Five-year Strategic Plan

Brookhaven
National Laboratory

Center for Functional
Nanomaterials

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EXECUTIVE SUMMARY

The Center for Functional Nanomaterials (CFN) is a Nanoscale Science Research Center operated for the U.S. Department of Energy (DOE) at Brookhaven National Laboratory (BNL). As a national scientific user facility, the CFN provides users a research experience supported by top-caliber scientists and with access to state-of-the-art instrumentation. The CFN mission is advancing nanoscience, by being an essential resource for the worldwide scientific community and by carrying out transformative nanoscience research to support the energy, economic, and national security of the United States. Strategic partnerships are crucial to CFN mission success, including the strong synergy with the National Synchrotron Light Source II (NSLS-II), also located at BNL.

The CFN Five-year Strategic Plan provides a vision with flexibility and nimbleness, to capitalize on new opportunities and most effectively serve a nanoscience community with evolving interests and needs. The foundation of the Plan is composed of three nanoscience Themes, which define a unique CFN identity reflecting the technical expertise of the staff and guiding development of new, state-of-the-art facilities.

The subject of Theme One is *Nanomaterial Synthesis by Assembly,* which has an ultimate goal of realizing a design strategy for synthesis of new materials with targeted functionality by assembly of nanoscale components into precise architectures. CFN research on self-assembly focuses on devising new strategies for interaction- and process-controlled assembly of components, discovery of the governing principles underlying self-assembly, and understanding assembly pathways using advanced *ex-situ* and *in-situ* characterization and computational methods. The recognized expertise of CFN scientists in self-organization of nanomaterials with soft matter molecules (*e.g.*, DNA and polymers) is crucial for developing novel approaches for self-assembling arrays and clusters based on molecular recognition, and for devising new ways of probing assembly phenomena *in-situ*, in real-time, and at multiple spatial and temporal scales.

Efforts are focused on developing nanomaterial synthesis-by-assembly methods and realizing functional material designs from polymer, nanoparticle, biomolecule-based, and 2D material components. Automation of synthesis-by-assembly processes will provide parallelism and reproducibility, facilitate assembly of increasingly complex architectures, provide control of assembly pathways, and allow incorporation of real-time feedback during processing. Advanced characterization and new methods to probe structure include nanoscale coherent X-ray beams at NSLS-II and 3D imaging of nanostructures by cryo- and in-liquid electron microscopy. Theory and simulation complement the experimental effort, including developing effective self-assembly strategies, assessing the inherent stability of resulting morphologies, and mapping the advantages and limitations imposed by kinetics.

The focus of Theme Two is *Accelerated Nanomaterial Discovery*. While historically the discovery and development of new materials has followed an iterative process of synthesis, measurement, and modeling, suitable integration of advanced characterization, robotics, and machine-learning can potentially radically accelerate this process. The CFN has an established record of discovering nanomaterials by applying new materials synthesis strategies, advanced characterization, and machine-learning. Integrating these efforts will enable autonomous platforms for iteratively exploring material parameter spaces, which have potential to revolutionize materials science by uncovering fundamental links between synthetic pathways, material structure, and functional properties.

CFN scientists are conducting research and developing instruments toward accelerating the material discovery loop. Realizing this vision requires advancing and automating all aspects of the discovery process, including: implementing combinatorial libraries and real-time synthesis platforms; improving multi-modal characterization and analysis of complex datasets; and using machine-learning to drive experiments.

Theme Three emphasizes the study of *Nanomaterials in Operando Conditions*. Interrogating materials at the nanoscale to derive atomic-level information on physicochemical processes under operating conditions remains a forefront and evolving nanoscience research field. The CFN continues to augment its comprehensive suite of instruments for *operando* studies of nanomaterials such as catalysts, photocatalysts, and battery electrodes. The CFN will increasingly integrate its *operando* capabilities together, with complementary facilities being developed at NSLS-II, and with data management and computational resources for advanced data analytics at the BNL Scientific Data and Computation Center, to strengthen BNL leadership in multimodal *operando* nanomaterial studies.

CFN users, working independently or collaborating with CFN staff, use combinations of *in-situ* and *operando* capabilities at high temperatures and variable pressures to understand catalytic reaction mechanisms. Aberration-corrected transmission electron microscopy with high spatial and energy resolution illuminates reaction pathways and structural changes in energy storage systems. Scanned-probe microscopy, infrared reflection absorption spectroscopy, and X-ray photoemission spectroscopy provide details on the elementary reaction steps through coordinated studies of model catalyst systems. Computational methods link atomistic structures to specific spectroscopic signatures and with catalytic functionality.

The CFN Strategic Plan is grounded in foundational Pillars of an expert staff, an engaged user community, and a portfolio of strategic research partners — all working safely and supported by excellent operations and a portfolio of state-of-the-art nanoscience facilities. The CFN will strive for higher levels of user engagement and diversity, through strategic partnerships with larger initiatives with synergistic goals, technical workshops customized to communities with specialized needs, and by more visibly promoting user science accomplishments. During the next five years, the CFN will invest in new instrumentation, make major upgrades to distinctive capabilities, and develop new data-analytics and data-management methods to maintain its status as a cutting-edge user facility.

A high priority is continuing to enhance the partnership between CFN and NSLS-II, through: investing further in four partner X-ray nanoscience instruments; working together to identify and capitalize on opportunities to create unique, new capabilities; and advancing joint projects with NSLS-II staff and users that exploit the complementary properties of X-rays and electrons to collect multimodal information on the same samples.

1. MISSION AND VISION

The Center for Functional Nanomaterials (CFN) is a state-of-the-art nanoscience facility with the dual mission of enabling the research of external users and carrying out transformative basic research to discover, understand, and implement nanomaterials. The combination of scientific staff expertise, portfolio of distinctive nanoscience capabilities, engaged community of users, and strong partnership with the National Synchrotron Light Source (NSLS-II) at BNL, make the CFN unique among nanoscience centers worldwide.

The rapidly changing scientific and technological landscape makes the CFN an essential element of the U.S. scientific research infrastructure. The tightly integrated state-of-the-art nanoscience facilities and the deep scientific expertise of the staff allow CFN users to pursue complex projects involving nanomaterial synthesis/fabrication, advanced characterization, and understanding of nanomaterials — all under one roof.

Since beginning operations in 2008, the CFN has become a vibrant, open hub for nanoscience research, where engaged users and expert staff use the most advanced tools for breakthrough discoveries of new materials and phenomena that manifest at the nanoscale. Deep research partnerships are an important element of CFN operations: e.g., by maximizing the value of co-location with NSLS-II and the complementarity of the two user facilities.

2. INTRODUCTION

In more than thirteen years of nanoscience operations, the CFN has fostered sustained growth of a large, productive community of users who benefit from both the state-of-the-art nanoscience facilities and the scientific expertise of CFN staff. The CFN supported 571 users in 2021, with users continuing to express high satisfaction with their CFN experience. The 2021 survey of user satisfaction indicated that 88% of respondents were either highly satisfied (73%) or satisfied (15%) with the service provided by CFN staff. The number of CFN publications was 376 in 2020, with 50% appearing in journals with impact factor >7, and 41% of them being the result of collaborations between users and staff.

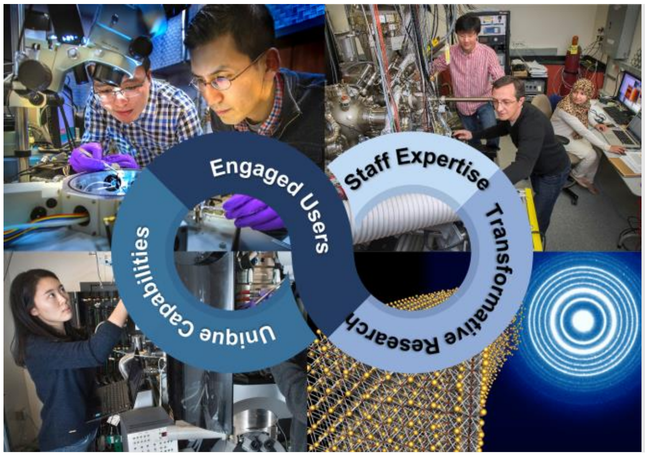

Figure 1. CFN users and expert staff work in collaboration or independently, taking advantage of unique capabilities to address complex scientific questions and technological challenges in nanoscience. The staff is recognized for its international leadership in DNA-mediated and block copolymer nanomaterial self-assembly, X-ray scattering, and catalysis, among other areas. Advanced CFN capabilities in *operando* probes, aberration-corrected electron microscopes, and high-resolution lithography stand out.

While the CFN user community is both geographically and topically diverse, many user projects cluster around themes addressing key scientific questions and technological challenges. These themes correlate with the distinctive facilities and expertise of CFN scientists. As a result, the CFN has fostered a strong community of user and staff researchers working in pursuit of new understanding in catalysis, energy conversion and storage, and nanomaterial self-assembly, among other areas.

The CFN operates in a rapidly changing scientific world. For example, recent, intense worldwide interest in quantum information science has increased demand for new synthesis facilities for quantum materials, and advanced characterization capabilities for understanding quantum coherent phenomena. New classes of materials have emerged for this and other applications, such as the expanding palette of ultrathin two-dimensional materials derived from layered van der Waals solids. Heterostructures created from 2D building blocks can possess new types of electronic structure that hold promise for quantum information science. The demand to characterize materials in *operando* has grown, for example in efforts to bridge the gap between catalysts operating under industrial conditions and model catalysts probed under idealized conditions. Experiments and instruments are becoming increasingly complex, especially when measurements under variable conditions (*e.g.*, pressure and temperature) are added to those of static parameters. New ways of collecting information about the effects of those variables have led to an explosion of scientific data. CFN users utilize multiple facilities, including advanced characterization by electron and photon probes. The CFN conducts a focused program of internal research and continually develops new facilities and expertise to take advantage of opportunities. For example, the CFN continues to develop instrumentation in the strategic area of *operando* nanoscience probes. As a result, the CFN is extremely well positioned to realize its vision and accelerate the DOE basic science mission.

This Strategic Plan is the roadmap that guides CFN actions for the next five years. Progress on this Plan will be gauged periodically using criteria detailed in the Metrics section. Fully implementing the Plan requires prudently allocating Resources for upgrading facilities, installing new capabilities, and building a world-class staff equipped with the appropriate technical skillsets.

3. OBJECTIVES AND THEMES

The objectives of the Strategic Plan follow directly from the two-fold CFN mission:

- The CFN will be an essential resource for the scientific community, enabling user projects that address fundamental questions and outstanding technical challenges in nanoscience.
- The CFN staff will carry out a program of internal research that produces transformative nanoscience breakthroughs.

Mission success rests on the expertise of the CFN staff and the state-of-the-art nanoscience capabilities they develop and operate. Naturally, staff and facilities support a broader range of user nanoscience projects than the narrower scope of the internal research program. However, many projects carried out by CFN users can be grouped into categories that strongly overlap topics underpinning the in-house research program, and which the CFN facilities are well-suited to address. This alignment is very positive and provides an efficient approach to complex, multi-disciplinary science, and ultimately determines the CFN identity.

The scientific questions underlying many CFN projects fall under three themes:

- Nanomaterial synthesis by assembly;
- Accelerated nanomaterial discovery; and
- Nanomaterials in *operando* conditions.

These themes utilize the leading expertise of CFN staff in nanomaterial synthesis, block copolymer self-assembly, and DNA-mediated nanostructures. They uniquely leverage the CFN capabilities for *in-situ* imaging and spectroscopy, and X-ray based techniques both at the CFN and at NSLS-II. The successful development of these three Themes rests on five sustaining Pillars, which are:

- Conducting research with Safety and Operational Excellence;
- Hiring and mentoring an Expert and Diverse Staff;
- Fostering and supporting an Engaged and Diverse User Community;
- Providing State-of-the-art Facilities for nanoscience; and
- Engaging in Partnerships for broadest impact.

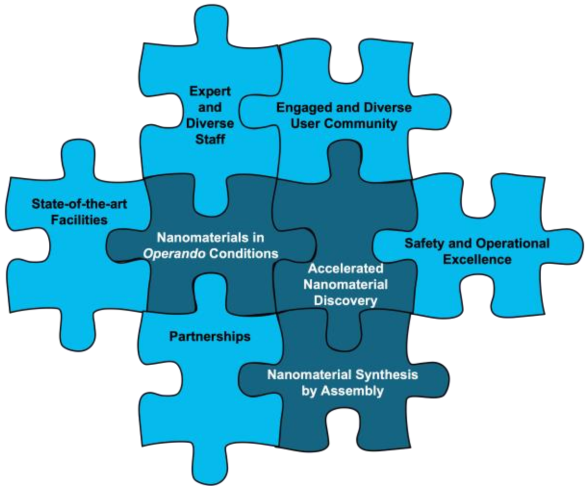

Figure 2. The CFN Strategic Plan provides a roadmap for making the CFN an essential resource for the nanoscience community and producing scientific breakthroughs in nanomaterials. Three overarching scientific Themes are supported by five Pillars.

3.1 Theme 1: Nanomaterial Synthesis by Assembly

The CFN is pursuing fundamental research into nanomaterial synthesis by assembly. This theme encompasses self-assembly, traditional and emerging thin-film and bulk processing methods such as nanofabrication and 3D printing, nanoscale templating and combinations of ‘top-down’ and ‘bottom-up’ methods. Advances in chemical and biological synthesis have generated a wide palette of structurally diverse and property-specific nanoscale components, including: complex polymers, supramolecular complexes, designed proteins and DNA constructs, and sculptured nanoparticles. This material toolkit provides a rich opportunity to establish a new nanomaterial synthesis paradigm based on *by-design* assembly of components into targeted functional architectures with prescribed spatial organization and composition. The longstanding CFN research program in self-assembly develops approaches based on assembly of for fabrication of prescribed materials from these components, illuminates the governing principles and establishes practical methods. A long-term goal is developing an *inverse design* strategy, wherein assembly procedures are defined for achieving specific structures with targeted functions.

CFN staff are pursuing research in a broad range of self-assembling nanomaterials, including: nanoparticles, DNA, biomolecular constructs, polymers, zeolites, and 2D materials. Staff and users have used these components and systems for generating novel nanomaterials and devices. For example, DNA assembly methods have been applied to create novel optically and chemically active materials. Similarly, block copolymers have been used to create antireflective optical coatings, and water-repellent surfaces.

The CFN will advance the science of synthesis by assembly by integrating self-assembly methods and nanomaterial synthesis with robotic platforms and autonomous experimentation. Automation of the synthesis-by-assembly process will provide parallelism and reproducibility, facilitate assembly of increasingly complex architectures, enable control of assembly pathways, and allow incorporation of real-time feedback during processing. Using AI and machine learning methods for multimodal characterization will facilitate insights about assembly processes and efficient principles for system design.

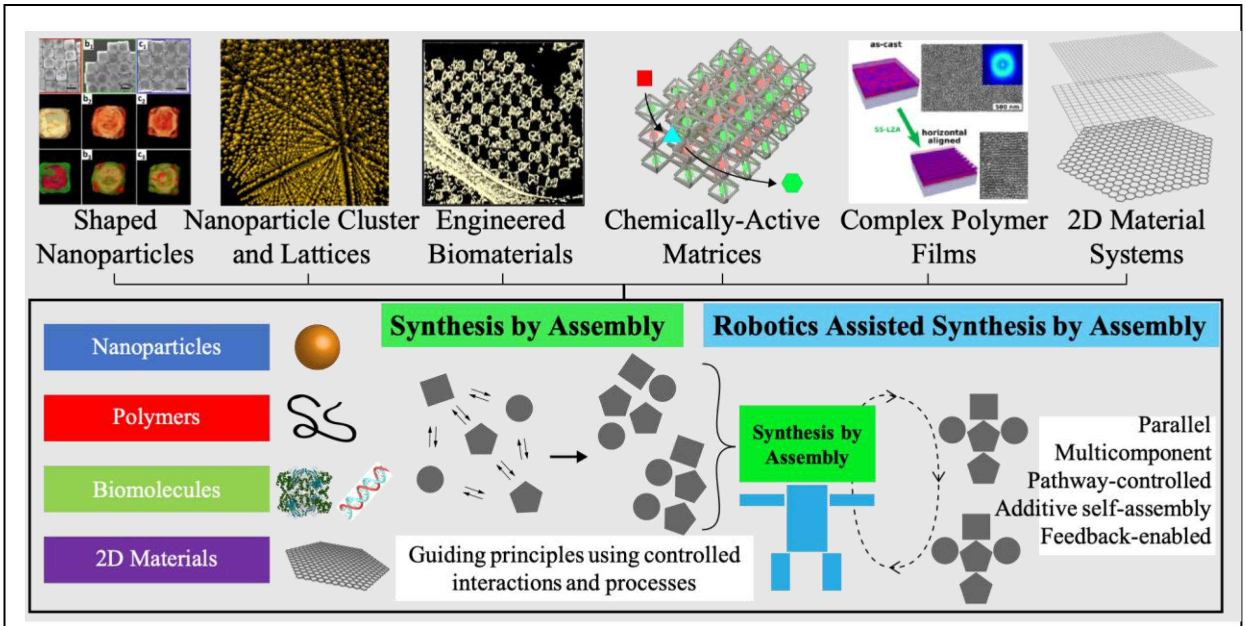

Figure 3. Nanomaterial synthesis-by-assembly is realized through discovery of guiding principles of self-assembly by design, and development of effective structure-formation processes. Robotic-assisted synthesis-by-assembly platforms will address challenging aspects of structure formation for creating a broad range of nanomaterials with targeted architectures and functions.

This effort relies on cutting-edge characterization methods such as X-ray scattering, electron microscopy, single-molecule optical detection, spectroscopy, and photon- and electron-based tomography — and will ultimately integrate *in-situ* characterization into the synthesis process for real-time structure/property monitoring and feedback.

In the next five years, scientific efforts will be focused on development of nanomaterial synthesis-by-assembly to achieve architectures with multiple levels of structural and compositional complexity, designed and built to enable target functionality. Major directions include:

- Nanoparticle periodic arrays with predefined organization and composition for controlling collective properties, including optical, magnetic, and catalytic properties. Nanoparticles with directional bonds will enable formation of lattices with prescribed symmetry and multiscale internal organizations through synergetic connections of experimental, theoretical, and computational efforts.
- Incorporating functional biomolecular complexes (enzymes, proteins) into nanostructured materials and gaining control over building designed biomaterial systems. Nano-biomaterials can enhance biochemical reactions, promote interactions of biomolecules with inorganic, organic and biological matter, and provide new sensing modalities.
- Developing versatile modeling and design tools for realizing rational nanomaterial assembly. Numerical simulations will be used for optimizing kinetic pathways of self-assembly, as well as the structural properties, response to stimulus, and other functions.
- Using liquid handling robotics for automated fabrication by assembly. The robotic system will integrate DNA-based assembly platforms with component libraries of nanoparticles, proteins, and other biomolecules, with rapid on-deck characterization, for programmable design and fabrication of nanomaterials.

- Pathway assembly control of self-assembled nanostructures on different length-scales. By exploiting non-equilibrium behavior, it will be possible to manipulate different length scales during self-assembly. For example, photo-thermal stimulation will be used to pattern block copolymer films at the microscale, with molecular-scale self-organization.
- Controlled fabrication of sculpted nanomaterials with atomically defined planes. CFN will continue developing methods for controlling nanoparticle growth mechanisms and will use high-throughput synthesis and multi-modal, in-situ characterization using optical and X-ray scattering and spectroscopy.

To advance these scientific directions, major methods and capabilities will be developed:

- In-situ and time-correlated X-ray scattering analysis methods for quantitative descriptions of ordered, weakly ordered, and cluster nanoscale organizations and their dynamic behavior at size scales ranging from one nanometer to hundreds of nanometers, and timescales as short as milliseconds.
- X-ray and electron tomography methods for revealing structural and chemical 3D local organization of nanoscale assemblies using photon- and electron-based approaches with sub-nm spatial resolutions of micron-scale structures.
- Micro-beam synchrotron light scattering for multiscale and phase mapping of nanostructured multi-component materials on a range of scales, from molecules to macroscopic dimensions.
- In-situ nano-mechanical characterization of hard, soft, and hybrid materials, for both bulk and surfaces, combined with high-resolution imaging by electron microcopy.
- Single-molecule optical methods to probe local optical fields, energy transfer and transduction processes, polarization signatures and material heterogeneity.

3.2 Theme 2: Accelerated Nanomaterial Discovery

Modern materials are increasingly complex. Formed from a wide range of components, they exhibit structural order at multiple length scales (atomic, molecular, nano, meso, micro, macro), are synthesized using elaborate processing pathways, and are frequently non-equilibrium. The functional demands on new materials are increasing, as they are designed for performance improvements in next-generation applications (e.g., energy materials, quantum information science, enhanced optical or mechanical properties). Historically, new materials development has followed an iterative process of synthesis, measurement, and modeling. Tightening this discovery loop has potential for radically accelerating the design of new materials and revolutionizing materials science. Guided experimentation using data analytics and combinatorial sample libraries enables data-driven material discovery. Autonomous platforms leveraging robotics, advanced characterization, and machine-learning can iteratively explore material parameter spaces and uncover fundamental links between synthetic pathways, material structure, and functional properties. Ultimately, we envision real-time steering of material synthesis.

This theme builds upon core CFN strengths in synthesis, characterization, and theory/analytics, which have contributed to discoveries of materials with novel properties. For example, *synthesis* of hybrid polymer/inorganic materials *via* infiltration of organometallic precursors provides continuous tuning of composition, and thus control of functional properties, including chemical, mechanical, electrical, and quantum. *Characterization* via X-ray absorption mapping contributed to the discovery of a quantum material with the ability to manage and cloak thermal radiation. The CFN has demonstrated a *theory/analytics* approach to extracting local atomic motifs from X-ray spectra, by applying machine-learning to scientific datasets.

During the next five years, CFN scientists will conduct fundamental research to further accelerate the material discovery loop. Realizing this vision requires advancements and automation of all stages of material discovery: synthesis, especially by implementing combinatorial libraries and real-time synthesis platforms; characterization, including multi-modal *in-situ*/*operando* measurements; and understanding, through theory/analytics and use of machine-learning to drive experiments. CFN efforts will be guided by key questions that currently limit the implementations of automated discovery platforms:

- *How can functional nanomaterials by synthesized parametrically*? The CFN will pursue combinatorial methods for synthesis of inorganic, soft, and hybrid nanomaterials. For example, flow reactors can be used to generate a continuous stream of distinct synthesis environments, exploring the conditions for nanoparticle synthesis, functionalization, and assembly. The CFN will advance synthesis methodologies for desired functionalities, especially in hybrid, hierarchical, and quantum nanomaterials. A primary target will be platforms that provide *in-situ* property measurement during synthesis.
- *How can the material discovery loop be more efficient*? The CFN will continue to advance its expertise in advanced nanomaterial characterization, especially *in-situ* and *operando* probes, enabling real-time studies *during* synthesis and processing. These characterization methods will be integrated with physics-aware data analytics and decision-making algorithms to enable machine-guided experimentation. Modern human-computer interaction methods will be leveraged to provide experimenters with sophisticated controls.
- *How can theory/data analytics uncover fundamental materials physics*? The CFN will advance the modeling of materials by improving existing theories and coupling these to modern machine-learning methods. For example, physics-constrained neural network models are being developed that can extract meaningful structural descriptors from spectroscopy data.

These studies will provide improved materials knowledge and new, robust platforms for studies of

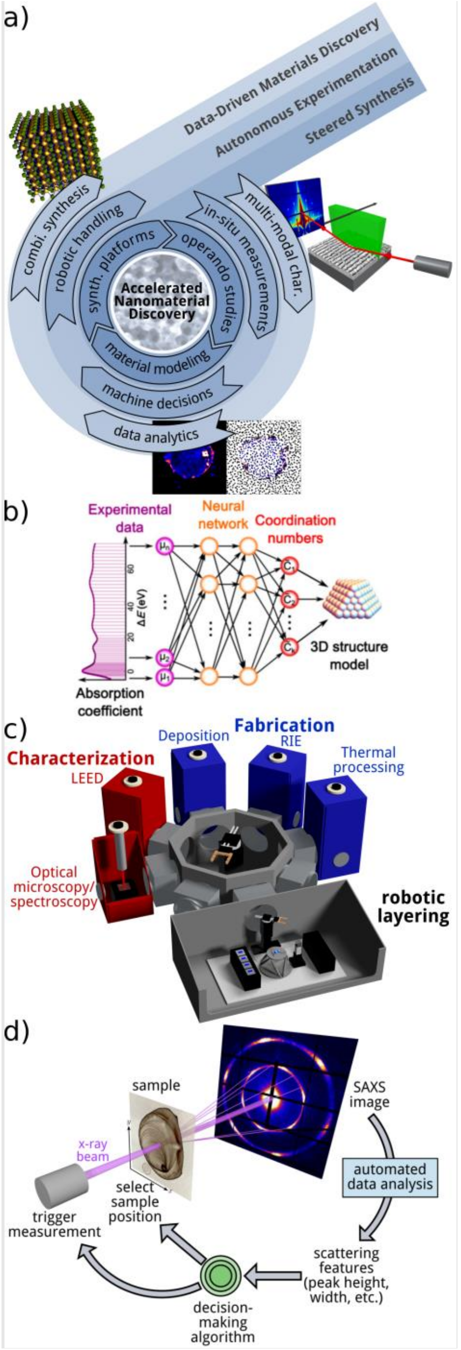

Figure 4. Accelerated Nanomaterial Discovery. (a) The discovery loop can be tightened by close coupling of automated synthesis, measurement, and analytics. (b) Machine-learning has been used to convert from X-ray spectra to local structural motifs. (c) The QPress is an automated platform for constructing 2D material heterostructures by assembly. (d) Autonomous experiments have been implemented at synchrotron X-ray scattering beamlines.

nanomaterials during synthesis, assembly, and processing. These platforms will benefit broad nanomaterials communities by empowering a host of searches for novel materials. Some ongoing and targeted developments are:

- Improved algorithms for control of experimental platforms. We are developing a portfolio of machine-guidance algorithms applicable to a wide range of problems, including general-purpose methods (using Gaussian processes) and material-specific approaches (using reinforcement learning).
- The Quantum Material Press (QPress) cluster tool is enabling automated exfoliation and characterization of 2D materials, and assembly into stacked heterostructures with tailored quantum properties. This automated platform will enable fabrication of quantum devices with previously impossible complexity.
- A suite of combinatorial methods for synthesis and processing of inorganic, soft, and hybrid materials. This includes flow-coating and electrospray for composition and thickness gradients, photo-thermal annealing for processing gradients, flow reactors for nanoparticle synthesis, lithographic sample arrays, and a new ultrasonic spray platform for combined deposition, solvent processing, and infiltration synthesis.
- Specialized X-ray spectroscopy studies, coupled to machine-learning models and databases of computed and measured spectra, for establishing spectral-structural assignments and uncovering local atomic motifs. For example, this enables understanding of how structural motifs at interfaces influence catalytic activity, and discerning whether catalytic operation changes material structure.

3.3 Theme 3: Nanomaterials in *Operando* Conditions

Characterizing nanomaterials during operation in their native environments is vitally important to modern materials science. External stimuli, including pressure, temperature, light, and voltage, lead to dynamic material changes, which may only be observable under functional conditions. Discovery of structure-function relationships hinges on *in-situ* material investigations. For example, synthesis of new catalysts hinges on identifying active phases and reaction mechanisms, *as they emerge at the elevated pressures and temperatures under which catalysts operate*. Environmental transmission electron microscopy (ETEM) and local area electron energy loss spectroscopy (EELS), scanning probe microscopy (SPM), low energy electron microscopy/X-ray photoemission microscopy (LEEM/XPEEM), vibrational (IR and Raman) spectroscopy, and ambient-pressure photoemission spectroscopy (AP-PES) comprise an experimental toolset capable of *operando* nanomaterial interrogation. Atomic scale theory links key structural motifs to complex spectra and observed characteristics.

The CFN has assembled a comprehensive suite of tools for understanding materials as diverse as catalysts, photocatalysts, and battery electrodes. *In-situ* and *operando* implementations of these core techniques are a frontier area of research, as is their integration for multimodal characterization. Time-resolved perturbations by modulation excitation spectroscopy (MES) can provide additional kinetic data. The CFN will incorporate MES into both X-ray and vibrational spectroscopy and electron microscopy, and advance data science methods for making use of the resulting complex spectrokinetic datasets. In the next five years, the CFN will further advance its capabilities for *in-situ* and *operando* measurements and integrate them with others being developed at NSLS-II. A universal sample holder and vacuum suitcase will facilitate sample transfers for multimodal material studies. The CFN will develop accompanying methods in data

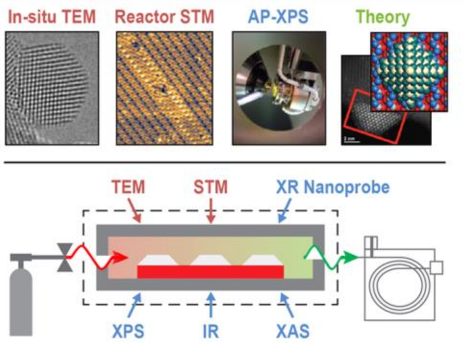

Figure 5. Nanomaterials in *Operando* Conditions. The CFN portfolio of *in-situ* and *operando* techniques using electrons, ultrafast photons, and X-ray radiation, coupled with theory, creates a unique setting for studying the link between structure and function of nanomaterials. A key element of this approach is correlating multimodal imaging and spectroscopy with real-time reactivity measurements. This can be done using portable high-pressure reaction cells in which the same materials are interrogated in *operando* by electron probes at the CFN and complementary photon probes at NSLS-II or using a vacuum suitcase for transferring samples among ambient pressure systems and reactors.

analytics to extract and interpret the rich information derived from multidimensional datasets. The goal is strengthening BNL's position as a world leader for multimodal *operando* studies of nanomaterials.

CFN users and staff are interested in catalysts for greenhouse gas-to-fuels and biomass conversion, with metals dispersed on nanoporous materials such as zeolites and metal organic frameworks or integrated inside them. Chemistry in nanoconfined spaces can affect the formation of active centers and reaction pathways and can change the required reaction conditions. Copper-based catalysts, for example, show promise for hydrogenation of CO_2 to methanol and direct conversion of methane into alcohols. Stabilization of active Cu ensembles in nanoporous materials with local effective high pressures can facilitate their detailed study under *operando* conditions.

In-situ characterization is important for understanding photocatalytic water splitting. Photocatalysis requires separation of photoexcited charge carriers on ultrafast timescales, to avoid recombination losses. Ultrafast optical spectroscopy is uniquely suited to quantifying recombination losses and understanding how they can be suppressed through nanoscale material and interface design. Catalytic steps occur on longer time scales, and changes in the local structure of active centers can be understood using spectrokinetic XAS, AP-PES, and IR measurements under *operando* conditions.

A third area where it is crucial to probe materials in complex environments is energy storage. Creating electrochemical batteries with both high storage capacity and high cyclability requires understanding electrochemical processes at electrode/electrolyte interfaces, including structural and phase changes, electron/ion insertion mechanisms, and the dynamical evolution of the interface structure. CFN users studying Li-ion (and other alkali-ion) batteries are interested in resolving the reaction pathways and unlocking the potential of novel materials such as disordered layered oxides as battery electrodes. The combination of synchrotron-based techniques with electron microscopy provides critical insights into these processes.

In the next five years, CFN scientists and users will exploit this wealth of capabilities and expertise to understand nanomaterials under *operando* conditions, working on:

- Elucidation of catalytic reactions using a multimodal approach combining Reactor STM, AP-PES, IR, Raman and ETEM, all operating at elevated local pressure and temperature;
- Optimization of separation of photogenerated charged carriers at nanostructured hybrid organometallic/oxide interfaces of photocatalysts using *in-situ* ultrafast optical spectroscopy

and light-modulation excitation spectroscopy and X-ray spectroscopy, including *operando* XAS at NSLS-II;

- Investigations of reaction pathways in the electrode structure of energy storage systems using TEM-based methods with high spatial and energy resolution.

A unique CFN strength derives from multimodal integration of complementary methods to probe the *operando* behavior of nanostructured functional materials, and will be further enhanced by major instrument and method development, namely:

- The CFN will design and commission a unique environmental, monochromated, aberration-corrected scanning transmission electron microscope (E-STEM) designed for high-resolution studies of structural and chemical evolution of nanomaterials in working environments;
- The CFN will extend its AP-PES capabilities by designing and commissioning a new instrument with a multicolor tender X-ray source. This new instrument, combined with new liquid cells, will enable studies of liquid/solid interfaces, including during nanomaterial synthesis and electrochemical processes;
- Developments at lab-based AP-PES and AFM/IR spectroscopy instruments will guide upgrades at existing and new capabilities at NSLS-II;
- A suite of portable gas/liquid reactor cells will make it possible to interrogate the same catalyst or battery material under identical environmental conditions by TEM, AP-PES, and other X-ray spectroscopies at NSLS-II beamlines. These cells will measure gas pressure, reactant/product composition, temperature, and pH. Holders will be standardized across the portfolio of surface science instruments for sample transfer in vacuum among CFN and NSLS-II facilities, glove boxes, and synthesis facilities;
- Computational methods, including improved first-principles approaches and machine learning, will link atomistic structures to spectroscopic signatures obtained from simulated materials or structures on databases, and to unravel the controlled growth of thin films and catalytic reaction pathways from data obtained under synthesis or reaction conditions.

4. PILLARS

Achieving the Objectives of this Strategic Plan requires an expert and diverse staff, an engaged and diverse user community, and a set of strategic research partnerships. World-leading research by staff and users must be conducted safely and supported by both excellent operations and state-of-the-art facilities — including those developed and offered in partnership with NSLS-II. Implementing this Plan requires strengthening these essential pillars, as detailed here.

4.1 Safety and Operational Excellence

Excellence in operations and a strong safety culture are central to CFN success. The CFN emphasizes the importance of communication among the staff and users with diverse backgrounds and experiences as the best approach to achieving safety compliance. A robust operational and administrative infrastructure supports the research experience from concept to project completion. Safety is fully integrated into all aspects of the work. Regularly reviewed and updated course modules and on-the-job training play an essential role in safe and productive research. Prior to independent use of CFN facilities, every new user is trained in all general and work specific CFN operations and safety procedures. New users are paired with an expert mentor for guidance on operations, hazard identification, and response. Specific focus areas for operations and safety include:

- More tightly integrating safety standards and guidelines, operational procedures, and administrative support for planning and executing research;
- Implementing a project planning process to instrument acquisition, installation, commissioning, and operations that reduces and controls hazards;
- Implementing a team-oriented work planning and control process to enhance efficiency and safety in service and maintenance of CFN instruments;
- Developing a facility master space plan to guide new instrument installations and workspace allocation;
- Improving the user proposal submission, review, and allocation process for better usability and coordination with other BNL facilities;
- Integrating web-based modules designed for efficient user and staff training, into a robust system to ensure that requirements are met prior to granting access to CFN facilities;
- Improving records management and data storage/access for operations and scientific activities;
- Implementing a "cradle to grave" materials safety concept, placing equally-high emphasis on nanomaterials and chemical safety from project inception, through project execution, to final material disposal.

4.2 Expert and Diverse Staff

Recruiting, developing, and retaining a diverse group of the highest-quality scientific, technical, and administrative staff is central to fulfilling the CFN mission. The CFN strives to create and maintain an environment where all staff feel respected as team members whose professional skills are essential to advancing the CFN mission. Key components of the CFN strategy include:

- Recruiting and retaining a diverse workforce of the most talented professionals at all levels;
- Creating a positive, inclusive work environment by promoting collaboration and fairness;
- Providing thoughtful mentoring and professional growth opportunities for all staff members at all career stages;
- Mentoring scientific staff members toward an appropriately balanced effort between support for CFN users and pursuing their internal research;
- Seeking internal and external recognitions for staff professional achievements;
- Maintaining vibrant postdoctoral researcher and graduate student programs (in coordination with the BNL Association of Students and Postdocs), which increase the scope of the CFN research program and provide opportunities for staff to develop supervisory and mentoring skills;
- Fostering an environment in which scientific staff augment their research by capitalizing on external opportunities, while maintaining their commitment to the user program. Examples include: BNL Laboratory Directed Research and Development program, DOE Early Career Awards, DOE Energy Frontier Research Centers, Small Business Innovation Research Programs, and partnering with users in other DOE initiatives.

4.3 Engaged and Diverse User Community

To fulfill its mission of serving a satisfied and productive user community, the CFN continuously engages past and current users and actively seeks communities of new users. Users are integral to the CFN culture and planning, well beyond facility usage and staff collaborations. The CFN strives to identify and eliminate barriers hindering user research, to more effectively support the broadest community of users, through activities that include:

- Fostering an Engaged Users' Executive Committee (UEC). The CFN UEC provides an organized framework for communicating user needs to CFN and BNL management. The UEC promotes and encourages effective use of the CFN by providing forums for discussions among users. Together with the NSLS-II UEC, the UEC organizes the annual Users' Meeting, with assistance from the CFN. The CFN regularly engages the UEC for input on how to best support a diverse user community.
- Enhancing the User Experience. The CFN continues efforts to optimize the entire user experience, from proposal submission and review/allocation, instrument scheduling, data collection, and dissemination of research findings. Current focus areas include improving responsiveness to user inquiries and providing coordinated access to all BNL user facilities.
- Advancing Remote@CFN. The CFN is rapidly expanding its ability to support users who are not physically present at the facility. This program builds on existing support for remote user access to computing resources. Remote@CFN has facilitated remote engagement of users with CFN staff, remote operation of instruments during experiments, a mail-in process for user sample analysis, and off-site access to experimental data and CFN-supported analytics software.
- Promoting User Science. The CFN promotes user science through BNL newsroom stories, social media, the *iCFN* newsletter, research highlights, and at scientific meetings and conferences.
- Engaging in Strategic Partnerships. CFN staff will continue to engage with user teams when there is a clear alignment of scientific interests, *e.g.*, Energy Research Frontier Centers and Small Business Innovation Research (SBIR) projects.
- Reaching New and Diverse Users. The CFN is resuming efforts to expand the diversity of users by deploying staff as ambassadors at scientific meetings and conferences. An element of this strategy is understanding and reducing barriers for underrepresented groups to access CFN facilities.
- Providing Technical Workshops. Instrument/technique training and user development workshops are excellent ways of linking CFN staff and experienced users with targeted potential users over topics of shared interest. These events and complementary tutorials serve as an effective outreach instrument to expand the user community and strengthen engagement.

4.4 State-of-the-art Facilities

The CFN facility is envisioned with the entire process of materials research in mind (synthesis and fabrication, advanced characterization, and understanding), such that users access an integrated set of tools for a complete research experience under one roof. The CFN operates advanced instrumentation in nanolithography, materials preparation, electron and photon probes, including those located at NSLS-II, and computational resources with diverse software tools for nanoscience theory, simulation, and data analytics.

The CFN portfolio of instrumentation is strategically refreshed to provide cutting-edge facilities, internationally attractive to high-impact users. The CFN will continue its sustained initiative to develop and operate new, unique nanoscience tools in partnership with NSLS-II. We will upgrade existing major CFN capabilities and acquire key new instruments. The Plan envisions investments driven by the needs and trends in materials research demand. For example, in the next five years:

- The CFN will extend its lab-based AP-PES capabilities by designing and commissioning a new instrument with a multicolor tender X-ray source for studies of liquid/solid interfaces.
- The CFN will upgrade the aberration-corrected low-energy electron microscope / photoemission electron microscope (AC-LEEM/XPEEM) operated at the Electron Spectro-Microscopy (ESM) beamline at NSLS-II for cryogenic operation and with a new analyzer to deliver world-record XPEEM energy resolution.
- The CFN will continue to invest in the Complex Materials Scattering (CMS) and Soft Matter Interfaces (SMI) beamlines, partner X-ray scattering endstations at NSLS-II. Modes of high-throughput and autonomous experimentation will be further developed through advanced software tools and novel real-time materials processing platforms, including photothermal thin film processing and flow reactors for solution-phase synthesis and assembly.
- Reactor cells, universal sample holders and vacuum suitcase transfer are enabling an increasing scope of multi-modal experimentation, including probes by X-ray and vibrational spectroscopy, and transmission electron microscopy. New machine-learning-based data analysis approaches are being developed to link spectra and images to structural motifs.
- In collaboration with Lawrence Berkeley National Laboratory and the BNL Computational Sciences Initiative, the CFN will install a unique, ultrafast direct electron detector on the Titan aberration corrected ETEM. This will be a first-of-its-kind capability for atomic-scale, *operando* studies of material dynamics on microsecond timescales.
- The CFN will install a state-of-art electron microscope for high-resolution studies of electron-beam sensitive materials with *in-situ* capabilities. An ultra-bright, cold field-emission gun with aberration correction and monochromator will provide atomic resolution imaging at low voltage and low electron dose, as well as high energy-resolution of EELS at high speed.
- The CFN will install a new aberration-corrected environmental scanning transmission electron microscope (E-STEM). It will be a double-corrected monochromatic, ultra-high-vacuum (UHV) microscope with atomic spatial resolution in both TEM and STEM and state-of-the-art ~5 meV energy resolution electron energy loss spectroscopy while allowing gaseous environment at the sample for next-generation in-situ/operando experiments.
- The CFN is commissioning a revolutionary cluster tool (QPress) that automates handling of 2D materials, as well as the synthesis of layered heterostructures from such materials. The system features robotic exfoliation from van der Waals crystals, multi-modal characterization, machine-guided materials layering, and automated machine-learning analytics. Ongoing upgrades include advanced thermal control, and integration with other facilities via vacuum suitcase.
- The CFN is constructing an Integrated Multimodal Characterization and Processing platform for Quantum Materials (QM-IMCP), a unique facility for precise assembly, processing, and multimodal characterization of heterostructure quantum materials. The QM-IMCP platform will provide an integrated set of complementary cryo-probes, and support transfer of materials and analysis of the same feature or micrometer-size structure across the entire suite. These capabilities will be integrated with the QPress, allowing researchers to combine automated synthesis and multimodal characterization into a single workflow.
- CFN will further develop new capabilities to achieve spectrally, spatially, temporally, and polarization-resolved optical signatures on the single-molecule level. We will expand these

characterizations to cryogenic temperatures and field-controlled environments to investigate structure, charge, energy transfer, and quantum effects in nanomaterials.

- CFN will develop a novel scanning probe microscope capable of performing multimodal ultrahigh-sensitivity measurements at millikelvin temperatures that correlate spatial changes in magnetic, electronic, and dielectric properties of materials and devices with their corresponding functional properties.
- The CFN is establishing a liquid-handling robotic platform for automated synthesis-by-assembly of multicomponent systems including nanoparticle-based materials, DNA structures, and designed biomaterials. The system will incorporate in-line characterization and AI/ML guided feedback control. Instrumentation, automated liquid handling, and transfer of labware will enable synthesis, purification, and characterization of materials systems.
- Nanoscale indentation for direct measurements of mechanical properties of nanomaterials with simultaneous visualization using scanning electron microscopy will probe a broad range of materials including, hard, soft and hybrid systems for samples as small as one micron.
- Innovative software and data management, including the use of machine-learning tools, as well as development and applications of physical theory, will be supported by regular CFN investments in new high-performance and high-throughput computing capabilities, data storage, and communications in cooperation with the BNL Scientific Data and Computational Center and a range of access options to meet user needs.

4.5 Partnerships

Leveraging a culture of collaboration and innovation, the CFN will develop and strengthen strategic partnerships to maximize the impact of CFN research expertise and capabilities.

The CFN will continue to deepen the relationship with NSLS-II through investments in X-ray nanoscience instrumentation, joint projects, development of staff expertise, and streamlined access mechanisms. This relationship between facilities continues to benefit users and staff. Some examples include:

- The CFN has participated in developing new X-ray beamlines for nanoscience and maintains Partner User Agreements with NSLS-II to support users at four endstations. These joint ventures have established leading capabilities in X-ray scattering, photoelectron spectromicroscopy, and *operando* spectroscopy. CFN is contributing essential equipment and staff, helping nanoscience users to access unique experimental capabilities while exploring new partnerships in areas of mutual scientific and technical interest.
- CFN capabilities and expertise are applied to developing new capabilities at NSLS-II. For example, CFN nanofabrication is used to create high-performance X-ray optics, beam sensors, and reference samples for method development at beamlines.
- The CFN will continue to establish joint projects with NSLS-II staff and users. For example, CFN will exploit the complementary properties of X-rays and electrons to image the same catalyst under realistic operating conditions, to interrogate working photo-electrochemical systems, and for imaging and spectroscopy of soft and hybrid hierarchical systems.
- The CFN and NSLS-II are collaborating to develop and implement data analysis and machine learning software for large data sets collected in synchrotron-based and electron-microscopy experiments. This work benefits from ongoing BNL investments in the Computational Sciences Initiative.

The CFN partnership with NSLS-II

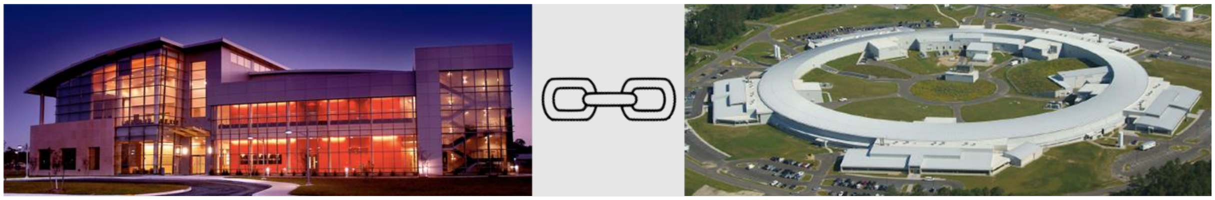

Figure 6. A strong partnership with NSLS-II is a key element of the Strategic Plan. Three illustrative areas are:

Theme 1: Nanomaterial Synthesis by Assembly

The CFN is a world-leader in DNA programmable self-assembly of heterogeneous nanomaterial lattices. CFN is developing methods using the advanced beamlines at NSLS-II to study how self-assembly can be used to organize targeted nanocomponents, especially optically and chemically-active species, into well-defined functional lattices.

Theme 2: Accelerated Nanomaterial Discovery

CFN is developing autonomous experimentation methods. Machine-learning methods have been created to automatically analyze X-ray scattering and spectroscopy data. Algorithms for autonomous experiment control have been developed in collaboration with the DOE CAMERA project. In partnership with NSLS-II, these concepts have been implemented at multiple beamlines for autonomous exploration of materials parameter spaces.

Theme 3: Nanomaterials in *Operando* Conditions

CFN and NSLS-II staff have built an ambient pressure photoelectron spectroscopy (AP-PES) endstation, operate a low energy electron/X-ray photo-electron emission microscope (LEEM/XPEEM) endstation, and have implemented cells and data analytics for hard X-ray absorption spectroscopy (XAS) at NSLS-II. Combining information from operando experiments at NSLS-II endstations and the CFN environmental transmission electron microscope (E-TEM), critical information from dynamic processes in catalysts can be interrogated.

The CFN will continue to establish partnerships with universities and industry in areas of strategic research overlap, with emphasis on proposal submissions to leverage DOE resources and deepen relationships. Examples include strong engagements with the DOE SBIR program, Energy Frontier Research Centers in catalysis and energy storage, the DOE Technologist in Residence Program, and emerging partnership opportunities in quantum information science. Intellectual property and technology transfer offer other ways to increase CFN impact and connect with industry researchers. Finally, partner users can provide investments of expertise and equipment that help CFN grow in new directions.

5. METRICS & RESOURCES

5.1 Metrics

The ultimate measure of success for this Strategic Plan is the extent to which the CFN supports a community of users and staff renowned for nanoscience breakthroughs and societal impact. Each year, the CFN reports its progress to DOE and receives feedback through the DOE Performance Evaluation and Measurement Plan (PEMP). The DOE reviews all aspects of CFN operations, user program, and internal research every three years. External reviewers assess the impact of internal research and user science, user satisfaction, and efficiency of facility operations. The CFN management team, supported by the external CFN Science Advisory Committee (SAC), regularly assesses progress in executing this Strategic Plan.

Internally, we will measure progress in mission-critical areas using trackable metrics:

- Operating a world-class nanoscience user facility
  - Have the scientific facilities operated at peak performance and been available to staff and users?
  - Have the scientific facilities been used by users and staff, and has their use contributed to scientific publications?
  - Have the scientific instruments and the entire CFN facility been maintained on a regular schedule, to keep them in optimal condition?
  - Has the CFN continued to evaluate and improve internal processes more efficient operations and better tracking of performance?
- Developing unique capabilities for the user community
  - Have the new capabilities been fully available to CFN users, and have users taken advantage of those capabilities?
  - Have those new capabilities led to impactful investigations not previously possible?
  - How satisfied are the users with the effectiveness of the facilities and with the support provided by CFN staff?
- Fostering the success of world-class scientific staff
  - Has the CFN internal research effort resulted in scientific breakthroughs, published in top journals and widely cited?
  - Has execution of the research plan generated new intellectual property?
  - Are CFN staff members in leadership positions within their respective fields, with recognition for their accomplishments by the external technical community?
- Being an essential resource for collaborative research
  - Is the CFN engaged in multidisciplinary research partnerships involving academia, other national laboratories, and industry?
  - Is the CFN considered an essential resource in large-scale scientific efforts in catalysis, energy storage, or quantum information science?
  - Are CFN scientists key members of collaborative teams in their areas of expertise?

5.2 Resources

Guided by user feedback, input from the UEC, and advice from the SAC, the CFN allocates resources (equipment, staff, and operating funds) to support each facility and ensure that high-impact research is carried out in each thematic area. CFN operations are primarily funded by a block grant from the DOE Office of Science. From this operating budget, the CFN invests a minimum of 10 percent each year in new scientific equipment, with additional funds used as contingency for facility upgrades. Projecting realistic future budget increases in line with past history, the CFN anticipates being able to successfully carry out this Strategic Plan over the five-year period described here.

To fully exploit the capabilities that will be developed in the scope of this Strategic Plan, the CFN will continue to add scientific staff members in strategic areas. Additional operations funding, beyond budget projections, will further enhance unique CFN capabilities.

If resources are more limited in the future, the CFN will adjust the scope of this Plan accordingly and adopt a conservative approach toward hiring additional staff. In such a scenario, the CFN will establish priorities based on progress among the scientific themes, growth of high-impact facility usage, and input from the SAC and the user community, to ensure that the CFN fulfills its core mission and continues to thrive.