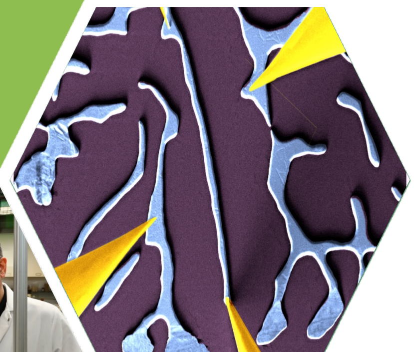
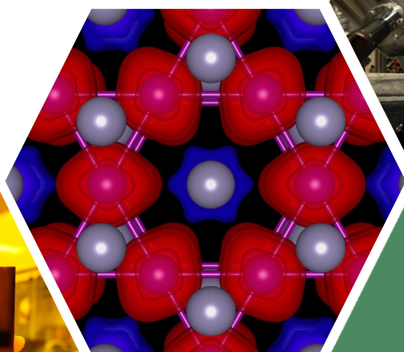
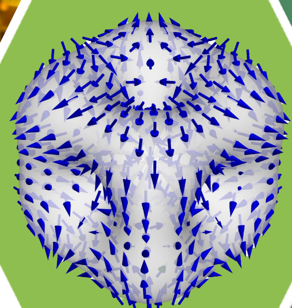
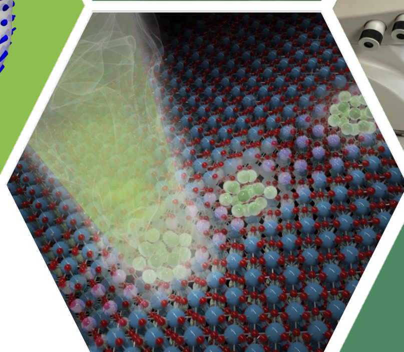


5-Year Strategic Plan for the **Center for Nanophase Materials Sciences (CNMS)**

FYs 2024-2028







updated March 2024

OAK RIDGE National Laboratory | CENTER FOR NANOPHASE MATERIALS SCIENCES

Executive Summary

The Center for Nanophase Materials Sciences (CNMS) is a Nanoscale Science Research Center 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. The CNMS research effort not only contributes to enhancing our mechanistic understanding of nanomaterial properties, but also leads to the development of advanced capabilities in synthesis, characterization, imaging, and theory, modeling, and simulation that will benefit future users. CNMS' research efforts are organized into three research themes that catalyze new nanoscience capabilities and knowledge for quantum information science, novel polymer synthesis, and multiscale dynamics, 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.

As CNMS and the broad research community continue to advance new research, novel capabilities must continually be acquired or developed to address new challenges and exploit new opportunities. CNMS leadership, working with its User Executive Committee and Scientific Advisory Committee, 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.

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, 2D materials, and the 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, the Spallation Neutron Source, and the High Flux Isotope Reactor.

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.

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Abbreviations/Acronyms

AFM	atomic force microscopy
AI	artificial intelligence
AML	Advanced Microscopy Laboratory
APT	atom probe tomography
AutoFlowS	autonomous continuous flow reactor synthesis
BES	Basic Energy Sciences
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
Cryo-EM	cryogenic electron microscopy
CVD	Chemical Vapor Deposition
DNA	Data NanoAnalytics
DOE	Department of Energy
DSC	differential scanning calorimetry
EELS	Electron Energy Loss Spectroscopy
ESH&Q	Environmental, Safety, Health, and Quality
F-AFM	Functional Atomic Force Microscopy
FHN	Functional Hybrid Nanomaterials
FIB	focused ion beam
GPC	gel permeation chromatography
HCMC	Harnessing Complex Macromolecular Conformations
HFIR	High Flux Isotope Reactor
HIM	Helium Ion Microscopy
HPC	High Performance Computing
HQM	Heterogeneities in Quantum Materials
ICE	Institutional Capital Equipment
IR	infrared
LEAP	local electrode atom probe
LDRD	Laboratory Directed Research and Development
LS	light scattering
Macro	Macromolecular Nanomaterials Group
MAC-STEM	monochromated, aberration-corrected STEM
MALDI	Matrix Assisted Laser Desorption Ionization
MBE	molecular beam epitaxy
MCC	Materials Characterization Core
MD	Multiscale Dynamics
MF	Molecular Foundry
ML	machine learning
MMÅ	Materials MicroÅnalysis
MPMS	Materials Properties Measurement System (Quantum Design)
MS	Mass Spectrometry
NC	Nanomaterials Characterization
NERSC	National Energy Research Scientific Computer Center
NMR	nuclear magnetic resonance
NRL	Nanofabrication Research Laboratory
NS	Nanomaterials Synthesis
NSRC	Nanoscale Science Research Center
NTI	Nanomaterials Theory Institute
NV	nitrogen vacancy
OLCF	Oak Ridge Leadership Computing Facility
OPO	optical parametric oscillator
ORNL	Oak Ridge National Laboratory
PFIB	plasma focused ion beam
PiFM	photoinduced force microscopy
PL	photoluminescence
PLD	pulsed laser deposition
PZ	polyzwitterion
QCM	quartz crystal microbalance
QIS	Quantum Information Science
QSC	Quantum Science Center
RHEED	reflective high-energy electron diffraction
RIE	reactive ion etcher
RT	room temperature
SA	Specific Aim
SAC	Scientific Advisory Committee
SAXS	small-angle x-ray scattering
SBMS	Standards Based Management System
SC	Office of Science
SEM	scanning electron microscopy
SIMS	secondary ion mass spectrometry
SMIM	scanning microwave impedance microscopy
SNOM	scanning near-field optical microscopy
SNS	Spallation Neutron Source
SP	spin polarized
SPE	single photon emitter
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
TACS	Theory and Computation Section
TEM	transmission electron microscopy

TGA	thermogravimetric analysis
TOF-SIMS	time of flight - secondary ion mass spectrometry
UEC	User Executive Committee
UHV	ultrahigh vacuum
UV-Vis	ultraviolet-visible
WAXS	wide-angle x-ray scattering

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, 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.

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

SC Office of Basic Energy Sciences (BES) Basic Research Needs Workshops and Roundtables provide strategic guidance to CNMS for planning research and illustrate many of the future needs of users:

- Neuromorphic Computing (October 2015)
- Quantum Materials for Energy Relevant Technology (February 2016)

[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.

- 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)
- Opportunities for Quantum Computing in Chemical and Materials Sciences (October 2017)
- Opportunities for Basic Research for Next-generation Quantum Systems (November 2017)
- Microelectronics (October 2018)
- Chemical Upcycling of Polymers (April 2019)
- Producing and Managing Large Scientific Data with Artificial Intelligence and Machine Learning (October 2019)
- Transformative Manufacturing (March 2020)
- Office of Science User Facilities Lessons from the COVID Era and Visions for the Future (December 2020)
- Physical Science Enabled by Cryogenic Electron Microscopy (May 2021)
- Foundational Science for Carbon-Neutral Hydrogen Technologies (August 2021)
- Foundational Science for Carbon Dioxide Removal Technologies (March 2022)
- BES Network Requirements Review (March – September 2022)

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 an important 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 also pursue their own scientific careers and leadership opportunities. Our in-house research effort, organized in the three scientific themes, 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, for example BES Energy Frontier Research Centers. CNMS also leverages 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, and equipment programs such as the Institutional Capital Equipment (ICE) Program. These efforts continue to play an important role in maintaining scientific diversity, providing leadership opportunities to early- and mid-career researchers, maintaining vibrant communities consisting of a balance of students, postdocs, and permanent staff, and investing in new equipment for the laboratory.

Modern materials science research increasingly relies on a very direct integration not only of theory, modeling, and simulation, 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.

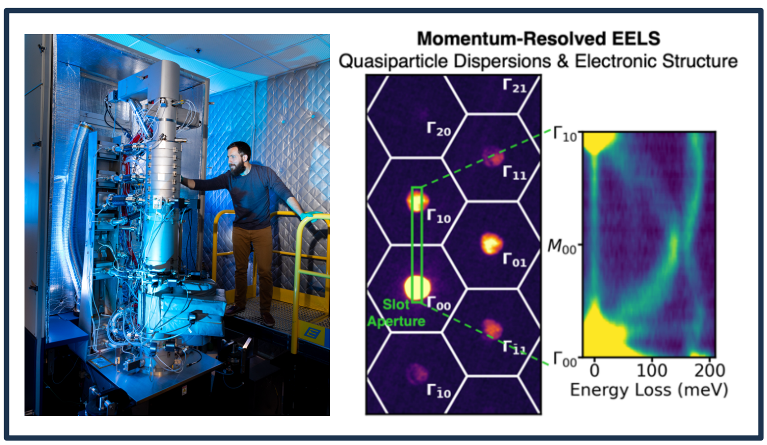
DOE fosters an environment in which the five NSRCs collaborate and coordinate activities, while at the same time critically evaluating each of the NSRCs in terms of well-defined metrics (primarily the number of users and number of publications). Each year CNMS supports >750 unique users from more than 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. Collectively, CNMS staff and users publish more than 400 scientific articles in peer-reviewed journals per year, with more than half of the publications appearing in journals with an impact factor > 7, which are considered high-impact journals by the BES Scientific User Facilities Division (SUFD). In addition, CNMS researchers were lead or co-inventors on more than >100 patents since 2006. Reflecting the impact of CNMS research, we note that more than 150,000 papers have cited CNMS publications since its inception.

2. CNMS Research Groups

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 and expert staff, with researchers from different disciplines working closely together.

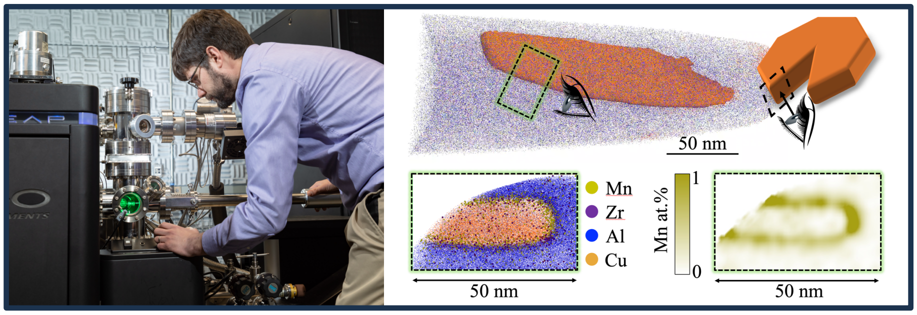
All researchers at CNMS are responsible for working with users and for performing in-house science within the framework of the DOE-reviewed CNMS program. Research staff are expected to split their SUFD-funded effort between in-house research and user support; technicians and technical staff generally play a large role in the user effort through their direct involvement with instrumentation, which is dedicated 80% of the time to the user effort.

Nanomaterials Characterization (NC) Section:

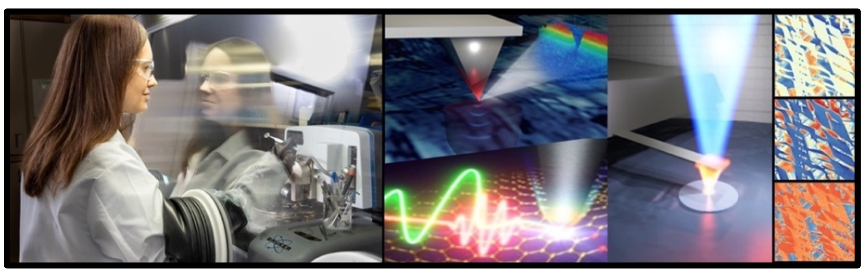


Scanning Transmission Electron Microscopy (STEM) *"advancing atomic-scale e-beam imaging and spectroscopy to enable new understanding of materials, quantum phenomena, and energy technologies."* The STEM group excels at understanding the structure, properties, and functions of materials at the nanoscale. This understanding is critical to advancing many areas of modern technology. STEM is a powerful characterization technique used to probe the structural, chemical, magnetic, and electrical properties of a material. We use the *Z*-contrast imaging mode to image composition at the atomic scale, Lorentz imaging mode to map in-plane magnetic textures and their response to various external stimuli, and ultrahigh-energy resolution electron energy-loss spectroscopy (EELS) to probe local vibrational, optical, and electronic responses at variable temperatures. When combined with theory, computation, and x-ray and neutron scattering capabilities offered by other groups at ORNL, the state-of-the-art CNMS microscopy facilities offer many complementary routes to analyze next-generation materials. Users are provided access to staff with considerable expertise in the capabilities of these instruments. This expertise, when added to access to high-quality electron microscopes, is an attraction for many users. Responsibility for maintaining the STEM instruments is shared with the Materials MicroÅnalysis (MMÅ) group.

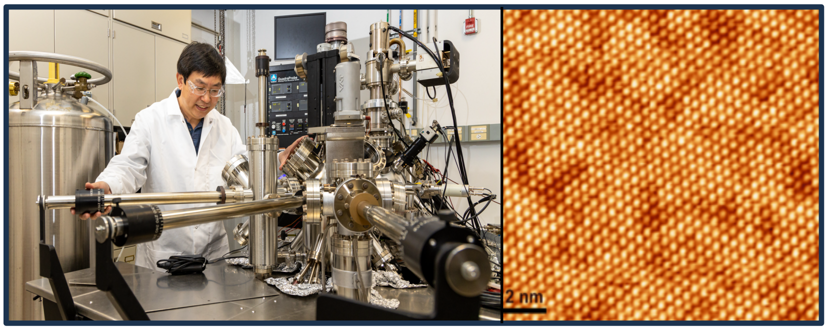
Materials MicroÅnalysis (MMÅ) *"understand the structure, chemistry, and function of materials through the development and application of advanced, state-of-the-art analytical microscopy, APT, in situ microscopy, and cryogenic electron microscopy (Cryo-EM)."* The MMÅ group specializes in the development and application of advanced analytical electron microscopy and APT to understand materials structure and chemistry at the atomic level. The extensive array of transmission electron microscopy (TEM)/STEM instruments available in both the MMÅ and STEM groups has enabled new studies specifically aimed at understanding the physical phenomena responsible for controlling materials performance. The STEM and MMÅ groups oversee and maintain the laboratories/facilities located on ORNL's central campus, including an advanced array of specimen-preparation facilities. MMÅ staff are developing new imaging, spectroscopy, and operando techniques necessary to enable new scientific discoveries. The group is responsible for providing users with access to state-of-the-art analytical STEM and APT capabilities as well as staff expertise—and recently, Cryo-EM—that can be coupled directly with synthesis and performance studies conducted at CNMS or to perform standalone experiments to characterize novel materials prepared at their home institutions.



Functional Atomic Force Microscopy (FAFM) *"advancing scanning probe microscopies (SPMs) and spectroscopies to capture the nanoscale origins of functional properties in materials for energy and information."* The FAFM group is dedicated to advancing the capabilities of SPM. Our team explores new ways to use the tip of the AFM to induce concentrated strain, electrical, thermal, electromagnetic, and other fields at the material's surface and characterize their response through minute displacements of the surface, current flow from the tip to the surface, or optical spectroscopy of light emitted from the tip–surface junction to learn about the behavior of materials at fundamental length scales from tens of nanometers to the atomic scale. We leverage and develop cutting-edge data acquisition and control systems to precisely deliver complex electrical, photonic, and thermal signals to the sample and to capture the response. By scanning the tip across the surface while performing measurements, we can create high-resolution maps of the behavior of advanced materials from multiple stimuli at multiple scales. This information can reveal the ways that material inhomogeneities influence macroscale behavior. In collaboration with the CNMS Data NanoAnalytics (DNA) group, we develop and integrate novel artificial intelligence (AI)-based approaches to improve the quality and efficiency of imaging and spectroscopy and to improve analysis of the data we acquire. We aim to deliver these capabilities and offer our expertise to the user community.



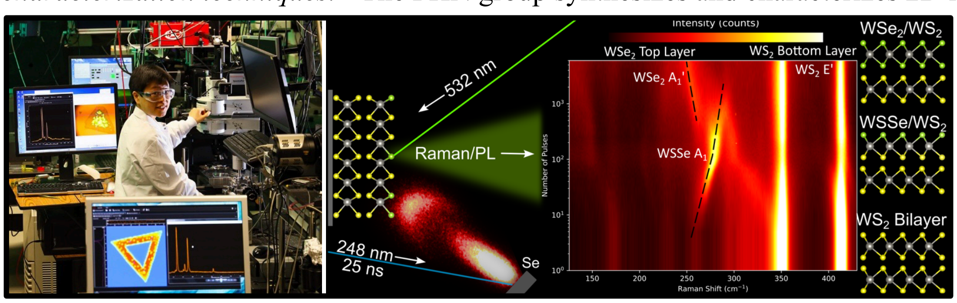
Scanning Tunneling Microscopy (STM) *"understand and control the correlations of atomic structure with electronic, magnetic, and transport properties in quantum materials through the development of STM methods."* The STM group specializes in atomic-resolution imaging, electronic



density of states mapping, nanoscale electron transport surveys, and atom and molecule manipulating through the development and utilization of STM techniques. Research is focused on establishing correlation of physical properties with atomic structures and controlling material processes at the atomic scale to tailor electronic wave functions and interactions. Unique capabilities include the only dilution refrigeration vector magnet STM instrument available in a user facility, the only spin-polarized four-probe STM instrument in the world, and a new scanning nitrogen vacancy (NV) microscope. These instruments cover a wide range of sample environments, including temperatures of 40 mK–300 K, magnetic fields up to 9 T, and ultrahigh vacuum (UHV). The STM systems are integrated with material growth capabilities by molecular beam epitaxy (MBE), pulse laser deposition (PLD), and substrate-assisted on-surface chemical reactions. Samples can be shared between microscopes via vacuum suitcases. These capabilities have enabled the first identification of skyrmions in a van der Waals magnet, the first direct evidence of spin–momentum locking in topological materials through the spin chemical potential, the first direct observation of single-vacancy-based nonvolatile resistive switching, and the first demonstration of single-molecule telegraphy across a surface.

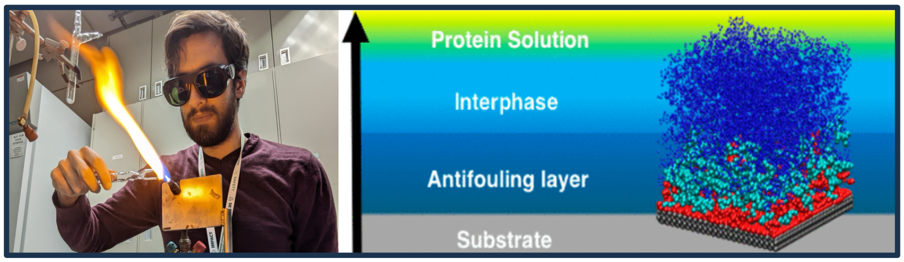
Nanomaterials Synthesis (NS) Section:

Functional Hybrid Nanomaterials (FHN) *"understand and control nanomaterials synthesis and assembly to elicit special functionalities by integrating real-time diagnostics and advanced characterization techniques."* The FHN group synthesizes and characterizes 2D materials, thin films, and

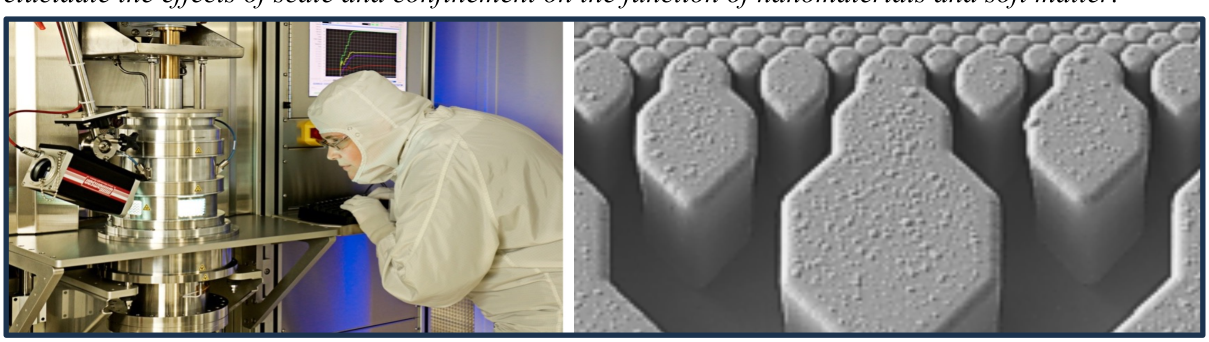


hybrid architectures using a variety of techniques. Synthesis and processing methods include PLD, chemical vapor deposition (CVD) and transport, exfoliation of 2D materials and atmospherically controlled stamping, and other techniques. The group specializes in optical spectroscopy characterization of nanomaterials and methods to employ these as *in situ* diagnostics of a material's structure and properties during synthesis or *in operando* in response to controlled environments, catalytic reactions, or optical and electrical stimulus. The *in situ* diagnostic data acquired during synthesis is used to develop growth models for carbon nanotubes, graphene, transition metal dichalcogenides, Janus monolayers, and thin films. The group is heavily involved in automating synthesis incorporating *in situ* diagnostics and characterization to enable the development AI and machine learning (ML) approaches for autonomous synthesis. Signature automated PLD facilities incorporate *in situ* reflective high-energy electron diffraction (RHEED), x-ray photoelectron spectroscopy/diffraction (XPS/XPD), Raman spectroscopy, and data acquisition. Raman, photoluminescence (PL), second harmonic generation, ultraviolet–visible (UV-vis) spectroscopy, and reflectance spectroscopies are used to map 2D materials and develop optoelectronic, sensing, and catalytic functionalities. Multimodal "lab on a crystal" quartz crystal microbalance (QCM)-based platforms are used to correlate electrical, optical, and gravimetric/viscoelastic properties of thin films with environmental effects using AI/ML. Raman and PL microspectroscopy are performed in several different environments, including rapid mapping capabilities, tunable laser excitation, low-frequency Raman modes, and cryogenic environments. Ultrafast laser microspectroscopy is used to measure quasiparticle dynamics of 2D materials and heterostructures, and cathodoluminescence (CL) spectroscopy with nanometer resolution is coupled with single superconducting nanowire detection for quantum correlation measurements of single photon emitters (SPE).

Macromolecular Nanomaterials ([Macro]) *"understand and design novel macromolecular and deuterated nanomaterials using cutting-edge synthesis and characterization methods."* Much interest is focused on investigating nanophase behavior in soft matter. This investigation requires exploration of new synthetic routes and controlled macromolecular microstructures. The group is focused on designing and developing novel macromolecules and deuterated nanomaterials driven by computation and AI/ML strategies using cutting-edge synthesis methods and characterization techniques. Utilizing computational methods from atomistic to coarse-grain methods, we can report on multiphase behavior and complex fluids to guide the synthesis and understand phenomena. A primary mission is to support neutron studies with deuterated probes and macromolecules. Advanced macromolecular characterization methods available in the Macro labs include nuclear magnetic resonance (NMR) spectroscopy, matrix-assisted laser desorption/ionization (MALDI), and solution characterization with multidetector gel permeation chromatography (GPC) and light scattering (LS). Solid-state and thermomechanical characterization utilizing differential scanning calorimetry (DSC), thermogravimetric analysis (TGA), dynamic mechanical analysis, rheology, and dielectric spectroscopy complement x-ray and neutron scattering and TEM for investigating macromolecular phase separation and morphologies. We are developing continuous flow reactions and polymerization methods driven by AI/ML workflows and operando reaction monitoring.



Nanofabrication Research Laboratory ([NRL]) *"advancing nanofabrication processes to evoke and elucidate the effects of scale and confinement on the function of nanomaterials and soft matter."*

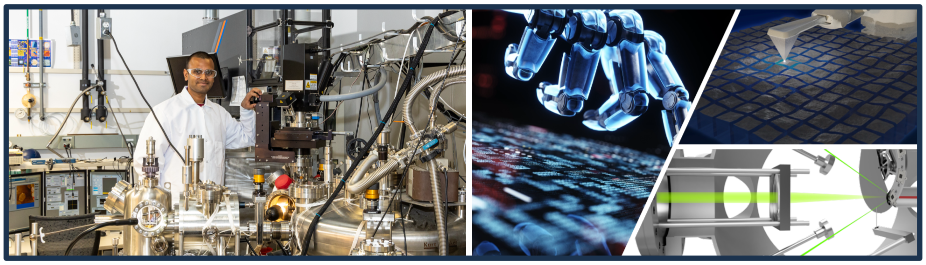


The complex work of unraveling and harnessing the unique properties of nanomaterials requires the ability to shape, manipulate, and integrate them across length scales. The NRL leverages a suite of tools, adapted from conventional semiconductor processing technologies, to develop workflows that translate materials discovery into functional architectures. We use these tools to address a broad range of scientific questions related to transport phenomena, light–matter interactions, nanomechanics, and bio-material interfaces. Moreover, pioneering work in understanding the use of ion and electron beam (e-beam) induced chemistry to shape and modify materials, as well as the continued pursuit of understanding material transformations that occur during integration and processing, is foundational to our research. The NRL endeavors to integrate top-down and bottom-up fabrication and synthesis strategies to integrate unique inorganic and soft matter materials into functional systems used in sensing, information processing, memory, and other energy applications. The NRL maintains a full suite of photolithography, e-beam lithography, direct laser writing lithography, wet and dry etch processes, and a variety of chemical and physical vapor deposition methods to process samples. Direct-write processes such as multiple ion species focused ion beam (FIB), e-beam-induced chemistry, and two-photon polymerization offer rapid 3D prototyping and direct materials property modification (e.g., defect introduction). To characterize these structures, the cleanroom houses optical microscopy, Raman spectroscopy, optical and mechanical thin film measurement, electron and ion microscopy, and AFM. New capability development is driven internally through basic research aimed at developing a more thorough understanding of the chemical and physical phenomena underlying material manipulation, processing, and integration at the nanoscale.

Theory & Computation Section (TACS):

Nanomaterials Theory Institute (NTI) *"deliver advanced theoretical, computational, and ML capabilities that can provide a foundation for fundamental understanding and prediction of materials and their function, and chemical processes at the nanoscale."* The NTI group delivers advanced theoretical, computational, and ML capabilities that can provide a foundation for fundamental understanding and prediction of materials, their function, and chemical processes at the nanoscale, and help interpret as well as steer novel experiments. NTI staff have expertise in a wide range of electronic-structure, atomistic, coarse-grained, continuum modeling, as well as AI/ML approaches. They lead scientific research topics in quantum materials, soft matter systems, and energy materials that enable explanations and predictions of novel phenomena in close collaboration with experimentalists. Our unique strengths lie in developing and using codes at scale to understand structure, properties, dynamics, transport, and reactions in response to external stimuli, predictive treatments, and understanding strongly correlated materials, especially in the presence of heterogeneities, and large-scale simulations of soft matter systems. In addition to experiments at CNMS, we work closely with experimentalists in the neutron scattering facilities. NTI staff maintain codes on the CNMS owned internal computational cluster and retain dedicated allocations on DOE's exascale supercomputers, specifically Perlmutter at the National Energy Research Scientific Computing Center (NERSC) and Frontier at the OLCF. They lend their expertise to CNMS users to make these capabilities available for user projects. Experimental users are encouraged to contact NTI staff to discuss possible theoretical support for their projects.

Data NanoAnalytics (DNA) *“accelerate the development of autonomous research tools and workflows capable of scientific discovery in nanoscale synthesis and characterization by combining simulations, physics-driven ML methods, and instrument automation.”* The DNA group is known for developing and implementing physics-driven ML experimental workflows to increase the capabilities of instruments for both nanoscale characterization and synthesis. To this end, we develop and apply ML tools to analyze multi-modal data streams, including imaging data from microscopy, hyperspectral data (e.g., STEM-EELS), and other forms of spectroscopy. We utilize these algorithms—alongside human-in-the-loop knowledge injection, automation, simulations, and edge computing—to steer autonomous instruments and assist in materials modification and optimization and, ultimately, in discovering new physics. We are well recognized for our physics-driven approach to autonomous science. We use these new capabilities to explore frontiers in nanoscience, including to study and perturb interfaces in ferroelectric and ferroelastic materials, explore structural phase transitions as a function of electrical stress and temperature, manipulate topological structures, rearrange individual atoms deterministically to form structures with designer properties in a microscope, and optimize growth of chalcogenides to enhance opto-electronic properties. We develop autonomous science tools by combining physics-informed ML methods with automation, simulations, and edge computing to realize new computational–experimental workflows capable of new types of experiments to characterize, synthesize, and manipulate material at the nanoscale.



CNMS Operations and Support

CNMS management is responsible for all aspects of the environmental, safety, and health 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 Across the CNMS

Some of the major capabilities that are offered to the user community at CNMS are briefly listed below. Additional details can be found at cnms.ornl.gov:

- **Imaging, microscopy, spectroscopy, 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 characterizing 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 techniques for resonance imaging, high data acquisition rates, μs time resolution, and spin-dependent transport. A 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, monochromated 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. Recently, ORNL invested in a fully equipped Cryo-EM facility that is managed by the CNMS. Capabilities for 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 multidimensional data sets, process and characterization histories, etc. CNMS strives to develop and provide consistent and traceable workflows that allow end-to-end conversion of instrumental data to 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 (NRL). Chemical and biological functionalization of nanoscale materials with a special emphasis on the directed assembly of 3D structures. Synthesis of 2D materials, hybrid structures, and epitaxial oxide layers by 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 PLD synthesis platform for quantum materials 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.
- **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**: Investigations 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.
- **3D 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, UV-vis spectrophotometry, electrical and optoelectronic characterization, ultrafast optical pump-probe dynamics measurements, ultrafast nm-resolution CL, quantum

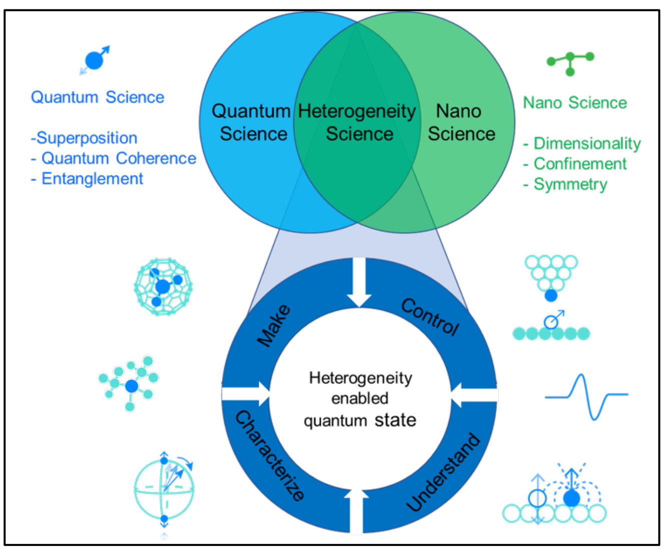
coincidence measurements, catalytic properties, magnetometry and magneto-transport, XRD including small-angle/wide-angle x-ray scattering (SAXS/WAXS), and QCM-based platforms tailored for gravimetric/optoelectronic and multimodal characterization.

3. CNMS Science Themes

CNMS has a dual mission to provide state-of-the-art capabilities and expertise in nanoscience to a broad user community and to conduct a leading nanoscience research program. These two activities are closely coupled: new capabilities that emerge from the CNMS theme science program are made available to the user community. In fact, the CNMS science themes are specifically designed to enable the development of new capabilities that will continue to attract a world-class, multidisciplinary user community in strategic science areas, namely:

1. **Heterogeneities in Quantum Materials** (HQM): Theme Lead - An-Ping Li
2. **Harnessing Complex Macromolecular Conformations** (HCMC): Theme Lead - Rajeev Kumar
3. **Multiscale Dynamics** (MD): Theme Lead - Peter Maksymovych

Heterogeneities in Quantum Materials—The interplay of reduced dimensionality and heterogeneities in heterostructures, nanostructures, molecular compounds, and other complex systems bring materials into regimes dominated by the quantum properties of defects, interfaces, and disorder. Such heterogeneous quantum materials often host exotic states that promise the development of highly engineered, scalable, and functional quantum systems for applications in computation, sensing, and communication. Despite the revolutionary opportunities presented, great challenges remain in tailoring structures, controlling delicate interactions, and protecting quantum states from decoherence to enable encoding and transduction for QIS and technology.



The overarching goal of the HQM theme is to understand the formation, behavior, and coherence of quantum states in low-dimensional quantum materials via nanoscale control over defects, interfaces, and disorder. This goal will be achieved by executing three specific aims (SAs):

SA1. Reveal quantum states associated with heterogeneities.

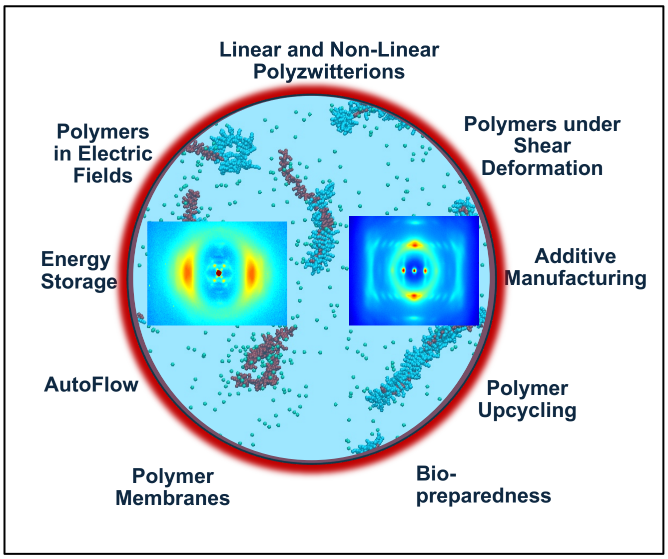
SA2. Understand quantum interactions in heterogeneous quantum systems.

SA3. Establish and enhance coherence and entanglement by controlling heterogeneities.

Our guiding hypothesis is that nanostructured materials possess novel quantum properties that can be introduced and controlled by heterogeneities and harnessed for robust, high-performance energy and QIS applications. This research theme builds directly on our strengths and expertise across atomic-level measurement, control, and modeling of low-dimensional materials via innovative development and utilization of scanning probe and electron microscopies and spectroscopies along with low-frequency Raman and ultrafast laser spectroscopies, enhanced with first-principles calculations facilitated by ML approaches. Once realized, our research will enable the design of new generations of quantum materials by harnessing heterogeneities to catalyze a paradigm shift of QIS. The development of unique capabilities will offer users a first-principles approach to quantum materials by interrogating individual heterogeneities, understanding their interactions, and building them up as new materials with desirable quantum functions.

Harnessing Complex Macromolecular Conformations—After a century of macromolecular science, the development of materials by controlling conformations of polymers is now feasible. Building on established connections between chain conformations and macroscopic properties at and near equilibrium conditions, we hypothesize that we can access new regimes of material properties by incorporating zwitterionic groups, creating nonlinear polymer chains, and using external fields to manipulate chain conformations. Access to the new regimes will significantly expand the design space for developing responsive polymer systems with behavior inaccessible to linear neutral polymers.

Using closely integrated experimental and computational studies, the HCMC theme will achieve the overarching goal of providing a predictive understanding of the link between chain conformations of zwitterionic polymers and macroscopic structure, properties, and responses to external stimuli.



To achieve this goal, efforts will focus on three SAs directed at understanding conformations of polyzwitterions (PZs) in solutions and dry states while making correlations between electrostatics, topological effects, and processing:

SA1. Understand the conformations of PZs in complexes and copolymers.

SA2. Control and evaluate topological effects in films containing PZs.

SA3. Elucidate the role of processing and rheology on structure.

These three SAs will provide critical understanding of the underlying entropy-driven processes that underpin technology development in applications for energy storage, flexible electronics, polymer upcycling, 3D printing, and antifouling membranes.

Multiscale Dynamics—Multiscale dynamics are crucial for understanding the behavior of functional materials throughout their life cycle, i.e., from the moment of synthesis that then proceeds through exchange of matter and energy with the environment, and ultimately ends with its decomposition or recycling. These dynamics occur at various energy, time, and length scales, and they influence the synthesis, processing, function, aging, failure, and recycling of materials. From the perspective of future, efficient, and sustainable materials, a lack of scalable and quantitative frameworks to capture dynamic processes has so far limited the efficiency and scope of materials design efforts, whose primary emphasis has been on material stability rather than its interaction with the environment and ensuing dynamics.

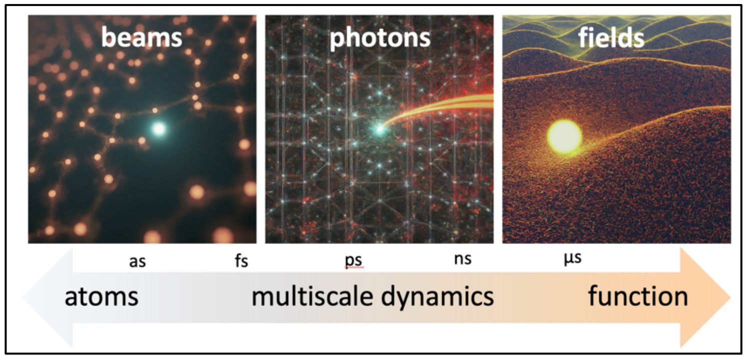
The overarching goal of the MD theme is to accelerate the design of functional materials by developing theory-guided discovery and autonomous approaches to discover, quantify, and control multiscale dynamic processes within the nanoscience paradigm. The nanoscale is uniquely suited to probe dynamic processes because of accessibility to far-from-equilibrium conditions in confined geometries. Recent advances in computational techniques enable the scope of nonequilibrium dynamics to be captured with increasing accuracy.

New methodologies will be pursued in three SAs to capture, quantify, and eventually predict the motion of specific degrees of freedom:

SA1. Understand the collective response of order parameter fields under far from equilibrium conditions.

SA2. Induce nonadiabatic atomic-scale motion and chemical reactions under energetic beams.

SA3. Couple slow and fast dynamics by multiscale stimulus.



Methodologically, the main emphasis in the MD theme will be on increased temporal resolution of traditionally slow microscopy techniques, increased scale and scope of accurate modeling frameworks, and development of autonomous methods for material dynamics. Research will stimulate the development of distinct new capabilities, including multimodal and autonomous SPM and STEM, nanoscale imaging in the proximity of phase transitions under direct action of thermal and photonic fields, ionic patterning of materials, and multiscale computational methods to capture the dynamic response from multidimensional and multimodal information. These topics leverage specific expertise by the CNMS staff in well-defined model material systems where nonequilibrium dynamics can be observed and quantified. We anticipate a broad impact of these on emerging DOE priorities in microelectronics and energy storage initiatives.

The CNMS science themes 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 Reviews, recommendations from the user community, as well as through direct interactions with the other NSRCs, all contribute to the planning of our future directions.

4. Near-Term Plans for Future Capabilities and Upgrades

CNMS has successfully leveraged numerous opportunities to better target research and investments toward specific and emerging fields of study, including quantum materials, polymer science, autonomous experiments enabled by AI/ML, and microelectronics, e.g., infrastructure investments via LDRD and ICE programs in addition to CNMS operating and capital funds. We have exceeded DOE's 10% budget allocation mandate for recapitalization every year through careful planning and prioritization. These efforts are reflected in, and strengthened by, our three theme science areas (HQM, HCMC, and MD) that pave the way for developing new approaches and capabilities needed to enable science discoveries that will benefit the broad user community. To be successful in the future, CNMS must continue to prioritize strategic investments in new capabilities that will not only provide the user community with access to best-in-class instrumentation but will also enable new science and accelerate discovery of nanomaterials with targeted functionalities. This will be accomplished by continuing to leverage institutional investments whenever the opportunity is presented, and importantly, continue to allocate CNMS project funds toward re-capitalization of the center.

Many of the planned facility improvements and research directions are determined as part of the strategic goals set by the CNMS research groups and sections, and these objectives serve as the starting point to prioritize equipment and staffing needs. Based on these plans, we have collectively identified the focus areas and new research directions we will prioritize for investment in terms of equipment, science strategy, and staffing in this 5-Year Strategic Plan for the center, but priorities change, and we are continuously re-evaluating our strategy in the context of user needs, immediate center concerns, annual budgets, and long-term opportunities. Fortunately, the CNMS 5-Year Strategic Plan is a living document that is updated every year to reflect priority changes. We have identified the CNMS priorities in terms of capabilities, enhancements, and upgrades that will enable the groups to implement strategic goals that build on our existing strengths and expertise and also create world-leading facilities for the scientific user community.

The **key capability areas** currently offered to the CNMS user community are listed below, which reflects group needs and section synergies, and are presented with future strategies in these areas; the planned equipment, upgrades, and replacements within each of the key areas are provided with details on cost and timeline. The equipment listed here are items we expect to acquire within the next 2-years and are either instruments to deliver new science in line with the key strategic areas, are replacements for aging equipment, or are upgrades to add capabilities to existing equipment.

Imaging, microscopy, spectroscopy, and nanoscale characterization: Four groups comprise the NC section, FAFM, STM, STEM, and MMÅ, with expertise and instrumentation centered around particular microscopy platforms. Researchers in these groups have expertise that combines extensive knowledge about different classes of materials and phenomena with knowhow and inventiveness to push past existing limits of instrumentation. The overarching goal for the section is to enable understanding about why materials behave as they do and provide insights that will lead to improved performance. Strategies for achieving this overarching goal include not only identifying and acquiring the best instrumentation for revealing nano- and atomic-scale phenomena, but also continuously improving instrument performance and expanding the phenomena that can be investigated. The highest priority strategic directions for NC are:

1. *Integrating optical characterization with electron, ion, and SPM to excite and observe new phenomena*. Photons are among the best means to elicit and extract properties and dynamic behaviors of materials. However, limits imposed by electromagnetic wavelengths often prevent their use in nano- and atomic scale characterization. Notably, this limit applies primarily to "photon only" measurements but can be circumvented by combining optical measurements with electron, ion, and scanning probes; their combination can provide spatial resolutions approaching probe volumes (Ås–10s nm), temporal resolutions of optical measurements, and information about electronic, magnetic, optical, thermal, and related properties. Successful examples of this synergistic approach at CNMS are evident in CL-SEM, scanning microwave impedance microscopy (SMIM), nano-IR, AFM-Raman, and the recent introduction of laser excitation into STEM instruments. We aim to push further with investments into specialized laser-coupled AFMs, scanning NV microscopes, and light and RF-coupled STM. Combining tunable lasers with STEM-based spectroscopies (4D-STEM, high-resolution EELS) will provide our world-class electron microscopes with even greater capacity to expose the inner workings of light-matter interactions and non-equilibrium thermally driven phenomena and also create a platform for atomic resolution CL spectroscopy. Through continued expansion, instrument acquisition, and in-house development of optical systems combined with targeted hiring of new staff with expertise in optics, we will force-multiply our nanocharacterization capabilities to drive and perceive a wider class of nanoscale phenomena central to materials and devices for quantum and energy applications.

2. *Cryogenic microscopy across microscopy platforms.* Interest in cryogenic microscopy techniques, e.g., STEM, STM, in the physical sciences (materials and chemistry) has recently observed a major spike in interest due to the ability to capture physical properties, behaviors, processes/interactions, phase transitions, and other critical phenomena that emerge at lower temperatures. New stages, holders, and fast cameras have enabled operation of microscopes at liquid nitrogen temperature and many of these platforms have been extended down to the mK-temperature range (the mK-STM, for example, and the liquid-He-cooled MAC-STEM scheduled for delivery in FY 2026), facilitating new understandings of materials (quantum, polymers, beam-sensitive materials) at relevant temperatures. CNMS is prioritizing low-temperature microscopy by building the infrastructure for cryogenic specimen preparation (Cryo-EM and specimen preparation facilities, the new cryo-plasma-FIB [PFIB]) and the ability for cryo-transfer across multiple microscope platforms.
3. *Advanced controls to enable flexible user-friendly interfaces, autonomous operation, and manipulation of materials at the nanoscale.* Over a decade of efforts in the different microscopy groups have demonstrated that microscopy can be used to induce targeted changes. Nanoscale probes and the concentrated strain, thermal, and electrical fields they generate can force materials across phase transitions, through non-linear regimes, and into new conformations. This ability to induce local transformations can be harnessed as a nanofabrication and synthesis tool if suitable control and feedback systems are put in place to apply stimulus and react quickly, thereby providing the means to guide, characterize, and understand material dynamics in new ways. The MD theme is focused on better understanding these behaviors. As knowledge grows, the ability to explain nanoscale phenomena increases and this can be applied to extend existing synthesis and fabrication capabilities to length scales much smaller than accessible through traditional nanofabrication approaches.
4. *Coordinating and combining measurements across platforms to leverage complementarity*. Two specific areas where CNMS is well-recognized – STEM and SPM – are independently highly successful but can be even more so when integrated given their high degree of complementarity. The potential for synergy includes ability of STEM to see through material vs. SPM's surface sensitivity, the power of EELS to identify elements and chemical states vs. STM/S, and other voltage spectroscopies with extreme sensitivity to interrogate local electronic structure and bias-dependent phenomena, the wide span of energies (μeV – 100 keV) and temperatures (10s mK vs. 100s K), and direct measurements of transport vs. the ability to map strain, phonon, and plasmonic behaviors. Coordinating and co-locating these powerful methods along with ever improving chemical imaging methods, e.g., time of flight – secondary ion mass spectroscopy (ToF-SIMS), APT, will be vital and we will bring different forms of nanoscale characterization together via two paths: (1) improving sample compatibility across platforms and coordination of experiments between groups where success will be measured by the ability to routinely identify and probe the same atom across multiple microscope platforms; (b) close collaboration with the NTI and DNA groups to develop methods to seamlessly merge data from multiple information channels to realize new understanding. This powerful combination of tools will enable multifaceted studies of materials and behaviors of individual defects and interfaces and their roles in quantum systems, as centers of catalytic behavior, and as critical players in ion- and photon-based energy materials. <u>Few places have the potential for this level of coordinated measurement, analysis, and insight as available at the CNMS and the aim is to realize the potential synergy of coupling multiple characterization methods with theory to accelerate discovery.</u>

Equipment priorities for Imaging, microscopy, spectroscopy, and nanoscale characterization:

- *The most pressing need in the center is to replace the Cameca Instruments Local Electrode Atom Probe (LEAP) 4000X HR with a modern atom probe, the Cameca Instruments LEAP 6000 XR.* The LEAP 4000X HR is 15 years old, and while it is currently running well, Cameca (now Ametek) recently announced "obsolescence of the LEAP 4000 product line and termination of service contract

support in 2025," which has huge implications for the user facility. CNMS is the only NSRC with atom probe capabilities and APT is a huge draw for the center (the LEAP is one of the most over-subscribed instruments). The advanced capabilities available on the LEAP 6000 XR are orders of magnitude improvement over the LEAP 4000 and will open new materials research opportunities for CNMS. Major improvements include combined voltage and laser pulsing to improve signal to noise, detection sensitivity, and overall data quality; a nearly 2X improvement in detection efficiency allowing for the collection of more atoms within a volume of material; deep UV laser will improve our ability to analyze large band gap insulating materials that cannot be run on the LEAP 4000; when combined with the recently installed cryo-PFIB, APT analysis of hydrated materials, soft matter, and possibly organic materials could be realized. ORNL has been at the forefront of APT since the 1980s (under the leadership of Michael Miller) and spearheaded the use of annular FIB milling to prepare the site-specific needle-shaped specimens required for APT. APT capabilities at CNMS are no longer state-of-the-art, and to maintain and solidify CNMS's leadership in the field will require replacement of the current instrument, which will enable new materials science research and attract new users.
Timeline - the LEAP 6000 XR could be available to users by early FY 2026.

- *Cryogenic upgrade of the Qnami ProteusQ scanning NV microscope purchased in FY 2023*: the microscope is a complete quantum microscope system and is the first scanning NV microscope for analysis of magnetic materials at the atomic scale. By combining NV magnetometry and SPM techniques into a single instrument, scanning NV magnetometry provides simultaneous acquisition of the sample's topography and its surface magnetic fields with nanoscale resolution. The instrument currently supports operations at room temperature (RT) since a cryogenic stage was not yet available. Plans are to trade-in the RT instrument for the low-temperature system, which will be necessary to study magnetic properties at relevant temperatures that will complement other STM and STEM techniques.
  Timeline – complete acquisition by end of FY 2024 or early FY 2025.
- *Spectrometer and camera upgrade for the JEOL NeoARM STEM*: a GIF Continuum 1069HD with high-speed Stela hybrid-pixel camera to replace the current EELS system. Cryogenic STEM is set to be a key area in future energy and quantum materials research, with CNMS already equipped with the foundational expertise and equipment. The acquisition of a direct-electron camera for EELS on the NeoARM will greatly enhance CNMS's role as a leader in the fields of cryogenic STEM for energy and quantum materials. The spectrometer's current CCD camera does not allow EELS analysis at cryogenic temperatures due to its slow speed, which fails to counter the instabilities during cryogenic experiments and the current EELS does not have the required sensitivity or the multi-frame acquisition capabilities to enable atomic resolution EELS analysis, impeding the correlation between structure, chemistry, and charge of materials.
  Timeline – upgrade is planned for FY 2025.
- *Molecular Vista VISTA 75 photoinduced force microscope (PiFM) plus scanning near-field optical microscopy (SNOM) and functional AFM*: instrument integrates true nanoscale non-destructive and non-invasive chemical characterization with sensitivity down to the single molecule level with correlative nanoscale functional and optical nanospectroscopy characterization tools for understanding of compositional-functional studies in materials sciences.
  Timeline – acquisition is planned for FY 2025.

Synthesis and nanofabrication: Creating and putting materials together—especially on the atomic to nanoscale—is not simply about synthesizing and assembling the "right" materials; it requires a deeper understanding of how materials can be combined and shaped into functional architectures, i.e., not only how materials *form* but also how they *transform* during integration and operation. The capacity to predict and tune how architectures respond to stimuli, environmental changes, processing steps, and extended operation must be developed. Moreover, understanding the influence of interfaces on macroscopic behavior is critically important. Three groups comprise the Nanomaterials Synthesis (NS) section: FHN, NRL, and Macro, and each group views that the challenges regarding the precise design, creation, integration, and operation of advanced nanomaterials are linked with their functionality. Feedback between theory, synthesis, and characterization is critical at every step of the integration process to realize targeted functional architectures. The high priority strategic areas for the NS section are described below.

1. *Automated and autonomous synthesis.* A deeper understanding of how materials form and transform during integration and operation will emerge within an infrastructure that coordinates feedback between theoretical models, data analytics, characterization, and materials synthesis/integration. In situ diagnostics integrated with growth and synthesis chambers combined with extensive ex situ optical and electrical characterization and modeling has been a hallmark of the center's research for decades, which has enabled the precision synthesis of low-dimensional and soft matter materials, providing a foundation for automated and autonomous synthesis platforms for optimized scalability. The development of integrated workflows for multimodal analysis and higher throughput functional assessment has generated massive data sets that are required to harness the power of AI/ML for driving autonomous material synthesis. These efforts are embodied through the development of automated systems (toward autonomous) for the growth, characterization, and transfer/stacking of 2D materials as well as efforts to expand our use of flow chemistry (autonomous continuous flow reactor synthesis, AutoFlowS) for the scalable precision synthesis of small molecules, deuterated compounds, and polymers.
2. *Hybrid material interfaces.* Tailoring surface topology and chemistry of inorganic materials combined with modifying the surfaces with polymers or 3D printed polymers with precise composition and topology enables the creation of unique interfaces for applications in biology and flexible electronics. Coordinated efforts across the section are enabling fundamental investigations into the role of local surface structure, confinement, and chemistry on changes in polymer organization and dynamics at interfaces, which is also a major focus of the HCMC science theme. Unique NRL capabilities are being leveraged to create nanostructures compatible with characterization modes that provide multi-scale, multimodal, and dynamic information about chemical reactions, changes in structure, and hydration at the inorganic-organic and solid-liquid interfaces, in real time within dynamic environments. By making these model interfaces compatible with characterization techniques that include neutron reflectometry, non-linear optical spectroscopy, AFM, and electron microscopy, and leveraging AI/ML workflows and theoretical models, it will be possible to provide a wholistic interfacial picture that links atomic level structure and chemical reactivity to dynamic macroscale behavior.
3. *Scalable integration and fabrication for next generation computing with low-dimensional materials.* In situ characterization as well as interface design and manipulation are enabling the directed formation of low-dimensional materials into precise arrangements with tunable architecture, strain, and functional defects. With precise placement and tunable properties, integration into addressable arrays for optoelectronic and quantum-based sensing and computing become attainable. Work is focused on unresolved scientific and technological challenges in the wafer-scale growth of 2D materials, a critical step for reliable creation of functional systems derived from these materials. Working across the section, efforts are centered on leveraging these advances to create 2D networks and vertically stacked layers of atomically thin 2D materials to realize architectures with unique opto-electronic, microelectronic, and quantum behaviors. Additionally, expertise in direct write processing allows us to leverage the unique chemical and energetic processing regimes made accessible through FIB and e-beams, and surface chemistry driven by them. Low-dimensional material response to energetic direct write, functional defect generation, and minimally invasive shaping and confinement are points of emphasis. Ultimately, understanding the range of transformations that occur during integration is essential to realizing functional architectures for next generation sensing and computing.
4. *Soft matter synthesis and characterization*: Cutting-edge synthesis and molecular level characterization methods are used to design novel macromolecules and deuterated nanomaterials driven by computational strategies. We will use advanced computational methods from atomistic to

coarse grain to understand dynamics and phase behavior. This is most important as we envision a greater understanding of dynamic phenomena at the interface (and surface) and in the presence of charged electrolyte behavior. Multi-phase behavior at interfaces and complex fluids will be investigated to guide future synthesis goals and understand phenomena. A primary mission is to support neutron studies with deuterated probes and macromolecules, taking advantage of collaborations with the neutron community. We are developing continuous flow reaction chemistry and polymerization methods (AutoFlowS) driven by AI/ML workflows and in-operando reaction monitoring to guide the design and optimization of the process. This will highly impact the current techniques used for free and controlled radical polymerizations (solution, emulsion, dispersions); anionic, cationic, metathesis polymerization reactions, controlled ring-opening polymerization to synthesize well-defined polymeric materials; multi-step organic reactions using metal-mediated and enzyme-catalyzed reactions, including synthesis of novel monomers, precursors, and nanomaterials; and deuteration and isotope-enhanced reactions (deuterated monomers and building blocks for neutron studies). AutoFlowS will be the future of optimized chemistry and polymerization. This will allow the group to contribute and utilize ORNL strengths in computation, advanced manufacturing, isotope production, and bioderived manufacturing and recycling initiatives.

5. *Functional characterization of nanomaterials*: Advanced characterization is the focal point for users and facilities and techniques need to keep pace with developing materials to discover their unique functionalities. A multidimensional ML data acquisition/fitting approach that was developed to reveal a material's multimodal properties response (structure, optical, physical, chemical reactivity) to changing environmental (gas, temperature, humidity, light, fields) conditions presents an opportunity to discover hidden functionalities of 2D or other materials that will be expanded to other characterization platforms. The high-resolution, tunable laser sources and Raman spectroscopy capabilities at cryogenic temperatures, should be enhanced to include magnetic fields for magneto-Raman spectroscopy. CL extending into the telecom (near-IR) band is a key area for SPE development and quantum communication/sensing, which will be developed through tailored materials and new detection capabilities. Ultrafast laser spectroscopy plays a crucial role in understanding the photophysics of materials by revealing quasiparticle dynamics, e.g., exciton lifetimes or exciton-phonon scattering processes, so new techniques such as multidimensional coherent spectroscopy and THz spectroscopy should target the processes, e.g., interlayer exciton formation, cooperative states in twisted 2D moirés, that are emerging in biased 2D device structures, and are crucial for future quantum or energy applications.

Equipment priorities for synthesis and nanofabrication:

- *Replacement of the Zeiss Merlin low-voltage SEM*: this SEM is more than 15 years old and Zeiss will discontinue service contract support in 2025. Access to an SEM is essential for many synthesis and nanofabrication operations and CNMS has prioritized replacement by FY 2025.
  Timeline - replace in FY 2025 or FY 2026.
- *Replacement of the Oxford Plasmalab Flourine- and Chlorine-based Reactive Ion Etching (RIE) instruments*: essential user tools in the cleanroom to etch Si-based films and materials and metals, respectively; service support for the RIE tools will end in FY 2025. New capabilities, such as laser end-point detection for ultrathin layers, will be included.
  Timeline – we expect to replace these two instruments in FY 2026 or FY 2027.
- *Femtosecond Laser System Replacement/Upgrade with optical parametric oscillator (OPO)*: new femtosecond laser to replace aging system; new system is enclosed and is super stable and environmentally capable with significantly more power (2 W vs. 9 W).
  Timeline – expect to replace system in FY 2025 or FY 2026.

Theory, modeling, and simulation: Theory and computation play a central role in CNMS's user and theme science programs due to the focus on foundational theoretical, computational, ML, and data sciences capabilities that can provide a means for fundamental understanding and prediction of materials and their function and chemical processes at the nanoscale. The expertise and capabilities within the two groups, NTI and DNA, of the TACS will continue to attract a diverse and growing user community as the approaches used are engrained in all aspects of nanoscience and many can be accessed through a virtual environment. TACS efforts enable investigating, understanding, and design of novel material structures, compositions, and processes, which also allows for probing situations or conditions that may not be readily accessible in experiments, i.e., to answer the "what-if" questions such as: What are properties of metastable states and how can we access them? What are chemical or physical deconstruction or transformation pathways? Moving forward, new capabilities to better address the multiscale dynamics (length, time, and energy) of many materials and chemical processes will be developed. Here, integration of theory with AI/ML approaches, the so-called "theory in the loop" for autonomous design and discovery, will be key toward enabling this capability. Other key developments will include routine treatments for nuclear degrees of freedom and entropy; accurate inclusion of solvation effects; equal prediction accuracy of molecular and macroscopic (bulk) properties; automated approaches for finding new reactions and synthesis pathways; and autonomous synthesis and characterization. These capabilities and expertise will enable CNMS to pursue science directions such as obtaining a deeper understanding and mastering of the origin of quantum phenomena; understanding and tailoring excitations (beam-matter, electric field) and transport (energy flow across scales); elucidating how functionalities emerge at interfaces; understanding and controlling materials defects and disorder to yield new properties; understanding heterogeneous dynamics and metastable states; and enabling predictive, atom-precise materials synthesis. The strategic priorities for TACS are:

1. *Science and user-driven data analysis workflows.* The wealth of instrumentation available at CNMS yields large volumes of scientific data in the form of multidimensional data sets and process and characterization histories. CNMS strives to develop and provide consistent and traceable workflows that allow end-to-end conversion of instrumental data to physical and chemical descriptors and models and providing science-driven outcomes. The DNA group is involved with furthering the center's developments of autonomous experiments via automating instrument platforms, connecting them with algorithms and computation for *active decision-making* tasks, thereby enabling autonomous operations. The DNA group manages important computational equipment: a cloud server with multiple CPU and GPU-enabled nodes to enable data analysis, simulations, and model training, and a DGX-2 GPU server dedicated to running algorithms for autonomous experiments at the edge. This enables us to build the self-driving laboratories of the future. Key areas of future development include:
2. *Improved data/software pipelines for analysis and acquisition*. All ML models begin with collecting and appropriately labeling volumes of data, requiring data infrastructure and standardized data models. The DNA group will continue to develop data pipelines from the instruments to centralized data stores at ORNL, including setting up automation pipelines for data pre-processing and automated data entry into federated databases to enable easy and rapid access by users and staff, regardless of location. We will also further develop our software analytics packages (pycroscopy). A particular focus will be on improving and diversifying our software to analyze data from synthesis tools, such as those derived from PLD or AutoFlowS.
3. *Algorithms, theory, and simulations in coupled workflows*. Virtually all autonomous experimental workflows operate in a data-driven manner, resulting in significantly reduced efficiency and limiting effectiveness. We propose to realize theory-coupled experimental workflows, where both theory and experimental pieces are run in an intricate closed loop, feeding back on each other to improve both the experimental exploration as well as the development of the appropriate theoretical model. DNA will work closely with NTI to realize digital twins in which AI agents can be pre-trained to enable higher impact autonomous experiments. This will require algorithmic development with methods such as multi-fidelity Bayesian optimization, curiosity-driven RL, and structured Gaussian processes and deep kernel learning. Finally, we plan to investigate large language models trained on scientific

data to leverage priors from the literature in active experimental workflows and to facilitate experimental and simulations setup and planning.

4. *Scientist-in-the-loop autonomous workflows*. An emerging trend in self-driving laboratories is that fully autonomous workflows can be sub-optimal, either due to 'AI Alignment' issues, lack of reasonable scientific priors, or imposed limitations to the experimental parameter space. Recent research indicates that human-AI coupled workflows are superior to either alone. We will develop metrics for monitoring the progress of autonomous workflows and allow for human guidance of autonomous synthesis and characterization tools that will better steer trajectories towards desired states. We will also develop methods to better incorporate human feedback to realize combined AI-human steered autonomous synthesis.

Overall, these developments comprise a holistic approach to advancing autonomous experimentation in nanoscience, leveraging cutting-edge technologies and methodologies to maximize efficiency, effectiveness, and alignment with scientific goals.

5. User Trends - 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 and patents. A significant effort in increasing efficiency in our user operation will be needed to maintain and grow our user numbers. Increased automation and remote access, 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.

CNMS continues to seek ways to increase the number of users that participate in experiments and to this end, CNMS has embraced the ability to interact with users remotely. Remote collaborations have been a successful model for working with theory users for many years and since the COVID pandemic, remote access has proven to be a viable way to increase the number of users without necessarily increasing the number of experimental sessions since it allows more users to participate in single experiments. CNMS expects the number of remote users to grow significantly in the future (we currently work with ~37% of our users remotely). This remote model does not only imply that a single user is operating an instrument from a remote location (although this is possible for highly trained users); rather, most user sessions now involve larger groups of users from the same institution that participate in the same experiments via teleconferencing/video. For example, user experiments conducted prior to COVID would typically involve 1-3 users working on-site with their technical contact or conducting experiments on their own – this was the "normal" operating mode for users. Remote access, on the other hand, allows more people to participate in the experiments from their home institution via teleconferencing whether a primary user is working on-site with some remote participants, or all the users participate remotely. In this way, more users are exposed to the experiment and can contribute their ideas and learn from active engagement. Importantly, CNMS has been investing in making additional capabilities remotely accessible and operable, and this number is growing every year.

CNMS is positioned to grow user communities in strategic areas that are also research priorities for ORNL and SC. These key areas are commensurate with the three CNMS science themes, e.g., quantum materials, polymer science, AI/ML, and microelectronics. CNMS is not only building an infrastructure of unique capabilities in these strategic areas, but our staff are recognized scientific leaders in their fields. Users seek to collaborate with these CNMS staff members, and it is this combination of cutting-edge instrumentation and knowledgeable staff that attract users to CNMS.

CNMS continues to improve its outreach to neutron scattering users and seeks to develop greater visibility within that growing community. This is exemplified by recent efforts to advertise and better incorporate CNMS deuteration capabilities for neutron scattering experiments. CNMS offers its users an opportunity to request neutron scattering time as part of a CNMS user proposal and an equivalent mechanism has been implemented to provide streamlined CNMS access to the neutron user community as part of the neutron proposals.

We are confident that the alignment around these future growth areas in combination with targeted CNMS science themes, a highly motivated and exceptionally skilled workforce, and the ability to engage more users remotely and build a larger user community, will enable the CNMS to continue to excel at recruiting new user communities and provide novel resources for a highly satisfied user community.

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. The NSRC Recapitalization Project is providing a mechanism for CNMS to acquire a one-of-a kind liquid-He-cooled MAC-STEM (expected delivery in late FY 2025) and recently a cryo-PFIB was installed at CNMS.

With the cost of liquid helium and the reliability of its supply being significant issues, we will be installing a He-recovery system for STM instruments located in Building 4515. CNMS will also be able to access the He-recovery system at SNS for smaller, local needs.

A major expansion of the Advanced Microscopy Laboratory (AML) has been approved by ORNL; construction is expected to start in early FY 2025. This extension will add two microscope bays to the existing AML and will be constructed in time to locate the liquid-He-cooled MAC-STEM when it is delivered in FY 2026. A He recovery system will also be included in the AML extension project.

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 Cryo-EM facilities, the low-temperature 4-probe STM, microelectronics infrastructure, 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 and soft matter.

7. Plans for Future Capabilities

In addition to the new equipment or equipment upgrades listed in Section 4, which are for near-term acquisitions that are based on the immediate needs of the center, CNMS maintains a prioritized list of equipment requiring long-term planning that also tries to reflect a balance between upgrading or replacing existing (aging) major capabilities with investments in key equipment that will provide new capabilities for the user community. This list of prioritized capital equipment (costing more than $500,000) for the center is provided below.

Item	Description	Timeline
Functional Atom Force Microscopy Group		
Asylum Research Vero ES AFM	Ultra-high sensitivity AFM microscope with unique signal to noise ratio. Includes BlueDrive and fast force map and current sensing.	FY 2026
NeaSPEC Time resolved THz Time Domain Spectroscopy	New capability – Microelectronics at 6G band – quantum materials (polaritons, plasmons)	FY 2026
Scanning Tunneling Microscopy Group		
Scienta-Omicron Polar STM	Upgrade for aging VT-SPM for operation at 1 K and up to 5 T field; compatible with other components on SPM	FY 2027
Scanning Transmission Electron Microscopy & Materials MicroÅnalysis Groups		
Thermo Fisher Scientific Spectra Ultra STEM (or similar)	New aberration-corrected STEM to replace the aging FEI Titan for materials science research	FY 2027 – FY 2028
Nanofabrication Research Laboratory		
Thin Film Sputter Deposition System	Current tool is aging, and significant upgrades include RF, reactive sputter, pulsed dc sputter, and evaporation module.	FY 2027
Plasma Enhanced Chemical Vapor Deposition System	Current tool is likely to be sunsetted in next few years. New capabilities such as high temperature and doping will open new fab workflows.	FY 2027
Atomic Layer Etcher	Key new technology for microelectronics and quantum where low to zero ion damage is acceptable.	FY 2027
Functional Hybrid Nanomaterials Group		
Roll-to-Roll CVD system	Needed to scale the production of 2D materials in support of new directives in Transformative Manufacturing and Microelectronics, and to supply large single-crystal samples of 2D materials including wafer scale transfer.	FY 2026
Attocube/AttoRaman Magneto-Optical Cryospectroscopy system	9 Tesla cryo-spectroscopy will provide the first variable field spectroscopy capability for the CNMS and is essential to quantum efforts. Low-temperature (cryo-) Raman in high magnetic fields to understand quantum phenomena; will address CNMS-predicted Raman spectra indicating 2D magnons.	FY 2027 -FY 2028
Nanomaterials Theory Institute and Data NanoAnalytics Groups		
1 PB Data Storage	Needed to store data from theory and experiment	FY 2026
GPU-nodes for CNMS Compute Cluster	GPU nodes will form a partition from current CPU-only nodes; required since many codes have a GPU-implementation	FY 2026
Nvidia DGX Box	Second edge computer for autonomous instrument operation	FY 2027

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 theory, modeling, and simulation, 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.