Center for Nanoscale Materials Strategic Plan: 2015–2019

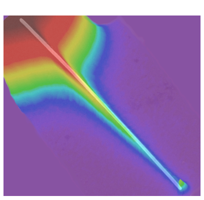
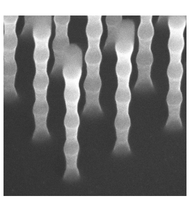
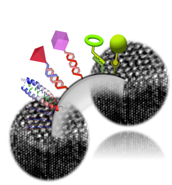
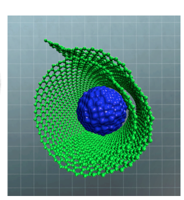
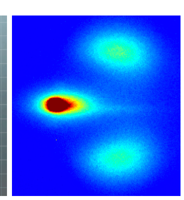
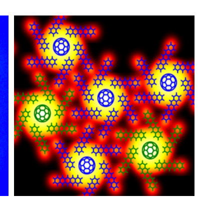
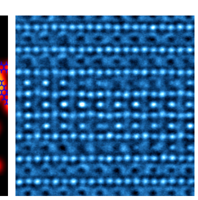








TABLE OF CONTENTS

Executive Summary

The Center for Nanoscale Materials (CNM) is at the forefront of discovery research that addresses its cross-cutting scientific theme of ***Energy and Information Transduction at the Nanoscale.*** The CNM is an integral part of the National Nanotechnology Initiative and, as a DOE Nanoscale Science Research Center (NSRC), supports its scientific user facility mission by enabling research that addresses grand challenges in energy and information transduction, while furthering the DOE missions of energy generation, storage and efficiency. Specifically, we are exploring ways to tailor nanoscale interactions, by discovering, visualizing and manipulating the behavior of hybrid nanomaterials for energy needs. The focus of harnessing the increased complexity of hybrid nanomaterials to yield desired, targeted functionalities is at the core of DOE's scientific mission to discover new materials and design new chemical and physical processes for energy production, conversion, waste mitigation, transmission, storage, and energy efficiency. The CNM research vision also guides and shapes our vibrant and high impact user program. In order to maintain and extend the CNM at the leading edge of nanoscience, we will pursue innovative research programs and develop new capabilities within seven thematic research groups. Research at the CNM is strongly linked to Argonne's strategic initiatives in Materials for Energy, Hard X-ray Sciences, and Leadership Computing, enabling us to leverage developments across the Lab to enhance the world-leading capabilities available to CNM users.

The nanoscience portfolio created by execution of the CNM's user program will also advance the foundation required to develop cutting-edge hybrid nanoscale structures that harness increasing levels of complex functionality via interactions of different nanomaterials in physical proximity. The past decade of nanoscience was dominated by understanding how the properties of materials change when scaled down to the nanometer regime. Significant technological successes that employ nanoscience have resulted from these research efforts. The success of nanoscience for future decades will depend on understanding how to achieve successful hierarchical integration of these materials for advanced functionality and novel applications. A focus is therefore to control the behavior at interfaces in complex nanostructured materials. These materials are aimed at supporting the nation's energy security in fields such as energy storage, solar energy, novel fuels, and energy efficiency.

The CNM mission for user support and staff science is executed through seven research groups that contribute to the cross-cutting scientific theme discussed above. The scientific research groups are described below:

- **Electronic and Magnetic Materials and Devices** – New electron- and spin-based materials and phenomena in constrained geometries are discovered, understood, and exploited for reduced power dissipation, improved efficiency of data storage by atomic and molecular level imaging, spectroscopy, and manipulation, spin current and electrical field-assisted writing, new atomic scale imaging methods, and enhanced energy conversion in organic photovoltaic devices.
- **Nanobio Interfaces** – Functionally integrated biomolecule-inorganic hybrid conjugates are developed, guided by nature's principles to create self-assembled energy gradient architectures capable of spontaneously separating charges upon applied stimuli, for energy and information transduction, catalysis, biosensors, energy conversion, storage, and advanced medical therapies.
- **Nanofabrication & Devices** – Non-traditional nanostructured materials, nanodevices, and nanosystems are fabricated by advancing state-of-the art techniques in nanopatterning, which incorporate hybrid self-assembly and lithographic approaches.

- **Nanophotonics –** Optical energy and its conversion on the nanoscale are controlled by combining the properties of metal, organic, semiconducting, and dielectric materials to create strongly coupled states of light and matter for enhanced chemical/catalytic reactivity, photonic circuits, new chemical and biological sensors, and enhanced optical nonlinearities.
- **Theory and Modeling** – Theory and modeling are used to enable the discovery and characterization of nanoscale materials and devices. Particular focus is placed on molecular conversion and transport at interfaces, the atomistic origins of thermal and mechanical properties, and optical and quantum phenomena.
- **X-ray Microscopy** – X-rays are utilized to probe the structure of new materials and phenomena at the nanoscale with sensitivity to crystalline order and strain, elemental and chemical makeup, and electron spin and orbital states. Development of new concepts and techniques for studying nanomaterials and nanostructures with nano-focused x-ray beams, particularly buried and *in-situ* structures under a range of environmental conditions, enables novel science at the Hard X-ray Nanoprobe (HXN) facility.
- **Electron Microscopy** – Electron microscopy and scattering are used to provide new insight into material structure and behavior at the atomic level, while development of new and innovative resources advances world-class capabilities in electron diffraction, imaging, and spectroscopy.

The CNM provides world-leading expertise and tools to its users, which enable design and creation of new nanomaterials through synthesis, fabrication, characterization, and theory/simulation, and enables insights into their behavior that unlock potential applications. Key CNM capabilities include: the unique HXN based at the adjacent Advanced Photon Source, novel ultrahigh vacuum scanning probe microscope systems, high-voltage electron beam lithography, complex oxide molecular beam epitaxy, a suite of electron microscopes, and a computational cluster. The CNM currently employs 66 staff, including those associated with the Electron Microscopy Center (EMC). The staff contributes unique and world-leading scientific programs in addition to supporting users of the facility.

The CNM's goal for the next five years is to enhance its position at the forefront of nanoscience. This will be achieved by emphasizing activities in two strategic scientific directions that we have identified as being of critical importance: **Hybrid Nanomaterials for Energy Conversion and Storage**, and **Tailoring Interactions at the Nanoscale**. We will make both strategic hires and cutting-edge-equipment procurements designed to optimize staff and user science impact in these areas. Foremost will be our commitment to continue to provide the highest quality experience for CNM users. We will further develop user science business processes to make record keeping and data queries as automated as possible. The innovative science being performed by the CNM staff scientists shapes the user program and guarantees that it will remain vibrant, while at the same time, innovative user science helps formulate future scientific directions for the CNM. The future success of the CNM will be assured by the continuation of efficient integration of user and staff science.

1. Introduction

In order to maintain our position at the forefront of nanoscience and nanotechnology, and to maintain our excellence as a world-class user facility, we need to continue to pursue innovative research programs and develop new capabilities that will strengthen user science at the CNM. We recognize that cross-cutting activities, which link the seven research groups within the CNM, lead to a whole that is greater than the sum of the parts, further enhancing our core science and thus our ability to attract high quality users. For this reason the CNM has identified *Energy and Information Transduction at the Nanoscale* as a research theme that encompasses our scientific vision. Within this theme we are focusing on two strategic scientific directions, *Hybrid Nanomaterials for Energy Storage and Conversion* and *Tailoring Interactions at the Nanoscale*, which will be elaborated in Sec. 2: *Vision for Scientific Growth*. We link the critical pillars of materials discovery, visualization and manipulation on which our scientific vision is built, and which guide in shaping the user program. Our research theme is closely linked to one of the supporting strategies developed in Argonne's strategic plan, namely *Materials for Energy*, and through the HXN we contribute to another of Argonne's strategic directions, *Hard X-ray Science*. These interactions enable us to leverage developments across the Lab to enhance the capabilities available to CNM users. The CNM represents one of a number of DOE-BES scientific user facilities at Argonne, which includes the Advanced Photon Source (APS), the Argonne leadership computing facility (ALCF), and the EMC (to be consolidated within the CNM during FY14, as discussed below). Where appropriate the CNM is collaborating with these facilities to provide a wider range of tools and expertise to our users and will continue to do so.

A key challenge for maintaining the high quality of the CNM user program will be striking an optimum balance between investment in essential, but routine, capabilities and infrastructure that are nonetheless necessary for the success of user science activities, and investment in world-leading capabilities that clearly position the CNM for cutting-edge nanoscience. Input for creating the CNM equipment plan has, and will continue, to come from several sources, as described below.

- Input from the CNM user community and staff, through CNM-sponsored workshops, the annual CNM Users' meeting, strategic science retreats and town hall meetings for the CNM staff, and via informal interactions with staff and users
- Suggestions from the User community that come to the CNM via the User Executive Committee
- Identification of user projects that require capabilities that do not yet exist in the CNM; also of user demands that exceed available resources.
- The CNM seeks advice from the Scientific Advisory Committee (SAC) on a regular basis. The SAC continues to provide valuable input on future scientific directions and on resources that are needed to support these scientific directions.

Together with the challenge of attracting new talent to the CNM and the tools and latitude they need to become world-leading scientists, comes the challenge of the retention of world-class scientific staff in a user facility environment. We believe that the innovative science being performed by the CNM staff scientists shapes the user program and guarantees that it will remain vibrant, while at the same time, innovative user science drives future scientific directions and capability development for the CNM. The success of the CNM depends on the demonstrated ability to integrate user science and staff science in an effective fashion.

Moving forward we will explore the possibility of offering a 'concierge system' of user support, in particular for projects that make use of our imaging and spectroscopy capabilities. In this model, users would submit proposals in which they lay out the scientific or technological challenge that they wish to address at the CNM, and we work with them to propose the most suitable instruments and effective strategies to address their needs. It is not a model that would work for all users, but we believe that it would help to lower the 'activation barrier' for those of our users who are not experts in the field of imaging and visualization, and would also help us to make the most appropriate use of our resources. We anticipate that this concierge system would be also especially beneficial for first time industrial users.

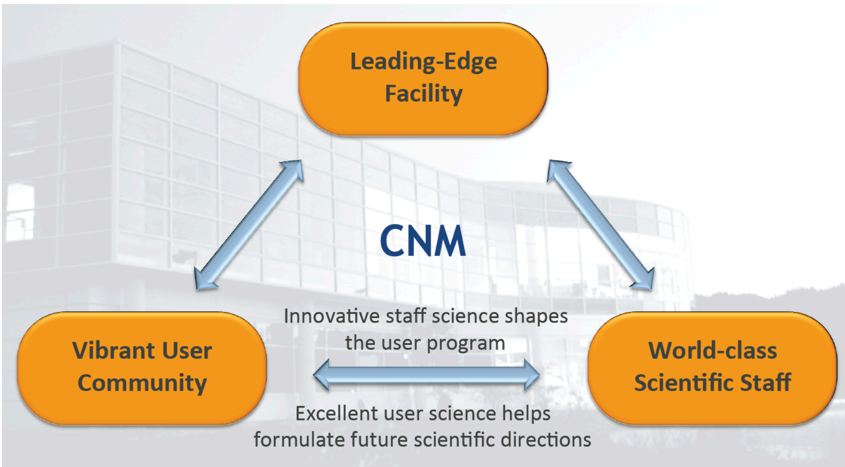


Fig. 1 Close link between CNM User Program and Core Science program

2. Vision for Scientific Growth

Nanoscience and nanotechnology are critical to the scientific and technological advancement of the nation. The past decade of nanoscience was dominated by understanding how the properties of materials change when scaled down to the nanometer regime. Significant technological successes that employ nanoscience have resulted from such research efforts. The success of nanoscience for future decades will depend on understanding how to manipulate hierarchical integration of materials for advanced functionality. The CNM and other NSRCs are a pivotal resource for Office of Science activities. For example, the mesoscale science initiative has a focus to build complex materials based on hybrid nanomaterial building blocks. The new properties that emerge will be based on interactions at the nanoscale that are tailored to yield 'designer' functionalities. We will develop expertise and tools aimed at solving scientific challenges in this area, and thus support new and continuing users. Overviews of the scientific challenges in two focus areas are outlined below, together with our management plan for supporting them. The key elements of the plan are continued outstanding hires of early- and mid-career scientists and technical staff, and acquisition of new state-of-the-art equipment. Going forward the CNM will employ a strategy for increasing its collaborations and partnerships inside, as well as outside, Argonne by increasing the number, impact and reach of Partner User Proposals. Furthermore, the Nanoscience & Technology Division in which the CNM is now housed will actively strive to increase the number of CRADAs and other funding sources, enhancing the CNM user program by leveraging the impact of such activities.

2.1 Hybrid Nanomaterials for Energy Storage and Conversion

We will address the challenges that arise from synthesizing and characterizing complex hybrid nanomaterials formed by both self-assembly and top-down approaches. Discovery of new electronic, optical, or magnetic functionalities at the nanoscale, produced by closely coupling nanomaterials with different behaviors, is the goal of this work. We are ideally positioned to predict, design, create, and characterize (spatially and temporally) hierarchically-organized structures that exhibit new pathways for energy transduction, including propagation of excitons, charge, and spin. Within our proposed research program, just a few examples of hybrid nanomaterials under exploration include core-shell nanostructures that encourage separation of electron and hole wavefunctions (type-II structures), metal-semiconductor nanostructures to create mixed quantized/collective states for ultrafast nano-optical phenomena, and atom-by-atom assemblies of nanostructures made with our scanning probe tools. Our fabrication and synthesis approaches have demonstrated success in creating highly homogeneous nanomaterials, which is a critical requirement for utilizing (rather than being hostage to) size-dependent effects of nanoscale materials.

Combining nanomaterials with different functionalities enables us to explore the emergent behavior that arises and to create materials with increased complexity. Our unique characterization approaches include atomic scale probes, such as synchrotron X-ray scanning tunneling microscope (SX-STM) at the HXN facility, laser-coupled STM, as well as *in situ* structure-function correlations at the nanoscale performed at the APS. We will further make use of hybrid materials created by directed self-assembly to support novel lithography protocols aimed at developing features on the sub-10 nm scale. We intend ultimately to produce new opportunities in hybrid nanomaterials of relevance to the DOE mission for energy transport, conversion, storage, and efficiency that will inspire CNM's users to expand into this emerging field.

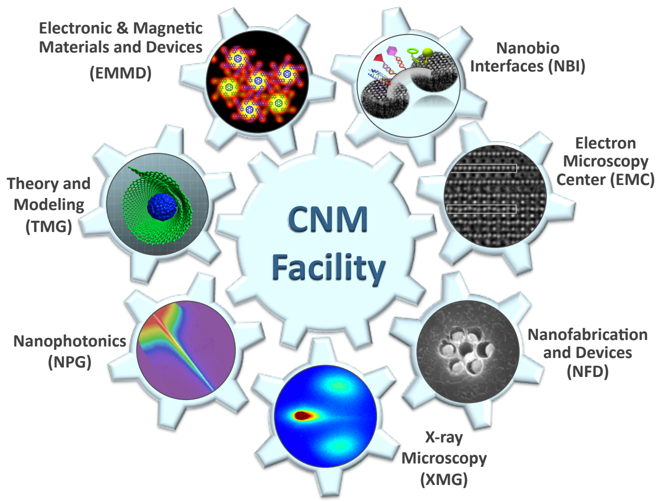
2.2 Tailoring Interactions at the Nanoscale

Tailoring nanoscale interactions is a new frontier in nanoscience that the CNM is well positioned to lead. Closely spaced nanoparticles can interact strongly to produce novel properties and opportunities that exceed those of the building blocks. An obvious example is natural systems, where coupled nanostructures produce long-range and hierarchical communication processes that underpin living organisms. Current technology makes it difficult to explore these nanoscale interactions in controlled artificial systems. In the CNM, we are developing important new approaches to fabricate, *and actuate*, nanostructure assemblies to produce tailored responses. A particular focus will be driving nanomaterials beyond a linear response into the nonlinear regime. This will open new directions in the areas of nonlinear optics and nonlinear mechanical behavior. Nonlinear mechanical behavior is a field that is barely explored even at a macroscopic level. Investigation of such driven, complex mesoscale systems creates a critical demand for quantitative characterization techniques capable of high-sensitivity visualization of chemistry and nanostructure, and of the response of the systems to *in-situ* nanoscale manipulation. Some examples are the use of micro-electromechanical (MEMS) structures functionalized with nanoparticles to electrostatically position the interacting nanoparticles with sub-nm separation control, and the use of ultrahigh vacuum scanning probe microscopes to position individual atoms with atomic precision to confine electrons. Separation variation alters the non-linear optical behavior through coupled plasmonic modes, with the possibility to control the degree of charge exchanged between the nanoparticles.

3 Expansion of capabilities

3.1 Integration of the Electron Microscopy Center

A key activity that enhances the experience for CNM and EMC users is the consolidation of the EMC within the CNM. This integration enhances scientific synergies, of which there are already many. It also makes it easier for users to access a broader set of tools and expertise via a single user-proposal system. We have already transferred the EMC into the home division of the CNM, the Nanoscience and Technology Division (NST). With this transfer, all the major functions of the EMC including management, safety, IT, and building operations are administered within this new division. The merger of the EMC's proposal system has been established, and the first combined call for proposals using a single system has been successfully completed. The combined proposal system is being administered by the CNM User Office. The EMC website is now administered and updated by the same IT staff that maintains the CNM site. Full integration will be accomplished by the end of CY2014.



The EMC staff will form a seventh group within the CNM officially beginning October 2014, although this part of the integration is essentially already complete. We propose to keep the EMC name for this group because of its name recognition and stature within the established electron microscopy user community. From a management standpoint, since the EMC and CNM are both within the NST division, the EMC leader, Dean Miller, is a member of the NST management team, and attends CNM group leader meetings.

3.2 Improved User Office capabilities

It is not a surprise that overall user efficiency and satisfaction increases with ease of access and minimized paperwork. Currently the CNM has a well-established business process and a good level of automation in order to evaluate, approve and authorize user work. Going forward we will further develop our User Office business software. Specifically we will improve the user publication tracking process to rapidly measure and evaluate our performance and output at any given time. Another planned improvement is a new tool scheduler, and web-based logbooks that can be integrated into a usage report.

Within the next five years the goal is to move to a paperless User Office where all signatures, door access, user agreements, *etc.* are web-based and real time accessible. This will simplify procedures for both users and CNM staff, and will readily enable access to performance metrics, such as publications, tool usage and user numbers.

3.3 NEMXIS: A New Dedicated Beamline for X-ray Nanotomography and SX-STM

We propose to meet the evolving imaging challenges of our frontier nanoscience programs by developing a Nanoscale Energy Materials X-ray Imaging Station (NEMXIS). It will be situated at the Sector 26 Bending Magnet line of the APS. NEMXIS will provide a dedicated home for both transmission X-ray microscopy (TXM) and SX-STM. These capabilities currently share the Sector 26 Insertion Device beamline with the nano-diffraction program. Separating the three capabilities into three dedicated experimental stations will enable a tripling of the available beamtime for user access while taking full advantage of the APS Upgrade project that is underway. A dedicated SX-STM station will provide a permanent home for complex instrumental developments, such as the planned low temperature SX-STM. It will also provide access to soft x-rays, to a few hundred eV. This will open the door to atomic and molecular scale elemental, chemical and spin-sensitive resonant experiments, including for magnetic and organic systems containing transition metals, The APS Upgrade will provide an opportunity for dynamic imaging of real materials under real conditions in real time. This opportunity will be met by creating a next-generation platform for *in-situ* imaging of chemically and electrically active nanoscale energy materials.

3.4 Enhancing Cleanroom Capabilities

In order to continue providing our users with timely and efficient access to state-of-the-art nanofabrication equipment, we will upgrade existing lithography and etching capabilities. Our JEOL 9300 electron beam lithography system is one of the most popular tools in our cleanroom. This system is close to reaching ten years of continuous use and it will require a significant upgrade in order to provide reliable and reproducible sub-10 nm patterning capabilities.

We have recently incorporated a used ASML step-and-repeat lithography system that will help to accommodate the needs of users requiring wafer scale patterning of micro and nanostructures. While the current system will support a large number of projects with critical dimensions around 500 nm, a more precise stepper capable of large scale patterning of features close to 100 nm will be essential to achieve very large scale integration (VLSI) of nanostructures. Conventional DUV steppers (248 nm laser) can be used for this purpose, and we foresee an important demand in the next five years. Furthermore, since step-and-repeat lithography is the standard patterning technique used in the semiconductor industry, availability of this technology will be attractive to users interested in commercializing nanotechnology.

4 Key Staffing Decisions

We will make strategic hires across the CNM in support of our strategic scientific directions, and to enhance the capabilities, expertise and support available to our users. In accordance with our previous five-year plan, we opened two junior staff positions to rehire in the Nanophotonics and the Theory and Modeling groups; we will make offers to candidates before the end of 2014. The planned new hires include both scientific PI staff and scientific support staff who will also work to enhance the user program. There is a close tie between our strategic plan for equipment acquisitions and our staffing plan for FY15–19, both of which tie in with our vision for scientific growth for the future, presented in Sec. 2 above. The plans are of critical importance to our user community and to address key challenges of nanoscience in the future.

4.1 Scientific Staff

- Our most pressing need is for a junior staff scientist with a physics background and expertise in developing models and theory in the areas of transport, nanomagnetism and spintronics. The CNM has strong experimental programs in exploring spin and charge transport in hybrid materials, both through synthesis and through advanced characterization, and we would like additionally to be able to provide the third pillar of theory and modeling to provide our users with comprehensive expertise in this field and bring the theory group back to its original staffing. Interviews for this position are in progress.
- We also have an urgent need to make a junior scientific staff hire in Nanophotonics. This is a direct replacement for a staff member who has recently left CNM to take up a faculty position. Our goal is to hire a scientist with expertise in photo-catalysis, time-resolved spectroscopy and single-photon emission from hybrid nanomaterials, to provide user support in this burgeoning field and to carry out best-in-class staff science that will benefit our user community. Candidates for this opening are currently being interviewed.

- Additionally, recognizing that a strength of the Electron Microscopy Center is *in-situ* transmission electron microscopy (TEM), we plan to hire a junior staff scientist with expertise in this broad area, who will complement the existing scientific expertise in *in-situ* microscopy of nanomaterials across the CNM and EMC. *In-situ* electron microscopy is a critical technique to visualize the behavior of hybrid nanomaterials and the way in which their functionality is related to nanostructure and composition.
- Our fourth proposed key hire is a junior staff scientist to support our oxide MBE program. The oxide MBE system at the CNM is unique as a user facility tool and is key to our ability to synthesize layered oxide hybrid nanomaterials with atomic precision. This capability enables us to engineer interfacial electronic behavior with exquisite control, and thus to tailor the interactions between adjacent layers. However, this capability is currently staffed only at the 25% level as a result of staff relocations. We would like to replace the 25% support by a full-time staff scientist to grow a science program and to enhance user support via digitally synthesized samples and value-added expertise.
- Our fifth key hire is a junior staff scientist with expertise in electrochemistry to support the experimental program in the development of gradient architecture materials for energy storage. This is an area of expertise that is currently lacking at CNM, despite our successful programs in tailoring nanoscale interactions in nanomaterials for batteries.
- Our sixth key hire is a junior staff scientist with expertise in x-ray microscopy and/or spectroscopy, to develop and support the NEMXIS beamline with its SX-STM and TXM programs. The anticipated growth, as well as the scientific benefits of dedicated efforts in these two areas, requires a full-time staff scientist. This program will significantly expand access to the nanoscale surface and 3D x-ray microscopy instrumentation for both CNM users and staff.

4.2 Support Staff

In addition to expanding our complement of staff scientists, we recognize that the user program requires substantial scientific support. We are indeed fortunate to have the high level of scientific and facility support that we are able to provide to our users, but the increase in user numbers, particularly in the clean room, has stretched this support to its limits.

- We would like to hire a support scientist with expertise in cleanroom technologies who would provide instrument support in the clean room and also support users with basic activities.
- The second scientific support position that we believe to be strategically important is a software developer for the HXN facility, who would provide new, high-level control software to facilitate automated operation of the HXN instruments, and to package existing analysis routines and algorithms (*e.g.,* for reconstructing Bragg ptychography data) to provide rapid data analysis and visualization during experiments.
- Finally, there is a need for an additional IT staff to support the User Office in further developing and automating the user office business software, which will be critical because of the increasing user numbers and ever-increasing demands for quick retrieval of database information.

5. Key Instrumentation Decisions

To support the strategic scientific areas discussed above, and to keep the CNM as a world-leading user facility in nanoscience, we see an opportunity to develop further visualization capabilities with temporal and spatial resolution for exploring the *local* behavior of complex nanosystems. Monitoring and understanding the transfer of energy, charge, and/or spin in hybrid nanomaterials, for example across interfaces between dissimilar nanoparticles, relies on being able to probe behavior in response to external stimuli (*e.g.* light, magnetic and electric fields, and chemical environment) with appropriate spatial and temporal resolution. *Controlling* this energy transduction additionally requires a detailed understanding of the way in which behavior is influenced by structural and compositional parameters.

Ultimately our goal is to develop capabilities that will enable users access to 5D visualization capabilities, which include the three spatial, one temporal, plus an external-stimulus dimension. An important part of this activity will be to develop *ab initio* approaches to enable real-time determination of atomistic configurations in nanostructures from x-ray absorption and inelastic scattering measurements. These approaches will use a genetic algorithm in conjunction with *ab initio* calculations of spectra and energies. We will continue our interactions with the Mathematics and Computer Sciences Division at Argonne to develop methods to visualize and mine the complex data sets that we produce. Integration of the EMC into the CNM provides an opportunity to exploit the synergies present between the imaging and spectroscopy capabilities in the two facilities.

We will work closely with the Advanced Photon Source to ensure that our strategic directions for development of the HXN at the Sector 26 Insertion Device beamline are synergistic with the MBA Lattice envisioned for the APS Upgrade, and the two-orders-of-magnitude greater brightness it will deliver. This will directly translate to 100x greater nano-focused flux on a sample, enabling transformative science with the scanning probe HXN instruments. Nano-resolved studies of spin, charge, and orbital ordering in systems such as complex oxides will become feasible in scanning nano-diffraction mode, as will trace element analysis in nano-biological systems in scanning fluorescence mode.

Instrumentation developments that we plan to pursue at the CNM include:

- Upgrades to our unique HXN facility, including the capability for *in-situ* temperature and electric/magnetic field manipulation, and implementation of a diamond phase retarder for generation of polarized x-rays for nanomagnetism experiments.
- Development of the NEMXIS beamline to provide a dedicated facility for rapid, *in-situ,* 3D imaging of nanoscale materials by TXM for energy harvesting, transduction and storage, and for nanometer-resolved surface physics by SX-STM.
- A lab-based X-ray microscope for 3D characterization by nano-tomography and to “pre-screen” samples, and *in-situ* sample holders for the HXN. This will enable us to devote time on the HXN only to worthy experiments, making best use of this precious and competitive resource.
- An electron paramagnetic resonance (EPR) system because EPR has the ability to unambiguously identify the constituents participating in light-induced charge separation. A requirement for high quantum efficiency of light conversion is fast separation of charges and very slow back reaction. It is proposed that spin dynamics alters the longevity of charge separation, and with the proposed instrument we would be able to addresses this challenge through direct observation of temporal dependence of spin dynamics during the course of photo-induced charge separation.
- Development of a time-resolved photoelectron spectrometer for directly monitoring the temporal evolution of electron energies in hybrid nanomaterials, thereby revealing key excitonic interactions and charge separation phenomena that are highly complementary to information acquired through transient absorption or emission studies.
- Development of novel scanning probe tools and techniques that allow user research at milli-Kelvin temperatures and under high magnetic fields, tailored to the energy resolution necessary for exploring quantum phenomena. User access to such techniques is essential for exploring the unique spatially-variant electronic and magnetic states anticipated in hierarchically-structured mesoscale systems.

- Combining optical excitation with a controlled environment and enhanced time resolution *in-situ* in the TEM to explore self-assembly and the tailored response of hybrid nanomaterials to external stimuli.
- Development of an aberration-corrected scanning transmission electron microscope (STEM) optimized for analytical characterization, including spectroscopy. The proposed instrument would be configured to enable the simultaneous collection of structural, chemical, electronic, and property data of single nanoscale particles, in addition to allowing multi-modal probing of hybrid nanostructures. Of importance are parameters, such as the bonding across interfaces in these hybrid structures, that can be directly probed by electron energy loss spectroscopy, and that controls behavior such as charge transfer across the interfaces
- Acquisition of a DUV step-and-repeat lithography system capable of VLSI of nanostructures

Computation and modeling are part of many user projects carried out at CNM, in addition to forming a integral part of much of the CNM's core science program. The CNM computing cluster facilitates a significant fraction of the CNM user science. The cluster is a combination of nodes from several acquisition phases, the earliest of which were procured seven years ago. A replacement of the earliest phase is in progress in FY14, albeit at a comparably small scope. To maintain competitiveness and feasibility of our staff- and user-driven modeling efforts, we envision a replacement of the next oldest phase over the course of the next two years, thereby increasing our compute capacity while improving power efficiency.

In the longer term we believe that it is critical to keep our nanofabrication capabilities state-of-the-art in order to maintain our leadership position and provide state-of-the-art capabilities to our user community. In this area we plan to develop novel, high-throughput nanofabrication methods for top-down fabrication below 10 nm; this will allow us to build nanostructures into mesoscale assemblies in which we can tailor interactions by informed positioning of the individual components. Critical to this activity is the need to deposit thin films in a controlled manner, and the purchase of a dedicated electron beam evaporator is envisioned. Secondly, as the nanostructures that we fabricate become more complex, so the need to image them at different stages of the fabrication process at high spatial resolution becomes more critical. Therefore, a state-of-the-art focused ion beam system will be needed to replace the older model. Finally, we see a need to upgrade our electron-beam writing capabilities with a state-of-the-art system (2 nm beam size or better). The e-beam writer is a key nanofabrication capability, which will need replacing as its ten years of continuous (basically 24/7) use nears. Tools that are ten years or older often cannot be covered under a service contract, hence their maintenance costs could become prohibitive.

6. Infrastructure Upgrades

Keeping the CNM as a center of excellence for nanoscience research is stretching the infrastructure of the building and IT systems to its limits. Thus our strategic plan includes mitigating goals. The CNM building is Silver LEED certified, and we continue to strive for energy efficiency and sustainability.

In order to save on waste disposal costs we would like to purchase a waste compactor to reduce the volume of non-hazardous solid waste generated by CNM. Since our waste disposal is billed by volume, this would decrease our costs and reduce our contribution to land-fills. We also intend to explore the option of timers and/or motion sensors for the office and lab corridor lighting system to conserve energy and to save on utility costs.

The CNM currently has a 3,000-gallon reservoir for liquid nitrogen (LN_2). The boil off from this reservoir supplies high purity dry nitrogen to laboratories in the CNM facility. The CNM uses approximately 10,000 gallons per month. There is no backup supply for this utility and so a second 3,000-gallon reservoir is needed for sufficient supply and backup.

Another purchase that is strategically critical is for a helium liquefaction system. The CNM has developed considerable expertise and unique capabilities in the exploration of nanosystems at low temperatures via scanning tunneling microscopy. As our range of systems in the user program increases, so the demand for liquid helium increases. Spiking prices and tightening supplies threaten the future of scientific research at low temperature. Therefore, we are planning to acquire a helium liquefaction system as a joint effort with APS, which also has a large *l*-He usage.

The generation rate, volume, and complexity of data being produced in CNM already fall into the "Big Data" category. Examples are real-time tracking of nanoparticle self-assembly by TEM, all-atom molecular dynamics simulations, and x-ray Bragg ptychography with the HXN. We envision the need to prepare for significant increases in these directions going forward. We will need to invest in new data backup, storage, and real-time analysis capabilities to meet our expected requirements, which extend to PB data volumes and GB/min data transfer rates.

Finally we would like to upgrade the user seating area by enclosing part of the second floor gallery space to reduce the noise coming from the foyer and the eating area. Additionally, the CNM will be able to use this room for special events including user meetings, conferences and staff meetings.

7. User Program and Outreach Activities

The CNM user program continually strives to attract the highest-impact users possible, including researchers from across the country and around the globe. Many of our capabilities and staff expertise are one-of-a-kind – people want to work at the CNM because it houses the most sophisticated and unique instrumentation together with scientists who can contribute intellectually to their endeavors. Therefore the main outreach goal is to make an ever-widening circle of researchers aware of the CNM's potential in order to push the frontiers of nanoscience. Towards this goal, CNM scientists will continue to comprise a critical component of CNM outreach by publishing high-impact papers, delivering invited talks, and organizing symposia and conferences. The user office manager also will continue to promote the CNM's scientific achievements through the website, newsletters, and social media, to suggest ideas for workshops and other events based on user feedback, and to explore new avenues for such promotion. With greater awareness will come increased requests to access the CNM. We will ensure that as many projects as possible of the highest potential impact are awarded access by means of fair and equitable processes.

A critical component of a healthy and diverse user community is a representative percentage of industrial users. In 2013 and 2014 the CNM managed to establish a CRADA, a Work-For-Others, as well as started its first proprietary user proposal, with two more companies interested in working in close collaboration with the CNM in the coming year. We also explore possible partnerships for cross-facility participation by industrial partners together with the APS and ACLF at Argonne. Furthermore, we will expand efforts to collaborate across Argonne's scientific (non-user facility) divisions to ensure that CNM is well integrated in the Lab's strategic initiatives, such as in the Imaging Institute, Applied Materials and Novel Devices, and Big Data, as they evolve.

8. Refocusing of Research Topics

The CNM is refocusing a large part of its research efforts on hybrid nanomaterials and on controlling interactions in these systems. We recognize that there are concurrently research areas that we will no longer pursue at the same level as in the past few years. For example, although we will continue to synthesize single nanoparticles, our main internal focus will be on incorporating these into hybrid structures.

Similarly our theoretical studies of catalyst systems will focus more on photo-catalysis and on catalysts composed of earth-abundant materials, such as transition metal oxides, rather than on precious metals. This will mesh well with our proposed experimental program in photo-catalysis.

We have had an active program exploring the propagation of surface plasmon polaritons. Our refocus will be on active plasmonic structures derived from nonlinear materials, and on incorporating them into nano-mechanical devices, strongly coupled metal nanoparticle systems, or hybrid nanomaterials. Such systems can be tailored to provide greater enhancement of the local electromagnetic field and modulation of the local refractive index than isolated nanoparticles.

We are planning to move the emphasis for the near-field scanning optical microscope (NSOM) away from visible wavelengths and into the infrared spectral region in order to explore evanescent fields in two-dimensional nanomaterials that are increasingly studied by users and staff. The microscope is still available to users for diffraction-limited optical microscopies. We are also pushing forward on this tool to offer transient absorption microscopy (TAM), which complements our already successful transient absorption efforts to include high spatial resolution and time-resolved imaging opportunities. Each year we will assess our research areas and continue to refine our strategy and make the necessary adjustments.

Summary

In summary, the CNM will focus its efforts on the big challenges in nanoscience that require large and diverse teams, keeping our research at the forefront of nanoscience and our tools state-of-the-art. This will ensure CNM's continued leadership in nanoscience and attract best-in-class users.