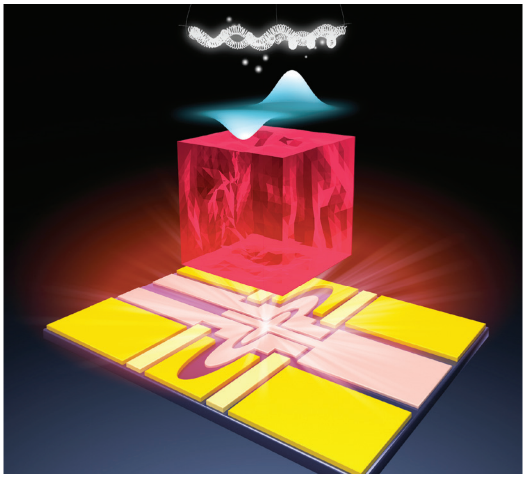
www.anl.gov

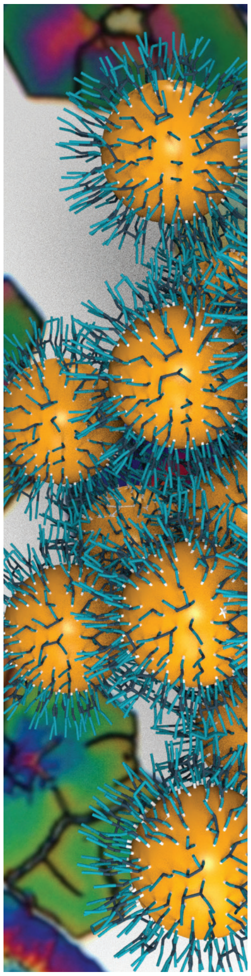
Argonne NATIONAL LABORATORY

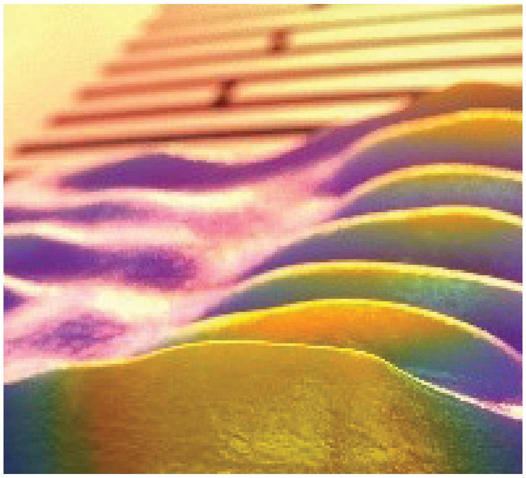
CENTER FOR NANOSCALE MATERIALS

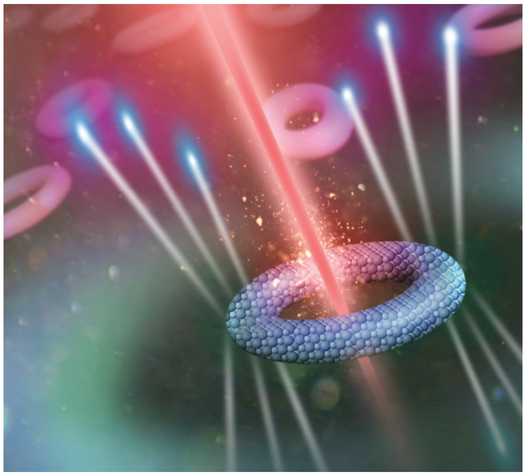
STRATEGIC PLAN

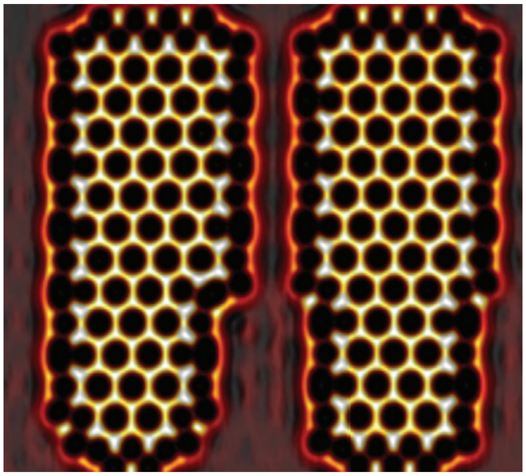
Fiscal Year 2023







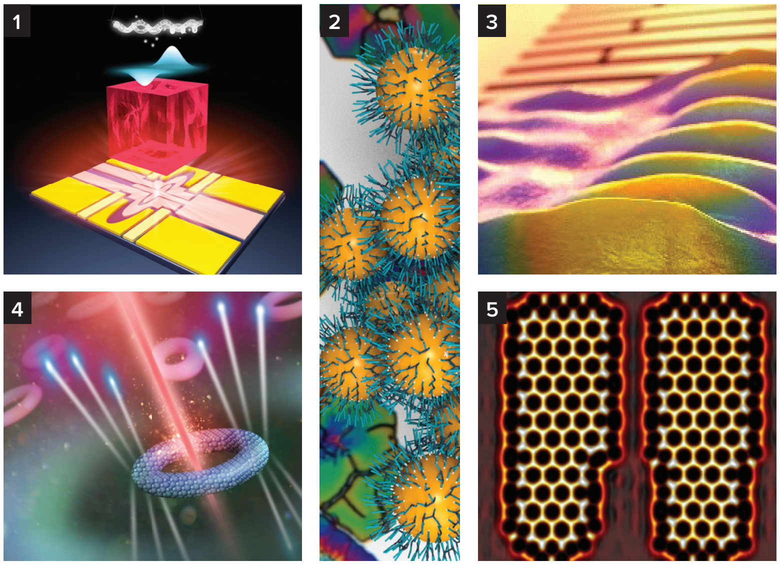




U.S. DEPARTMENT OF ENERGY

Argonne National Laboratory is a U.S. Department of Energy laboratory managed by UChicago Argonne, LLC.

ON THE COVER



1. A schematic is shown of a single-electron qubit trapped on a solid neon surface and interacting with a superconducting coplanar stripline resonator. (*Nature* **605**, 46 [2022])
2. Coarse-grained molecular dynamics simulation showing superlattice of nanoparticles (yellow) stabilized by ligands (blue). Background image of nanoparticle superlattice crystals taken by optical microscopy. (*Nanoscale* **11**, 10655 [2019])
3. Three-dimensional rendering of time-varying lattice strain caused by focused surface acoustic waves used to manipulate spin defects in the quantum material silicon carbide (SiC). Image created in collaboration with University of Chicago at CNM/APS Hard X-ray Nanoprobe. (*Nature Commun.* **10** (1), 3386 [2019])
4. Depiction of a cadmium selenide (CdSe) quantum ring emitting light in one axis. Optical transition dipole moments measured with angle-resolved photoluminescence spectroscopy and high-order scanning laser microscopy, conducted in part at CNM. Empirical tight-binding calculations of the wave functions also performed in part at CNM. (*Nature Commun.* **10**, 3253 [2019])
5. A scanning tunneling microscopy (STM) image of an artificial graphene nanoribbon is shown. The nanoribbons are assembled using STM to manipulate the positions of carbon monoxide molecules on a copper surface. These assemblies are being explored as artificial quantum coherent systems with applications to quantum information science. (*ACS Nano* **16**, 10, 16085 [2022])

Table of Contents

1. Introduction and Overview

Our vision for the Center for Nanoscale Materials (CNM) at Argonne National Laboratory is to continue to enhance its role as a world-leading research center in nanoscience, while serving and leveraging a large user science community to produce basic science discoveries that impact the needs and critical technologies of the United States. These include, for example, discoveries that impact clean and sustainable energy, quantum information science (QIS), artificial intelligence/machine learning (AI/ML), and the circular economy. Achieving this impact requires the discovery, integration, and characterization of materials across different scales at the cutting-edge technological extremes of temporal, spatial, and energy resolutions. We organize our research collectively into three cross-cutting scientific themes, which we describe here in our five-year Strategic Plan. The themes inform our strategies for new scientific directions and for investments in staff, user engagement, equipment, and infrastructure. Brief summaries of each of the CNM themes are given below:

I. **Quantum Materials and Sensing**. The goal of this theme is to combine CNM's expertise in synthesis, fabrication, characterization, and theory on nanometer length scales to discover fundamental mechanisms and materials for quantum information and sensing. This theme includes discovery of quantum bit (qubit) and quantum optical materials, precise placement and characterization of quantum emitters or qubits, photonic cavity control of quantum excitations, spin-photon couplings for transduction, and optically and electrically accessible defects in low-dimensional and bulk materials for the study of quantum coherence and entanglement.

II. **Manipulating Nanoscale Interactions**. The goal of this theme is to study the mechanical forces and the electromagnetic interactions between nanoscale constituents at length scales that vary from the atomic to the sub-micron. These include manipulating and coupling nanomechanical elements or optical near fields, determining the origin of energy dissipation via friction at the nanoscale, simulating materials and defects from their inter-atomic interactions, and synthesizing hierarchical structures across different length scales. In this theme, new directions in autonomous synthesis and materials processing are underway, revealing novel materials and improved understanding of nanoscale interactions that impact energy and sustainability applications.

III. **Nanoscale Dynamics**. The goal of this theme is to study excitation-driven energy flow and structural transitions in nanoscale materials on femtosecond to millisecond time scales over angstrom to macroscopic length scales. This includes, for example, the evolution of optical and electrical properties in materials in response to stimuli, study of reaction dynamics, and probing of exchange processes between excitations on the nanoscale. This theme leverages the rapid instrumentation improvements that enable multidimensional parameter measurements, including high-resolution spatiotemporal imaging using excitations such as electrons or photons.

The CNM employs unique expertise and capabilities to maximize the impact of our research efforts. One example is our strong effort to develop and apply AI/ML for a large range of studies and applications that leverage exceptional Argonne-wide high performance computing (HPC) facilities. Efforts include (i) the enabling of rapid materials discovery to guide researchers and users more efficiently in experimental characterization efforts and (ii) autonomous synthesis to enable enhanced information extraction and analysis of experimental datasets. In these efforts, CNM scientists are developing (i) tools and methods to scan through synthetic and preparatory conditions faster, (ii) more accurate molecular modeling of materials and on-the-fly interpretation of electron and X-ray microscopy data, and (iii) autonomous synthesis/processing of nanoscale materials, including those that can contribute to easily processed microelectronics applications, and polymer materials that can potentially be recycled more easily as part of a circular economy.

A second example is our cutting-edge ultrafast electron microscopy (UEM) capability. We have already started growing a new user base in nanoscale dynamics by being the first Nanoscale Science Research Center (NSRC) to offer user capabilities (available as a general user tool in March 2021) for UEM. This tool, which we carefully designed, opens the door for a specialized technique that has to date been available only to a few research groups that specialize in technique development. The UEM represents a key experimental method that can offer insights to ultrafast (sub-picosecond) structural and chemical change to a wide range of materials systems with diversified potential applications, including understanding heat transfer through materials important for QIS and microelectronics.

A third example is in materials for quantum information, in which we have been investing since 2016. At this juncture, major user capabilities include ultralow temperature cryostats equipped with microwave spectroscopies (including the only dilution refrigerator available to users at any NSRC), two electron paramagnetic resonance spectroscopy tools, and extensive quantum optical microscopy tools. All are available for use and are producing nearly 20 publications per year in the area of QIS.

Research at CNM is closely tied to significant capabilities at the Advanced Photon Source (APS) and the Argonne Leadership Computing Facility (ALCF). Research collaborations among APS, ALCF, and CNM are important differentiators of CNM and are key factors in the development and implementation of our Strategic Plan. These distinguish CNM from the other four NSRCs operated by the Office of Basic Energy Sciences (BES) and other nanoscale science research efforts in the United States and abroad. Furthermore, researchers at CNM work closely with their counterparts in many Argonne divisions across multiple Argonne Directorates.

The research at CNM is integrated in its capabilities, staff, and users (Figure 1-1). Leveraging our relationships with some of the best research groups in the world, aggressively going after promising candidates, and recruiting diversely and selectively, we have been able to, and will continue to, attract world-class talent to CNM. These individuals bring fresh thinking to our scientific themes and user programs. Our users are the reason we are here. We listen to them carefully, and their feedback informs our capability acquisitions and helps us anticipate emerging scientific directions in the nanosciences to support US science and technology.

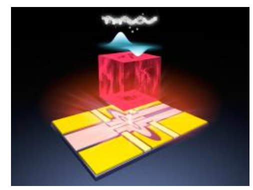
Figure 1-1: *Integrated approach to research at CNM reflecting close links between the CNM user program, staff expertise, and state-of-the-art facilities for nanoscience and nanotechnology research, providing basic science advances that support key national scientific priorities.*

In this Strategic Plan, we provide an outline of our science and technology, as well as our capabilities. This will be followed by a description of our operations and strategic directions for enhancement, including in safety, cybersecurity, data management, and staffing. We close with a description of our user community and strategic plans for enhancing this community to include diversity, equity, and inclusion for our users and staff.

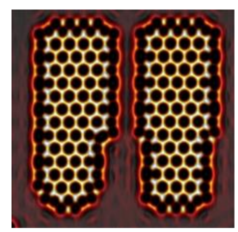
2. Science and Technology

As introduced above, CNM staff and facility users conduct research consistent with our three major themes: (i) Quantum Materials and Sensing, (ii) Manipulating Nanoscale Interactions, and (iii) Nanoscale Dynamics. CNM is equipped with unique and comprehensive capabilities in cleanroom-based nanofabrication; electron, X-ray, and scanning tunneling microscopies; optical and transport physics; and computational materials science.

CNM's scientific activities within the three scientific themes are carried out by five research groups, which interact closely with one another in carrying out their theme-based research. Each group is involved in research within all three themes. The group leaders are responsible for leading the scientific vision of their groups, for maintaining high standards for the staff, and for ensuring outstanding user research programs. Each group leader has line management responsibilities for the staff, laboratories, and safety. The groups and the respective group leaders are described below.

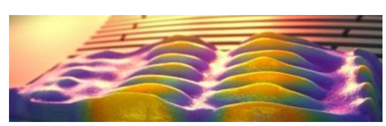


The Nanofabrication and Devices Group (NFD), with Group Leader Anirudha Sumant, specializes in the fundamental science behind the development of micro- and nanoscale systems with the goal of achieving unprecedented control in the fabrication, integration, and manipulation of nanostructures. This includes the incorporation—under cleanroom conditions—of materials and active submicron elements that couple mechanical, optical, and electrical signals to produce working nanofabricated structures.

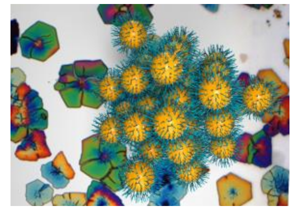


The Quantum and Energy Materials Group (QEM), with Group Leader Nathan Guisinger, focuses on the synthesis and fundamental characterization of molecules and materials on nanometer to atomic length scales. This group employs molecular epitaxy, physical deposition, and chemical synthesis methods and uses a powerful suite of scanning probe capabilities, X-ray probes, transport, and optical measurements—in some cases, *simultaneously*—to develop next-generation nanostructured materials to address challenges in energy and QIS.

The Nanophotonics and Biofunctional Structures Group (nPBS), with Group Leader Richard Schaller, seeks to understand and control light–matter interactions in nanomaterials. It does so by studying the dynamics of photo-active processes through time-resolved spectroscopies and microscopies over multiple contrast mechanisms and energy ranges. The group also studies the interaction of light with biological assemblies for nature-inspired research in energy transduction and sensing. Through basic science advances in light–matter interactions, the group seeks to impact technologically important areas that include QIS, energy conversion, and biosensing.



The Electron and X-ray Microscopy Group (EXM), with Group Leader Martin Holt, performs research to achieve local understanding and control of the structure and dynamic behavior of quantum- and energy-related materials at the atomic scale to the nanoscale via the use of advanced electron and X-ray imaging, diffraction, and spectroscopic techniques coupled with data science-based approaches. Their research bridges the unique capabilities offered by the newly developed ultrafast electron microscope (UEM) and an ultrafast hard X-ray nanoprobe (HXN) that will become even more powerful with the forthcoming APS-Upgrade. EXM also draws upon traditional strengths in high-resolution and in-situ quantitative electron microscopy.



The Theory and Modeling Group (TMG), with Group Leader Subramanian Sankaranarayanan, works on large-scale molecular dynamics, high-level electronic structure theory, quantum and electrodynamics theory, and multi-scale modeling and data science–based approaches to understand and predict a wide range of phenomena, including nanoscale tribology, thermal and charge transport, and quantum-entangled systems.

We describe here our specific strategic plans for each of the three themes. It is not our intent to comprehensively list all the projects that could be carried out within CNM, but rather to focus on broader research directions within the three themes. The five different groups interact and engage to accomplish the objectives of the themes.

Theme I—Quantum Materials and Sensing

Over the next five years, we will focus on leveraging the unprecedented characterization and control that have been achieved through modern nanoscience to develop a deeper understanding and new experimental platforms for QIS. Our approach to QIS research and the development of user science capabilities involves three areas: qubit discovery, deterministic placement and characterization of qubits, and controlled manipulation and quantum coherence.

Qubit discovery deals with the discovery of solid-state systems that enable the creation of coherent quanta of information for sensing, computing, or communication. These systems can take many forms, such as defects or dopants in a host matrix, or semiconducting particles, which are nanostructured for quantum confinement. Either way, these systems are intrinsically nanoscale in nature, and their development will benefit immensely from the strategic deployment of nanoscience experimental and theoretical methodologies, expertise, and instrumentation. The second area—deterministic placement and characterization of qubits—addresses the need to place qubits precisely (at the nanometer level) for many QIS applications. Precise placements enables the initiation of entanglement between qubits. However, the degree of entanglement relies on many details, such as relative energy levels, relative orientations of neighboring qubits, and the timescale of quantum coherence (such as spin coherence) for each qubit. Thus, extensive characterization efforts to optimize qubit performance and the ability to place qubits precisely are both needed to realize the promise of QIS. The third area addresses the need to not only achieve quantum coherence, but to manipulate and control quantum coherence. This requires a pathway to use an external input, such as light, that can be coupled to the quantum coherent excitations. We are developing advanced means, such as through photonic cavities or superconducting circuits, to increase the degree of coupling between photons and quantum excitations, thereby providing a pathway to external control of quantum states, and to also transduce information from a qubit in a quantum material to propagating photons, thereby transporting quantum information via a network. We describe our research and user science plans for the three target areas below.

Our qubit discovery research centers on the rapidly increasing interest in the scientific community (and to future users) in solid-state qubits. This is due to their anticipated scalability and the rich physics they enable. Here, we explore the underlying materials science and physics of solid-state qubits based upon spin, charge, and optical photons. We develop these systems while keeping in mind the need for pathways to transduction of information coherently between different qubit systems. We envision a compact solid-state quantum information system as one where coherent excitations are transducted between solid-state spin qubits operating at a circuit level and single photons, which then carry the information over longer distances. This research direction thus examines the science of deterministic single-photon sources, charge and spin-based qubits at microwave energies, and quantum transduction between the two.

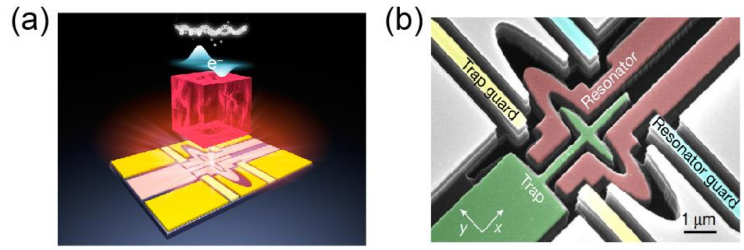


__Figure 2-1:__ (a) Schematic of a single-electron qubit trapped on a solid neon surface and interacting with a superconducting coplanar stripline resonator. (b) Scanning electron microscopy image of the device around the electron trap and photon coupling region. Future work will target continued improvement of quantum coherence properties to further enable QIS applications (Nature 605, 46 [2022]).

In one example, we are developing a fundamentally new kind of qubit based on isolated single electrons trapped on an ultraclean solid-neon surface in vacuum (Figure 2-1) and reported the initial discovery in a journal article (*Nature* 605, 46 [2022]). Using on-chip integration of an electrostatic trap and a superconducting quantum circuit, we achieved, for the first time, strong coupling between a single electron and a single microwave photon. Our single-electron qubit platform takes advantage of state-of-the-art circuit quantum electrodynamics architectures to realize qubit gate control. Relaxation times and readout fidelities are already near state-of-the-art as a charge qubit.

Our next steps are to realize real-time entanglement between multiple electron qubits as well as strong coupling with other types of quantum excitations such as phonons or qubits such as superconducting qubits. Our planned activities cover approaches that offer superior quantum environments for long coherence times, scalability based on chip technologies, and controllable interaction via integration with platforms such as silicon photonics.

New capability development is also a strong component of qubit discovery. To date, work on solid-state spin qubits has centered on defects or dopants in highly environmentally controlled environments or buried deep inside near-perfect crystals, significantly curtailing their potential utility and limiting the ability to control and place these systems with precision. We are addressing this challenge through the development of the Atomic Quantum Information Surface Science (AQuISS) lab, awarded by DOE for QIS research. With this lab, we plan to open a new frontier in nanometer-scale control over *optically active spin quantum systems* by combining quantum control (both microwave and optical) and advanced techniques of surface science, including scanning probe microscopy characterization and manipulation, on controllable ultrahigh vacuum (UHV) surfaces (see schematic, Figure 2-2a). Experimental efforts will be guided by first principles theory and modeling. In addition to near-surface bulk defects, we envision that many promising new systems can also be developed at surfaces, including surface defects, dopants, molecules (Figure 2-2c), and engineered van der Waals materials (Figure 2-2b).

In the second QIS focus area, deterministic placement and characterization of qubits, we are developing newly commissioned tools, also awarded by DOE for QIS research. Given that all quantum emitters share the property of scale—from single atoms to several nanometers—understanding the impact of the local structural or electronic environment on the properties of quantum emitters offers technological pathways for engineered properties, such as deterministic emission and controlled coherence. The Quantum Emitter Electron Nanomaterial Microscope (QuEEN-M) is a platform that integrates cathodoluminescence (CL) / photoluminescence (PL) spectroscopies with a probe-corrected scanning transmission electron

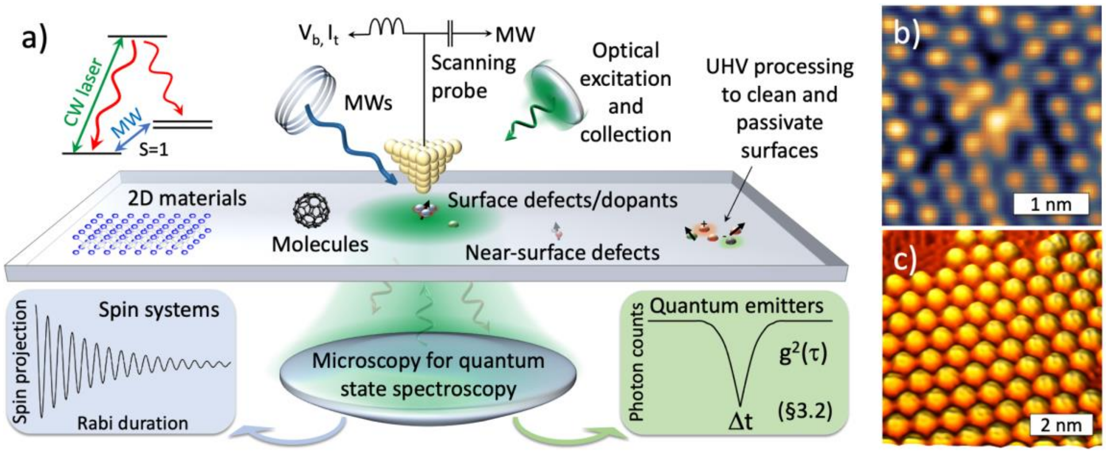


Figure 2-2: *(a) Overview of the planned studies with the Atomic Quantum Information Surface Science (AQuISS) lab, using ultrahigh vacuum (UHV) scanning probe microscopy (SPM) combined with an optical microscope for quantum state spectroscopy (spin and $g^2(t)$ indicated), showing opportunities for understanding and control at surfaces. Images from UHV SPM atomic-scale characterization of (b) point defect in bilayer WSe_2 (J. Phys. Chem. C 125, 14056 [2021]) and (c) self-assembled C_{60}-pentacene heterostructure on Cu (ACS Nano 7, 3086 [2013]).*

microscope (STEM), which enables real space atomic-resolution imaging and electron energy loss spectroscopy, along with multimodal CL/PL spectroscopy with high spatiotemporal resolution through the use of ultrafast pulsers (Figure 2-3). It enables the investigation of optically active defects and dopants with unprecedented spatial resolution, as well as photodynamic studies, with the goal of revealing the complete picture of the electronic structures of quantum emitters by nanoscale CL correlative microscopy and time-resolved CL for lifetime and coherence measurements beyond the optical diffraction limit. Due to the difficulty of imaging buried quantum emitters embedded in three-dimensional (3D) materials, atomically thin or few-layer two-dimensional (2D) material systems will enable nanoscale CL correlative microscopy with atomic-resolution STEM.

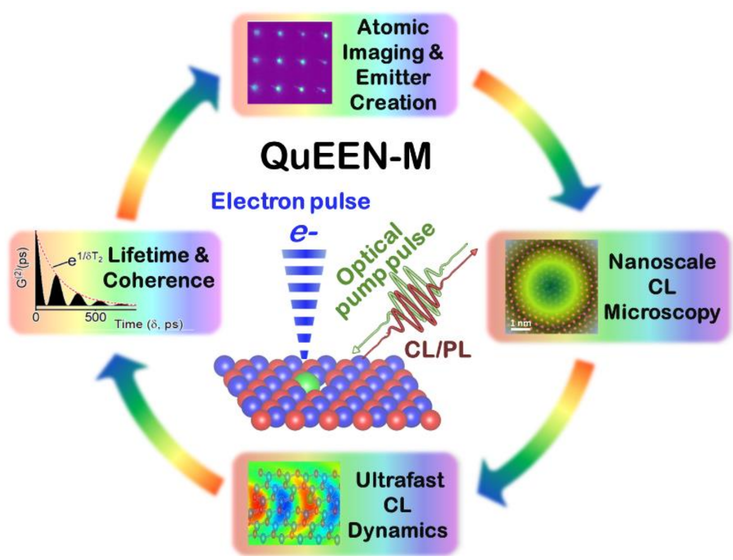


Figure 2-3: *Schematic of Quantum Emitter Electron Nanomaterial Microscope (QuEEN-M). This platform integrates CL/PL spectroscopies with a probe-corrected STEM, which enables atomic-resolution imaging and electron energy loss spectroscopy, along with multimodal CL/PL spectroscopy with high spatiotemporal resolution.*

In-situ formation, processing, and measurements of quantum emitters will be studied using the QuEEN-M. High-energy electron beams will be exploited to rearrange atoms in a sample (e.g., reversing the Si-carbon bond and placing of single atoms), although this is still an uncontrolled process under most circumstances. Other samples (e.g., freestanding diamond membranes) will be studied for deterministic placement of nitrogen vacancy centers in diamond.

Additional advanced approaches to quantum defect and qubit characterization will be undertaken. For example, we will continue to use our deep expertise in X-ray and electron microscopy to understand the microstructural and chemical basis of coherent quantum information flow. For example, the manipulation of strain near isolated point defects and engineered structures provides a route to controlling a solid-state qubit environment without introducing stray electromagnetic fields. The degree and nature of strain coupling to local properties, such as energy level degeneracy, are poorly understood. Using CNM's unique HXN capability at the APS, we have recently developed a technique of 3D Bragg projection ptychography. Building upon promising results already obtained in showing how sound (using surface acoustic waves) can modulate a SiC defect qubit (*Nature Phys.* 15, 490 [2019]), we intend to use this technique extensively to visualize near-defect strain directly in quantum materials in the coming years.

In the third QIS research area—manipulating and controlling quantum coherence—we are motivated by the science of quantum transduction. Here, we study how quantum information can be coherently transferred from one format to another without converting to classical information. Quantum information in different excitations or formats may operate in different frequency regimes: microwave photon, optical photon, electron spin, nuclear spin, phonon, etc. The ability to achieve quantum transduction between these formats and frequency ranges is thus an important topic of interest in QIS.

One of our targets in this area is hybrid magnonic systems, where microwave photons, optical photons, and mechanical phonons can coexist and interact with collective spin excitations (i.e., magnons) with large tunability and very low loss. Hybrid magnonics uses the coherent information exchange between strongly coupled magnons and microwave photons for quantum information processing. This type of mode hybridization opens opportunities for controlling quantum excitations at microwave frequencies using magnons, which introduce unique properties such as nonreciprocity and tunability that are not easily achievable in other types of information carriers. We plan to use the nanofabrication capability at CNM to bring conventional magnonics to the nanoscale and study the fundamental physics of hybrid magnonics in the quantum regime.

Another direction that we will explore in the coming years is to harness high-frequency piezomechanics for achieving bidirectional quantum state conversion between microwave and optical frequencies (Figure 2-4). This has become an urgent and very important topic in QIS because it holds the key to the realization of a global quantum network. So far, the leading platforms for quantum information processing (quantum computing) operate at microwave frequencies, but the best quantum communication carrier is the optical photon in the telecom band where the propagation loss and environmental noise are minimized. The ability to transfer quantum states between microwave frequencies and optical light would enable new technologies, such as distributed quantum computing and sensing.

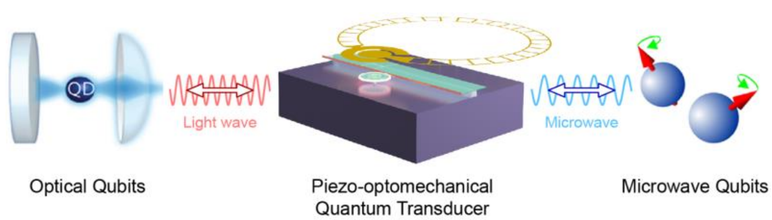


Figure 2-4: *Schematic of planned piezo-optomechanical quantum transducer, bridging optical and microwave qubits for realizing a quantum network.*

Our experimental systems used for these studies, which broadly enable characterization of electronic, optical, mechanical, and spin properties of various quantum materials and devices at both cryogenic and room temperatures, are available as user tools to serve the larger QIS community. These tools will allow users, for example, to manipulate and interconnect microwave and optical qubits to enable experimental research on new devices. Theory and simulation are essential both to enable the experimental QIS outlined in this plan and drive new experiments and directions. We will also take advantage of machine learning to accelerate challenging quantum science measurements.

Theme II—Manipulating Nanoscale Interactions

A central motif of this theme is to study and control the forces, electromagnetic interactions, and energy dissipation between nanoscale constituents at interaction lengths that vary from the atomic scale (~0.1 nm) to distant (~100 nm). These interactions can be collective and dissipative, making it challenging to predict and control them. This is the case in a large variety of nanoscale systems, such as the nonlinear dynamics of nano-electromechanical systems (NEMS) and metasurfaces (10–100 nm), the fundamentals of friction at the nanoscale (1–100 nm), the molecular dynamics simulations of materials (1–10 nm), and the synthesis of heterogeneous materials (0.1–100 nm).

The use of computational methods to predict materials properties and/or to support experimental efforts is a major part of our approach to this thematic research. One example of our plans addresses interactions with structural defects. First principles computational modeling, such as density functional theory (DFT), allows accurate prediction of the thermodynamic and electronic properties of point and extended defects, and more recently, ML capabilities have been developed to accelerate the prediction and design of defects. Defects play an important role in key materials and technologies, such as semiconductors for photovoltaic, optoelectronic, and quantum applications. In photovoltaics and optoelectronics, native and non-native point defects control carrier concentrations and lifetimes, and extended defects such as grain boundaries and dislocation cores affect impurity segregation and carrier recombination, effects which are critical for determining power conversion efficiency. Another example regards quantum applications, where the thermodynamics and charge and spin properties of defect complexes are crucial for next-generation defect qubits. This strongly impacts the Quantum Materials and Sensing theme, and is one example of how our themes complement and enhance the other themes of the center.

In one example of the power of this approach, it is known that grain boundaries and dislocation cores play an important role in controlling power conversion efficiency in thin film photovoltaic materials such as CdTe. Passivation of grain boundaries reduces recombination, but the choice of stable, effective passivants is challenging. Using realistic dislocation core structures obtained from STEM, we have shown different passivants/co-passivants and perform DFT calculations to understand their properties (*Solar Ener. Mater. Solar Cells* 232, 111279 [2021]). By understanding the origin of the midgap states in different dislocation cores due to DFT analysis, we plan to impact the design of appropriate passivants and co-passivants to optimize performance. In a second example, the approach is further extended to perovskite halides both for improvement of defect properties (*J. Mater. Sci.* 57, 10736 [2022]) and for the design of perovskite halide (ABX_3) materials with different A and B site cations that are promising for photovoltaic applications. In general, computations leveraging DFT+ML are a promising pathway towards materials design that we plan to leverage in the coming years (Figure 2-5).

Molecular dynamics studies are another area of our nanoscale interactions research plans that are also strongly guided by computational efforts informed by AI techniques. This work will feed into all three of our science research themes by providing broad materials discovery and materials design guidance. Here, CNM's goal is to apply ML and data science approaches in concert with first principles physics and/or experimental data to develop a suite of significantly more accurate (compared to what is available today), yet computationally efficient approaches for simulating the interaction between atoms for molecular

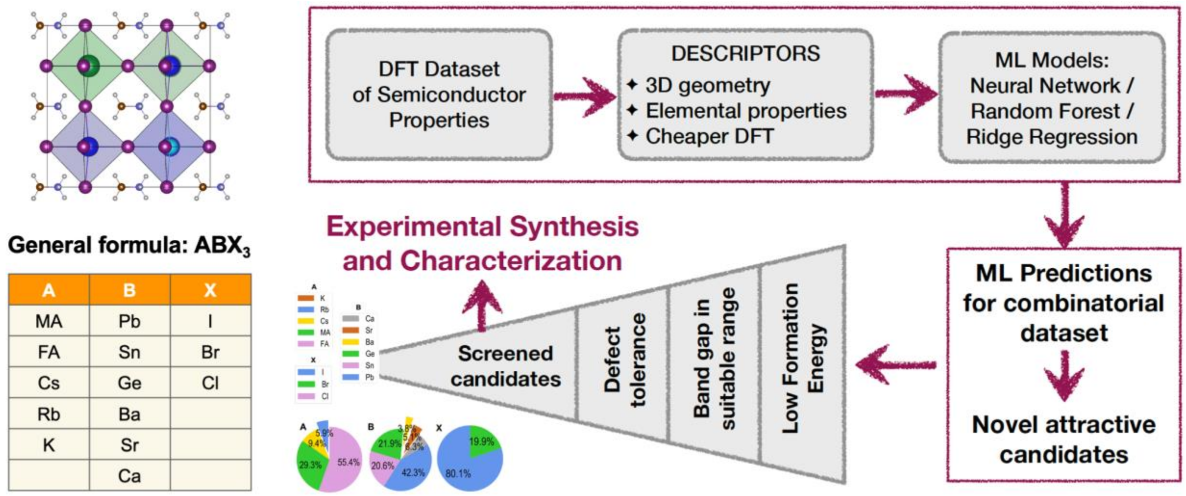


Figure 2-5*: A computational framework powered by high-throughput computations and machine learning, which we utilize for understanding defect interactions, and which we plan to continue to apply to technologically important materials for energy, quantum science, optoelectronics, and other areas. This example shows the approach for the design and prediction of mixed cation halide perovskite alloys (Energy Enviro. Sci. 15, 1930 [2022]) of interest for solar energy conversion.*

dynamics calculations. This work builds upon remarkably promising results obtained by us over the past few years (see, for instance, *Nature Comm*. 10(1), 379 [2019]). Our efforts over the next five years will include the development of a set of user tools for molecular dynamics simulations of materials informed by AI. The use of AI for the physical sciences is of great interest to the scientific community and also has emerged as a focal point of emphasis for DOE's Office of Science. We, therefore, anticipate significant user interest in this area over the next five years.

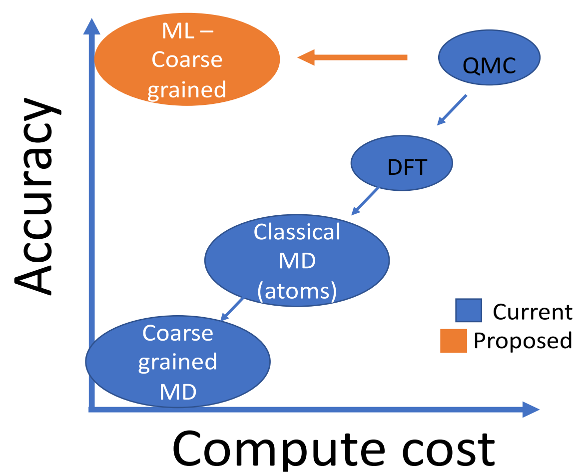


Figure 2-6: *Our ML framework combining the speed of coarse-grained molecular dynamics (MD) potentials with the accuracy of quantum Monte Carlo (QMC) simulations. This treatment will enable high-fidelity, long-timescale simulations of multimillion-molecule systems to probe nucleation and growth phenomena in hierarchical materials.*

We will explore new means of determining flexible force fields (neural networks as well as physics-based models from symbolic regression) for large-scale molecular dynamics simulations capable of describing reactive catalytic processes, disorder, and phase transformation much more accurately and efficiently than is possible today. We plan to leverage active learning strategies that we have already developed successfully to sample computationally expensive but accurate training data efficiently. Such strategies can train models against sparser datasets compared to conventional training procedures.

Our end goal is to design inexpensive, yet accurate ML molecular models (Figure 2-6) that will be available to our users and will accelerate materials design and discovery in at least two ways. First, they will be combined with AI-based decision tree approaches to explore the structural phase space and perform "inverse design" to predict optimal structures for user-defined target properties. Second, they will be combined with computer vision and deep learning approaches to build and train deep (convolutional) neural network classifiers for supporting microscopy experiments.

Synthesis of nanomaterials across scales represents a broad, traditional area of strength for CNM and influences research in all of our three scientific themes. Here, we have made significant progress in the control of zero-, one-, and two-dimensional materials synthesis using a variety of colloidal and solution chemistry, biochemical, and vacuum deposition methods. We will continue our efforts in this direction of synthesis, and over the next five years, we will increasingly use these capabilities to focus on integration and functionality. Examples include the incorporation of plasmonic nanostructures for metasurface engineered flat lenses (discussed below), the synthesis of hybrid nanomaterials as solid lubricants for superlubricity applications, and the synthesis of single-photon emitting quantum materials.

We are also pursuing autonomous synthesis, a state-of-the-art experimental paradigm based on a modular robotic system driven by AI/ML-based methods, to efficiently search large, complex parameter spaces for emerging classes of nanomaterials discovery. In the past two years, PolyBot, a ML-integrated modular robotic material discovery platform, was developed at CNM. This autonomous synthesis platform includes advanced synthesis, processing, and characterization capabilities, as well as an AI/ML methods embedded software package. We have already demonstrated a closed-loop optimization of highly conductive polymer nanofilms via autonomously turning formulation and processing conditions. In the next five years, we will continue to upgrade the capabilities of this autonomous synthesis platform with more experimental functionalities (e.g., synthesis capabilities, analyzation tools) and AI/ML methods for different staff and user projects. More importantly, we will use this newly developed platform to accelerate the discovery of electronic functional polymers for eco-friendly electronics applications, peptide synthesis with desired sequences, and inorganic nanoparticles with desired properties. An example of one direction that we expect to be fruitful in the coming years is the development of robust and stretchable polymeric materials to be used as sensors for medical diagnostics and other applications (Figure 2-7).

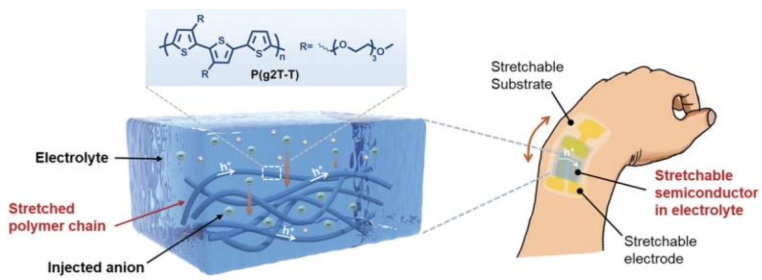


Figure 2-7: *Schematic of the mixed ion and hole transport within a functionalized polythiophene film, to be developed for medical diagnostics applications (Adv. Mater. 34, 2201178 [2022]). We are targeting autonomous synthesis and processing of polymer films for a range of additional impacts (including clean energy, microelectronics, and the circular economy) over the next five years.*

Computational molecular simulations and synthesis contribute to another significant research direction in the CNM—the science of tribology and superlubricity at the nanoscale. We harness 0D and 2D nanomaterials to carry out discovery science in solid-state lubricants for superlubricity (state of zero friction) under realistic conditions. Via collaboration between nanomaterials synthesis experts, molecular dynamics experts, and experimentalists working on nanoscale tribology, we have carved out a unique niche for CNM in this space. Obtaining a fundamental understanding of the atomistic-scale dynamical processes at tribo-interfaces is crucial for the design of functional lubricants. Our recent experimental studies over the past three years have shown that 2D materials (including graphene, MoS_2, and hexagonal-BN), when combined with nanoscale diamond particles, undergo tribochemical reactions at the nanoscale, leading to formation of onion-like-carbon “bearing” structures at the tribological contact during the sliding process. This yields superlubricity at the macroscale and is supported by simulation of the onion-like bearing structure (*Adv. Mater. Interfaces* 6, 1901416 [2019]). The central idea of this work is to develop a generic lubricant that can work in multifarious environments. We will build on our work over the next five years to broadly develop the field of superlubricity at the macroscale using nanomaterials engineering. Our studies will include the visualization of tribochemical modifications in real time at the tribological contact down to the atomic level to better understand the mechanism(s) of superlubricity.

We further note that our research on superlubricity has received particular attention from industry. We have already been successful in receiving recent funding from DOE under Technology Commercialization Fund (TCF) grants in collaboration with John Crane, Inc., and Magna. In these efforts we have been able to develop new solid lubricants based on 2D materials and other nanomaterials for dry gas seals and carried out successful test trials at the industrial scale. We expect our research into nanoscale tribology and superlubricity, supported by computational simulation and prediction, will continue to attract industrial users over the next five years and beyond.

An additional area of focus is synthesizing, understanding, and utilizing the interactions of hierarchically assembled biomolecular materials for functional capabilities in optical energy conversion and sensing. For example, we are assembling zero-, one-, or two-dimensional peptide assemblies that can incorporate multiple chromophores (co-factors) for adding function while still demonstrating morphological control, with one recent example published in a journal article (*Nature Chem*., doi.org/10.1038/s41557-022-01055-3 [2022]). Functional capabilities can further be found through cell-free synthetic biology to create nano-bio composites that, when combined with photocatalytic nanoparticles, we can use to produce hydrogen or reduce CO_2. We further plan to develop our nano-bio composites for highly specific and sensitive nanostructured sensors, ultimately to be used in concert with 2D waveguide arrays for on-chip sensing in collaboration with the Quantum Materials and Sensing theme. These efforts will employ AI/ML methods for predictive peptide materials and sequence optimization of opsin-based proteins to accelerate the materials discovery process, much like the Polybot plans described above. Finally, these efforts are also expected to impact bio-preparedness through deterministic binding of target analytes combined with nanoparticle support structures that provide robustness over a range of environmental conditions.

Our research under this theme rounds out with our efforts in the science of metasurface engineering, where we harness collective interactions at the ~10-nm length scale to study and create metasurface- and NEMS-based miniaturized optical systems. All structures are surrounded by fluctuating electromagnetic fields due to thermal and quantum fluctuations of the charge and current density at their surfaces. Immediately outside the bodies, this electromagnetic field exists partly in the form of propagating electromagnetic waves and partly in the form of evanescent waves that decay exponentially with distance away from the body’s surface. We intend to implement reliable methods to probe, control, and manipulate nanostructures by controlling these near-field forces. Furthermore, metasurface-based optical elements enable wave-front engineering by locally controlling the properties (amplitude, phase, etc.) of the incident illumination. They hold great potential to promote a new generation of “ultra-flat” lenses and thin optical systems for imaging and sensing. At CNM we are exploring ultrathin (thinner than wavelength) metasurfaces based on dielectric nanoresonators, using ML as a guide. This research will continue over the next few years to take full advantage of the strong interactions between nanoresonators to understand and implement novel metasurfaces for manipulation of visible light and novel sensing platforms.

Theme III—Nanoscale Dynamics

Today, there is significant need for interrogating, visualizing, and understanding time-dependent phenomena in materials at the nanoscale at ultra-short (femto- to nanosecond) time scales. This includes understanding energy flow and loss in energy conversion materials, evolution of metastable materials and lattice phase changes, non-equilibrium mechanical responses, plasmonic processes, and the dynamics of the interaction of materials with various excitation quanta (such as optical, magnetic, or electronic) on such time scales. Constrained by the capability of experimental equipment, our window into this world has been limited so far. However, recent advancements in instrumentation are enabling us to change this by probing new contrast mechanisms at greater spectral ranges with high temporal resolution. We believe we have, therefore, made a timely decision to identify nanoscale dynamics as a key theme for staff and user science at CNM. In the next five years, we aim to offer new tools, methods, and approaches to target central problems in condensed matter physics, novel optical phenomena, and chemical processes, with impact on the science of catalysis, energy conversion, and the circular economy.

Beginning with metastable materials, such materials are far from just an anomaly. A better understanding of the dynamics of metastable phase formation would greatly improve knowledge of the mechanisms of many synthesis pathways. As a result, we are pursuing a joint theory and experimental effort that essentially seeks to establish the metastable phase diagram in varied synthesis processes. Conventional phase diagrams are invaluable tools for material synthesis and provide information on the phases of the material at any given thermodynamic condition (i.e., state variables such as pressure, temperature, and composition). However, conventional phase diagrams only represent a reduced set of phases observed at distinct thermodynamic equilibria. In contrast, materials during their synthesis, operation, or processing may not reach their thermodynamic equilibrium state but, instead, become trapped in a local (metastable) free energy minimum that may exhibit desirable properties. The discovery of these metastable states is often by serendipity. Mapping these metastable phases and their thermodynamic behavior is, therefore, highly desirable but currently lacking due to the vast configurational landscape.

We are working to establish an automated workflow that integrates first-principles physics and atomistic simulations with ML and HPC to allow rapid exploration of the metastable phases of any given elemental composition. A first example appeared in *Nature Comm.* 13, 3251 [2022] (Figure 2-8). We use a representative material, carbon, that exhibits a vast number of metastable phases and demonstrate an automatic ML-based construction workflow to generate a metastable phase diagram that maps hundreds of metastable states ranging from near equilibrium to those far from equilibrium (within 500 meV/atom). We validate our predictions of synthesizability of new metastable phases in high temperature and high pressure experiments using a diamond anvil cell on graphite sample coupled with high-resolution transmission electron microscopy (HR-TEM). Our approach is quite general, and we plan to extend this effort to other materials, including multi-component alloy systems. Future plans are also based on the fact that some of the described ML/simulation-based workflow is done in conjunction with our users on collaborative AI materials design efforts (*Nature Comm.* 13, 368 [2022]; *npj Comput. Mater.* 8, 124 [2022]; *Science* 375, 533 [2022]). These efforts further serve as the foundation for future development of digital twins towards autonomous synthesis and characterization for accelerated materials discovery.

We are continually updating our experimental research capabilities to build upon our strong, productive, and traditional efforts in ultrafast optical physics, which benefit from our current range of time-resolved contrast mechanisms, such as optical photons for electronic transitions, infrared photons for vibrational modes, THz photons for transient conductivity, or X-ray photons for structural dynamics. Additionally, we seek to improve our ability in the optical regime to provide spatial resolution with dynamical capabilities. This will complement the dynamical capabilities currently present, but which we plan to develop further

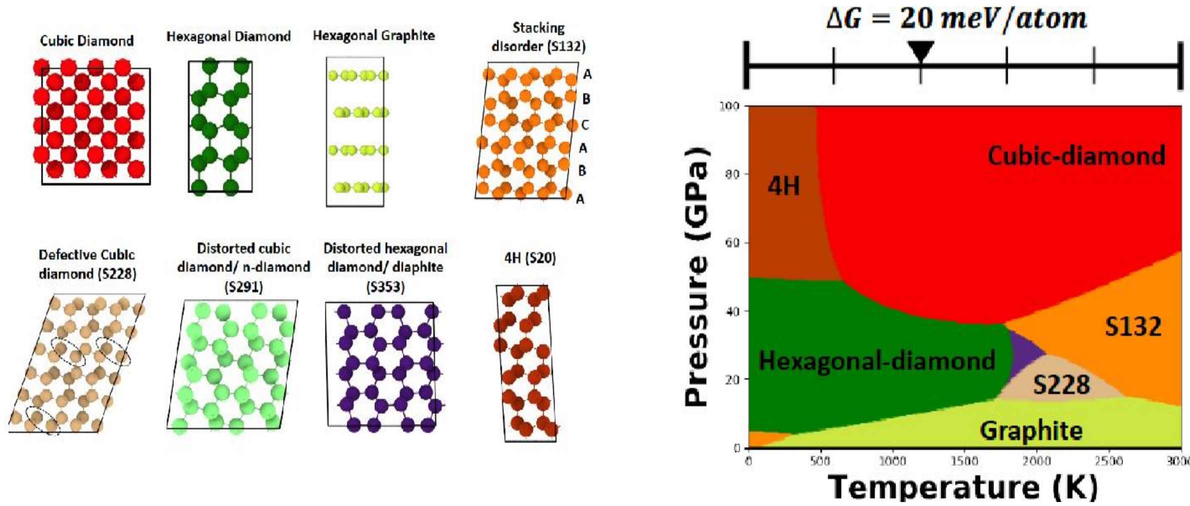


Figure 2-8: *Representative metastable phases of carbon and a corresponding phase diagram showing the phase space in pressure and temperature at which they are likely to occur. The ΔG of 20 meV/atom represents the degree of non-equilibrium relative to the most stable phase, i.e., graphite. This figure serves as one example amongst future plans for developing ML/simulation workflow for predicting metastable phase formation in many classes of materials. This effort will also impact work on digital twins for autonomous synthesis and characterization, ultimately producing accelerated materials discovery.*

for electrons and X-rays via, respectively, the UEM and HXN. Each of these plans for optical, electron, and X-ray dynamics is described briefly below.

Our research plans for ultrafast optical physics expand into ultrafast optical microscopy (UOM) and newly offered 2D transient terahertz spectroscopy. CNM currently offers an encompassing range of spectroscopic capabilities with femtosecond and picosecond time resolution spanning ultraviolet through far-infrared; however, we lack high-spatial-resolution spectroscopic methods. To offer such capabilities, we will continue to develop transient absorption microscopy and time-resolved emission microscopy with simultaneous high temporal and spatial resolution, each of which complements our efforts with UEM. Our UOM approach will enable reconfiguration so that we can also offer momentum resolution via Fourier plane imaging. These optical microscopies will be used to address several areas in the transient response of materials, including effects that are triggered inhomogeneously, such as energy migration between domains in polycrystalline solids and transient excitations of materials that involve thermal dissipation.

Multiple classes of materials problems and sample types prompt staff and user interest in UOM. Chief among these are small lateral extent or spatially varying samples with inhomogeneities or variations in electronic properties (*Nature Comm.* 11, 4442 [2020]). Often such inhomogeneities play key roles in the local nucleation and triggering of events, such as charge separation, energy funneling, or phase transitions, including metal-to-insulator transitions. UOM offers the spatial and temporal resolution needed for such studies. Characterizing the time scale and channels into which energy flows upon impulsive excitation is fundamental to a wide range of nonequilibrium phenomena. Fourier plane imaging approaches permit examination of mechanical motion, such as phonon generation and thermal dissipation, which can otherwise be challenging to evaluate because electron–phonon scattering can occur on the femtosecond to picosecond time scales. Our interest here lies in gaining a fundamental understanding and control of dynamic electron–phonon processes, with the broader goals of tailoring energy flow within nanostructured materials and controlling spectral evolution of carriers and phonons. Concomitantly, we will perform theoretical modeling of nonequilibrium electron–phonon dynamics, using a combination of many-body perturbation theory, Boltzmann transport equation, time-dependent DFT and first-principles molecular dynamics approaches. In particular, we aim to understand how electron–phonon dynamics are impacted by nanoscale heterogeneity, anisotropy, interfaces, and proximal phase transitions.

Our ultrafast microscopy research centers around an electron column-based UEM that was commissioned as a user instrument in March 2021. It combines a state-of-the-art, high-repetition-rate, tunable femtosecond laser with selectable pump wavelength from 325 to 2,000 nm with a synchronous laser-pumped, pulsed TEM that is outfitted with high-sensitivity cameras and electron energy filtering. In the stroboscopic UEM mode, the instrument delivers up to 470-fs temporal resolution and 0.5-eV energy resolution, and delivers 1–1,000 electrons per pulse (depending on time-resolution needs). This tool creates a unique microscopic perspective on the local origins of transient and nonequilibrium material response, complementing ultrafast spectroscopic and momentum-resolved techniques both for CNM staff and the broader user community. The UEM provides the means to evaluate sample changes spatially (with sub-nanometer resolution) and temporally with regard to real-space local structure, reciprocal space (via electron diffraction), charge distribution, and local electric field on ultrafast time scales. By flexibly tuning these microscopic contrast mechanisms at a fixed time slice relative to a dynamic process, the UEM can deliver insights on ultrafast structural and chemical changes to a wide range of systems. Due to its complexity, this type of system to date has been available only to a few research groups that have specialized in technique development. However, we have the opportunity to harness the potential of this investment on behalf of a broad user community and staff scientific research, advancing in parallel many areas of nanoscience through the understanding of transient processes, such as in exciton localization, short-lived metastable phases, photo-induced segregation, dynamics in topological materials, plasmonic systems, molecular motors, and magnetic fluctuations, to name a few.

We will work in the coming years to develop novel sample environments and routes of sample excitation of fundamental and device relevance, by developing ultrafast mechanical and electrical triggering mechanisms in addition to a variable-delay pump-pump-probe mode. Combined with ultrafast probing, this will permit insights into non-equilibrium phenomena in an electric field and under strain. Structural distortions that exist upon formation and disturbance of quantum material systems will be targeted, as will in-operando nano-enabled transistors, memories, NEMS behavior, and microstructure response to ultra-high strain rates.

CNM is also planning to upgrade our HXN for time-resolved nanobeam Bragg ptychography to fully utilize the upgraded source parameters of APS-U and create a unique visualization tool for time-resolved microscopy at high spatial resolution. We will be synchronizing observed dynamic material behavior with 3D visualizations created by scanning X-ray diffraction microscopy using 100-ps synchrotron X-ray pulses. Our goal is to create a 4D ptychography approach capable of imaging strain volumes with nanoscale (~20-30 nm) real-space voxel resolution at 100-ps time resolution. We will be able to detect, for example, time-resolved strain induced by ultrafast electrical, acoustic, or optical stimulation of defects in materials. The first steps in this development using the pre-upgraded APS source have recently been demonstrated at the HXN in collaboration with scientists from Argonne’s Materials Science Division (*PNAS* 119, e2118597119 [2022]), establishing the potential of this approach for a nanoscale understanding of ultrafast transition pathways for optically driven phase transitions. This capability can more broadly contribute to the understanding and control of dynamic electron–phonon processes in energy materials, as well as dissipation and decoherence in quantum materials, and enable us to study the roles of defects or inhomogeneities in triggering materials phenomena within large rendered volumes. Our further planned development of AI-enabled correlative methodologies that broadly combine our in-situ time-resolved electron and X-ray microscopy capabilities will uniquely enable progress in these areas through correlative imaging of chemical heterogeneity with structural phase and strain to understand local perturbations in energy conversion within complex materials for quantum transduction, energy storage, harvesting, and catalysis.

3. Capabilities

CNM provides an array of capabilities, expertise, and tools to its users. These include optical spectroscopy from ultraviolet to terahertz at the extremes of spatial and time resolution; an electron microscopy center with a wide range of electron microscopes that includes the UEM; a full suite of variable-temperature scanning tunneling microscope capabilities; comprehensive nanofabrication capabilities in a newly expanded 18,000-ft^2 cleanroom; the Carbon supercomputing cluster, which will be upgraded specifically for data-intensive ML workloads; quantum science capabilities designed to study single-photon as well as charge- and spin-based coherent systems; and the HXN jointly operated by CNM/APS. For a description of our many major capabilities, please see **Appendix 1.a.** For a complete list and description of all CNM capabilities, please visit our website at https://www.anl.gov/cnm/cnm-capabilities.

Here, we discuss our approach to upgrading our toolset. CNM continuously analyzes the suite of capabilities available to users and adds new instruments and enhancements as needed. It also removes capabilities that have become obsolete or display little to no usage. Our main mechanisms to upgrade our toolset is the recapitalization of a portion of our yearly funds. We are also currently acquiring equipment through the NSRC Major Items of Equipment (MIE) project, as well as NSRC QIS-specific projects.

Recapitalization refers to the use of up to 10% of CNM annual operating funds each year to procure new tools or to upgrade aging tools. It is the mechanism that enables the largest quantity of new tools and upgrades each year. Our process for recapitalization incorporates suggestions from both our users and our research staff.

Another process through which new capabilities have been acquired is through the NSRC QIS infrastructure grants. The CNM has received three of these grants. The first, entitled “Photon qubit entanglement and transduction,” provided major new capabilities in quantum optics, particularly enabling time-correlated single-photon counting microscopes in both the visible and near-infrared. The near-infrared microscope includes single-photon detectors of ultrahigh-efficiency superconducting nanowire. Additionally, a magneto-optical microscope was constructed that is equipped with a microscope cryostat and performs spin-dependent optical emission studies of quantum materials. The equipment from the other two grants, as well as the planned scientific and user impact, were given previously in this document. The AQuISS lab was described in Figure 2-2 and surrounding text. The QuEEN-M was described in Figure 2-3 and surrounding text. Each of these tools is expected to be completed in FY 2023.

The NSRC MIE project extends across all five NSRCs and represents a major DOE commitment to further the nation’s capabilities in nanoscience research. The project, funded at $80M, enables investment in major capabilities that are too large to be accommodated through NSRC recapitalization funds. The CNM will acquire four new tools via this project. The dynamic double-aberration corrected scanning transmission electron microscope (Dynamic DAC-STEM) as well as the transient photoelectron and cathodoluminescence spectroscopy (TPCS) tool will significantly add to our nanoscale dynamics research capabilities. The Dynamic DAC-STEM will be complementary to the UEM, by virtue of having higher spatial resolution, but will have longer time-scale dynamics capabilities (tens of microseconds and beyond). The TPCS tool will be placed at APS sector 29, with a vacuum highway between the TPCS instrument and other beamline tools, to enable the same sample to be studied for structure and dynamic function correlations. The TPCS tool continues our effort to be able to study dynamics with different contrast mechanisms. Since this tool can directly extract carrier energy dynamics, it is expected to complement our time-resolved optical capabilities in cases where the optical spectrum is too dense with many transitions or in systems that lack significant optical extension such that time-resolved optical studies are not possible. Our plasma focused ion beam will enable nanostructuring without concern for gallium ion doping issues, which can limit electronic performance of a nanostructured device, and will also enable much faster materials preparation for TEM studies. Finally, the millikelvin scanning tunneling microscope (STM) will for the first time give our users access to mK temperatures with STM, complementing our other variable temperature STMs and enabling detailed quantum state and spin state studies that were not possible before.

For additional detail and timelines, the tools that we plan to acquire through the three processes of recapitalization, NSRC QIS projects, and the MIE project are given in **Appendix 1.b**.

4. Operations

Here, we briefly outline our plans for key operations areas, particularly staffing, safety, cybersecurity, and data management.

Staffing

The CNM opened for operations and began welcoming users in the fall of 2006. Since this time, the CNM has grown as a facility, with approximately 40 regular staff (research and technical support) members and more than 700 users annually. A facility of this size also requires dedicated personnel to support operations. These personnel, numbering approximately one dozen, provide for user program administration, IT (addressing computer administration, data management, and the development of operations software), safety, administrative professionals, building/facilities management, and senior facility managers (director and deputy director). A detailed and updated organization chart can be found at the CNM website here. CNM staffing is rounded out with approximately two to three dozen temporary appointments, which include postdoctoral fellows and visiting graduate students, as well as interns during the summer months.

Overall, CNM's continued success and leadership will depend, in large part, upon: (i) our ability, along with our users, to continue to perform world-class science; (ii) investments in differentiating and leading-edge research tools that attract the best scientific users; (iii) recruitment and retention of talented scientific staff; and (iv) importantly, our ability to anticipate and influence strategic areas that will define future nanomaterials research. These four objectives need to be accompanied by a realistic staffing plan and an investment plan that is compatible with projected budgets and turnover. Going forward, we plan to add research staff hires in key national strategic areas that link to our thematic research in each of the following areas: clean energy, AI/ML, sustainability, biopreparedness, advanced characterization, and QIS. Additionally, as our number of users continues to grow, we also intend to make new technical staff hires in the capabilities areas with the heaviest use that map to our strategic growth areas. The approximate rate of these hires during the next five years is 1 FTE (full-time equivalent) for research staff and 1 FTE for technical staff per year. Finally, we note that the CNM believes in a supportive and diverse work culture, and believes it maximizes our research and user science output and impact. More details on our Diversity, Equity and Inclusion (DEI) efforts are given in **Appendix 2**.

Safety

CNM has responsibility for environment, safety, health, and quality assurance (ESHQ) aspects of the facility's operations and, through policies and procedures, defines how responsibilities are delegated from the director through line managers to technically competent staff members supporting user research activities. The CNM program complements Argonne's laboratory-level safety program by incorporating methods, controls, and a work approval approach tailored to the risk characteristics of a user facility and the materials, instruments, and processes that constitute CNM operations. The specifics of the program are periodically updated to ensure compliance with evolving standards, Argonne safety program evolution, and emerging information on hazards. A certified ESH professional is dedicated to help CNM better ensure research productivity in conjunction with efficient implementation of the applicable ESHQ standards and requirements. The CNM staff collaborate on important ESH projects, such as the Hazardous Gas Response Team in the Nanoscience and Technology Division. This team developed their activities and formalized them in a guide and job aids, which are maintained by division safety and operations personnel.

The guide and aids define appropriate and safe paths assuring prompt and effective control of risks signaled by the facility's toxic gas monitoring system. The team has a balanced combination of safety, operations, and research experience to appropriately respond to gas monitoring system alerts. CNM continues to employ a precautionary approach where there is uncertainty about the hazard potential of new chemicals, including nanomaterials. This concept guides the conduct of hazards analysis and specification of precautions when handling nanomaterials. In this way, CNM contributes to a better understanding and management of ESH concerns associated with nanomaterials and nano-enabled products.

Cybersecurity

The CNM's cybersecurity operations are driven by Argonne's cybersecurity posture, which is a required component of the Lab's Authorization to Operate, issued by DOE via the Argonne Site Office. The cybersecurity plan for the Lab is presented in a National Institutes of Standards and Technology-driven framework documented in Argonne's Cyber Office's Cyber Security Program Plan (CSPP). This is a 375+ page document covering perimeter protection, internal segmentation/separation, network/system monitoring, active response, incident response, configuration management, system management, best practices, auditing/self-assessments, documentation, physical controls, etc. The CNM implements the components of the CSPP to meet the requirements. In addition, CNM augments the existing policy with additional controls, like lab system protection using micro-segmentation to provide additional protections for systems that may, due to necessity, deviate from best practices, in order to meet the scientific mission, such as for systems attached to research equipment locked to a particular operating system.

Data Management

CNM recognizes effective data management has the potential to increase the pace of scientific discovery and promote efficient and effective use of funding and resources. To advance this principle and others defined by the DOE Policy for Digital Research Data Management, CNM has established the following research data management goals:

1. Reliably capture and describe scientific data in a way that facilitates preservation and reuse.
2. Create quality metadata for data discovery, preservation, and provenance.
3. Design and implement interfaces for users to obtain and interact with data.
4. Preserve collected data for long-term users.

Data are stored and curated in compliance with Argonne's data retention policy for scientific research data. This includes the use of granular controls and security measures necessary to keep data safe until it is intended to be shared. The CNM also has a data-retention policy for user-generated data, which is described in our User Access Program (link) documentation.

5. User Community

The CNM strives to attract a diverse user community that performs high-caliber, impactful research. Our user community includes researchers from across the country and around the globe. Figure 5-1 displays the diversity of CNM users as a function of their affiliation, showing that nearly half of the approximately 700 users per year are from U.S. academia, with representation of non-Argonne users from international, industrial, and other government organizations as well. Figure 5-2 displays the diverse fields of research represented by CNM users.

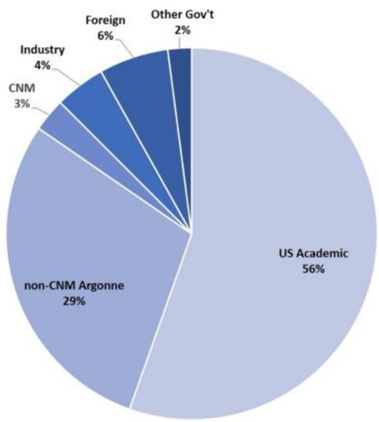


Figure 5-1: *Institutional affiliations of CNM users by affiliation during FY 2022.*

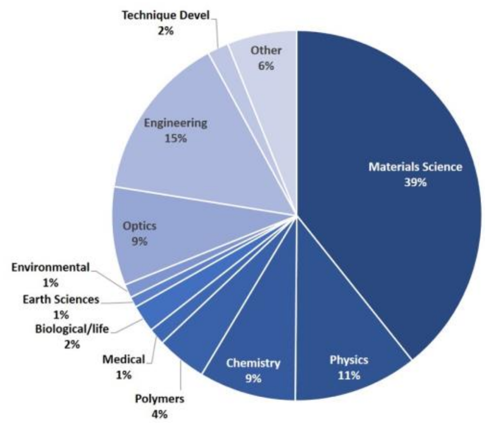


Figure 5-2: *Fields of research identified by CNM users during FY 2022.*

Our users, together with CNM's research/scientific support staff, are continuing to publish in peer-reviewed journals at a high rate: FY 2019 = 337; FY 2020 = 334; FY 2021 = 327; and FY 2022 = 289. In FY 2022, 54% of our publications were in journals with an Impact Factor greater than 7. The global COVID-19 pandemic that began during FY 2020 impacted our ability to host our usual complement of users. However, demand was still strong, and we were able to accommodate the work of 631 unique users that year. Furthermore, in FY 2020 and FY 2021, we invested significantly in remote-access capabilities, which enabled users to remotely access more than 30 tools during this period. This effort resulted in the CNM's ability to host more users in FY 2022 than during the pre-pandemic period. We are planning for a growing remote access effort, which will be a means to leverage our research impact and reach more users in the coming years.

The objective of the CNM user program is to provide the user community with access to equipment, facilities, and expertise supporting CNM's overall focus on nanoscale materials. User time is allocated through a proposal submission and review process in which CNM principal investigator staff, management, and external reviewers all have key roles regarding feasibility, scheduling, and scientific merit. The goal is to operate a process that is open and based on the scientific and technical quality of the proposals.

Determining a technical strategy that will maximize our impact to science and benefit our users requires us to anticipate future user needs, as well as help shape future areas of focus in the nanosciences. Our future capability and facility upgrades are based upon prioritization of our science, staffing, infrastructure, and equipment needs. The needs of our scientific themes are a starting point for setting our prioritization directions and are adjusted by taking into account any new initiatives driven by national need. Discussions with our Scientific Advisory Committee, our User Executive Committee, and science strategy discussions during CNM's annual strategic retreat further inform this prioritization. Emerging areas of research in the nanoscience community are also strongly considered, for example, as defined by national research agency funding directions and strategic documents (for instance, the Office of Science Basic Research Needs Workshops and Roundtables), which provide an indication of future user needs. Feedback from an annual user survey administered at the end of each proposal project is another mechanism that we use.

CNM is also actively expanding the scope of its science toward future industrial impacts, seeking opportunities beyond its industrial user program. Recently, an Industrial Collaboration Committee formed and organized a successful workshop, CNM–Industry Collaboration Opportunities, at the CNM/APS annual user meeting to reach out to industry users. The purpose was to let them know about the capabilities and expertise available at CNM and accelerate new collaborations with industry users. This type of outreach to industry will continue. Current industrial partners include major organizations such as John Crane, Inc., Magna International, and Boeing. Smaller partner companies include Euclid Tech Labs, Sentient Science, Axion Technologies, QDIR, Lam Research, Ragaku, Microtech, Frore Systems (a Silicon Valley startup), and Iris Light Tech (a spin-off from CNM).

We note that AI for science feeds into all three of our themes and has the potential to revolutionize user experience and efficiency. Here, CNM scientists are developing tools and methods for faster and more accurate molecular modeling of materials, more efficient interpretation of electron and X-ray microscopy data, and new approaches that will combine ML with a physics basis. Exciting new opportunities are also resulting from combining automatic synthesis robotics with AI/ML. We are deploying AI/ML tools to understand and control both the kinetics and nonequilibrium thermodynamics to enable autonomous synthesis of materials. Example targets for materials processing lie in mechanical or electrical conductivity of polymers, where robotic processing is combined with automated characterization in a feedback loop to improve the targeted property. Research on "digital twins" will allow users to explore characterization experiments and conditions before arrival at the CNM, enabling users to efficiently target the experiments most likely to succeed.

Finally, we emphasize that CNM seeks to provide a welcoming environment for diverse users and staff across different career stages, institutions, and nationalities (**Appendix 2**). We recently established a diversity, equity, and inclusion committee that will further enhance feedback between users and CNM staff, to continue to ensure a positive and respectful workplace culture. We are also planning additional outreach to minority serving institutions, as well as planning to establish formal opportunities for students at such institutions to work at the CNM.

Appendix 1.a: Major CNM Capabilities

The CNM maintains a large suite of capabilities, while regularly upgrading or procuring new capabilities. A full list of capabilities can be found at our website, link. Given here are detailed descriptions of major CNM capabilities that have been developed by our staff, where additional descriptions are warranted to aid users who may be interested in accessing them.

Quantum Information Science (QIS) Capabilities

For the past several years, the CNM has added several new tools focusing on QIS research, dramatically impacting our activities within the Quantum Materials and Sensing theme. The CNM is the first user facility among the NSRCs that offers comprehensive capabilities for studying coherent interactions in solid-state optical and spin-based qubits for QIS. Examples of the new tools and capabilities follow.

Ultra-low-temperature, dilution-refrigerator (DR)-based experimental systems are essential to conducting QIS research but are expensive and thus beyond the reach of many users. To attract a broad range of users and collaborators, CNM set up a new low-temperature laboratory dedicated to research covering most qubit platforms: from superconductor to semiconductor qubits and defect centers to single electrons. This laboratory opened to users in September 2019. It is the first millikelvin lab within the NSRCs. The system is equipped with microwave spectroscopy capabilities for spin-based studies, and soon, a femtosecond ultrafast laser system will be integrated with the DR system. A second tool, the adiabatic demagnetization refrigerator (ADR), became available to users in the October 2021 user proposal call. The ADR offers faster cool-down times than a dilution refrigerator, with a slightly higher base temperature of 30 mK. It is also equipped with microwave spectroscopy capabilities for studying spin excitations in nanomaterials. Collectively, these systems will enable studying ultrafast quantum dynamics, single-molecule imaging and sensing, collective spin waves (magnons), and single-atom/molecule electron-spin-based QIS research.

We have also developed strong quantum entanglement and transduction (QET) capabilities. These is a comprehensive suite of user tools being developed for the study of quantum optics and hybrid quantum networks linking photons, spins, and magnons. The QET capabilities will enable single-photon correlation spectroscopy, magneto-optical spectroscopy, optically detected magnetic resonance spectroscopy, magneto-electrical spectroscopy covering microwave and optical frequencies, and new QIS computational modeling environments. Already commissioned and available for users is a photon correlation microscope that enables time-gated photon correlation (and a 9-T magnet equipped with a microscope cryostat) for magnetic field studies of spin-dependent energy levels. The instrument has continuous-wave and femtosecond pulsed lasers covering wavelengths from 370 nm to 1300 nm, as well as visible and near-infrared cameras and photon-counting detectors for imaging. It can be used to conduct static and time-resolved spectroscopic studies under external magnetic fields. Our theory and modeling capabilities in QIS play a key role in informing and advancing our experimental capabilities. In particular, we model the effects of dissipation and noise via quantum master equation approaches, use ML to accelerate quantum measurements, and develop quantum algorithms for materials modeling.

Ultrafast Electron Microscopy (UEM) Laboratory

The UEM was installed in spring 2019 and began full user operations in 2021 as the first user tool for UEM within the NSRCs. This tool, in its final form, allows users to access time-resolved stroboscopic imaging, diffraction, and spectroscopy/spectral imaging capabilities, built using a combination of commercial vendors for transmission electron microscopes (TEMs), lasers, detectors, and column integration. The system enables researchers to observe dynamic reversible processes in events that are optically triggered via laser excitation at wavelengths selectable over a wide range from 325 to 2000 nm, and it is being further developed for electrical and mechanical triggering of complex material phenomena. These capabilities will be significantly enhanced over the next three years through a unique correlative integration of ultrafast electron and X-ray microscopy coupled with ML approaches for multi-modal multi-platform data synthesis.

Artificial Intelligence (AI) for Materials Science Capability

CNM has set forth a comprehensive software and hardware upgrade plan, which leverages recent advances in AI to accelerate our computational materials science capabilities. The upgrades include developing user-friendly tools and capabilities and the ability to process large volumes of data effectively. These are highlighted below:

FANTASTX (Fully Automated Nanoscale to Atomistic Structure from Theory and eXperiment)

In this project, we are developing a computer vision-based software tool that will enable close to real-time interpretation of images generated from electron and scanning probe microscopies. FANTASTX will ingest experimental images as input and make comparisons with thousands of atomistically simulated crystallographic or morphological structures to identify an "optimally matched structure" in a manner analogous to facial recognition software. The optimization is carried out using genetic algorithms, where the cost function includes both the degree of match as well as minimization of the energy of the configurations. To start, this capability will be deployed in the EXM group.

BLAST (Bridging length-scales via atomistic simulation toolkit)

Molecular modeling is a powerful tool today. Historically, however, a gulf exists between the handful of research groups that develop new interatomic potential models for materials modeling (often involving several years of effort) and the increasingly large user community that applies these models. Users currently do not have the flexibility to adapt these predefined potential models to problems of their interest. BLAST, a computational workflow tool, will overcome this barrier by allowing users to create their own models by providing a simplified framework that permits users to handle various types of training data, optimize potential functions using evolutionary algorithms, and cross-validate their model predictions. BLAST users will be able to select functional forms available in popular molecular dynamics codes, as well as apply combinations of global and local optimization schemes to generate force fields for molecular simulations. Such a general-purpose tool holds promise for identifying structure-property-processing relationships in various material classes.

Our tasks also include development of a user-friendly, common-platform graphical user interface (GUI) for FANTASTX and BLAST, with GUI development being subcontracted. As of October 2021, we had a beta version of BLAST available to expert users and are currently in the process of licensing the software to

Sentient Science. We also built a GUI and are currently carrying out GUI integration with the backend. BLAST and FANTASTX are now available to general users.

Carbon Cluster

CNM is investing strongly in our Carbon Computer Cluster to handle all problems associated with experimental data analysis and theoretical modeling, with additional capabilities for handling AI problems. Built upon our existing cluster (Carbon), we are targeting 240 CPU (central processing unit)-only nodes and 40 GPU (graphics processing unit)-oriented nodes, each with two data center-class GPUs, all connected over a high-speed InfiniBand director switch and an in-cluster 2-PB storage system. Overall, the planned upgrades will triple the cluster's current compute capacity to 200 teraFLOPS on CPUs and will add 10 petaFLOPS of nominal GPU deep learning capacity. GPUs are natively suited for processing image data and certain ML algorithms, whereas CPUs can be used for all types of modeling.

Superlubricity Science Laboratory

For several years, CNM scientists have been exploring and demonstrating the remarkable result of true superlubricity (near zero friction) at the macroscale using nanoscroll-shaped solid lubricants of diamond nanoparticles wrapped with graphene. This work continues to grow and expand to other materials systems via partnerships with two major industry leaders through two DOE TCF awards that CNM received. Given CNM's leadership in this space, we are continuing to grow our capabilities in the area of superlubricity science at the nanoscale.

A Superlubricity Science Laboratory has recently been configured to include a multifunctional tribometer with integrated confocal microscopy and Raman spectroscopy. We added new capabilities that will allow us to: (i) carry out in-situ TEM of tribological interfaces and changes upon loading and (ii) assess the wear/friction behavior of two-dimensional materials and nanomaterials at elevated temperatures (~500°C), uncovering the detailed wear/friction behaviors of these materials that are unknown today. Combining these new unique capabilities will enable our user community and our own researchers to understand and study nanomechanical and wear/friction behaviors under realistic conditions and across the entire length scales of relevance (nanometer to microns) for the first time.

Cleanroom Facility

In 2017, through funding (~$9 million) from Argonne, the CNM cleanroom was expanded from 12,000 ft^2 to ~18,000 ft^2. CNM users have full access to the expanded portion of the cleanroom. Going forward, in agreement with Argonne management, the additional space will be used to house both CNM and non-CNM tools, and CNM will be responsible for managing the space. This expansion has solved our space crunch for cleanroom tools, and the extra square footage enables our planned modernization of the nanofabrication facilities. Several new tools and upgrades have been added in the CNM cleanroom, including a FastScan atomic force microscope for metrology, replacement of a critical point dryer for micro-electromechanical systems release, a thermal evaporation system from Angstrom, and others.

Appendix 1.b: Future Capability Enhancements at the CNM

As described in the strategic plan, the CNM acquires new tools primarily through three processes that include “recapitalization” with up to 10% of CNM’s operating funds each year, through external grants (particularly NSRC QIS infrastructure grants), and through the DOE Major Items of Equipment (MIE) project. The equipment we are planning to acquire is given in the table below, separated into time frames of near term (1-2 years) and long term (3-5 years). Many of these items address key priority areas such as AI/ML and autonomous synthesis, as well as replacing aging capabilities with upgraded, state-of-the-art tools. Some examples of the tools planned include an automated profilometer station that can be integrated into the autonomous Polybot (described in Section 2) to provide extremely accurate, repeatable, and reproducible metrology for surface characterizations. This is important because surface morphology and thickness are key characteristics for discovery of nanoscale thin film materials, which can be used for microelectronics and energy storage applications. Also contributing to this research direction is the planned acquisition of a Plate Reader Research Hub for automated materials database development. This is a suite of automated tools for basic, but systematic sample preparation and characterization in plate reader format. This would fulfill the needs of nano-bio users interested in high throughput sample preparation and characterization as well as fast database curation. We are also upgrading our computational cluster, Carbon, which not only contributes extensively to the AI/ML research direction, but also is a heavily used tool by remote users of the CNM. Some of the tools are chosen for continuing to build on our areas of strength, such as dynamics studies, which include the transient photoelectron and cathodoluminescence system and the transient absorption microscope equipped with a high repetition-rate laser system for probing small materials with low energy pulses at a high rate of signal averaging. Characterization tools heavily requested by users, such as the X-ray photoelectron spectroscopy tool, are also planned for procurement. Critical tools to continue to enable the high-quality fabrication of nanoscale structures are also planned, including focused ion beam tools, a rapid ion etcher, and a rapid thermal annealer. Additional details and motivations for many of these tools can be found within the strategic plan, particularly for the two NSRC QIS-funded tools (described in Section 2) and for the four MIE-funded tools (described in Section 3).

Tool	Source of Funds	1-2 years	3-5 years	Function
Oxford Instruments Cypher Atomic Force Microscope (AFM)	Recap	X		Advanced AFM placed in the conventional laboratory space to complement AFM in cleanroom
2D Transient IR Spectrometer	Recap	X		Varies pump and probe wavelengths for 2D infrared spectroscopy with temporal resolution
Rapid Thermal Annealing (RTA) System	Recap	X		Replaces aging RTA system for annealing of various films during cleanroom fabrication processes
Rapid Ion Etcher (RIE)	Recap	X		Replaces an aging RIE tool for cleaning and etching during cleanroom fabrication processes
Atomic Quantum Information Surface Science (AQuISS) Lab	NSRC QIS	X		Scanning probe tool for controlling and characterizing optically active spin quantum systems near surfaces

Quantum Emitter Electron Nanomaterial Microscope (QuEEN-M)	NSRC QIS	X		A TEM that integrates time-resolved cathodoluminescence to characterize the electronic structure of quantum emitters
Carbon Cluster Upgrade	Recap	X		Replacement of compute nodes and switch upgrades to keep Carbon Cluster at state-of-the-art
Fluorimeter	Recap	X		Replacement of aging CW fluorimeter
Gallium Focused Ion Beam (FIB) tool	Recap	X		Replacement of an aging FIB to be primarily used for TEM sample preparation
Helium Recovery and Liquefaction System	Recap	X		Needed for continuing operation of helium cryostat-based user tools, from both a supply and cost perspective
Dynamic Double-Aberration Corrected Scanning Transmission Electron Microscope (Dynamic DAC-STEM)	MIE	X		Provides high spatial resolution combined with longer-time dynamics capabilities complementary to UEM
Multibeam Ion Microscope	MIE	X		Complements Ga^+ FIB with state-of-the-art plasma FIB
Transient Photoelectron and Cathodoluminescence Spectrometer (TPCS)	MIE		X	Gives dynamics of carrier energies directly to complement optical measurements (placed at APS to enable structure-function correlation studies)
A Microscope Capable of Single Spin Imaging (mK-STM)	MIE		X	Gives access to milli-Kelvin scanning tunneling microscopy to complement our suite of variable temperature STMs
Magnetic Cryostat Transient Absorption System	Recap		X	Permits examinations of carrier spin dynamics
Transient Absorption Microscope with High Repetition Rate Laser	Recap		X	Permits examination of transient dynamics and spectral response of small samples
Plate Reader Research Hub for Automated Materials Database Development	Recap		X	Suite of automated tools for basic but systematic sample preparation and characterization in plate reader format
Automated Profilometer Station	Recap		X	To be integrated into Polybot to provide extremely accurate, repeatable, and reproducible metrology for surface characterizations
X-ray Photoelectron Spectroscopy (XPS) Tool	Recap		X	A frequently requested tool by users, which enables elemental composition studies and surface chemistry analysis

Appendix 2: Diversity, Equity, and Inclusion at the CNM

The CNM's diversity, equity, and inclusion (DEI) plan is in line with Argonne's core values and complements the DEI activities and initiatives of Argonne and the Physical Science and Engineering Directorate.

Creating a diverse, equitable, and inclusive environment is not a singular project or one-time initiative. It is an ongoing and constantly evolving effort that involves everyone in the CNM and includes all aspects of our user support, our research, and our development projects. Long-term success in advancing DEI values requires identification, understanding, and resolution of existing and potential problems.

To help address DEI challenges, the Nanoscience and Technology Division (NST) DEI Council was established as an advisory body to the NST/CNM leadership in 2021. (Note that NST is the parent division of the CNM.) The purpose of this Council is to create a safe and welcoming work environment for everyone, regardless of their race, color, national origin, religion, gender, gender identity, sexual orientation, age, or disability. In alignment with Argonne's four challenges in the Argonne DEI Strategic Plan, the NST DEI Council has set forth the following goals:

Goal 1: Ensuring employees have a sense that they are valued, supported, and included.

The day-to-day culture of NST/CNM should be one of allyship, fairness, and collaboration, where everyone is valued both personally and professionally, with the goal of creating a physically and psychologically safe work environment, as well as inclusion of diverse thoughts, opinions, knowledge, and expertise.

Goal 2: Ensuring all leaders are developed and have equitable opportunities for advancement.

NST/CNM staff at all levels should have equitable opportunities for advancement. To support this, each supervisor is dedicated to discussing staff goals several times per year and providing opportunities for development. When opportunities arise, supervisors will be active in nominating their staff.

Goal 3: Developing professional development opportunities for employees to grow.

We will take a proactive role in fostering professional career development and recognition of NST/CNM staff. We have instituted Deputy Group Leader positions to help train the next generation of potential group leaders. We will continue to nominate our staff for leadership opportunities across the Argonne campus, as well as external to the Laboratory. We will support training courses supported by the Argonne Leadership Institute, as well as external courses (including those that take place at conferences) that staff desire to take to advance their career development.

Goal 4: Creating a climate for diverse recruiting and equitable advancement.

NST/CNM will continue exploring multiple avenues for talent acquisition and development to promote fairness and diversity in the recruitment, support, and retention of an efficient workforce, with the long-term goal of surpassing the national averages in demographic statistics in STEM-related fields. We are committed to equity in hiring, compensation, and professional development.

Goal 5 (CNM-specific for users): Ensuring users feel valued, included, and supported.

As we grow our user community, we strive to promote equitable access to the CNM. Many underrepresented users and underserved academic institutions, such as minority serving institutions (MSI), may not even be aware of the resources available to them at these user facilities. The CNM plans to target outreach to MSIs by giving talks at specific universities and joining MSI-specific professional groups. We plan to provide information about the CNM, tools and resources available, guidance on how to summit a successful proposal, and funding opportunities for students. Two such opportunities at Argonne are the Science Undergraduate Laboratory Internship and the National Consortium for Graduate Degrees for Minorities in Engineering and Science Consortium.

Some actions that the NST/CNM DEI Council will take in order to advance the five goals discussed above are the following:

- Creating a CNM DEI Webpage for sharing all relevant information with staff and users (e.g., people, events, cultural observances, and strategic plan), which also includes an anonymous online suggestion box
- Regularly contributing to a DEI section in the CNM newsletter
- Collaborating with the NST/CNM hiring committees of any posted positions
- Conducting staff surveys
- Collaborating with the CNM User Executive Committee and the Scientific Advisory Committees
- Collaborating with the laboratory-wide DEI councils and Employee Resource Groups
- Promoting Argonne community outreach and educational programs
- Providing staff with information on DEI training opportunities and activities throughout Argonne

A diverse, equitable, and inclusive culture is associated with higher productivity, innovative solutions, employee engagement, and talent retention. It is the responsibility of everyone in the organization to do their part to create this environment. This plan provides actionable steps that we will take as a user facility and world-class research institution to advance these DEI goals.