exploratory data analysis of large multidimensional data sets, exploring and flagging potentially unusual behaviors that may signify the presence of new physical behaviors, complementing the observation and detection power of a human operator. While helpful for classical 2D modalities, these capabilities are absolutely critical for the multi-dimensional data sets that are simply not accessible to human examination. Complementary to this is the development of theory matching methods that allow interpretation of the multi-dimensional data in the context of generative physical models, including both the refinement of model parameters and selection between alternative models of material behavior.

Beyond this point, the rapidly developing ML and AI fields now offer fundamentally new types of connections between experiment, theory, and human-driven science fields. Including utilization of past knowledge via Bayesian methods and its offshoots such as transfer and deep kernel learning, multimodal data integration, etc. Here, it is important to note that the fundamental correlative paradigm of ML is inconsistent with the hypothesis driven nature of modern physical sciences. Hence, a broad collaboration with efforts in causal ML, Bayesian ML, and development of parsimonious physical models and descriptors are a natural challenge.

ML and AI for Accelerating Nanoscience

The exploration of new materials synthesis is fundamentally limited by man's ability to draw connections between the information gained from disparate characterization techniques with the ability to vary materials synthesis processes in real time based on complex chemical and structural processes. Autonomous-smart experiments merge understanding from physics-based models of *ex situ* characterization techniques with real-time *in situ* control of materials growth and synthesis processes. Modern synthesis is a complex ecosystem composed of heuristic design rules, theories and phenomenological models, and advanced characterization tools capable of observing synthesis processes across multiple spatiotemporal scales. Modern ML and AI have proven successful at optimizing massively complex systems, yet there are no existing materials synthesis platforms within the DOE complex or the scientific community at large where this ML/AI optimization has been integrated nor attempted. AI offers a unique opportunity to break through such barriers by driving the advancements through autonomous-smart experiments. By taking the human out of the loop, materials innovation will be accelerated as AI has the ability to find new relationships not apparent using the traditional 'cook-and-look' approach of science.

Autonomous-smart experiments sit at the nexus of the CNMS' core capabilities that have been evolving over the past five years including ML-driven image analysis, feedback and control of materials synthesis systems, and the nascent activities in developing physically informed ML routines for both physics extraction and experimental design. Critically, the use of such strategies within materials synthesis sets the NSRCs apart and positions CNMS to complement the ML/AI activities at the BES light and neutron sources, which are focused on materials characterization. CNMS' modern synthesis research incorporates a wide-range of design rules and theories alongside advanced characterization tools capable of observing synthesis processes on the relevant size- and time-scales at which they occur. The unoptimized and traditional modes of synthesis exploration cannot be scaled to handle the complexity inherent to many next-generation materials (exhibiting collective, emergent, and responsive properties), which are multi-component and hierarchical—incorporating molecular, nanoscale, and mesoscale components. Therefore, revolutionary new modes of synthesis are needed. A promising direction is autonomous-smart synthesis enabled by end-to-end ML/AI integration across all aspects of the materials discovery loop—from material preparation, through characterization, to data interpretation and feedback to minimize the experimental trials needed to achieve the desired properties. This will allow vastly more challenging materials problems to be tackled. The key is ML for automation of model-building and decision-making aspects of the experimental loops that will enable machine-guided synthesis, processing, and ultimately materials discovery. Such an approach can enable breakthroughs in the synthesis and control of novel materials/processes such as:

- *Metastable phases and materials that persist out of equilibrium:* Such materials enable access to a diversity of properties beyond the limits drawn by equilibrium thermodynamics. For example, optically driven processes could provide more control over chemical processes and lead to new materials such as metastable phases or new LD materials with dynamics controlled by in-plane heterogeneity rather than by layer stacking order. Another example is self-assembly, where transient (non-equilibrium) intermediate states frequently appear and control of assembly pathways can enable improved structural control. Modern characterization systems such as electron and scanning probe

microscopies also allow "bottom-up" fabrication of new structures that are meta-stable, allowing, for example, arrays of topological defects to be created with nanometer precision for desired properties.

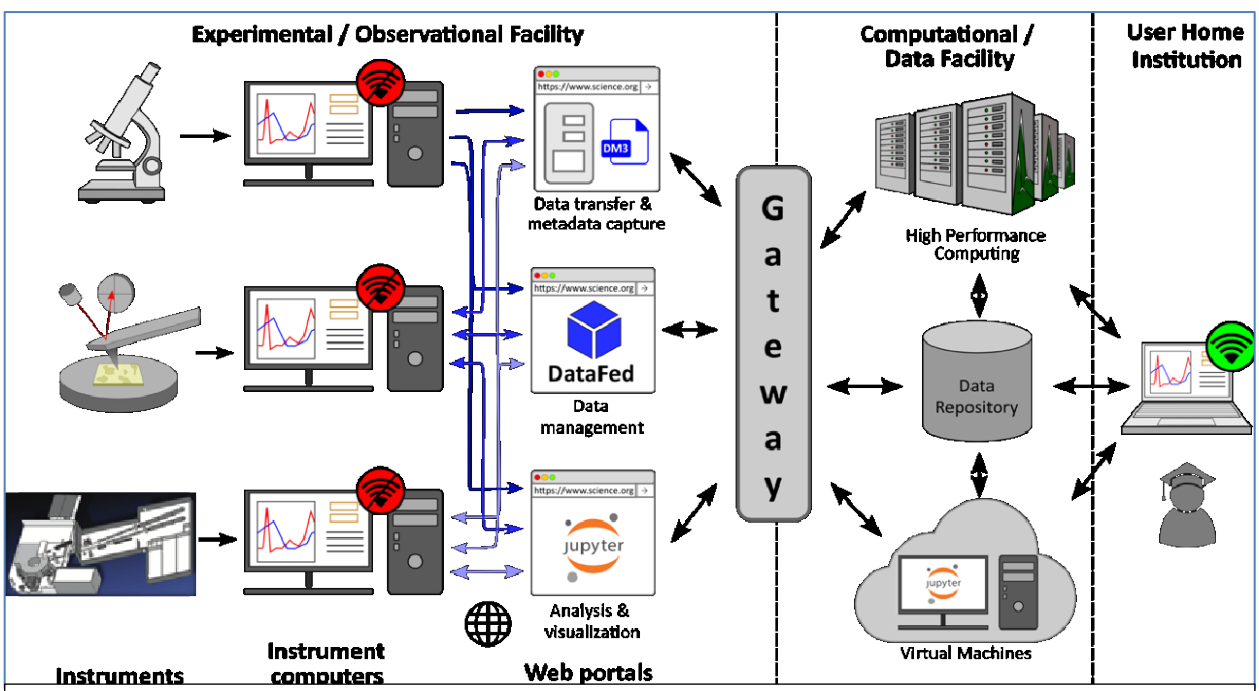
- *Interfacial processes and properties:* Controlling interfaces in materials often relies on precise control of atomic bonding at the 'joint' between two dissimilar materials. The ideal strategy to avoid performance-limiting defects is to minimize perturbation of the atomic order at the interface by preserving a high degree of crystallographic order, e.g., epitaxy. However, atomic-scale insights into grown structures presents significant inverse problems that can be tackled using combined physics-ML methodologies.
- *Materials for QIS*: These materials include potential solid-state qubits, photon sources, and quantum sensing systems. The CNMS layered materials stamping and new QIS physical PLD system will be generating rich structural, heterointerface, and functional property datasets.
- *Harnessing heterogeneity in complex systems*: Here, the aim is to broadly understand how population diversity influences growth and behavior, with the ultimate goal of creating a closed-loop materials property prediction, synthesis, and characterization loop, which will be accomplished using high-throughput nanomaterial synthesis and automated atomic-scale/multimodal characterization. By understanding and controlling heterogeneity, it may be possible to design multifunctional and self-regenerating catalytic systems.

Four technical challenges must be addressed to make autonomous-smart experiments a reality. These challenges address the need to link *in situ* and *ex situ* characterization techniques to active control of the materials synthesis process while addressing the barriers of big-data, data archiving, and high-speed decision making. These four challenges are edge computing and integrated experimental instruments, in situ multimodal analysis, big data and data curation, and automated smart characterization:

1. *Edge computing and integrated experimental instruments:* Computing at the experimental instrument(s) for on-the-fly analysis with feedback during an experiment must be implemented to maximize the information gained and exert efficient control. This will be particularly important for multimodal experimental probes that require analysis across different platforms. Edge computing for automating aspects of experiments, such as for ML/AI-assisted tuning of the environment, importance sampling, next-experiment recommendation, etc., will be critical. Additionally, on-demand pipelines to HPC for automatic spawning of jobs directly related to discoveries at the instrument is needed. This can be important for forming databases based on higher levels of ML models trained on simulated data, where the simulations would require an HPC environment. The goal is to provide fast on-the-fly analysis of 'streaming' experimental data.
2. *In situ multimodal analysis:* Implementing online analysis during an experiment will be important when more than one type of probe is being used. CNMS' characterization capabilities are constantly used for the determination of chemical composition, materials structure, physical properties, and to correlate functionality. In general, this involves: (1) an analytical step to confirm that the target chemicals and/or materials are produced; (2) characterization of the physical properties, morphologies, defects, and interfaces of the functional materials by using multiple probes/techniques; (3) characterization of the multi-functional properties, *in situ*/*operando*, in devices across a broad frequency range. Achieving acceleration will require integration of new analysis methods across all of these multi-information sources and platforms. *In situ* data analytics including cross-modeling

will be approached on two levels, with the first level being at the point of the experiment using edge computing and the second level being HPC. The ML algorithms will be incorporated as a part of *in situ* multimodal analysis that will lead to machine-guided decision-making algorithms for the selection of optimal experimental condition, minimal number of experiments, and reduced model errors.

3. *Big data and data curation:* Data must be labeled with all appropriate meta-data, stored and accessible from multiple different compute facilities, traceable, and all processes form acquisition to analysis to manuscript must be reproducible. Doing so requires substantial efforts at standardizing data formats, developing meta-data schemas, and installing gateway devices that process and shuttle data from instruments to centralized repositories at ORNL computational facilities, which can then also be accessed by authenticated users. We have focused on deploying an ORNL-developed solution termed 'DataFed' which provides a 'data backplane' capable of federating scientific data stores and enabling seamless access, rapid search through meta-data, and provenance tracking. This will enable links to other major facilities and capabilities such as OLCF, microscopy laboratories, HIFR, and SNS to characterize and fully understand the new materials. Development of workflows to integrate knowledge from across multiple facilities and the ability to create and draw on knowledge graphs to better inform modeling and propose new experiments will be developed.
4. *Automated smart characterization:* Use of active learning and Bayesian methodologies in combination with predictive modeling during characterization to efficiently explore heterogeneities in materials, to minimize uncertainty, and to maximize physics knowledge gain, as opposed to purely information theory-based metrics.



Data-analytics infrastructure development. Data is ingested from instruments and transferred to data repositories, which are federated via DataFed. Portals will be utilized to enable users to access data and spawn analytics servers on virtual machines with immediate access to data, as well as more extensive computational capabilities.

Opportunities in Quantum Information Science

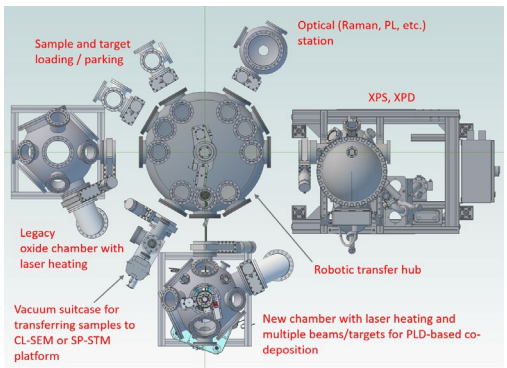
Quantum information science (QIS) is the exploitation of quantum phenomena for the storage, transmission, manipulation, and/or measurement of information. The National Quantum Initiative Act (December 2018) directs the President to implement a National Quantum Initiative Program to, among other things, establish the goals and priorities for a 10-year plan to accelerate the development of QIS and technology applications. The DOE is tasked to carry out a basic research program on quantum information science (National Quantum Initiative Act, Sec. 401) and DOE's SC to establish and operate National Quantum Information Science Research Centers to conduct basic research to accelerate scientific breakthroughs in QIS and technology (Sec. 402). The DOE selected ORNL to lead the collaborative Quantum Science Center (QSC) to develop quantum technologies that will realize the potential of topological quantum materials for manipulating, transferring, and storing quantum information.

The fundamental needs of QIS research have been documented in two BES roundtable reports on QIS – one focused on quantum computing, Opportunities for Quantum Computing in Chemical and Materials Sciences (October 2017) and Opportunities for Basic Research for Next-Generation Quantum Systems (October 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 communications, and significantly larger computational capabilities at speeds far greater than those possible in classical computing. However, realizing these advances requires breakthroughs 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. To paraphrase points made in a July 2016 interagency working group report on advancing quantum information science, "Much of the research in QIS has been conducted within existing institutional boundaries, meaning the next critical steps in QIS research and development will require increased collaborations across these boundaries and formation of teams with a diverse range of skills." This is precisely the type of environment that CNMS provides to its user community – a collaborative center where researchers from different disciplines and backgrounds work hand-in-hand on cutting-edge instrumentation in a setting that emphasizes the integration of TM&S into all steps of the studies.

It is thus not surprising that QIS is playing a rapidly increasing role at the 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, ORNL's computational resources, and the very direct interactions with ORNL's QSC and Quantum Information Sciences Group (within the Computing and Computational Sciences Directorate). In fact, interactions with this group have helped shape the CNMS direction in QIS and allowed the Center to build the bridge between materials manipulation and imaging, 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, DNT and HQM, have a direct link to QIS. In fact, some of the most remarkable achievements within the DNT effort, namely the direct placement and manipulation of individual dopants within a 2D lattice, were prominently featured in one of the

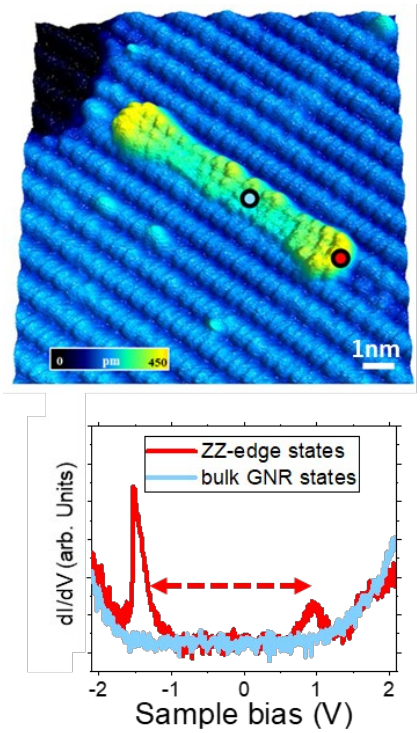
BES roundtable reports mentioned earlier. Combining such exquisite control over individual dopant placement with simultaneous progress in nanoscale 3D direct-write fabrication will lead to entirely new opportunities to create, and interface with, artificial quantum systems. The recently installed Raith Velion system at the CNMS combines focused ion beam (FIB) and scanning electron microscopy (SEM) and operates a variety of isotopically controlled ion sources for imaging, milling, and deposition. It offers new opportunities for creative material's synthesis and modification, particularly for the emerging areas of quantum systems such as creating superconducting resonator circuits. CNMS is actively planning investments in equipment that will not only enable higher throughput synthesis and characterization of a range of quantum materials with QIS functionality, but to directly integrate the somewhat dissimilar approaches of DNT and HQM by combining electron and ion beams with *in situ* monitoring and operando quantum observations.



High-throughput synthesis platform overview showing sample and target parking stations, optical and photoelectron characterization stations, conventional and co-deposition PLD synthesis stations, and vacuum suitcase for sample transfer to the CL-SEM and SP-STM. Samples are shuttled from synthesis to characterization using a robotic transfer hub.

Rapid synthesis and characterization are considered key to exploring many predicted materials with functionality for QIS. Moreover, it is imperative for an NSRC such as the CNMS to develop approaches that shorten the time-to-success for QIS users who come to the Center to develop new materials and test their performance. Combining these ideas, and therefore departing from the normal, lengthy materials development protocol, the CNMS, as part of a recent QIS Infrastructure project award, is developing a highly agile synthesis platform based on pulsed laser deposition (PLD) that will support ML approaches, rapid *in situ* diagnostics of key processing conditions, and high-throughput characterization of key materials parameters using X-ray photoelectron spectroscopy (XPS), X-ray photoelectron diffraction (XPD), and optical spectroscopies such as Raman and photoluminescence (PL) to guide and rapidly develop the synthesis of a wide variety of material systems relevant to QIS. Closing the loop – i.e., determination of QIS functionality – is supported by two other platforms under development in the QIS Infrastructure project, namely a quantitative cathodoluminescence scanning electron microscope (CL-SEM) modified for fs excitation of the field-emission-gun and an ultralow temperature scanning tunneling microscopy platform equipped with a vector magnetic field and modified for a spin polarized tip (SP-STM).

In situ manipulation and characterization of defects at the atomic scale is essential to developing an improved understanding of single photon emitters and spin qubits but characterizing the quantum state of these systems ultimately requires a photonic interface. CNMS recently installed a closed-cycle 3 K cryocooler with an integrated optical microscope for the characterization of single photon emitters and spin qubits at the mesoscale. A collocated set of superconducting nanowire single photon detectors for visible and telecom photons enable characterization of the purity and indistinguishability of photons emitted from a wide variety of defects in materials at temperatures from 3 to 300K. A new CL-SEM enables characterization of the quantum state of light emitted from defects under combined electron and laser excitation at temperatures from 8-300K. This microscope provides complete spatial, spectral, angle, polarization, temperature, and time-resolved characterization of the quantum state of defects. Moreover, it builds on existing CNMS research in 3D direct-write fabrication of plasmonic resonators around pre-characterized color centers to modify the optical properties of defects operando. Controlling the brightness, directionality, purity, and indistinguishability of spin qubits in this way will be essential to ORNL's QSC research efforts focused on spin-based quantum sensors for materials characterization. ORNL's QSC is centered on the development of topological platforms for QIS. Several CNMS researchers are contributing directly to the modeling and characterization of these quantum systems, but useful topological quantum devices will also require extensive nanoscale device design and fabrication that CNMS is well-equipped to support through user access.



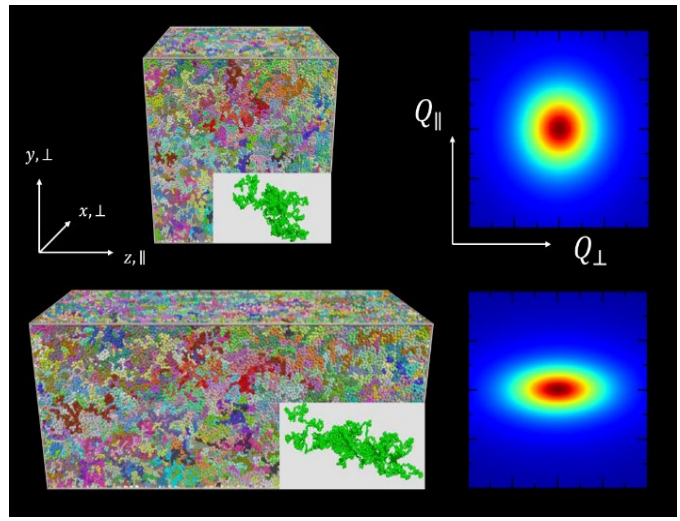
STM visualization of 7-atom wide armchair graphene nanoribbon on rutile TiO_2 (011) and tunneling spectroscopy reveal two resonances corresponding to zigzag magnetic end states that become entangled at finite distance.

Four-probe SPM approaches will similarly make it possible to manipulate dopants and observe the consequence on quantum behavior *in situ*. Developments of post-reconstruction data processes have 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 actively exploring ways to integrate quantum sensing into advanced measurement and imaging approaches. For example, CNMS recently teamed with ORNL's Quantum Information Science Group to demonstrate a quantum enhanced approach to SPM that utilizes quantum noise reduction in a squeezed light source to read out the displacement of an AFM microcantilever with sensitivity below the photon shot noise limit. Ongoing research targeting quantum enhanced optical and SPM is part of ORNL's QSC program and will rely on close interactions with the CNMS.

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 between all of our in-house science themes. The CNMS 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 are studied in the HQM theme and HCMC places a special emphasis on polymers with different topologies and architectures. These studies 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 projects.

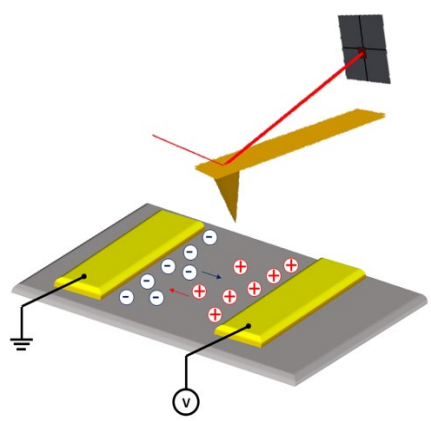


Snapshots of simulated entangled polymers in the equilibrium state (upper) and immediately after uniaxial stretching (bottom). Right frames correspond to simulated small-angle neutron scattering spectra, where Q⊥ is along the stretching direction.

Polymers are a versatile class of materials that often provide key advantages over other materials for energy applications: (i) light weight (built from the lightest atoms such as H, C, O, N); (ii) unique viscoelastic properties (only polymers exhibit rubber elasticity and unmatched ductility); (iii) extremely broad range of tunable properties (e.g., modulus from kPa to GPa, conductivity from insulators to conductors, selective permeability, etc.); and (iv) 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, 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, at CNMS 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.

New JEOL NEOARM STEM with low-acceleration voltage (30 kV), customized ultra-stable cryo-holder, low-dose imaging, enhanced annular bright field detector, large-solid-angle silicon drift X-ray detector, and a fast-pixelated detector (4000/sec).

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 installation 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 TM&S, 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 STEM 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 a 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.



Schematic of G-Mode SPM illustrating capabilities for G-Mode KPFM.

In late 2019, ORNL took delivery of a new SAXS/WAXS capability that is ideally suited for soft matter analyses, and recently came online. This capability has a newly developed metal jet X-ray source, which produces a beam as bright as a second-generation synchrotron and thus enables experiments with time resolution on the order of seconds over length scales that span several decades. Such time scales are relevant to many material studies, especially soft matter materials, under applied force fields, such as are found in application-relevant processes involving polymers

and other materials. This X-ray source also provides high flux for studies of materials on surfaces, such as epitaxially grown films and polymer brushes. The instrument will enable the development of specialized sample environments that are tailored to the requirements of ORNL researchers and that can also be used on the neutron scattering beamlines for maximum scientific impact. Importantly, the time-resolution that can be achieved with this instrument is comparable to that achievable with the neutron scattering instruments at SNS and HFIR. This new capability and its complementarity to neutron scattering provides CNMS researchers and the HCMC theme with an incredible instrumentation suite.

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 the SNS/HFIR). 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 architectures 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.

The Macro Group is also focused on developing new synthesis approaches, characterization tools, and fabrication methods to expand the understanding and utilization of macromolecules as both probes and new materials. The importance of leading the development of these capabilities is core to the mission of CNMS in empirical research and user access. This is reflected in our new capabilities in: 1) High-throughput polymerization using continuous flow chemistry. This is done by enabling controlled flow rate, temperature, and pressure conditions from otherwise batch-synthesis protocols, 2) New deuteration methods for improving yield and stereochemical control in designed neutron scattering molecular and macromolecular probes. The design of the molecular probes can be more tailored to the experimental approach and limitations of the instrumentation. 3) Use of novel rheological and dielectric characterization tools to understand the dynamic properties of blends and interfaces in complex fluids and extrusion processed compositions. 4) A newly established capability for 3D printing and additive manufacturing will enable the study of new polymer materials and unique environments for establishing structure-composition-property relationships. There is interest in collaborations with SNS on *in situ* and operando characterization of bulk vs. interfacial behavior during processing.

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

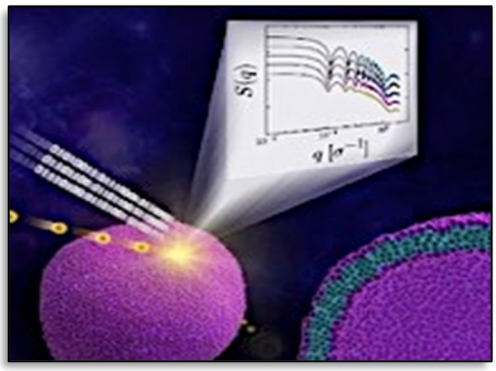
1. Provide expertise and capabilities for innovative polymer synthesis by enhancing our signature expertise in anionic polymerizations and directly utilizing the advantages of flow chemistry. The addition of flow chemistry, which uses channels or tubing to conduct a reaction in a continuous stream rather than in a flask, can provide chemists with unique opportunities to increase yield, enhance reactivity/selectivity, or in some cases enable new reactions.
2. Provide leading efforts for precision isotopically labelled materials synthesis, especially deuteration, enabled by exploring and understanding their influence on the physio-chemical properties.
3. Develop *in situ* soft matter characterization techniques and harness the power of predictive theory and modeling in synthesis.
4. Establish new methods of processing and fabrication of polymer materials and enable better structure-composition-property elucidation.

As an integral part of ORNL's research community, macromolecular research at the CNMS is well-positioned to achieve the above discussed goals. We have dedicated staff, engaged users, state-of-the-art in-house instrumentation, and co-located with core facilities at ORNL (supercomputing and neutron sources). Importantly, the vibrant CNMS in-house science themes directly integrate macromolecular science; in particular, the flow chemistry platforms we are 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 fields 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. 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, the CNMS and SNS/HFIR user offices worked to identify potential cross-facility users by integrating strategic questions in the facility proposals that staff could follow-up on following the proposal reviews; this new process has resulted in identifying several SNS/CNMS users each proposal cycle. Such administrative steps, including simplified access for CNMS users to SNS and HFIR and vice versa, have been implemented in the past year. Neutron–nanomaterials interactions are currently strongest in the



Artistic representation of new computational modeling capability for understanding dynamics of bilayer lipid membrances and for predicting neutron scattering results.

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 has continued 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 ORNL's 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 will 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 SPM-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 SNS as a part of an LDRD with 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 user work take advantage of quasi-elastic scattering on the SNS BASIS (Backscattering Spectrometer). Additionally, the SNS VULCAN (Engineering Materials Diffractometer) instrument provides 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 VISION (vibrational neutron spectrometer) uses neutrons to probe molecular vibrations while simultaneously collecting diffraction data and provides an opportunity for the study of *in situ* catalysis and studies 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, CNMS staff 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 quantum materials. This is particularly relevant in the context of increased interactions with the QIS community. On the synthesis side, 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 CNMS to take advantage of 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 ultrahigh energy resolution 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 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 maintains and further develops a host of complementary techniques that include NMR, MALDI-TOF-MS, Raman spectroscopy, light scattering and rheometry, 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 science 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.

Moving forward, we are co-investing in critical matrixed staff between SNS and CNMS to facilitate higher levels of common users and capability science. A new staff member will work directly at the liquids neutron reflectometer (NR) and the other new hire will focus on small molecule (biomaterials) deuteration. The new staff for NR will work closely with the liquids reflectometer team at SNS to develop a leading program in nanoscience focused on neutron scattering. A key component will be focused on championing developments on the liquids reflectometer including new sample environments and software developments to maintain and enhance the performance of the liquids reflectometer and expand the user base, especially in nanoscience. Additionally, providing support to SNS users by helping with experimental design and setup as well as serving as the local contact for beam line experiments will be an integral part of this position. This addition to CNMS will enhance our leadership in the area of nanoscience and neutron scattering.

The new staff for deuteration will develop a leading program for the synthesis of deuterated molecules, including de novo synthesis and hybrid biological as well as synthetic approaches for neutron scattering experiments that will be conducted using state-of-the-art techniques and at HFIR and SNS. A key component will be for user support, which includes growing the user program in the biomaterials area, user assistance as a local contact, helping with experimental setup, and experiment design and execution. This addition to CNMS will enhance our leadership in the area of deuteration of soft matter and greatly increase the ability to provide a growing user community with needed materials for deep-dive studies by neutron scattering that can uniquely characterize the spatial and temporal scales in these typically heterogenous systems.

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, some 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 that has been extended to spin-polarized transport probing, which offers a unique platform for characterizing and manipulating quantum states and dynamics. Similarly, band excitation and multi-dimensional 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 for curve fitting and a priori assumptions. Leading-edge chemical imaging methods that integrate ion beams and optical excitation and detection enable mapping the chemical and molecular species within materials at the nanoscale across a surface and in some cases, perpendicular to the surface. These capabilities have propelled CNMS to the forefronts of these fields and engendered a vibrant user program. The purchase order for a new STM operating at temperatures down to 40 mK with vector magnetic fields up to 9,2,2 T has been placed with Unisoku, which will open opportunities for measuring atomically resolved spin structures and excitations, including novel quantum quasiparticles. Similarly, ORNL’s acquisition of a true low-temperature Scienta-Omicron 4-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 that 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 Hermes MAC-STEM and aberration-corrected ultrahigh-energy-resolution EEL spectrometer, which will push the limit of achievable energy resolution to the sub-10meV regime. A cryo-stage upgrade (reaching liquid nitrogen temperatures) to ORNL's MAC-STEM is scheduled within the first half of 2021. The upgrade will allow us to better study quasiparticle coupling such as phonons, plasmons, and excitons in 2D materials, as well as explore the role of Moire potentials at LDs and to begin exploring the emergence of superconductivity and magnetic phases at the nanometer scale.

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 G-mode acquisition to higher speeds to examine the dynamics of ionic and electronic motion. Special attention here will be put towards the development of the workflows that allow consistent and traceable data analysis and allow for deployment in the form of user accessible notebooks
- continued development of chemical imaging methods that utilize either optical spectroscopy or mass spectrometry systems with highly localized sample volumes to provide insight into relationships between structural, chemical, and functional properties.

- 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, which will serve to strengthen interactions and collaboration with SNS and HFIR and enable advances in QIS, now being facilitated by the incorporation of the ultrahigh-energy resolution, aberration-corrected EELS on the MAC-STEM with world-leading energy resolution (approaching 5 meV) and a stable cryo-stage;
- 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 these methods accessible across the entire suite of CNMS STEM and APT instrumentation/facilities);
- ensure fully remote operation of selected instruments, including the local electrode atom probe, FIB, and two aberration-corrected STEMs, one with monochromation and cryo-capabilities;
- initiate operation of two specialized and dedicated Cryo-EM instruments - the new Thermo Fisher Scientific Titan Krios and Talos L120C Cryo-TEMs for low-dose, low-voltage imaging, single particle reconstructions, and spectroscopic analysis of biological materials, soft matter and energy materials;
- expand expertise in spin dynamics and 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;
- implement quantitative CL-SEM for improved understanding of single photon emitters and 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 and environmental chamber developments;
- 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 and environmental chamber developments;
- 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;
- Utilize advanced data analytics methods to increase the resolution of imaging modalities, increase the efficiency of data collection, and sample relevant regions of the parameter space to maximize physics knowledge gain. Here, methods including Bayesian optimization, Gaussian process regression and compressed sensing in combination with

data streaming approaches can provide the necessary toolkit to enable advances in all forms of microscopy;

- Applications at the edge and near-edge for immediate or near real-time analytics and feedback in imaging systems, such as utilization of trained models on edge hardware, instrument feedback based off of algorithms running on our DGX-2 server, or high throughput computations spawned from direct observations of materials structures;
- 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;
- enhance capabilities for sample sharing and transfer between different microscopes through controlled environmental or vacuum suitcases to allow for multi-modal functional imaging of the same samples.

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 MSED at BES through two ORNL Field Work Proposals (FWPs), through enhancing research capabilities on the Nion MAC-STEM and the Scienta-Omicron 4-Probe STM. These two instruments are ORNL resources that benefit multiple research programs including the CNMS.

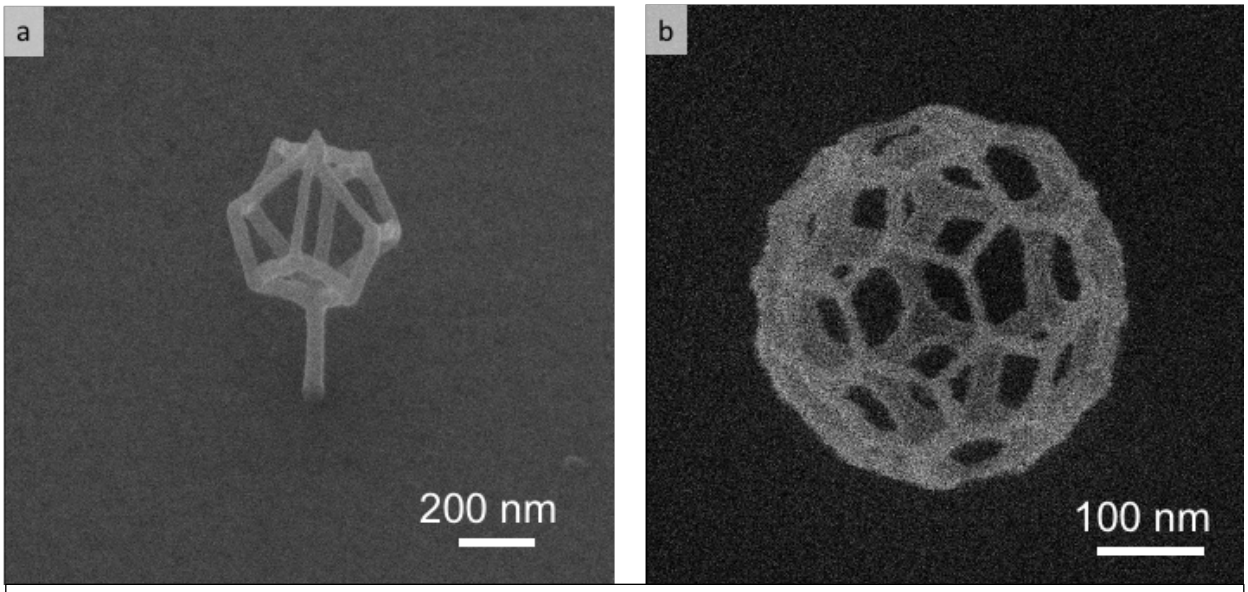
3D Direct-write Nanofabrication

The CNMS hosts several 3D direct write, or nanoprinting, capabilities in the NRL. These capabilities print at multiple length scales (micro– and nano-) while also offering cross-compatibility. Two–photon polymerization enables the micro/nano scale printing for host of polymers while charged particle induced nanoprinting, available on multiple instruments, makes is possible to deposit interconnected metallic/composite nanowires at the nanoscale.

Two-photon polymerization using the Nanoscribe at CNMS, provides a path to creating arbitrary 3D structures with feature sizes approaching 100 nm. Ongoing work emphasizes 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 creates novel opportunities for the user community in utilizing this tool.

3D nanoprinting using electron and ion capabilities have enabled user science in studies to interrogate plasmonic and magnetics responses in 3D nanostructures as well as the development of gas sensing elements not possible by conventional, in–plane nanofabrication. During electron and/or ion beam–induced deposition (EBID or IBID, respectively) a volatile organometallic precursor is controllably decomposed by charged particle irradiation, depositing material locally into a well-defined volume dictated by the beam spot size and dwell time. The CNMS, in an international collaboration with users, pushed EBID to the point where structures with exquisite

geometries and precise dimensions can be created, including those made of composites or high-purity metals.



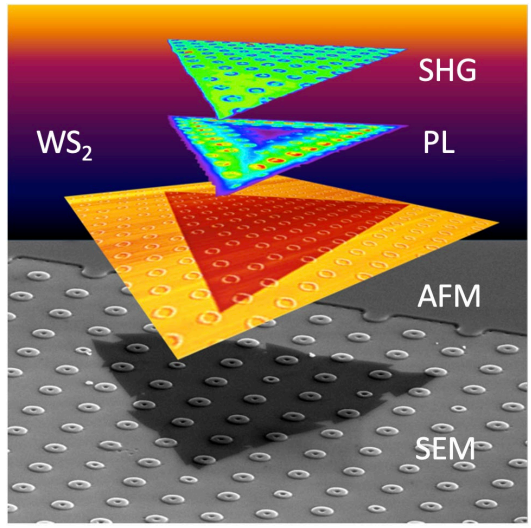
Complex 3D structures made with focused IBID in the HIM (a) deltahedron grown on a pillar and (b) a truncated icosahedron.

The capability is now fully compatible with the He ion microscope (HIM). 3D nanostructuring of metallic structures with focused (F) IBID in vacuum is possible using the HIM, with a resolution that exceeds the limit imposed using electrons. Further, HIM IBID can be carried out in the liquid phase using the Protochips® closed cell architecture, for metallic and polymeric deposition.

To remain at the forefront of 3D nanofabrication, the introduction of the Raith Velion ion patterning system provides the ability to expand into the foray of additional ion sources. With the integration of an SEM, FIB, probing stations, gas-injection, and interferometric stage, this system allows us to fully capitalize on the initial investment in the FEBID/FIBID 3D technology. The interferometrically controlled sample stage makes it possible to precisely integrate direct-write with complex wafer-scale elements or general cleanroom process flows. Ion induced deposits from Si ion species, while being Si device/substrate compatible, will be used to improve/tailor the deposit composition. Further, the improvement in deposit purity afforded by the use of ions relative to electrons will make it possible to enhance energy flow during deposition and avoid temperature gradients by improving materials quality via purity or, surprisingly, via reduced defects. Further efforts to use heavier ions like Si, Ge, and Au, that are available for this system will enhance the veracity and throughput necessary for other applications.

Nanomaterials Synthesis and Assembly

As described in the BRN reports for Synthesis Science and Quantum Materials (mentioned in Section 1), 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, 2D materials have emerged as a platform for theme science at CNMS because they are computationally tractable and each atom can be imaged individually. The explosion of interest in atomically thin 2D 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 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 field work proposal (FWP) addresses 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.

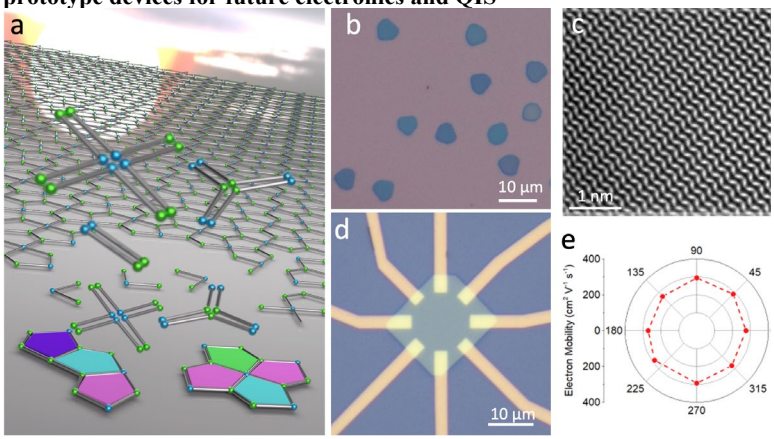


Strain in 2D materials can significantly impact growth and functionality. Fast and efficient characterization of strain distribution is important for their potential applications (in optoelectronics). A synergistic characterization strategy combines PL, SHG, Raman, and SEM to measure same location on a 2D crystal, which is able to reveal correlations between growth, strain, and optical properties.

Controlled processing and assembly of systems in an inert glovebox atmosphere for exfoliation and stamping of 2D materials has also been developed. Associated h-BN encapsulation, dry transfer, e-beam lithography, and other integration methods have been developed to characterize, manipulate 2D crystals, make quantum devices, and measure quantum transport in 2D layered and other quantum materials. This system allows us to exfoliate, transfer, and encapsulate air sensitive 2D materials, such as chalcogenides and chlorides, and build heterostructures for 2D electronics and quantum devices. This system is in high demand by users for the preparation of samples for functional testing and STEM characterization. It will also be critical for synergistic research with scientists working within the QSC at ORNL, which is focused on exploring 2D topological quantum materials. Ongoing efforts are focused on integrating a Raman microscope and AFM into the glove box environment to characterize the thickness and stacking orientation of 2D crystals and heterostructures *in situ*.

To meet the needs of users for the rapid exploration of atomically thin materials, the *in situ* diagnostic-guided approaches developed at CNMS that utilize non-equilibrium laser-based synthesis and processing of TMDs and other quantum materials will be expanded and emphasized to provide remote user capabilities, and the development of autonomous AI/ML-guided synthesis approaches. PLD and laser processing are especially well suited to the rapid exploration of atomically thin layers and thin epitaxial heterostructures. The new high-throughput PLD synthesis platform (see QIS section) will enable the synthesis of many new families of atomically thin layers, such as chalcogenides and chlorides, that are envisioned for emerging quantum materials. The system includes reactive/toxic gas handling, integrated load-lock 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 stoichiometric control.

Exploring the electrical properties of novel atomically thin 2D materials in prototype devices for future electronics and QIS



2D materials and heterostructures exist in many atomic, electronic, and quantum phases offering tremendous opportunities for future applications in next-generation electronics, photovoltaics, and QIS. Recently, we developed a CVD method for growing atomically thin $PdSe_2$, a novel noble metal dichalcogenide with an unusual pentagonal symmetry and a puckered layer structure (a-c). Laser spectroscopy measurements revealed strongly in-plane anisotropic optical properties. (d) To probe its anisotropic electrical transport properties, single flakes of $PdSe_2$ were addressed with electrical contacts via e-beam lithography into prototype field-effect transistors. These transistors displayed tunable ambipolar charge carrier conduction with electron mobilities as high as ~294 cm^2/Vs for a 9-nm thickness device and low electrical anisotropy (e), which is comparable to that of exfoliated $PdSe_2$.

Synthesis strategies that utilize the variety of pulsed laser sources and tunable wavelengths available 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, PL, and PL lifetime measurements, micro-absorption, second harmonic generation (SHG), and ultrafast pump-probe spectroscopy to characterize LD 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 atomic-resolution STEM 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 TMDs 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. In addition, to explore and understand the quantum properties of 2D topological quantum materials (1T' MoS_2, $MnBi_2Te_4$, and $PdTe_2$), Hall devices will be fabricated by the controlled assembly of 2D crystals. Integration with deterministic electrode patterns will be enabled by a combination of 3D direct-write nanofabrication and electron beam (e-beam) lithography in the NRL at CNMS. Hall measurements of the integrated materials will be performed in mK temperature and magnetic fields to understand the quantum spin Hall effect and quantum anomalous Hall effect in 2D topological quantum materials.

The characterization and control of ultrafast quasiparticle interaction dynamics in materials at scientifically relevant scales generally requires an ensemble of complementary microscopies and spectroscopies. The new CL-SEM (described in detail in section *Opportunities in Quantum Information Science)* is a unique tool for multi-dimensional characterization and control with sub-ps timing resolution, few nm 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. This newly installed microscope combines a Thermo Fisher FEI Quattro environmental SEM with a Delmic Sparc CL collection system and a Gatan cryostage. Ultrafast operation is enabled by a customized beam-blanking system (for sub-100 ps timing resolution) and a fs laser-pumped electron gun (for sub-ps timing resolution). Further, *in situ* material modification is enabled by EBID with a dimethyl gold precursor and e-beam patterning in a water vapor background. Together, these capabilities enable the characterization of spatially, spectrally, temporally, polarization, angle, and temperature resolved excitation energetics and dynamics in nanoscale material systems.

5. User Needs and Outreach

Anticipating and Accommodating User Growth

As a 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 2015, 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

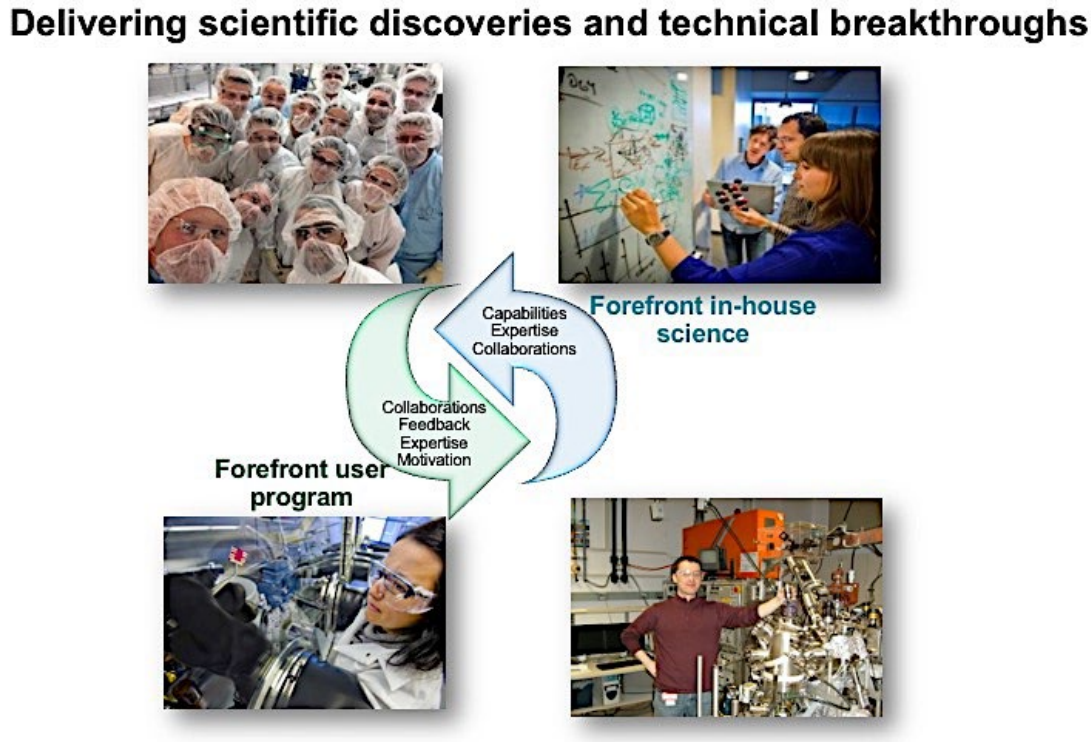
investing the resources into more specialized efforts). Despite the initial loss of "capacity" users, the combined program quickly re-established growth in user numbers and productivity: Over the time interval from FY2015 to FY2019, the user numbers grew to over 650 users in FY2019; the number of peer-reviewed scientific publications in regular journals increased to 403, 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 sustain in the long term 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) will 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 UEC, feedback received at the annual user meeting, and evaluation of our strategies with our SAC, 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 ML/AI approaches 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.

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.



The fundamental NSRC model of "make, characterize, understand" inspires vibrant engagement from the user community at CNMS and other NSRCs, leading to strong collaborations and feedback that guides forefront exploration in nanoscience and new developments (images are from various NSRCs).

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. The CNMS website (cnms.ornl.gov) has recently been revised to present information about the user effort in a more accessible way and the capabilities of all NSRCs are compared and searchable on the NSRC-common portal website (nsrcportal.sandia.gov); however, 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.

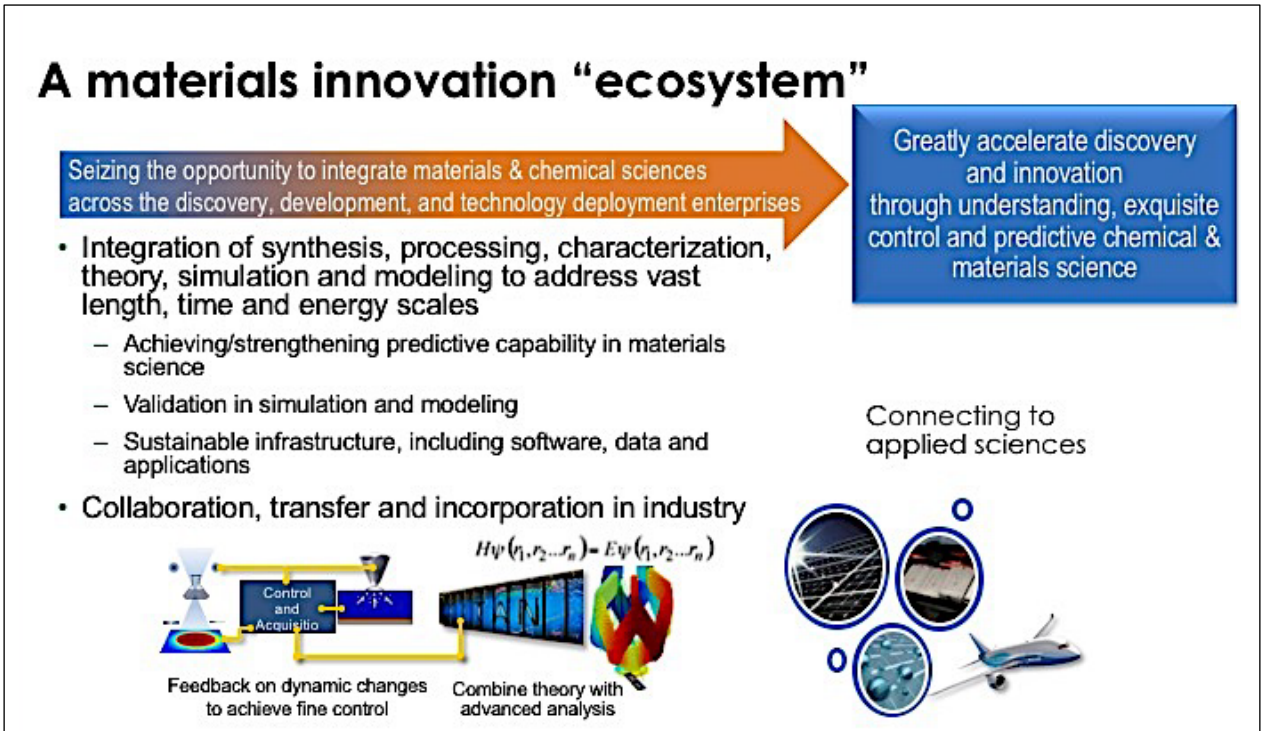
CNMS will continue to partner with the other NSRCs to hold specific events (such as the 1-day Presidential Symposium at the American Chemical Society Conference that featured NSRC user work in the area of ionic transport and prominent participation on various user facility panels, workshops, etc.) and feature booths at conference 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 CNMS 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 previous years, CNMS actively reached out to various institutions through targeted visits. Examples include events such as a mini-workshop at Georgia Tech (2017) focused 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 BES Roundtables on Next-Generation Quantum Systems (2017) and Producing and Managing Large Scientific Data with Artificial Intelligence and Machine Learning (2019), both of which included 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 ~5% of all on-site users), 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 industry 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 aspires to help solve industrially relevant basic science problems and we continue to grow the user base in this important area.

CNMS researchers continuously develop licensable intellectual property, as evidenced by 6 patents issued in FY2019 and several licensed technologies. The development of new intellectual property is consistent with the CNMS's position as a world-class research institution.

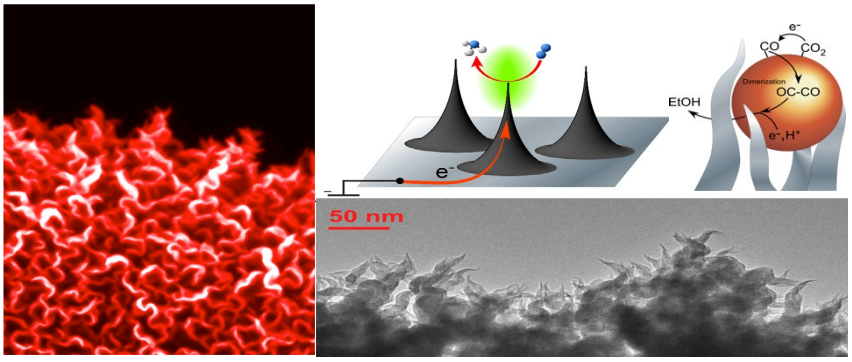
CNMS has several industry sponsored research projects and industrial collaborations. A few examples include:

- Proctor & Gamble (P&G) has worked with CNMS for 8 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 worked on understanding the factors that 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.

Enabling nontraditional catalytic materials for effiecent, scalable and renewable catalysis



Carbon nanospikes (CNS), formed from a PECVD process, can act as room temperature/pressure conduits for electrons to promote reaction with other chemicals, by creating high localized electric fields (green cloud). In an electrolytic cell, an electrode composed of CNS can enable catalytic reduction of common chemicals to high-value products (shown are nitrogen to ammonia and carbon dioxide to ethanol).

Despite these successes, CNMS leadership is determined to improve the accessibility of CNMS resources for industrial users since 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 including more industrial scientists in the proposal reviewer pool and the CNMS recently initiated an Industrial Liaison team to identify and work with potential industrial users to help them better understand and navigate the proposal submittal and review process. In addition, the CNMS and SNS/HFIR will host the 2nd Annual Industrial Workshop following the successful Industrial Workshop hosted by Brookhaven National Laboratory in December 2020. 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 re-emphasizing 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 intellectual property (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.

Unique Strengths for Specific User Areas

In addition to the strategic areas discussed in Sect. 4, the broad range of capabilities and resources at CNMS attracts users from many areas of research that are not (or not anymore) at the heart of our research activities. 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 lithography and dedicated APT sample preparation facilities (cryo-plasma focused ion beam (PFIB) instrument). The NSRC Recapitalization Project (currently undergoing CD1/CD3A review) is providing a mechanism for CNMS to acquire the cryo-PFIB for APT as well as a one-of-a kind liquid-He-cooled MAC-STEM. These two instruments are not expected to come online and be available to the user community for several years but are integral to the future growth of the CNMS.

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 before this, we continue to invest ~10% of our total annual funds into recapitalization.

With the cost of liquid helium and the reliability of its supply being significant issues, a plan for installing a site-wide He-recovery system is also being considered, including the NMR instrument. Recovering helium has become much easier because modern re-liquefiers require very little maintenance and staff attention. Possible use of laboratory overhead funds is being considered.

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, the low-temperature 4-probe SPM, and the SAXS/WAXS capability, 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 on important proposal initiatives (Early Career Award proposals, EFRC proposals, etc.) and to attend 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 funding 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, MSA, 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 and Group Leader positions. In 2019, two CNMS early career research staff received Office of Science Early Career Awards and another received a Presidential Early Career Award for Science and Engineering (PECASE).

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! 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
AAAS	American Association for the Advancement of Science
AC	alternative current
AFM	Atomic Force Microscopy
AI	Artificial Intelligence
ALCC	ASCR Leadership Computing Challenge
AML	Advanced Microscopy Laboratory
APS	American Physical Society
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
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
CPU	Central Processing Unit
CSGB	Chemical Sciences, Geosciences, and Biosciences (BES)
CVD	Chemical Vapor Deposition
DIB	Droplet Interface Bilayer
DNA	Data NanoAnalytics
DNT	Directed Nanoscale Transformations
DOE	Department of Energy
DSC	Differential Scanning Calorimetry
EAPM	Electron & Atom Probe Microscopy
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
F-AFM	Functional Atomic Force Microscopy
FDM	Fused Deposition Modeling
FHN	Functional Hybrid Nanomaterials
FIB	Focused Ion Beam
FPGA	Field-Programmable Gate Array
FTIR	Fourier-Transform Infrared Spectroscopy
FWP	Field Work Proposal
GPC	Gel Permeation Chromatography
GPU	Graphics Processing Unit
HFIR	High Flux Isotope Reactor
HIM	Helium Ion Microscopy
HNM	Hysteretic Nanomaterials
HPC	High Performance Computing
HQM	Heterogeneities in Quantum Materials
IBID	Ion Beam Induced Deposition
INCITE	Innovative and Novel Computational Impact on Theory and Experiment
IP	Intellectual Property
IR	Infrared
KPFM	Kelvin Probe Force Microscopy
LD	Low-dimensional
LDRD	Laboratory Directed Research and Development
MAC-STEM	Monochromated, Aberration-Corrected STEM
MALDI	Matrix Assisted Laser Desorption Ionization
MCC	Materials Characterization Core
MDN	Metastability in Driven Nanomaterials
MF	Molecular Foundry
ML	Machine Learning
MMÅ	Materials MicroÅnalysis
MPMS	Materials Properties Measurement System (Quantum Design)
MRS	Materials Research Society
MS	Mass Spectrometry
MSA	Microscopy Society of America
MSED	Materials Sciences and Engineering Division (BES)
NERSC	National Energy Research Scientific Computer enter
NMR	Nuclear Magnetic Resonance
NNI	National Nanotechnology Initiative
NOMAD	Nanoscale-Ordered Materials Diffractometer (SNS)
NR	Neutron Reflectometer
NRL	Nanofabrication Research Laboratory
NSRC	Nanoscale Science Research Center
NTI	Nanomaterials Theory Institute
OLCF	Oak Ridge Leadership Computing Facility
ORNL	Oak Ridge National Laboratory
PECASE	Presidential Early Career Award for Scientists and Engineers
PECVD	Plasma Enhanced Chemical Vapor Deposition
PFM	Piezoresponse Force Microscopy

PL	Photoluminescence
PLD	Pulsed Laser Deposition
POWGEN	Power Diffractometer (SNS)
PPMS	Physical Property Measurement System
QCM	Quartz Crystal Microbalance
QIS	Quantum Information Science
QSC	Quantum Science Center
R&D	Research & Development
SAC	Scientific Advisory Committee
SAXS	Small Angle X-ray Scattering
SBMS	Standards Based Management System
SC	Office of Science
SCEM	Scanning Electrochemical Microscopy
SEM	Scanning Electron Microscopy
ShaRE	Shared Research Equipment User Program
SIMS	Secondary Ion Mass Spectrometry
SLA	Stereolithographic Apparatus
SLS	Selective Laser Sintering
SNS	Spallation Neutron Source
SP	Spin Polarized
SPM	Scanning Probe Microscopy
SPP	Strategic Partnership Program
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
UHV	Ultra-High Vacuum
UV-Vis	Ultraviolet-visible
VISION	Vibrational Spectrometer (SNS)
VSP	Viscous Solution Printing
VULCAN	Engineering Materials Diffractometer (SNS)
WAXS	Wide Angle X-ray Scattering
XPD	X-ray Photoelectron Diffraction
XPS	X-ray Photoelectron Spectroscopy
XRD	X-ray diffraction