Exploring New Science Frontiers at NSLS-II

NSLS-II Strategic Planning Workshop

Stony Brook University and Brookhaven National Laboratory

October 21-23, 2019

Workshop Organizing Committee

Mercy Baez
Eric Dooryhee
Ignace Jarrige (Co-Chair)
Ricarda Laasch
Sean McSweeney
Ron Pindak
Qun Shen (Chair)
Jean Smiles
Eli Stavitski
Ryan Tappero
Juergen Thieme (Co-Chair)

Science Session Co-Leads

Quantum Materials:	Andrew Wray (NYU)
	Ignace Jarrige (BNL)
Soft Matter:	Dean DeLongchamp (NIST)
	Ron Pindak (BNL)
Materials Science & Eng.:	Paul Evans (Wisconsin)
	Eric Dooryhee (BNL)
Catalysis & Chemical Sci.:	Ayman Karim (Virginia Tech)
	Eli Stavitski (BNL)
Molecular & Cell Biology:	Sandra Gabelli (Johns Hopkins)
	Sean McSweeney (BNL)
Biosystems, Earth, Environment:	Tony Lanzirotti (Chicago)
	Ryan Tappero (BNL)

Special Thanks to:

NSLS-II User Executive Committee
Brookhaven Science Associates
Stony Brook Foundation

U.S. DEPARTMENT OF ENERGY

BROOKHAVEN NATIONAL LABORATORY

Stony Brook University

Table of Contents

Introduction

Entering the sixth year of its operations, NSLS-II has aggressively ramped up its scientific facilities and associated user programs, with 28 operating beamlines at the time of this report and 1 additional beamline under construction. More than 1700 distinct researchers have used NSLS-II for their research in 2019, and publications in 2019 have reached the level of one paper per day.

Given the current state of the NSLS-II facility, which is roughly half-built with its full capacity of ~60 beamlines, as well as the evolving science and technology landscape in the Nation and around the world, NSLS-II held a 3-day Strategic Planning Workshop, "*Exploring New Science Frontiers at NSLS-II*", October 21-23, 2019, at Stony Brook University and Brookhaven National Laboratory. The goal of the Workshop was to get the scientific community together to discuss the grand challenge questions and research needs in a number of scientific fields, and to identify new research capabilities that would be required and how NSLS-II may take part to advance the research in the fields.

The Workshop successfully drew 247 registered attendees. The format of the Workshop consisted of six parallel discussion sessions in the following science areas, each organized and chaired by two *Discussion Leaders*:

- Quantum materials research, *Ignace Jarrige (BNL), Andrew Wray (NYU)*
- Soft matter research, *Ron Pindak (BNL), Dean DeLongchamp (NIST)*
- Materials science and engineering, *Eric Dooryhee (BNL), Paul Evans (Wisconsin)*
- Catalysis and chemical sciences, *Eli Stavitski (BNL), Ayman Karim (Virginia Tech)*
- Molecular and cell biology, *Sean McSweeney (BNL), Sandra Gabelli (Johns Hopkins)*
- Biosystems & earth-environmental sciences, *Ryan Tappero (BNL), Tony Lanzirotti (Chicago)*.

In addition to parallel sessions, two plenary sessions were held on the first day of the Workshop to get everyone together for overviews of the respective scientific fields given by leading researchers in each of the six science areas. These were followed by short presentations by NSLS-II beamline program representatives that summarized the current scientific capabilities at NSLS-II .

The parallel sessions allowed more in-depth discussions in each science area through invited and ad-hoc short presentations. These discussions led to summary conclusions around four charge topics – grand scientific challenges, priority research directions, new capabilities that should be developed at NSLS-II, and potential impact of these new capabilities. At the plenary close-out session, a summary of the discussions was given by the Discussion Leaders in each of the six science areas.

This Workshop Report aims to capture the main conclusions from the 2019 NSLS-II Strategic Planning Workshop. It will serve as a forward-looking document that will inform our strategy on future facility developments in the next five to ten years; These developments include a set of possible new beamlines, new capabilities at existing beamlines and in support laboratories, as well as directions for future facility upgrades.

Executive Summary

This report on the NSLS-II Strategic Planning Workshop, "*Exploring New Science Frontiers at NSLS-II*", was completed immediately after the Workshop was held, October 21-23, 2019. However, active discussions following the Workshop and then the COVID-19 pandemic have led to fresh perspectives on the needs for future research and development in science and technology, as well as on the role NSLS-II should play in that future. These perspectives include:

Science and technology's critical role in the society. Science and technology will play an increasingly critical role in driving the global economy following the pandemic including innovation and advances in information technology and manufacturing, in clean and renewable energy, in maintaining sustainable environment, and in national security and human health and medicine.

Mission driven research enterprise and strategies. The complex nature of the grand challenges in science and technology increasingly require large-scale coordinated efforts with collaborations and participations of scientists and experts from multiple disciplines specializing in dedicated theoretical and experimental tools, often coordinated and supported from multiple sponsoring organizations.

Synchrotrons as critical resources in the nation's R&D infrastructure. Research and development capabilities and expertise at synchrotron light sources are widely recognized as key resources for tackling many of the grand scientific and technological challenges that society is facing; This requires synchrotron facilities to establish both the capabilities and the capacities to enable timely and nimble responses to meet the research and development needs in the society. Looking forward, these capabilities will increasingly be made available to researchers remotely.

This Executive Summary is structured as follows. We first summarize the main conclusions from the six science areas, including scientific and technological grand challenge questions, priority research directions, and the new capabilities that NSLS-II should consider developing. We then outline our preliminary strategy to develop the proposed new capabilities, taking into account the perspectives discussed above.

Science Challenges and Research Needs

The science discussed at the workshop is ultimately motivated by several large grand challenge questions the respective fields are seeking to address. The essence of these may be captured as follows:

- Can new quantum information systems and beyond-CMOS classical computing platforms be realized based on new quantum material systems with targeted functions at desirable ambient conditions?
- Can new sustainable processes be developed to create, synthesize, and manufacture advanced materials, based on strategic use of natural resources, to power the nation's industries?
- Can multi-scale heterogeneities and structural changes be controlled in catalytic systems to achieve the desired better chemical performance?
- How can we understand and mitigate the cycling of critical resources in natural systems and the impact of changing climate on these resources and the environment?
- Can we imagine new routes to drug discovery – based on molecular treatments and understanding how macromolecules function in biological cells in different physiological or pathological states?

To address these, and other, grand scientific challenge questions, Workshop attendees identified and recommended Priority Research Directions (PRDs) in each science area. The PRDs are captured in the respective sections in the next Chapter. Based on these PRDs, NSLS-II should enable:

- Precise measurements of electronic structure and magnetic properties within novel quantum phases, including studies of interfacial interactions, low-energy excitations, and spin-resolved electronic structures;
- Time-resolved studies of electronically driven structural phase transitions and order parameters in quantum materials, and imaging quantum phenomena and their dynamics at their native length and time scales in relevant electronic and magnetic systems;
- Real-time studies of out-of-equilibrium kinetics and intrinsic dynamics under in-situ/operando conditions during materials processing, synthesis, and manufacturing, and correlating to structures, morphology, function, and performance in post-processing materials;
- In-situ & ex-situ studies of phase inhomogeneities, phase boundaries, strain and composition gradients, defects, and interfaces in materials and catalysis systems;
- Precise identification of the active site in catalysts in terms of chemical states and their spatial distribution under reaction conditions, and transient studies to provide the critical link between dynamic structure and catalytic activity and selectivity;
- Research that bridges large range of length scales to model and understand the physics and chemistry of poorly constrained, heterogeneous natural systems and their complex interactions with biological organisms;
- Artifact-free imaging of biological cells in their disease states that are morphologically different to understand the impact of disease on cell morphology and function;
- Research that lead to understanding of the structure, function and dynamics of macromolecules in a multi-scale cellular context, including correlative and integrative analysis of multimodal datasets by complementary techniques.

New Capabilities and Potential Impact

During the discussions of these priority research directions, the participants identified a number of new capabilities and new beamlines that NSLS-II should consider developing. These are summarized below, grouped in five sections: soft X-ray scattering and spectroscopy, hard X-ray scattering and spectroscopy, X-ray imaging and microscopy, structural biology, and crosscutting capabilities.

Soft X-ray Scattering and Spectroscopy:

- *Infrared near-field nanospectroscopy*: Atomic force microscope-based near-field nanospectro-scopiy in a cryogenic environment is a new capability that would meet a number of priority research needs in quantum materials. This would a new IR beamline or a new endstation at MET.
- *Integrated laser-based ARPES at ESM*: Recent development of novel 2D materials characterized by flat bands demands ~<1-meV energy resolution for band dispersion measurements. This is not possible with synchrotron sources, but laser-based high-energy sources can now achieve bandwidths as narrow as 0.1 meV with fluxes as high as 10^{13}ph/s, albeit with a limited momentum range. Such a capability, coupled with the existing ARPES capability at ESM, would provide unprecedented access to the low-energy electronic degrees of freedom in quantum materials.
- *Spin-resolved ARPES*: Investigations of topological materials necessitates spin-resolved measurements of the electronic structure with high energy resolution. High spatial resolution, allowing measurements of smaller single-domain or device-scale surface regions, semi-confined states associated with nanostructure, magnetically polarized domains, and 1D topological states at step edges would also be beneficial. This would be additional capability at the ARI beamline.

- *Ultrahigh resolution ~1 meV soft RIXS*: RIXS is becoming a crucial complementary technique in the study of Quantum Spin Liquid (QSL) candidate systems amongst other quantum materials. Ultrahigh-energy-resolution RIXS is essential for a comprehensive understanding of such materials, and 1 meV RIXS should be the long-term upgrade plan for SIX.
- *Tender and hard RIXS nanoprobe*: There exists a growing need in XAS and RIXS between the soft and hard X-ray energy ranges, driven by interests in 4d transition metals and uranium quantum materials. Measurements at these edges using a sub-micron beam spot will provide insight into the orbital energies and electron occupancies and help address fundamental questions related to the valence of uranium and its spatial distribution in mixed-valent materials. This would require a new dedicated beamline.

Hard X-ray Scattering and Spectroscopy:

- *Multidimensional multimodal diffraction*: Many research areas require a multidimensional multimodal X-ray diffraction capability with a variable X-ray beam size of sub-mm to ~100 nm or below. Research interests include functional materials systems e.g. batteries, catalysts, interconnects, electronically driven structural phase transitions, and order parameters in quantum materials. This would be a new dedicated beamline or branch line at NSLS-II.
- *High pressure science with large volume press (LVP) and diamond Anvil cells (DAC)*: Dedicated high-energy x-ray diffraction instrument with high-pressure up to 35GPa using LVP and DACs, and variable temperatures from <10K to ~2,000K, is needed for studies on Earth minerals, ceramic, and quantum and functional materials at non-ambient conditions. Multimodal imaging and diffraction analysis bridging length scales (atoms to grains) and time scales (seconds to days) would also be required. This could be a new endstation at a HEX branch line.
- *High-throughput, high-resolution X-ray powder diffraction*: New materials discovery starts with a small amount of synthesized powdery substance of unknown structure and function. High-resolution X-ray powder diffraction plays an irreplaceable role in determining the structure of a new substance, providing feedback to materials developers to refine their process in order to achieve the desired properties. This would be a new 3PW beamline at NSLS-II.
- *X-ray diffraction & reflectivity for materials structure & processing (MSP)*: Much interest exists in the materials science and engineering community to access reliable, well-equipped x-ray diffraction and x-ray reflectivity capabilities for variety of studies important to industrial processes, including studies of strain, structure, grain size, and texture, and characterization of thin film and nanopatterned microelectronic devices. This would be a new 3PW beamline at NSLS-II.
- *Materials in radiation environment (MRE)*: A synchrotron facility optimized for the study of the structure and chemistry of radioactive materials is required to accelerate development and deployment of materials for nuclear systems. Information on crystal phase, mesoscale structures, and internal defects before and after irradiation testing in reactors provides unique critical data for the verification of simulations of nuclear systems. The new MRE beamline at NSLS-II will be an important part of the infrastructure to support the DOE's nuclear energy mission.
- *High-resolution hard X-ray XES, XRF, RIXS*: Activities in catalysts may exist in a number of chemical states and configurations with varying degrees of activity. High energy resolution fluorescence detection (HERFD) can resolve these chemical states and focus on a specific valence of interest. This would be a new undulator beamline at NSLS-II with synchronized scanning of ID gap and monochromator with sufficient resolution.
- *Advanced manufacturing processing (AMP) beamline*: Time-resolved coherent and incoherent X-ray scattering techniques are uniquely suited for in-situ/operando studies of the nanoscale material transformations under out-of-equilibrium AM processing conditions. Real world AM processing equipment and auxiliary characterization techniques require significantly larger sample areas than available at current beamlines. A new undulator beamline at NSLS-II would be needed to meet such research needs.

- *Processing and liquid surfaces (PLS)*: An independent processing and liquid scattering beamline is envisioned to provide a horizontal scattering instrument suitable for liquid surfaces and for polymer processing studies. These capabilities are coming online in FY21 as a time-shared facility at SMI but expected demand justifies a dedicated station. The plan is to develop the canted undulator beamline at SMI where the existing endsation has been developed.
- *Micro-beam X-ray transmission & scattering tomography*: Several scientific challenges in soft matter require spatially resolved structure information from complex solids. Micro-beam X-ray transmission and SAXS/WAXS tomography in both transmission and grazing incidence geometries will help to address these challenges. This could be a new endstation at SMI.

X-ray Imaging and Microcopy:

- *Soft x-ray STXM for soft matter and environmental science*: Many challenges in soft matter and environmental sciences require spatially resolved information on chemical structure, molecular orientation, and morphology at nanometer scales. With contrast provided by XANES, soft x-ray STXM at ~20 nm resolution can address these needs. This could be a new bend-magnet beamline, or an additional endstation at the Soft X-ray Nanoprobe beamline in the NEXT-II project.
- *Micro computed tomography (micro-CT) with XANES capability*: Microscale structure and morphology is often the determining factor for materials properties and function in natural and engineered systems where statistically meaningful information is required. Micro-CT would be an ideal 3PW beamline at NSLS-II to meet this research need, enabling 3D micro-imaging of real systems in-situ/operando – a capability currently missing in the NSLS-II beamlines suite.
- *Multimodal hard X-ray microprobe at ~μm resolution*: Similarly, there is considerable need for X-ray fluorescence and X-ray diffraction imaging and tomography at micron level resolution, to provide elemental, chemical, and structural information in a wide range of heterogenous materials systems and devices. Such capability would fill in the capabilities gap in the current NSLS-II portfolio of imaging capabilities. This would be a canted undulator beamline at SRX.
- *Tender X-ray 2-12 keV nanoprobe with cryogenic capability*: A tender-energy X-ray nanoprobe at 50-200 nm resolution with full cryogenic capabilities and low-background fluorescence detection is required for bio-geo-environmental investigations of the abundance, localization and chemical states of trace elements and nutrients in natural specimen in their preserved frozen-hydrated states. This would be an additional endstation at SRX.
- *Soft X-ray TXM to image frozen hydrated biological cells*: Soft X-ray cryo-tomography in the water window is a relatively new imaging modality to image intact frozen hydrated biological cells. It provides the critical information to address the science challenge on how macromolecule assemblies are organized to conduct specific cellular processes and how they are reorganized in different physiological or pathological states. This would require a new BM beamline with proper sample preparation capability at NSLS-II.

Structural Biology:

- *Automated MX for protein functional dynamics*: With serial micro-crystallography on micron-sized crystals at NSLS-II MX beamlines, new reaction trigger methods become feasible to initiative a biochemical reaction inside a micro-crystal, which enable room-temperature studies of wide range of enzyme kinetics in physiologically relevant processes at time resolutions in the μs range. To enable these new capabilities, extension to reaction initiation using the natural substrates needs to be developed for sample delivery at FMX/AMX, including microfluidic mixing jets, pH, oxygen, and temperature jumps, and capillary or droplet loaded systems. It also serves as the basis for a new highly-automated beamline.
- *Intrinsic anomalous MX for native macromolecules*: , those without any known analogs, require *de novo* methods for crystallographic phase evaluation. Single-wavelength anomalous diffraction (SAD) is being used for *de novo* structure determinations for completely novel structures. While native SAD by S or P

can succeed well on MX beamlines at ~7 keV, such experiments can be much improved using lower X-ray energies (3-7 keV) as well as microcrystals to reduce effects by absorption. This new development of native-SAD capability at NYX may have the potential to supplement Se-SAD for *de novo* structure determinations.

- *Automation from specimen prep to structure*: Robotic sample handling and automation has become the expected norm at synchrotron beamlines for macromolecular crystallography. Due to high brightness, the speed of dataset collection at the NSLS-II MX beamlines is much shorter than robotic sample handling, leading to the needs to further improve sample handling and delivery as well as automated intelligent decision-making during data collection & reduction in order to further improve the overall efficiency. This requires automated and efficient, reliable sample delivery, and the AI decision-making in specimen selection and data collection workflow.

Crosscutting Capabilities:

- *Time-resolved studies from milliseconds to nanoseconds scales*: Time-resolved studies are required for understanding and control of any materials processes. While atomic level changes and chemical reactions at fs to ps scales can be accessed at XFELs, the progression of the structural changes and reaction fronts are often affected by complexity and heterogeneities of the material system, leading to kinetics at ns to μs scales. Experiments at these time scales are well suited at NSLS-II, by taking advantage of single synchrotron pulses or via special X-ray optics & detectors with X-ray pulse-selection capability. These new crosscutting capabilities will have impacts on such studies as catalysis, microelectronics, and materials synthesis & processing.
- *In-situ & operando sample environment*: Application-specific in-situ/operando capability is a pressing need at NSLS-II beamlines for almost all science areas, such as (a) quantum materials as a function of temperature, pressure, electric and magnetic field, and lattice strain, (b) synthesis and processing of polymers, thin films, and additive manufactured devices, materials strengths and other properties under extreme conditions, and (c) catalysts performance in industrial-relevant temperature and pressure environment.
- *Sample preparation laboratories*: Dedicated sample preparations are required for biological, catalysis, and in-vacuum samples. These sample-prep capabilities should be established in user-support laboratories or at beamlines, such as (a) cryogenic sample preparation for artifact-free biological samples, (b) staging laboratory for catalysis research to allow users to benchmark their reactions in the sample environment to be used for synchrotron measurements, prior to actual beamtime, and (c) capabilities to handle or transfer air-sensitive samples into reactors or UHV, *e.g.* dedicated gloveboxes, sample suitcases or transport railway, and glovebox-at-endstation.
- *Complementary non-synchrotron techniques*: In all scientific fields, researchers make use of many complementary techniques to study materials and systems. These complementary tools include mass-spectrometry and gas chromatography, infrared, Raman, and UV-Vis spectroscopy, etc. They provide complementary information which, together with that revealed at synchrotron, provide a more complete picture about materials properties and processes for a given specimen. NSLS-II will develop these complementary methods either as multimodal capabilities at beamlines or as complementary off-line measurements in a support laboratory.
- *Data informatics, on-demand compute, AI and machine learning*: With better resolution and high flux photon beams at the sample, better detectors, and high-throughput capabilities, the amount of data generated during an experiment at NSLS-II has increased by many orders of magnitude. As such, new, intelligent, autonomous data analysis and management tools, with AI/ML and on-demand compute, are required to extract scientifically relevant information from the large amount of complex data. In addition to the newly acquired experimental data, the data strategy at NSLS-II should incorporate the experimental and theoretical & modelling data already available in the scientific literature and at materials databases. Establishing this new data-informatics approach at NSLS-II will require significant developments both in AI-guided data analysis codes as well as in data management workflows and infrastructure.

Strategy on Path Forward

NSLS-II would like to thank all Workshop attendees and all Discussion Leaders for participating in this Workshop and for many insightful input and recommendations. These discussions, as captured in this Workshop Report, provides a coherent framework to guide the future developments of the NSLS-II facility, including new beamlines and new capabilities at existing beamlines, new crosscutting technologies and support infrastructures, as well as potential future facility upgrade directions.

As discussed earlier, the COVID-19 pandemic has provided fresh perspectives on the needs for future developments in science and technology, including S&T's critical role in the society, mission driven research enterprise, and synchrotrons as irreplaceable resources in the nation's R&D infrastructure. As the nation's premier synchrotron facility, NSLS-II has the responsibility to establish and operate a wide range of user instruments to enable scientific research and technological advances. These instruments should aim to (a) offer unique and cutting-edge technical capabilities, (b) meet the mission needs of a variety of government agencies, and (c) provide sufficient capacity to enable timely responses to the research needs in the society.

With these perspectives in mind, NSLS-II has devised the following strategy on the development of new capabilities and new beamlines to meet the priority research directions identified by the scientific community. This includes an updated strategy to engage the community and potential agencies to pursue funding to develop new beamlines of mutual interest, and an approach to develop new capabilities at existing beamlines or new crosscutting capabilities.

New Beamlines

NSLS-II will consider new beamlines in three categories: enterprise beamlines, performance beamlines, and mission-focused beamlines, as stated below.

<u>*Enterprise beamlines with standard techniques*</u>: These beamlines will provide well established, widely used mature techniques and data analysis methods with high-throughput. They will complement existing high-end beamlines by running those research projects that do not, or not yet, require the high-end capabilities, and by providing preliminary or complementary measurements before taking beamtime at high-end beamlines. Overall, these enterprise beamlines typically support a large and experienced scientific user community, serve as regional capacity in the national synchrotron landscape, play a crucial role in enabling multimodal experiments, and in the end, make high-end beamlines more efficient and impactful. Possible enterprise beamlines may include:

- High-resolution X-ray powder diffraction (HRD),
- Micro computed tomography with XANES capability (MCT),
- X-ray diffraction and reflectivity for materials structure and processing (MSP),
- Micron resolution hard X-ray microprobe (MRX or SRX-II),
- Infrared near-field nano-spectroscopy (INF),
- Processing and liquid surfaces (PLS or SMI-II),
- Scanning transmission X-ray microscope (STX),
- Soft X-ray Tomography (SXT) to image frozen hydrated biological cells,
- Massive automated MX (MAX).

Because of the well-established standard techniques and communities, the developments of these enterprise beamlines are cost effective and may receive strong support from the broad scientific community as well as from a number of potential federal and state agencies that support acquisition of major research equipment for a broad community.

Performance beamlines with new cutting-edge capabilities: These beamlines will provide leadership-level, new cutting-edge capabilities, and fill the capabilities gaps in the existing NSLS-II beamline portfolio. As such, these high-end beamlines will bring in new scientific user communities and enable new research that cannot be done currently at NSLS-II, thus making high-impact to the relevant scientific fields. While at present NSLS-II already leads in a number of areas such as nano-imaging, coherent scattering, and high-resolution soft RIXS, possible new high-end beamlines may include:

- Tender and hard RIXS nanoprobe (TIN),
- Undulator-based high-resolution hard X-ray XES, XRF, and RIXS,
- Advanced manufacturing processing beamline (AMP),
- Multidimensional multimodal diffraction or a new nanoprobe dedicated for nano-XRD,
- Automated undulator based MX for protein functional dynamics (MAX-u),
- High-resolution PDF studies (HEX-II).

Aiming for world-class performance, these beamlines typically require special sources and photon delivery systems, plus one-of-a-kind endstations with specific in-situ specimen environment and sample handling for certain type of science. Development of these beamlines will require targeted outreach to potential funding agencies with their facility stewardship already exist for synchrotron beamlines.

Mission-driven beamlines with partner government agencies: As a DOE BES scientific user facility, NSLS-II is engaged in wide range of scientific research to understand and ultimately control matter and energy at the electronic, atomic, molecular, and nano- to micro-scale levels in support of the DOE stewardship missions in energy, environment, and national security. As part of our strategy, NSLS-II works with partner national laboratories and other federal and state institutions to develop new beamlines and new capabilities that meet the specific mission-driven R&D needs of the partners. Based on discussions at the Workshop and other ongoing dialogues with potential partners, such partner beamlines may include:

- Materials in radiation environment for nuclear materials (MRE),
- MARS sample return beamline facility,
- Materials research for defense threat reduction,
- Dedicated 3D nano-tomography for microelectronics (FXI-II),
- High-pressure tools for materials under stress (HEX-III),
- Metrology and instrumentation development (MID).

These beamlines all require specialized endstations that provide novel capabilities to accommodate difficult-to-handle samples in a secure environment, often in a satellite external building. NSLS-II will continue to work with the respective partner national laboratories and federal agencies to develop these new beamlines that are dedicated to meet the research and development needs of our society.

For each of the above categories of beamlines, NSLS-II plans to hold internal discussions to further digest the community's needs and prioritize among competing concepts, and to engage the scientific leaders in the respective communities and outreach to potential funding agencies about development of our high-priority beamlines. This may be followed by a community workshop for each of the beamline concepts to refine the science case and define the beamline scope and preconceptual layout. It is envisioned that a white paper will be produced to capture these discussions, including the science case, the beamline concept, the community, and the expected impact to the relevant research fields. The white paper will be shared with the interested funding agencies to engage in discussions of how the new beamline may be funded for development and construction.

Additions and Upgrades to Existing Beamlines, and Crosscutting Capabilities

A number of new capabilities proposed at the Workshop can be developed or accommodated as upgrades or additions at existing NSLS-II beamlines or at beamlines under development. These include the following:

- Integrated laser-based and spin-resolved ARPES at ESM or ARI,
- Ultrahigh energy resolution ~1 meV soft RIXS at SIX,
- Advanced fabrication and manipulation tool for layered materials at ESM,
- Tender X-ray nanoprobe for bio-environmental sciences at SRX,

- Soft X-ray STXM for soft matter and environmental research at SXN,
- Hard X-ray micro-CT for paleontology and engineering systems at HEX,
- Intrinsic anomalous MX for native macromolecules at NYX,
- Automation from specimen prep to macromolecule structure at AMX and FMX.

In addition, the following crosscutting capabilities as identified by Workshop participants are similar additions that can be located either at NSLS-II beamlines or in NSLS-II user laboratories:

- In-situ and operando sample environments to mimic real environmental and operating conditions,
- Sample preparation tools in laboratories and at endstations,
- Complementary non-synchrotron techniques.

NSLS-II will continue to allocate a small portion of its annual operating budget to support Facility Improvement Projects that will include some of the upgrades or additions to existing beamlines or laboratories. Since such funding is not expected to be large, NSLS-II will work with the scientific community to pursue additional funding opportunities at *e.g.* NSF, NIH, and DOE for acquisitions of major research instruments to be implemented and operated at NSLS-II. To ensure better coordination and programmatic alignment, NSLS-II plans to periodically outreach to the scientific community, outline the specific areas where additional major research instruments would be needed, and solicit interests from researchers in the academia or other institutions to jointly pursue funding for these instruments from respective agencies. In each of these cases, assuming funding is successful, a formal partnership may be developed in the form of a Partner User at a NSLS-II beamline or beamlines.

NSLS-II will take the stewardship role to develop the following crosscutting capabilities identified at the Workshop:

- Time-resolved studies from milliseconds to nanoseconds scales,
- Data informatics, on-demand compute, AI and machine learning.

The development of time-resolved capabilities requiring special NSLS-II storage ring bunch patterns will be coordinated through a newly formed Time-Resolved Studies taskforce to identify the specific research needs and the desired bunch patterns, as well as advanced detection schemes, in order to enable the targeted time-resolved materials research at NSLS-II. Once tested, such special bunch pattern runs, if needed, may become a part of the normal NSLS-II operating schedule moving forward in order to meet the needs of the research community.

Data analytics, on-demand computing, AI and machine learning is a very active and growing area of research and development at NSLS-II as well as at other light source facilities. In addition to the development of the most advanced flexible data acquisition and data broker architecture, NSLS-II staff have been awarded multiple BNL laboratory directed R&D (LDRD) projects in the area of data analytics and machine learning; These include: high-throughput data analytics on X-ray spectroscopy and diffraction data based on machine learning, community-curated data in literature and databases, as well as theory and modelling, AI-guided autonomous X-ray scattering experiments, physics informed machine-learning for X-ray spectro-imaging and microscopy, intelligent synthesis of quantum dots for high throughput targeted quantum materials discovery, and machine learning for real-time identification and removal of artifacts in coherent synchrotron data.

Recognizing its crucial importance, NSLS-II will continue to: (1) collaborate with the Computational Science Initiative (CSI) at BNL to advance the data management and data curation infrastructures at beamlines, at NSLS-II central, and at the future BNL data center, and (2) work with the scientific community as well as with the CSI other DOE light sources to develop modern, advanced, intelligent data analytics based on AI and machine-learning as well as curated experimental and theoretical data in the literature and the databases. In close coordination with other DOE BES light source user facilities, NSLS-II will pursue additional dedicated funding to support the development of these advanced capabilities and make them available to the broad scientific user community.

Science Session Reports

The 2019 NSLS-II Strategic Planning Workshop consisted of six parallel discussion sessions in the following science areas, each organized and chaired by two *Discussion Leaders*:

- Quantum materials research, *Ignace Jarrige (BNL), Andrew Wray (NYU)*
- Soft matter research, *Ron Pindak (BNL), Dean DeLongchamp (NIST)*
- Materials science and engineering, *Eric Dooryhee (BNL), Paul Evans (Wisconsin)*
- Catalysis and chemical sciences, *Eli Stavitski (BNL), Ayman Karim (Virginia Tech)*
- Molecular and cell biology, *Sean McSweeney (BNL), Sandra Gabelli (Johns Hopkins)*
- Biosystems & earth-environmental sciences, *Ryan Tappero (BNL), Tony Lanzirotti (Chicago)*.

The co-discussion leaders worked with the broad scientific community and organized the discussions in each of the six research fields to identify the scientific grand challenges, priority research directions and research needs, a set of new capabilities that should be developed at NSLS-II, and the potential impact of these new capabilities to the research field. A large amount of information was exchanged and discussed at these parallel sessions. The following are the six Science Session Reports intended to capture all the essential discussions from these Science Sessions.

Quantum Materials

Atoms and electrons are quantum mechanical objects, obeying principles such as Pauli exclusion and Heisenberg uncertainty that defy classical intuition. When many atoms come together, the emergent behavior of the collective system can exhibit remarkable and counterintuitive phenomena that extend up from the microscopic level to the scale of visible matter and devices. The quest to design and control such emergent quantum effects in materials to achieve a desired functionality is the foundation of quantum materials research.

Recently, the future of computing, both quantum and classical, has become a major driver of advancement in the field, as the industry and academy both look to quantum materials and their novel physics to drive innovations. These materials and their quantum coherent many-particle effects are indeed seen as an increasingly vital component of quantum information technologies, involving quantum computing, communication, and sensing. To provide some context, recent breakthroughs in this nascent area, including hints of vastly sped-up information processing in real-world quantum computers – referred to as 'quantum supremacy' – have had dramatic implications at many levels, including for basic science, consumer electronics, and national security. The importance of this area has been acknowledged by major funding initiatives within DOE, NSF, and DOD, as well as in the creation of new divisions within a range of private companies such as IBM, Microsoft, Google, Goldman Sachs, and many more. Besides quantum computing, quantum materials are also among the leading options for surpassing the limits of the existing silicon-based technology in classical computing, through a number of approaches relying on magnetism, electronic correlations, or the nontrivial topology of the electronic structure. Here, the end goal is to curb the decline of Moore's law by circumventing the energy inefficiency of the existing Von Neumann architecture. In addition to computing, quantum materials also serve as a platform for discovery and control of emergent properties in a range of applications including electronics, optics, thermoelectrics, energy harvesting, etc.

Besides promising applications, another indication of the richness of the field is reflected by the diversity of the quantum materials themselves. Quantum materials can be bulk materials, such as intrinsic superconductors, or can be engineered structures, such as nanophotonic structures and layered materials whose properties surpass the sum of their bulk constituents. The common thread in these materials and systems is that the relationship between quantum electronic properties and atomic structure is extremely complex since charge, spin, orbital, and lattice degrees of freedom are intertwined over length scales ranging from atomic to mesoscopic, which in turn yield emergent behaviors and intricate phase diagrams. As this complexity continues to fuel interest in this rapidly evolving field, the central questions over quantum materials remain the same: What quantum states are possible within bulk materials and engineered structures, and what emergent properties can arise from these states? How can quantum phenomena be tuned on-demand within functional devices? As discussed in this section, addressing these questions requires multiscale visualization of the microscopic quantum properties in solids, which necessitates an array of state-of-the-art probes of the electronic structure. Following an overview of the outstanding science questions and research priorities to address these questions, this section offers a summary of the capabilities that are currently missing in the beamline portfolio of NSLS-II to aggressively address these field-wide questions, and offer a unique suite of tools dedicated to advancing the state-of-the art in quantum materials research.

Science Questions/Challenges

Science Question #1: Can powerful new quantum information platforms be realized based on quantum materials?

Quantum information systems promise to revolutionize the future of computing technology, with the potential to profoundly outstrip Moore's law in some applications. However, as has been the case in the 70+ year development of our present transistor-based computing technology, there is no golden road forward by which one can envision the creation of a useful quantum computer with our existing materials science knowledge. Creating such a computer will require overcoming the most critical challenge for quantum information technologies, which is decoherence due to coupling with unwanted degrees of freedom. It is noteworthy that the approaches currently embraced as most practical by industry all rely on quantum material properties, such as superconductivity in transmon qubits (IBM, Google), and a combination of superconductivity with magnetism and quantum topology in Majorana-based approaches (Microsoft). Related material systems investigated by the workshop participants were even more diverse, driving home that the coming years will be a pivotal time for the identification of quantum materials, states and phases that can be incorporated to successfully advance the foundations of quantum information technology. The key scientific frontiers brought up in the workshop are described below.

Exotic superconductivity: The past few years have seen the emergence of new grand goals for superconductivity research, beyond the enhancement of the superconducting transition temperature. Recent heterostructure engineering and basic materials research increasingly focus on the realization of spin triplet and/or topological superconductivity, and recent theoretical proposals of Majorana fermion based quantum information platforms are generally modeled under the assumption that the material platform is a spin triplet superconductor. This type of superconductivity differs from the conventional type in that paired electrons have their spins aligned as opposed to the usual antiparallel state. Spin triplet superconductors are rare – of the many thousands of superconductors discovered, only a handful exhibit spin triplet pairing, and even here the experimental evidence has been controversial. Nonetheless, the physics of these superconductors, because the pairing interaction differs fundamentally from those in conventional superconductors, is far richer and more varied. A large class of spin triplet superconductors intrinsically incorporate nontrivial quantum topology, which is a more general prerequisite for fault tolerant Majorana fermion-based approaches to quantum computing.

Quantum spin liquids: A quantum spin liquid (QSL) is an exotic state of matter in which qubit-like quantum mechanical degrees of freedom of the electronic spin system persist even at absolute zero temperature, rather than freezing into a static ordered state (spin solid). The spins in a QSL are in a topologically-ordered state, in which electrons are behaving as if they are broken into several parts. An example of such fractional quasi-particles is called a Majorana fermion, which can be manipulated to carry out topological quantum computing. In other words, a QSL harbors the essential ingredient for building a topological quantum computer. However, a physical realization of a QSL is still elusive, and there is no consensus as to whether a robust QSL can be synthesized despite intensive materials research efforts to find a QSL material [1]. Geometrically frustrated magnetic insulators such as Kagome lattice antiferromagnet herbertsmithite have been studied extensively [2]. Another promising route is through honeycomb lattice magnets with bond-dependent Kitaev interactions arising from strong spin-orbit coupling [3]. Examples are alpha-$RuCl_3$ and various iridates [4-5].

Other stable excitations: Where current technologies manipulate the distribution of electronic charge to store quantum information, this tends to be insufficiently stable for preserving quantum coherence in a solid-state platform. The last few years have seen the incremental discovery of a series of more stable quantum systems based instead on other degrees of freedom such as spin at impurity sites [6-8], wavefunction phase distributions, or other charge-neutral modes that can be deemed "topologically stable" (including Majorana fermions). New classes of low energy collective modes are regularly discovered in quantum materials research, and represent the most stable forms that information can take within the given environment. Even when excitations do not readily achieve the high standards of stability needed for quantum information, as in the case of topologically stable magnetic skyrmions, they are often of great interest for next generation classical information and sensing technologies.

Science Question #2: Can quantum materials provide a promising approach for beyond-CMOS classical computing architectures?

The Von Neumann architecture forms the basis of modern CMOS-based computation, where logic and memory are separate steps, and suffers from an inherent bottleneck in the combined speed, efficiency, and heat dissipation that can be achieved. While this bottleneck is the root cause of the impending end of Moore's law, it has also given great impetus to the development of alternative concepts and devices for higher-efficiency computing, for which quantum materials have been proposed as building blocks. The most prominent efforts are summarized below.

Neuromorphics: The human brain is capable of supercomputing processing with a power consumption of just 20W by combining logic and memory in single steps. Efforts to mimic this neuronal logic at the device and hardware level are termed neuromorphic computing. At the heart of this architecture is the neuristor, a neuron-like transistor, which exhibits the spiking behavior of the neuron. A neuristor uses two coupled memristors, which are resistors with a history-dependent resistance providing a memory effect, and capacitors in series [9]. While memristors were first theorized by symmetry arguments in the 1970s [10], filamentary-based memristors were realized decades later [11]. For energy efficiency and industrial scaling purposes, less entropically expensive switching mechanisms are required than the breaking and reforming of fragile electroformed conductive filaments. As a result, strongly correlated oxides, where abrupt metal insulator transitions of several orders of magnitude can be triggered by the application of bias, temperature and stress are a promising area for developing the class of so-called "Mott memristors" for energy-efficient neuristors [12]. The key scientific question in this area is how can we rationally engineer the quantum mechanics of this class of materials to realize the desired switching properties?

Topological insulators and semimetals: Since 2006, there has been an explosive progression in the discovery of topologically ordered materials, and the distinction of important subclasses such as topological insulators along with Dirac, Weyl, and nodal line semimetals. These materials feature reduced scattering and strongly spin-polarized surface conductivity, properties that are leading to increased application in beyond-CMOS devices and other energy-conserving computing technologies needed to extend Koomey's law of transistor power consumption (a prerequisite of Moore's law). Topological field-effect switching is a considered a leading potential application, where the dissipationless conduction channels in the topological phase ('on') can be switched to non-conductive channels in the conventional insulating phase ('off') under the application of an electric field [13]. Another promising application utilizes the strong spin-charge coupling characteristic of topological matter to enable advanced spin-to-charge conversion, which can provide an efficient readout or control of the state of beyond-CMOS switches [14]. Further, materials exhibiting topologically protected quasiparticles or topologically non-trivial real-space spin textures are also inspiring novel spintronic devices aiming to be more stable and energy efficient [15]. While these developments look promising, viable technological applications will still require a better understanding of the fundamental physics of these systems.

Science Question #3: Can the physics of layered materials be harnessed for advanced electronic and optical devices?

Heterostructures built from stacked low-dimensional semiconductors have become a major element of global quantum materials research. The broad vision for these systems comes from their versatility and unprecedented layer-dependent electronic properties (transport, optical absorption and luminescence, magnetic and electrical susceptibility), which can be tuned through control of the individual layer composition, the relative crystallographic orientation between layers, or spatial patterning. Of particular interest is the van der Waals layering approach, which consists in placing atomically thin two-dimensional single-crystalline sheets on top of each other, and where van-der-Waals-like forces keep the stack together. This approach provides freedom from constraints on chemical compatibility between layers, atomic-scale interdiffusion, or strain, and enabling the fabrication of previously inaccessible material architectures with atomically-sharp interfaces between layers of vastly different properties. We note that a recently funded initiative will see the CFN at BNL develop a one-of-a-kind facility for automated synthesis of layered van der Waals heterostructure materials, called the Quantum Material Press (QPress), highlighting the potential for NSLS-II – CFN synergistic opportunities in this burgeoning field.

Microelectronics: 2D heterostructures possess unusual electronic properties that provide a fertile ground for both fundamental research and applications. Recent highlights include valleytronics, which relies on the controlled confinement of carriers in distinct valleys of the band structure as a way to achieve low-power information encoding [16], the realization of fully tunable topological valley transport robust against defects and impurities in materials [17], the development of designer topological materials based on Moiré networks [18], and even the emergence of unconventional superconductivity [19]. Central challenges in this area include the manipulation of the valley degree of freedom during the lifetime of the valley state and the transfer of valley information [20].

Optics and novel light-matter interactions: 2D materials such as transition metal dichalcogenides are endowed with unique optical attributes leading to rich optical phenomena. Examples include robust photoluminescence due to a direct-bandgap semiconductor structure [16], and single photon emitters from defect centers, which open up new applications in quantum optoelectronics [21]. Device concepts based on collective excitations in 2D materials have recently emerged as new platforms for exploring light-matter interactions at the nanoscale, such as the quantum-coherent coupling of electrons with light (polariton modes) [22], and the associated realization of topological photonic states [23, 24]. Open questions include: How to drive and manipulate coherent quantum effects with light and harness it at the nanoscale? Can we transfer the chirality and coherence of light waves to a solid-state system?

Priority Research Directions

Priority Research Direction #1: Achieving precise measurement of electronic structure and magnetic properties within desired quantum phases (Science Questions #1-3).

Basic characterization of electronic structure is already a primary focus of synchrotron research, and employs mature techniques such as angle-resolved photoelectron spectroscopy (ARPES), X-ray absorption spectroscopy (XAS), resonant elastic X-ray scattering (REXS) and resonant inelastic X-ray scattering (RIXS). There are prominent cases such as QSL and exotic superconductivity for which such basic characterization is the primary frontier of current research. QSL phases feature spins that are highly correlated with each other yet do not have a periodic alignment. Measuring the correlation and entanglement between disordered spins within a QSL phase is a prerequisite for unequivocal demonstration of the formation of a QSL ground state. It is however difficult with conventional synchrotron-based magnetic probes. One possible direction is to look for diffuse patterns of magnetic correlations in momentum space.

Exotic superconductivity also presents particularly broad characterization challenges as it emerges from a complex interplay of interactions. The candidate quantum materials for topological superconductivity incorporate topological and/or strongly correlated quantum materials physics, which lead to the unconventional coupling of electrons into spin-aligned Cooper pairs. The relevant electronic structure encompasses magnetic moments and atomic multiplet physics, transient correlations of spin and charge, and the momentum-resolved occupation of electronic states.

Exploring these states calls for *high energy resolution* since the quantum processes at work have energy scales of millielectronvolts, *control of sample environment* down to ultra-low temperatures (<1 K), and is benefited by control over additional parameters such as magnetic field. *Spatial resolution* is vital to isolate pristine regions for disorder-sensitive states (QSL and odd-parity superconductivity), and because many of the current key compounds (e.g. UTe_2) present inhomogeneous surfaces. Finally, *temporal resolution* up to 10 KHz is also key to gain information on the dynamics and fluctuations of the correlated charge and spin phenomena displayed by these systems.

Priority Research Direction #2: Imaging quantum phenomena at their native scales (Science Question #3).

There is an inherent, field-wide interest in probing quantum phenomena in real space, and understanding local properties in materials that control scattering, decoherence, and dynamical evolution of quantum states. One can also explore the interplay of exotic electronic states with nanoscale structure and disorder, such as the emergence of protected resonance states and edge modes in topological systems and graphene. Key knowledge gaps exist for many or all collective modes, such as the structure of polaritonic wavepackets in designer quantum heterostructures noted in Science Case 3 above, including chiral polariton modes that can be unidirectional and topologically protected against backscattering. Real-space observations of quantum phenomena are however extremely challenging as they require both nanoscale spatial resolution (10 nm or better) and high energy resolution down to ~1 meV while coupling to the low energy scales of quantum many-body physics. In other words, attaining this ambitious goal calls for *real-space nanoscale mapping of the dispersion of low-energy collective modes in frequency and momentum space*, such as real-space imaging of bulk or edge plasmons in nanoflakes and unidirectional chiral polariton propagation in thin-film edges.

Priority Research Direction #3: Studying emergence of electronic and magnetic properties arising from interfacial interactions (Science Questions #1-3)

The primary goal here is to selectively access interfacial states to extract information about interlayer interactions, proximity effects, and superstructure periodicity. This research direction is driven by the rapidly growing interest in superstructures, nanosheets, and artificial nanoscale structures in general, and also applies to studies of intrinsically inhomogeneous samples. This research direction is complementary to the previous one, in that it requires spatially resolved spectroscopic studies of the electrons and their associated dynamics. It would benefit from a *multimodal approach to measure the momentum-resolved electronic structure and the energy-momentum loss spectra at sub-micron-scale resolution* from the same spot within the same sample or device, in order to extract spatially dependent properties of the electronic quasiparticles and excitation spectra. Exploiting the resonance of the elements located at the interface would also be instrumental in enhancing interfacial selectivity.

Priority Research Direction #4: Measurements of collective modes in 4d transition-metal systems (Science Questions #1-3)

Owing to their combination of strong spin-orbit coupling and strong correlations, 4d transition metal compounds have become increasingly central to modern quantum materials research, and form the basis of some of the science questions discussed in the previous section. However, they represent a significant spectroscopic knowledge gap due to a lack of currently accessible core level resonances. These materials play a seminal role in the development of layered transition metal dichalcogenides (Nb, Mo, Ru) known for their unique electronic and optical properties, and functional metal oxides as a platform for novel electronics (Nb, Ru). Ruthenium compounds in particular have been in the spotlight in the context of Kitaev spin liquids, spin-triplet superconductivity, and for realizing solid state Higgs modes. The strong interest in the *4d* transition metals calls for *momentum-resolved spectroscopic measurements of their electronic structure and collective modes*. For resonant techniques, probing the valence states of these elements in a momentum-resolved fashion requires measuring at the $L_{2,3}$ edges. Measurements at these edges have specific optical constraints, described in the next section, which explains their scarcity.

Priority Research Direction #5: Measurement of low-energy excitation continua linked with emergent phenomena (Science Question #1).

Fluctuations with a continuum-like energy distributions are of increasingly broad interest. Fractional excitations such as the Majorana fermions expected to emerge within QSLs are likely to be most easily observed from associated magnetic excitation continua. Similar continua are speculated to be a generic feature of quantum critical points, which are of broad interest as generators of unique quantum material phenomena that can include exotic superconductivity. Another significant example is the recent observation of an anomalous continuum within superconducting cuprates, which may explain the "strange metal" behavior and associated quantum critical phenomenology that they exhibit. For QSLs, this research direction addresses a core question of the field, which is "how do we know whether we have a QSL or not?" The lack of magnetic order is one way, but proof of this is not easy to obtain. The most important tools for identifying QSL and directly probing Majorana fermions are momentum-resolving spin-coupled spectroscopy.

Regardless of origin, continua derived from electronic degrees of freedom tend to extend to large energy scales (100s of meV), and their impact on material physics is largely determined by how they extrapolate to the energy scale of thermal fluctuations (<30 meV).

Priority Research Direction #6: Measurement of spin-resolved electronic structure in topological materials and spintronics devices (Science Questions #1,2)

As is apparent in the Science Questions/Challenges subsection, topological materials are an extremely popular subject of investigation in quantum materials research. Because topological insulators are characterized by spin-polarized edge or surface states and Weyl semimetals by a spin-textured Fermi arc, measurements of the spin texture of the electronic structure are of central importance to these classes of materials. This can be done by adding *spin selectivity to angle resolved photoemission spectroscopy*, which has proven instrumental in studies of quantum topology and topological protection through measurements of spin-resolved band maps or momentum distribution curves at constant binding energy. Such spin-resolved measurements of the electronic structure are a prerequisite to demonstrating the practical importance of topological states of matter, such as in spintronics applications and spin-orbit coupling based quantum devices.

Priority Research Direction #7: Measurement of magnetization dynamics (Science Question #2)

Major candidates for beyond-CMOS technology include spintronics and nanomagnetic logic, which have the potential to give rise to ultralow-power, high-density computational architectures. The ability to visualize and control complex magnetization dynamics is vital to the development of both applications. For spintronics, the transport of domain walls has garnered increasing interest, with promises of current-controlled domain wall displacements for memory [25] and logic [26], and chirality-encoded domain wall logic [27].

For nanomagnetic logic systems, which rely on long arrays of single-domain nanomagnets, the main challenge is to overcome the effects of thermal fluctuations which can affect the level of ordering [28]. Also, the use of the domain walls has recently been proposed to improve interconnection performance [29]. Further progress in both applications calls for high spatial and temporal imaging of the dynamics of the domain walls, respectively down to ~50 nm and up to 10 KHz. This requires further improvements in the current state-of-the-art resolutions of x-ray photon correlation spectroscopy (XPCS), holography and Bragg topography.

New NSLS-II Capabilities

A core wish of the Quantum Materials Community, described through Capabilities #1-4, is to push the spatial, energy, and temporal resolutions to levels higher than what is currently offered at NSLS-II, or even planned with the NEXT-II project. There is also a manifest need to extend the range of accessible energies to the intermediate regime between soft and hard X-rays, as captured in Capability #5. Capabilities #6-8 respectively highlight the urgent need for close integration of specimen synthesis with the beamline environment, advanced multimodal sample environments, and development of new data analysis tools. Finally, Capabilities #9 and 10 address the capacity need for general-purpose characterization tools, and the desire to enable spin-resolved studies of the electronic structure. Time frame

Capability #1: Broadband near-field infrared nanospectroscopy. The development of AFM-based near-field nanospectroscopic probing and imaging in a cryogenic environment is proposed to support *Priority Research Directions #2 and #3*. The instrument uses apertureless scattering from an atomic force microscope (AFM) tip, simultaneously providing other proximal measurement modalities. The apertureless scattering technique is now a proven and powerful capability for studying quantum phase transitions and coherent optical properties in low-dimensional materials. The lack of laboratory-based IR sources having sufficient spectral radiance over the required frequency range normally limits the full applicability of the AFM probe as a spectroscopic tool. It is thus proposed to couple nano-optics with the high-radiance, ultra-broad spectral range of NSLS-II in the THz-IR region (2 to 1000 mm wavelength), to combine the merits of traditional broadband IR spectroscopy with nanoscopic resolution -- orders of magnitude better than the far-field diffraction-limit. This will allow measurements of the dispersion of collective modes in frequency over a broad range of energy scales and with a spatial resolution below 10 nm. The low-energy photons of THz and IR radiation are inherently sensitive to the elementary excitations and collective modes of electrons and the lattice, while the spatial resolution enables direct inquiry into the local electronic properties. A cryogenic environment (T~5K) will enable application to an extremely wide range of low temperature phases that can only be effectively studied with the high energy resolution of THz-IR photons. The combination of energy and spatial resolution coupled with the broadband photon source provided by the synchrotron will provide unparalleled real-space insight into the electronic properties of new quantum materials and quantum devices.

Capability #2: Laser-based sub-meV ARPES. The addition of a laser-based high-resolution ARPES capability to the existing ESM beamline infrastructure is proposed in support of *Priority Research Direction #1*. The recent development of novel 2D materials characterized by flat bands now routinely demands ~<1-meV energy resolution for band dispersion measurements. Although such ultrahigh resolutions are still unattainable for UV and soft X-ray synchrotron beamlines, laser-based high-energy sources can now achieve bandwidths as narrow as 0.1 meV with fluxes as high as 10^{13}ph/s, albeit with a limited momentum range. Therefore, the combination of the current synchrotron-based ARPES capability benefiting of large energy-momentum coverage at a 5-meV resolution with sub-meV resolution laser-based ARPES will provide unprecedented access to the low-energy electronic degrees of freedom in quantum materials. The commercial deployment of high-repetition Ti:sapphire femtosecond lasers a few years ago has contributed to a steep rise in the number of successful laser-based sub-meV ARPES set-ups. We note that such an addition to ESM will also enable round-the-clock operations (pre-alignment, evaluating cleaves, supplementary data sets, ARPES measurements while the synchrotron X-ray beam is switched to the PEEM branch etc). With some further work, this new laser-based capability could also be expanded to allow pump-probe time-resolved investigations.

Capability #3: Ultrahigh-energy-resolution soft X-ray RIXS. As reflected in the *Priority Research Directions #1 and #5*, the most important tools for identifying QSL and directly probing Majorana fermions are momentum-resolving magnetic spectroscopy methods such as inelastic neutron scattering (INS) or resonant inelastic x-ray scattering (RIXS). INS is well established, but RIXS is becoming a crucial complementary technique in the study of QSL candidate materials, especially for Kitaev QSL materials based on iridates due to the large neutron absorption cross-section of iridium. In addition, a recent theoretical study pointed out that polarization dependence in the RIXS spectrum could allow one to measure the energy-momentum relation of mobile Majorana fermions, which is not accessible to inelastic neutron scattering [30]. Therefore, ultrahigh-energy-resolution RIXS is essential to reach a comprehensive understanding of QSL physics in Kitaev materials such as alpha-$RuCl_3$ and Na_2IrO_3.

Based on the small energy scales of previously reported signatures of fractional excitations [5], a ~1 meV energy resolution would bring RIXS at the forefront of the quest for QSL and Majorana fermions. Such a revolutionary capability would also match the current energy resolution of ARPES and thermal neutron experiments, with the potential to redefine the cutting-edge in quantum materials research due to the bulk sensitivity and compatibility with small samples of RIXS. It is noteworthy that 1 meV RIXS would also play a leading role in determining the superconducting order parameter in exotic superconductors, and establishing the quantum mechanical details underlying unconventional superconductivity through determination of the electronic band structure and magnetic properties. While requiring major optical developments, a conceptual approach to push the current state-of-the-art of high-resolution soft X-ray RIXS towards 1 meV has been recently proposed [31].

Capability #4: Next-generation X-ray photoelectron correlation spectroscopy. Implementing *Priority Research Direction #7* requires pushing the current best-in-class capabilities of XPCS in terms of signal-to-noise ratio and time resolution. XPCS is a uniquely powerful tool to study fluctuations of collective magnetization dynamics at the nanoscale. Resolving the complex dynamic processes involving magnetic domains in spintronics and nanomagnet logic systems requires improving the time resolution by up to two orders of magnitude. Such improvement is not trivial, as it requires both faster detectors and an improvement of the signal-to-noise and signal-to-background ratios. The impact of such a major development would be felt beyond spintronics and nanomagnet logic, as it would also lead to new applications of XPCS to investigate exotic magnetic properties in relation with *Priority Research Direction #1* which span several orders of magnitude on the time axis, such as frustrated magnetism and diluted magnets [32].

Capability #5: Sub-micron resolution tender X-ray XAS and RIXS. *Priority Research Direction #4* highlights the growing need to bridge the gap between the soft and hard X-ray energy ranges, in order to cover the $L_{2,3}$ edges of 4d transition metals in the so-called tender X-ray regime. This will also provide access to the $M_{4,5}$ edges of uranium, an element of increasing importance in quantum materials research due to realizing spin triplet Cooper pairing of interest for topological superconductivity and quantum information. XAS measurements at these edges coupled with a sub-micron beam spot will provide invaluable insight into the orbital energies and electron occupancies of 4d-transition-metal based functional materials, superstructures and devices. It will also help address fundamental questions related to the valence of uranium and its spatial distribution in a wide range of mixed-valent materials studied in the context of exotic superconductivity. Due to the higher photon energies than soft X-rays, high-energy-resolution RIXS measurements at these edges will benefit from sizeable momentum transfers, enabling momentum-resolved studies of a host of low-energy electronic excitations in 4d-transition-metal and 5f-electron materials for the first time. Combined with a sub-micron beam spot, this RIXS capability could also be applied to nanostructures and devices associated with these elements, in line with *Priority Research Direction #3*.

We note that efforts to develop high-energy-resolution spectroscopic capabilities in the tender X-ray regime have been extremely scarce so far. Although the gap between grating (soft X-ray) and crystal (hard X-ray) monochromators has been closed around 2 keV some time ago, the tender X-ray region is outside the respective ranges of optimum performance of both types of monochromators. The gratings show a dramatic decrease in efficiency, while crystal-based monochromators suffer from heat-load related instabilities and stringent challenges associated with the fabrication of high-quality sapphire and quartz optics – whose use is imposed by the limited number of Bragg reflections for Si and Ge at low energies. Fortunately, recent optical breakthroughs are showing promise that high energy resolution in the tender X-ray regime is about to become a reality. A resolution of ~100 meV has already been achieved for RIXS at the Ru L_3 edge using crystal optics, with a path to approach 50 meV in the near future, and recent optimizations of multilayer-coated blazed gratings have also led to substantial increases in efficiency. These recent developments further attest to the timeliness of this suggested high-energy-resolution, submicron XAS and RIXS capability.

Capability #6: Advanced fabrication and manipulation tool for layered materials. The impact of the fast-expanding list of van der Waals layered materials is increasingly being felt across *all the Science Questions and Priority Research Directions* in this section. Despite their undeniable importance, research on these materials and the exploration of more complex heterostructures is severely limited by the random positions and shapes of exfoliated two-dimensional crystals which require a laborious manual process of optical microscopy-based search and mechanical transfer. Developing a user-friendly sample assembly station close to the beamlines dedicated to the exfoliation and stacking of these thin flakes often just a few microns in size will help NSLS-II play a leading role in the field. Current robotic carbon nanotube technology, as seen in 3D printing, laser engraving/ablation or microelectronic assembly, provides high-precision tools for shaping and manipulating micron-size specimens, which represent an appealing route for advanced layered sample preparation. Another option would be to build a simpler version of the CFN QPress at NSLS-II, including the software and standardized substrates to identify flakes. Given the sensitivity of 2D materials to air and moisture, this capability would benefit from running in an inert environment, and with provisions for isolated transport to beamlines using vacuum suitcases.

Capability #7: Advanced multimodal sample environments. Advanced sample environments are a pressing need for *all the Science Questions and Priority Research Directions* in this section. Most quantum phenomena of interest coexist with or result from competing electronic interactions resulting in complex phase diagrams, so the ability to carry out measurements as a function of temperature, magnetic field, and strain is necessary. In particular, the ability to achieve ultralow temperatures (<1 K) is essential to interrogate quantum ground states, achieve quantum coherence, and stabilize exotic phases of matter (e.g., spin liquids, topological phases, exotic superconductivity). Other nontraditional capabilities such as *in situ* transport characterization should also be impactful. Given the increasing need for using multiple probes on the same sample and under the same conditions, compatible sample environments, or compatible sample holders and vacuum suitcases for air-sensitive samples or surface-sensitive measurements, are strongly desired to enable multimodal studies at complementary beamlines.

Capability #8: Analysis and measurement software. New and upcoming beamlines at NSLS-II exploit high source coherence and multimodal measurement capabilities to enable richer and higher dimensional spectroscopic information than has previously been available. These expanded data sets call for new measurement software, such as to enable high quality rapid scanning data acquisition from samples that are damaged by a highly focused beam. Moreover, sparse and high dimensional data sets present an unprecedented visualization and analysis challenge for users, particularly when informed real-time decision making is needed during an experiment. Appropriate analysis software can also meaningfully expand the capabilities of a beamline, as recent studies have demonstrated that applying "big data" analysis algorithms to high-dimensional spectromicroscopy data sets can reveal novel experimental observables [33]. New classes of visualization software are reflected in the data presentation within recent spectromicroscopy publications [33,34], but these tools are not available within the spectroscopy community at large.

Capability #9: Characterization capability to advance synthesis and fabrication of new materials and systems. Quantum materials research relies increasingly on the growth of thin films of exceptional quality with new and complex stoichiometry, superstructures with atomic-scale layering control, and the fabrication of customized nanostructures. Routine characterizations of the structural, electronic and magnetic properties of these advanced materials and systems using synchrotron-based techniques have substantially accelerated improvement of their quality. However, in order to be meaningful, the interplay between the processes of making and characterizing the samples requires regular access to one or several beamlines, which is severely hindered by the oversubscription of the instruments of interest at NSLS-II. This calls for the development of a multimodal beamline for materials characterization, which would fill the current capacity shortage at NSLS-II, and provide all the required characterization tools on a single beamline, including a single-crystal diffraction setup compatible with crystal truncation rod studies, XAS, XMCD, and HAXPES. It should be noted that these capabilities can also be instrumental in providing fundamental support to the quest for complex, exotic states of quantum matter.

Capability #10: Spin-resolved ARPES. As described in *Priority Research Direction #7*, the investigation of topological materials is an important enterprise in current quantum materials research which necessitates spin-resolved measurements of the electronic structure. It specifically calls for a spin-resolved ARPES capability with high energy resolution. We note that this line of investigation can be bolstered by spatial resolution allowing measurements of smaller single-domain or device-scale surface regions. Smaller-scale features (<100nm) such as semi-confined states associated with nanostructure, magnetically polarized domains, and 1D topological states at step edges, are also of broad interest.

Impact of the New Capabilities

The capabilities proposed in this section will expand the spectroscopy, scattering, and imaging toolbox of NSLS-II, further advancing energy, spatial, and temporal resolution (Capabilities #1-4), while enabling major developments in sample preparation and environments (Capabilities #6,7). They will ignite substantial progress in our understanding of quantum states and phenomena and associated emerging devices, and accelerate research advances with the potential to spark the next big quantum leap, which has been envisioned to result in widespread use of technologies leveraging the principles of quantum mechanics. New perspectives will be gained on the future of computing, through advancing the frontiers of quantum information technologies, and addressing the pressing challenge of the semiconductor industry to find a "beyond CMOS" paradigm. Overall, this endeavor has a potential societal impact of the same magnitude as the development of the silicon transistor in the 20th century.

Besides quantum information and novel electronics research, these innovative and diverse capabilities will also offer transformative insights into the electronic structure of some quantum materials which are distinguished for their exceptional range of applications. Compelling examples are found in transition metal dichalcogenides, whose impact extends across the areas of environmental and biological analysis, catalysis, chemical and biomedical sensing [35]. Other good examples are numerous, such as subsets of correlated oxides that show promise for energy storage [36] and conversion [37]. More generally, the new tools will also contribute to the quest for novel quantum phases of matter and exotic electronic phenomena, which is at the foundation of fundamental research on quantum materials.

Further, the proposed capabilities will have considerable potential reaching beyond material science, to allow the study of a large body of samples found in environmental, geological, space, and bio-medical sciences. The world-class spatial resolution of Capabilities #1 and 3 will allow to interrogate catalysts under reaction conditions with respectively nanometer and sub-micron spatial resolution, and provide unparalleled chemical and mineralogical analysis of precious lunar, terrestrial, and interplanetary dust particle samples. The ultrabroad bandwidth offered by Capability #1 will facilitate bio-imaging at the unique multi-THz frequency range, where the photons are non-ionizing and considered safe for in vivo imaging applications [38], and for which the table-top systems do not yield high enough photon flux. The tender X-rays of Capability #3 will grant access to the K-edges of P, S, Cl, K, Ca, which are key elements in a range of disciplines, including energy materials and devices, biology for their functions in cells, and environmental and Earth sciences due to their fundamental role in plants, soil, and geological processes. The prospect of XAS and RIXS probes capable of studying these elements with sub-micron spatial resolution will undoubtedly appeal to these diverse user communities.

To summarize, the new toolbox proposed in this section promises to have a massive impact in quantum materials research, while also benefiting users across a broad range of communities, especially by offering spectro-imaging capabilities with cutting-edge spatial resolution, and advanced spectroscopic tools in the poorly explored tender X-ray range.

***References*:**

[1] L. Savary and L. Balents, *Quantum spin liquids: a review*. Rep. Prog. Phys. 80, 016502 (2017).

[2] T.-H. Han *et al.*, *Fractionalized excitations in the spin liquid state of a kagome-lattice antiferromagnet.* Nature 492, 406 (2012).

[3] A. Kitaev, *Anyons in an exactly solved model and beyond.* Ann. Phys. 321, 2 (2006).

[4] Y. Kasahara *et al.*, *Majorana quantization and half-integer thermal quantum Hall effect in a Kitaev spin liquid*. Nature 559, 227 (2018).

[5] H. Takagi *et al.*, *Concept and realization of Kitaev quantum spin liquids*, Nature Rev. Phys., 1, 264 (2019).

[6] H. Bernien et al., *Heralded entanglement between solid-state qubits separated by three metres*. Nature 497, 86 (2013).

[7] B. C. Rose et al., *Observation of an environmentally insensitive solid-state spin defect in diamond*. Science 361, 60 (2018).

[8] B. Trauzettel et al., *Spin qubits in graphene quantum dots*. Nature Phys. 3, 192 (2007).

[9] M.D. Pickett *et al.*, *A scalable neuristor built with Mott memristors*. Nat. Mater. 12, 114–117 (2012)

[10] L. Chua, *Memristor-The missing circuit element*. IEEE Transactions on Circuit Theory 18, 507 (1971)

[11] D.B. Strukov *et al*., *The missing memristor found*. Nature 453, 80 (2008)

[12] J.L. Andrews *et al*., *Building Brain-Inspired Logic Circuits from Dynamically Switchable Transition-Metal Oxides*. Trends in Chemistry, November 1, 711 (2019).

[13] J.L. Collins *et al*., *Electric-field-tuned topological phase transition in ultrathin Na3Bi*. Nature 564, 390 (2018)

[14] S. Maniputrani *et al*., *Scalable energy-efficient magnetoelectric spin–orbit logic*. Nature 565, 35 (2019)

[15] L. Smejkal *et al.*, *Topological antiferromagnetic spintronics*. Nature Phys. 14, 242 (2018)
[16] Q.H. Wang *et al.*, *Electronics and optoelectronics of two-dimensional transition metal dichalcogenides*. Nature Nanotech. 7, 699 (2012)
[17] M. Sui *et al.*, *Gate-tunable topological valley transport in bilayer graphene*. Nature Phys. 11, 1027 (2015)
[18] H. Yu *et al., Moiré excitons: From programmable quantum emitter arrays to spin-orbit-coupled artificial lattices. Sci*. Adv. 3, e1701696 (2017)
[19] Y. Cao *et al*., *Unconventional superconductivity in magic-angle graphene superlattices*. Nature 556, 43 (2018)
[20] S.A. Vitale *et al*., *Valleytronics: Opportunities, Challenges, and Paths Forward*. Small 14, 1801483 (2018)
[21] M. Koperski *et al*., *Single photon emitters in exfoliated WSe2 structures*. Nature Nanotech. 10, 503 (2015)
[22] G.X. Ni *et al*., *Fundamental limits to graphene plasmonics*. Nature 557, 530 (2018)
[23] J.C.W. Song *et al*., *Chiral plasmons without magnetic field*. PNAS 113, 4658 (2016)
[24] T. Karzig *et al*., *Topological polaritons*. Phys. Rev. X 5, 031001 (2015)
[25] S. Parkin *et al.*, *Magnetic domain-wall racetrack memory*. Science 320, 190–194 (2008)
[26] D.A. Allwood *et al.*, *Magnetic domain-wall logic*. Science 309, 1688–1692 (2005)
[27] K.A. Omari *et al*., *Toward Chirality-Encoded Domain Wall Logic*. Adv. Funct. Mater. 29, 1807282 (2019)
[28] G. Csaba *et al*., *Behavior of Nanomagnet Logic in the Presence of Thermal Noise*. Proceedings of the International Workshop on Computational Electronics, 1–4 (2010)
[29] F. Cairo *et al*., *Domain Wall Interconnections for NML*. IEEE Transactions on Very Large Scale Integration (VLSI) Systems 25, 3067 (2017)
[30] [25] G.B. Halász *et al.*, *Resonant Inelastic X-Ray Scattering Response of the Kitaev Honeycomb Model*. Phys. Rev. Lett. 117, 127203 (2016)
[31] J. Hill *et al*., *Future Trends in Synchrotron Science at NSLS-II*. J. Phys. Cond. Matt 32, 374008 (2020).
[32] S.-W. Cheong *et al.*, *Seeing is believing: visualization of antiferromagnetic domains*. NPJ Quantum Mater. 5, 3 (2020).
[33] E. Kotta *et al.*, *Spectromicroscopic measurement of surface and bulk band structure interplay in a disordered topological insulator*, Nature Phys. 16, 285 (2020)
[34] C. Kastl *et al.*, *Effects of Defects on Band Structure and Excitons in WS2 Revealed by Nanoscale Photoemission Spectroscopy*, ACS Nano 13, 1284 (2019)
[35] B.L. Li *et al.*, *Low-Dimensional Transition Metal Dichalcogenide Nanostructures Based Sensors*. Adv. Funct. Mater. 26, 7034 (2016)
[36] Y. Sun *et al.*, *Strongly correlated perovskite lithium ion shuttles*. PNAS 115, 9672 (2018)
[37] S. Lee *et al.*, *Strong Visible-Light Absorption and Hot-Carrier Injection in $TiO_2/SrRuO_3$ Heterostructures*. Adv. Ener. Mater. 3, 1084 (2013)
[38] P. H. Siegel, *Terahertz Technology in Biology and Medicine*, IEEE Transactions on Microwave Theory and Techniques (2004)

Soft Matter

The term *soft matter* encompasses a wide range of different systems. The main classes are liquid crystals, surfactants, colloids, and polymers [https://doi.org/10.3389/fphy.2018.00087]. *Liquid crystals* (LCs) are anisotropic shaped organic molecules, often with chiral, polar, or reactive groups that determine how the molecules self-assemble into positionally or orientationally ordered structures and how they react to applied stimuli. The LC units can also be molecular aggregates as in chromonic LCs. *Surfactants* are molecules with end-groups exhibiting different functionality (for example, hydrophilic and hydrophobic) that reduce the surface tension at interfaces and thereby result in stable systems of otherwise incompatible materials such as oil and water. The compatibilized systems can be isotropic emulsions or lyotropic liquid crystals in which changes in concentration drive phase transitions. *Colloids* are composed of nano- or micron-sized particles coated by organic ligands that solubilize the particles in a media (aqueous, organic solvent, liquid crystal). The constituent particles can be metallic, semiconducting, insulating, quantum dot, magnetic, with a uniform or anisotropic ligand coating that mediates their interparticle interactions. Recent progress has involved using DNA ligands with chemical recognition tuning the interparticle interactions. The colloidal particle composites can self-assemble into ordered structures. The fourth and probably largest class of soft matter systems are *polymers*. Polymers are linear or branched chains of monomer units that can be amorphous or semi-crystalline with conducting, semi-conducting, or insulating properties. The polymer chains can also incorporate reactive groups that can cross-link the chains into a network structure that exhibits a rigid or elastic structure that is thermally reversible or irreversible. The polymer constituents can also include liquid crystalline units that impart anisotropic and stimuli responsive properties. An interesting polymer construct are diblock copolymers comprised of two linked dissimilar polymers that self-avoid forming structures similar to those formed by surfactant systems. As a function of temperature or concentration, soft matter systems can be fluid-like in mechanical response or at lower temperature or higher concentration can exhibit a solid-like or gel state. Soft matter systems can be produced synthetically or are naturally occurring. Their fluid nature often requires confinement by solid substrates or channels, so the properties of the confining surfaces play a crucial role in some applications.

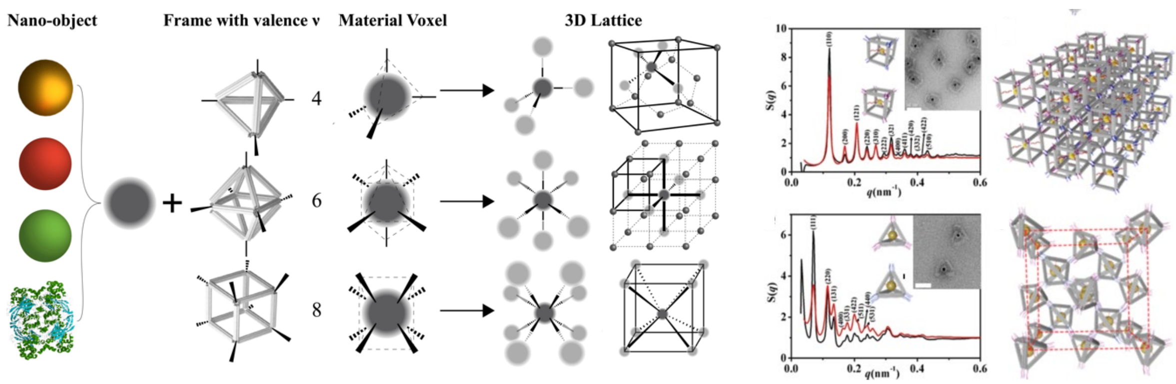


Figure 1. Adapted from (Y. Tian, et al, Nature Materials, (. https://doi.org/10.1038/s41563-019-0550-x. 2020) *Left*: Schematic of the DNA material voxels platform for assembly of 3D lattices from inorganic (nanoparticles) and bioorganic (proteins) nano-objects with DNA frames (~ 30-50 nm). Tetrahedra, octahedra and cubic have correspondingly valence v=4, 6 and 8, and their respective edge-lengths in this study are about 36, 29, and 29 nm. *Right*: Assembly of material voxels with 10 nm nanoparticles from DNA frames of different geometries (cube and tetrahedra) into BCC and cubic diamond superlattices. SAXS pattern and corresponding lattice models are shown.

Soft matter systems exhibit special properties that distinguish them from the solid state [https://doi.org/10.1103 /RevModPhys.89.025002]. Because the energy landscape between different soft matter phases is relatively flat, soft matter is commonly disordered with polydispersity prevalent and properties often governed by defects; it can be dissipative with multiple microscopic energy ‘sinks’; often out-of-equilibrium; heterogeneous with large fluctuations; sensitive to perturbations exhibiting a non-linear response such as shear thinning or thickening; self-assembles into structures that can be reconfigured by interactions of order kT; and forms hierarchical assemblies with a structure and collective dynamics that span length scales from Angstroms to mm and time scales from sub-microseconds to hundreds of seconds. The special features of soft matter open the opportunity to synthesize soft matter systems that are designed to optimally address specific needs: soft matter has the potential to be reconfigurable using minimal energy, be self-healing, be manufactured by low-temperature, energy efficient, and green-processes; and be biocompatible and recyclable. A number of new disruptive modes of manufacturing are under development that utilize the inherent self-assembly of soft matter to enable processing into high-performance films, fibers, and 3D printed components.

Another important research direction involves soft matter at complex heterogeneous liquid interfaces. For example, the replacement of mixtures of chemically synthesized surfactants in food, pharmaceutical, and cosmetics formulations by bio-surfactants depends upon a complex balance of interfacial interactions that are poorly understood. Liquid interfaces are used to model the complex heterogeneity of biochemical interfaces that underlie protein-lipid interactions and cell signaling, which are important for the development of functional biomaterials. In addition, dynamical processes on the molecular scale at liquid interfaces are critical to understanding ion transfer processes, such as solvent extraction, which are used to separate and purify technologically important rare earth elements and to clean up toxic and radioactive metals.

We grouped the most significant challenges for soft matter research and development in the next decade into five areas that were derived from the on-going research activities of our existing and anticipated user communities and based on input in the form of workshop presentations, sound-bites, or one-pagers. These areas best illustrate the research advances that will be enabled by the proposed enhancements to *existing* beamlines or *new* soft matter beamlines or endstations.

Science Questions/Challenges

A. Additive manufacturing from molecular deposition to 3d printing. Additive manufacturing (AM) is a disruptive manufacturing approach to take materials, including soft matter, from concept to product or device. The general challenges in AM stem from the processing of materials under far out-of-equilibrium conditions, from which the final material properties (structural/chemical/…) emerge and where the locked-in structure does not necessarily resemble an equilibrium state. AM of polymeric materials include polymers and polymer nanocomposites, reactive ink systems (e.g. printable ceramics), metal oxides and liquid crystal elastomers for cell scaffolds (responsive to external stimuli for cell growth) [http://dx.doi.org/10.1016/j.cad.2015.04.001]. Currently, material and process development are heavily relying on empirical knowledge and trial and error approaches, rendering the development cycle costly and lengthy. Shifting the exploration of materials and processing phase space from 'real world' to computational simulations would constitute a real paradigm shift. This requires an understanding of the materials' physics under processing conditions and at the relevant time and length scales, so computational models spanning from polymer to nanofiller to filament length scales can be built and verified against experiments. Challenges for this approach arise from the spatial and dynamic heterogeneities inherent to AM processes, time scales ranging from sub-ms dynamics (filler dynamics during extrusion) to hundreds of seconds (stress relaxation in extruded filaments) and length scales ranging from nanometers (polymer length scales) to hundreds of nanometers (filler particles) to hundreds of micrometers (filament sizes). Examples of specific scientific questions in this include: What is the connection between the processing conditions and the chemical structure of a complex material, its nano- to macroscale morphology and macroscopic properties (mechanical, optical, electrical, etc.)? How do these connections change when upscaling the processing conditions to industrial size AM like big area additive manufacturing (BAAM)? How to distinguish between different kinetic growth models for layer-by-layer deposition techniques?

B. Advanced materials synthesis and characterization at liquid interfaces. Liquid interface scattering is our newest soft matter NSLS II capability. Over the past several decades, x-ray scattering studies from liquid interfaces have led to key discoveries of polymorphic structures of two-dimensional molecular, biomolecular and nanoparticle films, surface ordering of organic and metallic liquids including surface freezing and surface alloying in liquid metals, the role of thermal fluctuations at soft interfaces and the distribution of ions at the charged and electrified interfaces. Such studies provide information that is unobtainable by any other technique and have helped address the challenge to develop a fundamental understanding of the unique phenomena found at liquid interfaces.

An emerging scientific challenge at liquid/vapor and liquid/liquid interfaces is to use these interfaces as a substrate to grow novel 2D polymer and inorganic materials. Since liquid surfaces are locally atomically smooth – no atomic steps - the possibility exists to synthesize materials to a perfection not possible on solid supports. For instance, the potential exists to fabricate large graphene sheets on liquid metal surfaces by CVD processes, yet challenges remain since adequate in-situ structural monitoring is required in order to control these processes [Guo, Wang, et al. "Rapid chemical vapor deposition of graphene on liquid copper." *Synthetic Metals* 216 (2016): 93-97]. In addition to graphene, liquid-liquid interfaces offer the possibility of making well-defined metal and semiconductor nanoparticles as well as very thin sheets of crystalline metals, some of which are relevant to catalytic processes [Scanlon, Micheál D., et al. "Gold Nanofilms at Liquid–Liquid Interfaces: An Emerging Platform for Redox Electrocatalysis, Nanoplasmonic Sensors, and Electrovariable Optics." *Chemical Reviews* 118.7 (2018): 3722-3751]. Opportunities also exist to explore 2D polymer films [Dallas, Panagiotis, and Vasilios Georgakilas. "Interfacial polymerization of conductive polymers: generation of polymeric nanostructures in a 2-D space." *Advances in Colloid and Interface Science* 224 (2015): 46-61]. In-situ structural studies during interfacial polymerization, will provide invaluable insight into how to make improved materials.

C. Frontiers of device science: nano-lithography, flex hybrid & organic devices. Soft matter is becoming increasing important both in the processing of conventional semiconductors as well as in organic electronics.

Electronic device technology is undergoing significant and continuous diversification, with different measurement needs depending on the technology and its end market. Conventional semiconductor electronics is challenged by limits to Moore's Law scaling as devices approach single-digit nanometer sizes. Challenges in this space include the measurement of process deviations and the detection of sparse defects at such small length scales. Future microelectronics development will require new nanolithography technologies based on soft matter, including Directed Self Assembly (DSA) [S-J Jeong, JY Kim, BH Kim, H-S Moon, SO Kim, "Directed self-assembly of block copolymers for next generation nanolithography." Materials Today 2013, 16, 468] and Extreme UltraViolet (EUV) lithography [Levinson, Harry J., and Timothy A. Brunner. "Current Challenges and Opportunities for EUV Lithography," in *International Conference on Extreme Ultraviolet Lithography 2018*, 10809:1080903. International Society for Optics and Photonics, 2018]. These new technologies require new metrology methods to measure chemical changes in soft material systems on the nanometer scale.

In flexible electronics [WS Wong, A Salleo, "Flexible Electronics: Materials and Applications." Flexible Electronics: Materials and Applications, Springer Science & Business Media, 2009], including hybrid and organic electronics devices, the pattern length scales are significantly larger and challenges stem from controlling the structure of material that is produced from printing and drying processes. In-situ methods to study the processing of flexible electronics formulations are required for science-based formulation and process development. In addition, it is necessary to assess several aspects of structure within the printed layers, such as long-range order and crystallinity, orientation in amorphous and liquid crystalline regions, the size, shape, and composition of nanoscale domains. Challenges revolve around making printing techniques (such as screen printing, ink jet printing, aerosol jet printing and others) more reliable, and to understand the nature of defects (cracks, voids, interfaces) impeding performance of the devices.

D. A new generation of soft matter: responsive, active, nanocomposites, nano-architectures. Designing the next generation of soft materials presents a number of research challenges. The first challenge is to understand at the molecular level how changes in molecular shape and the inclusion of chiral, polar, or reconfigurable entities impacts self-assembled macroscopic structure and function. In the case of colloidal nanoparticle architectures, the challenge is similar: to understand how nanoparticle type, shape, ligand coating and interactions can control the resultant self-assembled structure and function [https://doi:10.1038/natrevmats.2016.8, https://doi.org/10.1557/mrs.2017 .280]. The goal is to encode molecular information during the fabrication process to produce stimuli responsive materials that respond in a predictable way to light, temperature, humidity, chemical, electric or magnetic fields. The response can be a mechanical extension/contraction as occurs with liquid crystal elastomers, electronic as occurs in organic transistors or organic light emitting diodes, or orientational as occurs in liquid crystal displays. Blending nanoparticles into soft matter systems is another approach being taken to enhance soft material functionality or processability. Another interesting direction in materials design is to incorporate entities that are energy consuming/releasing that function as molecular motors (active or 'living' matter).

Equally challenging is to model the non-equilibrium, collective dynamics of these responsive, active, nanocomposite, nano-architecture systems under the relevant applied stimuli. Active matter exhibits unusual collective dynamical effects such as 'swarming', while the heterogeneity of the nanocomposite and nano-architecture systems leads to complex heterogeneous dynamics. Kinetic trapping in intermediate non-equilibrium states is a common occurrence and can be exploited to obtain a particular structure with desired properties.

E. Optimizing polymer film and fiber properties and recyclability. A primary challenge in polymer science and engineering is to design polymer-based materials with novel or enhanced properties (e.g., engineering or high-performance plastics). A key question is: how can we create polymeric materials that combine a traditionally incompatible or disparate set of properties? Examples of such materials under active investigation include: recyclable polymers with both the shape performance of thermosets and the easy remodelability of thermoplastics; biocompatible, stretchable, and tough hydrogels for applications in tissue engineering; stacked two-dimensional polymers and covalent organic frameworks for high-strength films and coatings; self-healing, high-dielectric layers for high-temperature and high-voltage polymer-film capacitors. Common to all these systems is the formation of hierarchical structure which exhibit different degrees of positional, orientational, and morphological order over multiple length scales. These structures in turn originate from an intricate balance between different types of intra-, inter-, and supramolecular associations, including covalent, ionic, H-bonding, hydrophobic, and van der Waals interactions. Fundamental understanding of how the hierarchical order emerges from these complex interactions and how its response to external stimuli and perturbations gives rise to target properties, or undesirable failure, is essential to addressing this challenge.

Another major challenge for polymer research is to understand processing-structure-property relationships. Be it industry-scale manufacturing or lab-scale fabrication, the formation of polymer-based materials relies on a variety of processing techniques (e.g., injection molding, extrusion, film blowing, fiber spinning, roll-to-roll or slot-die coating, solution-casting, and self-assembly) as well as post-processing (e.g., thermal or solvent-vapor annealing). Mechanistic understanding of processing-structure-property relations is critical to controlling and optimizing processing conditions toward specific target properties. This is particularly demanding for polymeric materials because the material and process parameter space of relevance is enormous and because their structure and properties are highly dependent on process history, with the entanglements and restricted mobility of polymer chains contributing to slow kinetics. Thus, addressing this challenge requires intelligent parameter-space exploration based on *in-situ* characterization of structural evolution kinetics and constituent dynamics *during material processing*. Of particular interest to industry is a fundamental understanding of polymeric material behavior under extreme conditions encountered in manufacturing processes, such as ultrahigh shear and tensile strain rates, and sudden thermal quenches.

Priority Research Directions

A. Additive manufacturing from molecular deposition to 3d printing.

The priorities in the area of additive and advanced manufacturing are in-situ/operando experiments of materials under real manufacturing conditions [https://doi.org/10.1021/acs.macromol.9b01620, https://doi.org/10.1021/acslangmuir 9b00766, https://doi.org/10.1021/acsami.9b12908] and post-processing characterization of AM manufactured parts.

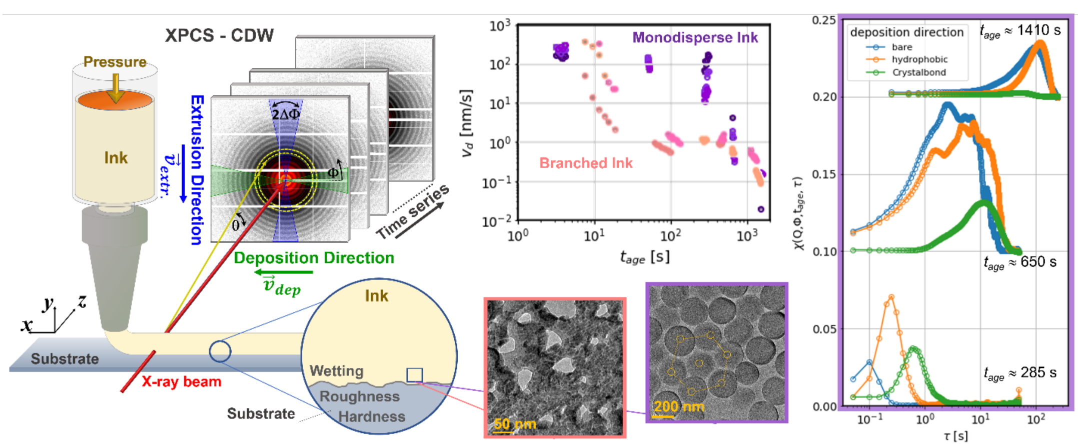


Figure 2. From [M. Torres Arango *et al.*, *Materials Today Physics* (2020) https://doi.org/10.1016/j.mtphys.2020.100220]. *Left*: Schematic of the XPCS operando experiment during filament deposition. *Center, top*: Displacement velocity of the colloidal particles during structural relaxation. Ink formulations used two type of colloids, monodisperse 250 nm Silica particles ("monodisperse ink") or aggregates of ~7nm Silica particles ('branched ink'). The slowing down of the structural relaxation with "age" (the time after initial filament deposition) is associated with a jamming process evidenced by the XPCS experiments. *Center, bottom*: Ex-situ electron microscopy showing the material structure at the interface with the printing substrate for the 'branched ink' (*left*) and 'monodisperse ink' (*right*); *Right:* The normalized variance of the correlation times measured by XPCS provides a direct measurement of dynamic heterogeneities which might be related to the formation of meso-scale structural defects. Occurrence and magnitude of these heterogeneities are related both to the bulk structure of the ink and its interaction with the printing substrate.

Soft Matter

1). The *in-situ/operando experiments* require scattering techniques such as (coherent) SAXS/WAXS with 'simultaneous' multimode capabilities, i.e. collecting as much of the scattering simultaneously. In particular, this calls for WAXS detectors at SMI and CHX that can collect scattering at high Q with a 270° coverage while passing the beam for simultaneous (coherent) SAXS. On the instrumentation side, in-situ/operando experiments require customized equipment such as 3D printing platforms, which are fully integrated with the beamline controls and analysis [https://doi.org/10.1080/08940886.2019. 1582285], enabling autonomous experiments. Autonomous data analysis is required for large (coherent) scattering datasets in order to efficiently process the high dimensional data and provide timely feedback to the experiments. Non x-ray techniques, such as Raman, need to be integrated with the x-ray experiments to obtain complementary information and to develop proxy techniques for the factory production floor. For big area additive manufacturing (BAAM), as well as for in-situ material growth with layer by layer deposition techniques, a state-of-the-art scattering beamline capable of handling much larger sample environments than the current beamlines is essential.

2). *Post-processing characterization capabilities* that are needed include STXM with chemical, magnetic, orientational information on the length scales relevant to polymer nano-composites. The obtained information can be linked to the scattering data that can be collected throughout the entire processing cycle. Instrumentation such as tensile testers and rheology integrated with x-ray scattering beamlines are needed to close the loop between processing and final properties of manufactured parts.

B. Advanced materials synthesis and characterization at liquid interfaces.

1). *Liquid interfaces presents an ideal platform for the synthesis of 2D soft materials* including inorganic 2D materials, metal-organic frameworks (MOFs), nanoparticle (NP) superlattices, and monolayers or thin films of polymers [Dong, Renhao, Tao Zhang, and Xinliang Feng. "Interface-assisted synthesis of 2D materials: trend and challenges." *Chemical Reviews* 118.13 (2018): 6189-6235], due to the ease of 2D motion and benefit of interfacial liquid species transport. These materials are flexible, free-standing, tunable with distinct electronic, optical, mechanical and catalytic functions. Current challenges lie in poor understanding of fundamental interfacial processes and mechanisms, such as precursor organization, nucleation, mineralization and polymerization. Establishing liquid interface-based synthesis is highly advantageous since it may overcome limitations in the material growth for targeted structures and properties. New in-situ approaches are needed to monitor and quantify the role of the liquid surfaces in the synthesis of the 2D soft materials.

2). *While polymer crystallization has been extensively studied, crystallization at liquid/liquid interface has rarely been investigated due to the complexity of the system*. However, controlling polymer crystallization at flat liquid/liquid or liquid/air interface could lead to asymmetrical membranes with tunable structure, morphology, and porosity [Fahrenkrug, Eli, and Stephen Maldonado. "Electrochemical Liquid–Liquid–Solid (ec-LLS) Crystal Growth: A Low-Temperature Strategy for Covalent Semiconductor Crystal Growth." *Accounts of Chemical Research* 48.7 (2015): 1881-1890]. One fundamental question raised in this research direction is how the chemistry, viscosity, surface energy and curvature of the interface collectively affect polymer phase behavior and crystallization.

3). *Room temperature semiconductor electrodeposition* provides an alternative to standard high temperature vacuum or gas phase deposition. Electrodeposition on a liquid metal substrate provides a potentially powerful alternative to solid supports. As an example, the electrodeposition of Ge at a liquid Hg/aqueous interface appears to be influenced by the presence of species from the electrolyte adsorbed on Hg prior to and perhaps during the electrodeposition stage [Staub, Mark C., and Christopher Y. Li. "Confined and directed polymer crystallization at curved liquid/liquid interface." *Macromolecular Chemistry and Physics* 219.3 (2018): 1700455]. In other systems, anecdotal observations further suggest that Pourbaix and bulk phase diagrams are insufficient to describe the resultant materials formed. Microscopic understanding will not only lead to better resultant materials but should also inform our basic understanding of how crystals nucleate and grow.

C. Frontiers of device science: nano-lithography, flex hybrid & organic devices

1). *Approaches to characterize the shape and size of the latent images produced by hot photoelectrons in exposed, patterned EUV photoresists.* These photoresists are composed of primarily organic compounds. Initially, approaches that work on Bragg structures should be sufficient, but eventually non-Bragg structures will require measurement with high (~5 nm or less) spatial resolution. Approaches that include in-situ EUV exposure, with calibrated EUV exposure tools, and available EUV lithography capabilities at a light source may be required to accelerate EUV photoresist material development.

2). *In-situ tools for studying solidification of printed electronics materials.* Such tools will provide fundamental information about the solidification mechanisms, delivering direct insight into the effects of formulation and process choices, and opening the way for science-based formulation and process design.

3). *Ex-situ tools for studying defects and interfaces in printed electronics materials*. Performance in these devices is typically held back by unanticipated electronic effects from defects or interfaces. Measurements to characterize the composition and structural distribution of printed solids are required to understand their heterogeneous nature and how these aspects of structure affect electronic properties. Interface measurements of composition, orientation/order, and electronic quantities such as work function and valence orbital structure are required to tailor interfaces, particularly in devices where interfaces form semiconducting junctions.

D. New generation of soft matter: responsive, active, nanocomposites, nano-architectures

1). *Determining molecular structure and correlating it to macroscopic texture.* Ordering at the molecular level produces characteristic defects [ISBN:9783527307258] (e.g. bond order resulting in a star-disclination defect in hexatic LCs) or characteristic morphologies or textures (e.g. the helical packing of the B4 LC phase producing a variety of helical filamentary textures [https://doi:10.1039/c9mh00089e]). Understanding textures is important since they provide visual cues to underlying order, especially the existence of novel types of order, and are often detrimental to device performance as the chevron-defect in ferroelectric LC displays. Understanding the correlation between structure and characteristic defects and morphologies requires SAXS and WAXS covering a wide q-range, *at low- and high-q*, to measure positional order and changes under stimuli; polarized resonant x-ray scattering (soft, tender, hard) is needed to achieve elemental specificity and determine nanoscale orientational periodicities; micron-sized beams are needed to probe spatial inhomogeneities and defects. Since the resultant macroscopic textures have features from nm to mm in size, high-resolution optical and x-ray microscopies are needed, simultaneously when feasible.

2). *Measuring non-equilibrium dynamics*. Since some soft matter systems do not exhibit long or quasi-long range order, tracking the dynamics requires use of x-ray photon correlation spectroscopy (XPCS) and tracer additives [https://doi.org/10.1146/annurev-matsci-070317-124334]. The widespread use of nanoparticle additives in products (health-care, foods, inks, adhesives), makes XPCS amenable to industrial studies since tracer particles are inherent to the products. For soft matter systems that exhibit positional (orientational) order, time-resolved (resonant) SAXS/WAXS with fast detectors can be applied to study dynamics in-situ under applied stimuli. The GISAXS(WAXS) geometry provides comparable information on surfaces and interfaces. For small molecule LC systems, neat or with nanoparticle additives, the dynamics can be sub-microseconds requiring a new generation of detectors. Equally important is the engagement of theorists to model the complex non-equilibrium dynamics for predictive control of processes.

3). *Efficient synthesis of nanoparticles with desired structural properties* can be achieved by combining micro-fluidic synthetic approaches *in-line* with real-time x-ray structural feedback and response feedback to guide optimization of the synthesis route. The next level of complexity is using liquid robotics to fabricate different nanoparticles into 3D architectures using similar in-line feedback approaches guided by artificial intelligence.

E. Optimizing polymer film and fiber properties and recyclability

Polymeric materials, including polymer (nano)composites, exhibit multi-scale, hierarchical structure that depend on a multitude of material and process parameters in a highly history-dependent manner. The nature of the hierarchical structure and its response or susceptibility to external perturbations give rise to the desirable properties and performance of these materials as functional components in practical applications. The following has been identified as priority research directions for polymeric materials:

1). *In-situ, time-resolved characterization of ordering kinetics and constituent dynamics under out-of-equilibrium conditions*, including extreme conditions relevant to industrial processes and product testing (e.g., shear/stretching at ultrahigh strain rates, temperature quenches, …).
2). *Effects of processing conditions and pathways on the evolution of multiscale structural features*: (semi)-crystallinity, anisotropy in molecular and domain/grain orientations, phase coexistence/ polymorphism, temporal heterogeneity of constituent dynamics, spatial heterogeneity (e.g., arising from high gradients).
3). *Effects of interfaces and confinement* on the nucleation and growth, molecular and morphological orientation
4). *Ancillary characterization of non-structural properties* accompanying in-situ, time-resolved x-ray measurements (e.g., stress-strain and rheology measurements during mechanical deformation; calorimetry during thermal processing; …)
5). *Development of online processing instrumentation with beamline-integrated controls* that can achieve manufacturing-relevant processing or product-testing conditions
6). *Development of machine-guided material processing and characterization methods* tod facilitate intelligent exploration of material-process parameter spaces.

F. Cross-cutting AI/ML approaches

Development of AI/ML-enhanced online experimentation capabilities that encompass material synthesis/processing, x-ray and ancillary characterizations, and real-time data analysis at beamlines, provides an opportunity to greatly expand our ability to explore soft materials' vast parameter spaces and understand their complex constituent-processing-structure-property relations. The priority research directions for AI/ML approaches are:

1). *Further refinement and deployment of autonomous decision algorithms to guide material synthesis/processing and structure/property measurements at beamlines*. Recent demonstrations of autonomous experiments at the CMS and SMI beamlines exploited "physics-agnostic" decision algorithms that are independent of experimental techniques used [M. Noack et al., Sci. Rep. 9, 11809 (2019)]. Despite the advantage of broad applicability, the physics-agnostic algorithms ignore existing knowledge about the material system or phenomenon under study, which the human experimenter exploits in traditional experimental planning. Thus, an important future direction is to develop "physics-informed" methods to incorporate system-specific expert knowledge into the autonomous decision process, without sacrificing the generality of the decision-making platform. The prior expert knowledge that could guide beamline experiments includes: theoretical or simulation-based models; materials database; the results of complementary measurements (e.g., in a multimodal setting). Ideally, the new algorithms to be developed should be designed to enhance data-theory interactions such that theoretical models can be improved iteratively during autonomous experiments at beamlines.

2). *Building up an arsenal of AI/ML methods for online data analysis at beamlines*, including instant data classification, data feature recognition, and analysis pipelines that streamline the extraction of physical quantities from x-ray data. The need for the latter is particularly acute for XPCS measurements of out-of-equilibrium dynamics in heterogeneous materials under processing conditions. AI/ML-based analysis methods are expected to significantly enhance beamline productivity by enabling adaptive experimental planning during limited beam time, as well as facilitate real-time data analysis needed as part of the autonomous experimentation platform.

3). *Further development of common online instrumentations for in-situ material synthesis and processing that are fully integrated into beamline controls.* Examples that are already in place and available to general users include: a prototype 3D printer platform and a photothermal annealer. Future possibilities, either under development or consideration, include: a heavy-duty 3D printer platform, a blade or slot-die coater, a microfluidic solution mixer, and heavy-duty tensile and shear stages. The integration of these instruments into beamline controls is a prerequisite to their incorporation into the autonomous experimentation platform.

New NSLS-II Capabilities

Existing beamline instrumental and software enhancements: A number of enhancements were identified in the 2020 NSLS-II Strategic Plan (pages 22-25) and endorsed by workshop attendees. These included: (CHX) a wide band-pass multilayer monochromator, sub-microsecond area detector (VIPIC or similar), a WAXS detector with 270 deg. coverage autonomous, AI/ML enhanced XPCS analysis software (possibly within XiCAM), machine-learning methods for efficient data reduction; (CMS) refined autonomous experimentation software, open floor-to-ceiling experimentation space; (IXS) enhanced flux and counting efficiency; (SMI) enhanced WAXS detector with 3x wider horizontal collection, polarization flipper, and a polymer processing and liquids scattering (PPLS) endstation.

At the workshop, several additional enhancements were identified: in-line complementary high-resolution optical microscopies (polarized, conoscopic) and spectroscopies (fluorescence, raman, UV); in-line rheometer; in-line microfluidic and robotic synthesis platforms; L-shaped WAXS detector at CHX for simultaneous SAXS XPCS and incoherent WAXS; a USAXS capability; a pulsed Diamond Anvil Cell for studies of transient states; and a wider variety of complex in-situ environmental chambers to probe process-structure relationships, failure modes, and aging.

Programmatic and infrastructure enhancements: It was recognized that beam time would soon become critically oversubscribed, which could be partially mitigated by more efficient data acquisition and analysis software, a mail-in program, remote access, more effective block allocation groups and lower-threshold partner user programs. Such programs could also provide more timely access required by industrial users. However, additional staffing is essential for implementing all these programs and, even if implemented and well-staffed, NSLS-II would still be missing beamline capabilities and capacity for soft matter research within the existing suite of beamlines. For example, the single SMI beamline will be shared for studies requiring microbeam SAXS/WAXS, time-resolved SAXS/WAXS, resonant SAXS/WAXS, liquid scattering, and open-platform polymer processing – an unsustainable path-forward. It was also recognized that the volume of data being generated exceeds capabilities of conventional data transfer systems, modeling and high performance computing approaches are often necessary for the interpretation of measurements on complex soft matter systems, better outreach is needed to industrial users and emerging research communities, and current hutch space is often too restrictive to accommodate industrial processing equipment for in-situ studies.

Essential new soft matter beamlines: Workshop participants identified the following new soft matter beamlines as essential tools for researchers in the field.

Independent Processing and Liquids Scattering (PLS) beamline -- Previous NSLS-II scientific justifications for liquid interface studies focused on model systems to investigate the statics and dynamics of self-assembly, interfacial orientation and forces, molecular interactions, molecular recognition, and chemical reactivity as was the focus of the successful SMI proposal in 2011 packaged as part of the NEXT beamlines. Here a liquids instrument was to be constructed along with the grazing WAXS/SAXS instrument. However, due to insufficient project funding the liquids endstation at SMI was not pursued. In 2016, the scientific case for liquids at NSLS-II was updated in a proposal to construct an independent processing and liquids scattering (PLS) beam line using a canted undulator at SMI that utilized much of the existing infrastructure. Although a fully independent, canted undulator was never constructed, as a consequence of the PLS effort the Polymer Processing and Liquids Scattering (PPLS) endstation is now being constructed at SMI. It is expected to enter user operations in 2020. Of note, there is only one other synchrotron-based liquids scattering instrument in the US (APS), in contrast to at least 5 capable instruments in Europe.

An independent liquids instrument at NSLS-II is also envisioned to provide a platform for polymer processing, similar to the PPLS vision. Processing often involve liquids, requires an open platform that is not available at SMI's SAXS/WAXS endstation, and benefits from a horizontal scattering geometry, all of which are provided in a liquid scattering station. Micron-sized vertical beams are essential, currently not possible at the PPLS endstation with its present optical layout. CRLs with a secondary source aperture sufficiently upstream is essential to achieve the required focus. For liquid/liquid studies, 30 keV is advantageous and this requires a high-performance, nitrogen cooled undulator. Specialized liquid/liquid cells for electrochemistry, high temperature liquids, and polymer and NP synthesis are essential along with ancillary diagnostics. The PPLS crystal deflector will also be enhanced to a two-crystal deflector enabling the liquid surface to remain stationary during scans.

Advanced Manufacturing Processes (AMP) beamline -- X-ray scattering techniques such as (coherent) SAXS/WAXS are uniquely suited to obtain in-situ/operando information during AM processing on the materials' relevant time- and length scales. Time-resolved coherent scattering contains all the structural information of time-resolved incoherent SAXS/WAXS, plus the out-of-equilibrium and anisotropic dynamics, including dynamic heterogeneities from e.g. stress relaxation, information about strain in amorphous materials via speckle tracking and plastic/elastic deformation via speckle echo techniques and enhanced sensitivity to weak ordering/alignment in materials via angular cross-correlation. While (U)SAXS/WAXS provides access to materials length scales from Angstroms to micrometers, microbeams offer the necessary spatial resolution to map the spatial heterogeneities inherent to AM processes and deliver imaging-like capabilities for structure and dynamics under processing conditions. Pairing these state-of-the art scattering techniques with AM in-situ/operando capabilities requires significantly larger sample areas than available at current beamlines and a consolidation of techniques for true 'in-situ multimodality'. Therefore, the concept for a new 'Advanced Manufacturing Processes (AMP)' beamline has been proposed.

AMP would combine the capabilities of (coherent) (GI)(U)SAXS/WAXS with microbeams and a large dual sample stage. One sample stage would be optimized for additive manufacturing, including the capabilities to host and operate large scale extruders used in BAAM and deposit materials at speeds up to ~1m/s. The second sample stage would be optimized for positioning of large growth chambers (up to 1.5x1.5x.8 m^3, 1 ton) with the capability for setup in 'ante room' outside of the experimental hutch and transfer of the operating chamber into the beam to eliminate unproductive setup time. The science enabled by this beamline include: in-situ/operando studies of AM processes including BAAM, combination of x-ray and complementary techniques in processing setups, combination of x-ray scattering techniques with industrial standard mechanical testing (e.g. tensile tester), accommodation of large growth chambers for studying advanced layer-by-layer deposition processes, as well as the study of aging/reliability of AM parts via automated/robotic measurements. Key design parameters for this beamline include an undulator beam (8-25 keV) tunable in focal size and degree of transverse coherence at the sample position without moving the beam position, micro-beam and ≥12 m sample-to-detector distance to resolve speckles and enable USAXS. A WAXS detector with 270° coverage will enable simultaneous time-resolved SAXS/WAXS. The available sample space along the beam direction would be between 1.5 and 0.5 m, adjustable without breaking vacuum to minimize the amount of beamtime lost for configuration changes.

<u>*Advanced soft matter endstations:*</u>

Soft X-ray Scanning Transmission X-ray Microscopy for Soft Matter (SSS) – Many soft matter challenges require the measurement of chemical structure, molecular orientation, and morphology at the nanometer scale. Soft x-ray STXM can potentially address these needs. With contrast provided by near-edge x-ray absorption fine structure (NEXAFS) spectroscopic principles, soft x-ray STXM can provide elemental sensitivity by the core shell that is accessed, molecular orbital information by the spectral peaks that appear, and molecular orientation information by the sensitivity of the peak intensity to electric field vector. STXM delivers these capabilities in a nanoscale-resolved microscopy measurement using a focusing zone plate (~20 nm) and rastering approach to create a real-space hyperspectral image. NSLS-II has considerable capabilities in soft x-ray spectroscopy and scattering, but a soft-matter x-ray STXM is a gap in its portfolio.

Significant beamline capabilities are required to address the scientific challenges. STXM using tender x-rays in addition to soft x-rays, would extend the measurement to biologically-relevant elements such as Sulfur and Phosphorous and important semiconductors such as Silicon. Most measurement needs would benefit from polarization control that could be provided with an elliptically polarized undulator. The STXM should be capable of studying inorganic magnetic domains as well as organic materials. Various focusing zone plates with different resolutions, as well as a drop-in attenuator scheme, would provide a capability to balance concerns about sample damage with the resolution needs of a specific measurement. Ideally, some tomographic capability would be desired to address measurement needs in samples for which simple projection images result in an ambiguous result.

Several of the scientific challenges require advanced environmental capabilities within the STXM. Directed cell growth experiments using liquid crystal elastomers require sample temperature control and a capability to study mechanical/magnetic effects on sample. Other materials studies would require the ability to apply external stimuli such as mechanical strain, UV illumination, and exposure to liquids and gases. Broad classes of scientific challenges require the ability to handle and measure delicate biological samples, which may further require liquid lines into the vacuum chamber sample stage for in-situ liquid supply/replenishment and potentially to enable combinatorial or autonomous studies. Electrochemical capabilities would be important for sensors and energy materials. A key concern for many of these scientific challenges is the question of beam damage, which leads to a strong recommendation of cryogenic (liquid nitrogen temperatures) sample capability for STXM, in analogy to cryo-electron microscopy. Many of these environments could use sample environments and holders that are interoperable between STXM, transmission electron microscopy, and resonant soft or tender x-ray scattering.

Data analysis for conventional STXM is straightforward for simple samples, such as two component blends. However, in more complex system such as biological materials, it can be difficult to analyze the results. There are opportunities for advanced machine learning for data analysis, particularly in unsupervised spectral deconvolution, where contributing component spectra may be extracted from hyperspectral images without the need for reference spectra.

Soft x-ray STXM would be a powerful addition to NSLS-II capabilities, but even more scientific challenges could be addressed by achieving higher resolution than STXM can provide. A nascent extension of STXM, soft x-ray ptychography, may be able to provide similar hyperspectral imaging capabilities with resolutions ~10x finer than STXM. An investment in STXM at NSLS-II should keep in mind the potential ease of eventually adapting the instrument to ptychography or developing an adjacent instrument suitable for ptychography. Investments in vibration isolation might be important, as would an upgrade path to a highly coherent beam, and a chamber design that could someday accommodate an imaging detector.

Moderate-resolution X-ray transmission and scattering tomography – Several scientific challenges, such as printed electronics technologies and digital 3D nanomaterials, require spatially resolved structure information from complex solids. Moderate-resolution x-ray transmission and scattering tomography, in both transmission and grazing incidence geometries, and in both wide- and small-angle x-ray scattering modes, may address some of these challenges. These capabilities could potentially be built at existing NSLS-II beamlines such as CMS or SMI. Some implementations could use nano- and microbeams with flexible optics based on beryllium lens transfocators for variable resolution. A critical challenge in this measurement is data analysis; the light source should be prepared to provide significant user assistance in data management, algorithm code, and computational capabilities for tomographic reconstruction. Tomography is also highly amenable to automation and autonomous experimentation, so investments in robotics would be appropriate.

EUV photolithography – Several challenges in photolithography may be best addressed by having EUV exposure accessible at the NSLS-II, with calibrated EUV exposure tools, and available EUV lithography capabilities such as interference lithography. Given the enormous expense of a standalone industrial EUV source (roughly $120M), synchrotrons have a unique opportunity to address this need by harnessing EUV-rich wavelengths directly. Should an in-situ capability be contemplated, multiple beams would be required, and the combination would presumably be fixed. However, a beamline providing only EUV wavelengths could be used to perform EUV exposures in controlled ways to create samples that could be studied ex-situ on a variety of other instruments. Such a capability could prove quite popular in multi-beamline proposals as a first stop to create unique samples. It could also be harnessed to fabricate new x-ray optics such as zone plates.

Impact of the New Capabilities

A. Additive manufacturing from molecular deposition to 3d printing. Optimizing existing scattering beamlines to obtain as much information as possible simultaneously during in-situ/operando experiments of AM processing or materials under mechanical testing or other stimuli, in combination with auxiliary techniques (e.g. optical spectroscopy) will advance the understanding of the structure-processing-property relationships that are key to the future development of soft materials including AM. A state-of-the-art scattering beamline optimized for hosting large sample environments and providing space for auxiliary techniques would be key in closing the gap between fundamental science and industrial applications by enabling the study of materials under relevant processing conditions.

B. Advanced materials synthesis and characterization at liquid interfaces. Synthesis at liquid interfaces provides the possibility for materials to grow from species which can be transported from both sides of the interface that is not possible with interfacial films formed at solid supports. New liquid surface capabilities at NSLS II will provide a micron-sized beam, higher x-ray energies as well an improved liquids spectrometer with a fixed sample height geometry. Together, these capabilities will provide the necessary diagnostic tools required to investigate the in-situ synthesis of soft and hard materials at liquid interfaces which in turn will help enable the development of new metal, hybrid and polymer films which cannot be produced on solid supports.

C. Frontiers of device science: nano-lithography, flex hybrid & organic devices. For conventional semiconductor electronics, the proposed capabilities would result in accelerated development of EUV photoresists and exposure tools, higher yields in EUV processes, and an improved competitiveness of U.S. semiconductor industry in world markets, as U.S. companies are best positioned to benefit from success of EUV. For printed electronics, the capabilities would enable the move from printed circuit boards to light-weight flexible substrates, with applications in developing wearable electronics for medical and fitness monitoring as well as monitoring the stress state of first responders or military personnel. Science-based formulation and process design would permit such devices to move from research demonstrators to full industrial production.

D. New generation of soft matter: responsive, active, nanocomposites, nano-architectures. New capabilities will provide the characterization tools needed to achieve a better understanding of how molecular level changes impact material response and macroscopic function. This understanding will accelerate the development of mechanical stimuli-responsive materials with a far-reaching impact in soft robotics, optical instrumentation, smart surfaces, and medical devices. If prepared from biocompatible materials, mechanical stimuli-responsive materials can be used in scaffolds to mimic muscle activity and direct the growth of stem cells. Molecular level understanding will also impact the development of enhanced organic electronic and opto-electronic materials for applications in energy-efficient OPVs, OLEDs, and high-speed opto-electronic processors – alla capable of being fabricated on flexible substrates.

The new capabilities, which incorporate large open and flexible space for processing set-ups, will also enable the development of a new nanofabrication platforms with active structural and functional feedback for the preparation of optimized nanocomposite inks for 3D printing or the assembly of different nanoparticles or biomolecules into targeted nano-architectures that integrate biotic and abiotic materials or function as phononic or photonic metamaterials. These advances will improve competitiveness of U.S. nanofabrication methodologies.

E. Optimizing polymer film and fiber properties and recyclability. The capabilities consolidating coherent XPCS and incoherent (U)SAXS/WAXS, at the new AMP beamline will advance our understanding of dynamics, structural kinetics, and processing-structure-property relations in polymeric and polymer (nano)composite films and fibers by significantly enhancing *in-situ* time-resolved, multi-length scale, and multimodal measurements of heterogeneous, hierarchical structures under processing and material testing conditions. The open platform capabilities, envisioned for the AMP and PLS beamlines, are essential to accessing extreme far-from-equilibrium conditions relevant to industrial manufacturing processes as well as interrogating the scalability of lab-scale to industrial-scale fabrication processes. The PLS beamline's horizontal scattering geometry and micron-scale vertical beam will be uniquely suited to understanding the interfacial and confinement effects on polymer films during their formation processes (e.g., roll-to-roll or slot-die coating). The elemental sensitivity of soft and tender x-ray STXM to chemical bond types and bond orientation will be critical to characterizing nano- to mesoscale heterogeneities prevalent in polymeric materials with limited electron density contrast, especially for amorphous or poorly ordered blends. Machine-guided online material processing, characterization, and data analysis capabilities will accelerate the discovery of novel polymeric materials and processes by enabling intelligent exploration and optimization of complex processing-structure-property relations in vast parameter spaces.

Materials Science and Engineering

Materials Science and Engineering (MSE) has benefited from significant developments underpinning advances in energy, electronics, environment, and health care. The impact of the field continues to grow and increasi ngly incorporates multi-disciplinarity in both the science of understanding materials and in the development of novel applications. The impact and importance of x-ray techniques is already crucial across all areas of materials science and all areas of applications. Novel approaches to materials research include a close coupling and feedback between theory, computation, synthesis, and characterization and give rise to a new set of opportunities and capabilities at NSLS-II, both existing and planned. The MSE session of the NSLS-II '*New Science Frontiers*' workshop and this report summarize the potential for new contributions of NSLS-II in MSE. The report highlights the scope of the opportunity by focusing on three areas: energy storage, advanced manufacturing, and quantum information processing. These point to x-ray science research priorities using *in situ* techniques providing insight into heterogeneous materials, methods exploiting the optical coherence of x-rays, and the integration of machine learning and artificial intelligence in experimental design and data analysis. There are several opportunities for NSLS-II to contribute through the refinement of highly impactful nearly standard techniques and through the development of completely new capabilities. This in turn can be underpinned by a common set of advances that allow multimodal characterization and rapid and impactful data analysis that are important areas of development and investment.

The expansion of the scope of applications and diversity of functional materials has driven major breakthroughs across a vast range of technologies and will continue to be at the root of the technological progress and sustainable development.[1] MSE has two components: (i) The scientific fields of understanding how materials behave, their optimization for specific applications, designing and manufacturing materials; and (ii) The development of practical technological applications of new materials. An emerging feature of materials research has been to incorporate the evaluation and the mitigation of the multiple consequences of the invention, production, use and disposal of the materials, *e.g.* including the reliability, sustainability, and environmental impact as key design considerations. Environmental acceptability, reduction of pollution, sustainability, energy security and autonomy and the economic impact are increasingly being considered in the fundamental design of materials. MSE is a centerpiece of civil infrastructure, transportation, communications and electronics, medicine, energy and renewable energy ranging from the hydrogen economy to advanced nuclear reactors, energy conversion and storage, and quantum information.

A powerful emerging paradigm for materials research is to employ a continuously improving understanding at the atomic and molecular levels to guide the design of new materials. At the heart of this approach is a feedback loop in which new functional materials are synthesized, characterized at the atomic level (often during their operation, *in operando*), and the results are used to inform the computational modeling. Ultimately the approach allows scientists and engineers to develop design rules that lead to new materials and structures.

Materials science involves multiple length scales (from the tiny size of nanomaterials and thin films through the meters of engineering applications), complex assemblies (self-assembled materials, composites, hybrids, multilayered structures), and kinetic or dynamic phenomena with timescales from picoseconds to year. Next-generation engineering materials emerge from the fundamental research on the structure and properties of the materials and how they evolve in operating conditions, *i.e.* under mechanical, electric, thermal, radiation or chemical stress, to name just a few.

[1] Robert Cahn. The Coming of Materials Science (Pergamon, Elsevier Science, Oxford 2001)

The experimental, theoretical, and computational tools available to materials researchers are rapidly evolving and are transforming all fields of materials, ranging from construction materials to alloys, polymers, ceramics, and biomaterials. Across all these fields there is a drive for greater functionality and reduced cost and environmental impact.[2] Materials research has been transformed in particular by advances in characterization and computational tools and more importantly their direct coupling. Modeling of the materials at multiple length scales is at the core of this strategy. For example, when the length scale characterizing the microstructure is the size of magnetic domains, or the size of defects such as dislocation lines. Or alternatively when the physical dimensions of the component itself are conductive of strongly nonlinear interactions and where the response of the material is affected by the presence of surfaces and interfaces. DOE missions in energy, environment and national security, rely on advances in materials research and are reflected in the goals of the DOE Basic Energy Sciences division of MSE.[3]

Materials research is rapidly becoming increasingly interdisciplinary, including developments in computational methods, artificial intelligence, quantum information, biology and biomaterials, and economic drivers. The new technologies serving MSE also bridge gaps with other scientific disciplines and include developments in robotics, machine learning, and remote sensing. Large-scale facilities are among the places where this connectivity among scientific fields can thrive and succeed.

This report and the workshop on which it is based capture trends, methodologies, and characterization needs in MSE. The workshop solicited feedback from the communities and experts in various MSE fields, and hence was aimed at identifying those capabilities that NSLS-II could contribute that can have a transformative impact on materials research. The report focuses on important topical examples, spanning fundamental processes (bulk and surfaces) through synthesis and applied materials science and manufacturing.

Science Questions/Challenges

Materials research faces a range of scientific questions and challenges that cut across the range of applications, methods, and compositions. The presentations and discussion at the workshop highlighted numerous challenges that will continue to be addressed, at varying levels, in all of the sub-fields of materials research. Among the key challenges presented was the understanding of materials within complex multicomponent thermodynamic phase fields and the use of this understanding to select the elemental and nanoscale components of the materials. Beyond the thermodynamic picture, significant scientific opportunities remain in mapping reaction pathways, and determining the role of intermediate configurations. The desirable products themselves are often metastable in their final, applied, form so the question of metastability more generally, remains a key challenge. Synchrotron x-ray methods provide unique insight because of the unambiguous connection between diffraction, spectroscopy, and imaging data and thermodynamic and kinetic variables.

A further range of challenges arises in understanding structural features ranging from the atomic scale to the scale of entire functional units: from single atoms to entire macroscopic objects. X-ray science has contributed crucially to resolving atomic scale issues, including the symmetry and atomic positions within idealized crystals. Other structural features, however, remain significant scientific challenges for which novel approaches are needed. For example, the structure of surfaces and interfaces in complex heterogeneous environments, such as solid/solid, solid/liquid, liquid/liquid, and amorphous/crystalline interfaces all remain mysterious in many critical ways. The highly dynamical structure of liquids, confined liquids, amorphous solids, and molten salts is not yet known to a sufficient degree to provide sufficient insight for many applications. Finally, the structures involving multiple interacting point and extended defects and their dynamics remain a significant challenge to both experimental and computational efforts.

---

[2] Field, F., Clark, J., & Ashby, M. (2001). Market Drivers for Materials and Process Development in the 21st Century. MRS Bulletin, 26(9), 716-725. doi:10.1557/mrs2001.198

[3] DOE-BES MSE division's objectives and scope at https://science.osti.gov/bes/mse/About. A variety of science highlights is available at https://science.osti.gov/bes/mse/Highlights.

Beyond the positions of the atoms and nanoscale building blocks, MSE increasingly offers control of other degrees of order. These parameters range from familiar chemical phenomena such as the ionic oxidation state and elastic distortion of bond lengths to magnetization, electrical polarization, and superconducting wavefunction. The synthesis of materials that offers control of these order parameters requires understanding of how the structure can be used to tune them and increasingly, how multiple order parameters can coexist and interact. The last of these points is the key to phenomena including multiferroicity, in which a single material possesses multiple interacting functional order parameters. A similar challenge arises in the understanding of defects and doping, which in many cases are the fundamental origin of desired functionalities.

The heterogeneity of important materials has long presented a challenge for materials researchers. The properties of materials measured at large scales provide information that is averaged over a range of structural and chemical variations over distances smaller than the probed volume. The averaged information, in turn, poses a challenge for understanding effects of variations within the ensemble. This challenge can be overcome by the development of methods that enable the characterization at the relevant length scales, often individual particles, crystalline grains, or quantum dots. The development of characterization techniques includes, for example, precise evaluations of local elastic distortion and chemical state that underpin advances in integration in nanoelectronic devices.

Additional challenges and opportunities arise in the time domain. Materials phenomena include rare events: nucleation, crack formation, and domain motion, that define processing, elastic, and ferroic phenomena. Matching length scales and time scales, particularly far from equilibrium is an important frontier in both theory and characterization. Chemical reactions, high mechanical loading rates, high fluxes of particles or radiation, and rapid heating/cooling all pose challenges for which advances in materials characterization techniques will make important contributions.

The characterization challenge is also *dynamic* in a way that has been difficult for existing techniques. The materials underpinning the functionality of batteries, solar thermal hydrogen generators, fuel cells, and other applications all go through dramatic cycles with large stoichiometric variance. How materials withstand and self heal in these cycles is a key area of research.

This section highlights these cross-cutting ideas in examples drawn from three important areas: energy storage, electronic materials for quantum information, and advanced manufacturing.

Materials Design in Energy Storage. Realizing new materials for applications in energy storage is a challenging problem involving often competing constraints of functionality, stability, manufacturability, and total cost. The scope of materials is being expanded to meet these challenges using approaches combining on the computational prediction of polymorphs and on the experimental resources of a synchrotron facility. In his presentation at the workshop, David Ginley (NREL) emphasized how *in situ*/operando synchrotron-based research can contribute to determine the 'synthesizability" of novel materials, including metastable forms. The design of these materials can be guided by theory and selected to have desired properties. A key consequence of being able to do relevant *in situ*/operando science is understanding the behavior during the dynamic cycling of the materials where large stoichiometric change in composition and defects can occur. For energy storage, such properties underpin solar fuel synthesis and photovoltaic applications (Fig. 3). Further extensions of this approach apply to the design of polar materials specific optical or ferroelectric properties.

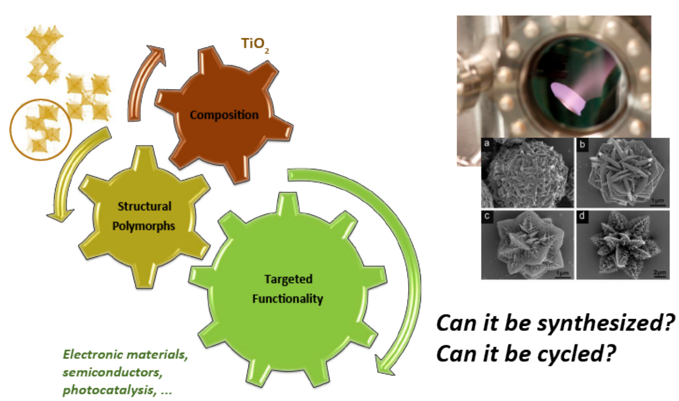


Figure 3: Functionality-driven Materials Design and Predictive Synthesis: case-study of TiO_2. Courtesy of D. Ginley.

An example of functionality-driven materials design is within families of similar metal-oxides compounds. The various polymorphs of these materials can be synthesized through a variety of methods (*e.g.* pulsed-laser deposition, sputtering, hydrothermal, solution, sol-gel, and chemical vapor deposition). These structures can be tailored to diverse applications (*e.g.* wastewater treatment, catalysis, sensors, supercapacitors, solar thermal hydrogen, fuel cells, batteries). The use of design principles in the selection of properties, the structures providing those properties, and the synthesis routes employed to create these materials is illustrated in Fig. 3.

Extending Electronic Materials towards Quantum Information. The physical implementation of devices for quantum information science is underpinned by the creation of devices for the manipulation of individual wavefunctions. Devices for quantum computing host and manipulate wavefunctions in superconductors, semiconductor quantum dots, specifically selected and positioned dopants in semiconductors, or in emerging quantum materials such as those exhibiting Majorana Fermion phenomena. The challenges faced in fabricating and optimizing quantum computing devices are significantly different from those that have been addressed over the last 40 years of electronic materials research. At the workshop Christian Lavoie (IBM) presented a range of issues in interface stability, thin film formation, and materials selection that uniquely impact superconducting quantum bits. What is presently known about the relevant materials system is often insufficient or, in some cases, beyond what can be learned from available techniques. Advances in synchrotron radiation methods promise to resolve questions pertaining to superconducting qubits, including stress effects, the mobility of atoms during thermal cycling, and ultimately how structural and quantum phenomena are coupled. As is the case uniformly across all of these highlights, interfaces and defects play a large role and there are key unifying themes associated with understanding how these structural features evolve during processing and operation.

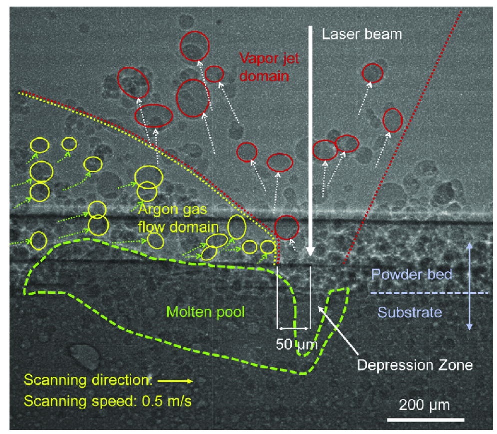


Figure 4: X-ray image of powder spattering during laser single track scanning.[1]

Advanced Manufacturing. Advanced manufacturing promises to speed the development of products, improve their functionality, and reduce the costs associated with making, transporting, and supporting them. The broad impact of advanced manufacturing approaches and techniques has attracted significant attention from industry,[4] has been the subject of a DOE series of Basic Research Needs workshop,[5] and has led to the formation of organizations enabling its progress in the US and international science and engineering research communities.[6]

Within the spectrum of advanced manufacturing technologies, L. Levine discussed the impact of additive manufacturing on the creation of metal, plastic, ceramic, and biomaterials components rapidly, in new geometries, and with low materials waste. The production of metal parts via laser powder bed fusion additive manufacturing (AM), as shown in the x-ray radiograph in Fig. 4, is rapidly growing. However, the transition of this technology from production of prototypes to production of critical parts is hindered by a lack of confidence in the quality of the part. A key outcome of research in this area is to establish a comparison of the properties of materials produced by additive manufacturing to those created by conventional means. Confidence can be established via a fundamental understanding of the physics of the near-melt pool processes, achieved through measurements, modeling and simulation. There are significant challenges in physics, computation, and materials science

4 2019 Additive Manufacturing Market Outlook and Summary of Opportunities https://www.smartechanalysis.com/reports/2019-additive-manufacturing-market-outlook/

5 Basic Research Needs workshop for Transformative Manufacturing was held March 9 -11, 2020. The focus of the workshop was to identify the basic science research priorities that could accelerate innovation to transform manufacturing in the future. https://science.osti.gov/bes/Community-Resources/Reports

6 https://www.nist.gov/ambench/organization

stemming from the broad range of length and time scales and temperature ranges associated with the process.[7, 8] The AM-Bench organization[6] (60 organizations, 83 members) was created in 2015 to allow modelers to test their simulations against rigorous, highly controlled AM benchmark data. This is an opportunity for NSLS-II to contribute its expertise within a large partnership that includes simulation software companies, AM industry, AM machine manufacturers, end users of AM products and academia.

Among the many chemical, metallurgical, and kinetic phenomena impacting materials created by additive manufacturing, local stress is a critical factor for all devices at all steps. The magnitude and distribution of stress and how the stress impacts processing, assembly, and functionality have been difficult to measure within nanoscale (< 50nm dimension) volumes throughout extended 3D volumes of complex materials or devices.[9] There is presently a lack of experimental tools to provide powerful direct tests for computational models of materials structural evolution with point-to-point maps of evolving structure and defect distributions, including resolved strain maps.[10] This kind of information is precisely what is needed to test emerging computational models and move MSE closer to predicting materials properties from composition and processing history. The synchrotron instrument should enable one to nondestructively map crystal structures and defects in three-dimensional volumes of arbitrary samples with resolutions of tens of nanometres within tens to hundreds of cubic micrometer volumes.[7]

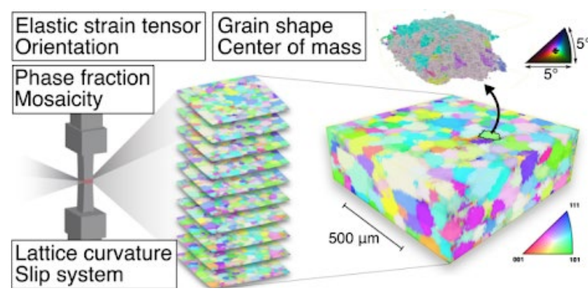


Figure 5: Near-field 3D X-ray Diffraction Microscopy (1 μm resolution, 1 mm3 FOV). Courtesy of A. Bucsek.

Priority Research Directions

Advances in the areas of materials research described above depend in turn on solutions to important challenges in materials characterization using x-ray methods. NSLS-II is already a crucial resource for materials research, and will become increasingly valuable as its capabilities are expanded. This section highlights three key areas of x-ray research that were identified in the workshop: developing methods for solving structural problems in materials incorporating hierarchical complex structures, exploiting x-ray coherence to move from ensemble averages to studies of small-scale heterogeneity, and taking advantage of advances in machine learning, artificial intelligence (AI), and high-throughput data analysis to make experiments far more efficient. There is an increasing need to apply all of these approaches *in situ* and *operando* because a snapshot of the material does not provide sufficient insight. All of the research directions described here must be adapted to get dynamic information. Doing this is key to the development of desired phases or interfaces and their stability during their functional operation.

Priority Research Direction #1: Three-dimensional in situ imaging of structural and chemical diversity

The metals and ceramics relevant to engineering applications often have a complex and hierarchical microstructure with a large number of crystallographic grains and multiple structural or chemical phases. The functionality of these materials depends on controlling the structural and chemical variation precisely. In structural materials, for example, mechanical properties vary significantly within materials of the same composition when processes yield different orientations, sizes, and elastic distortion within the crystalline grains. Similarly, transformations in shape-

[7] W. E. King et al. 'Laser powder bed fusion additive manufacturing of metals; physics, computational, and materials challenges'. Applied Physics Reviews 2, 041304 (2015); https://doi.org/10.1063/1.4937809

[8] T. Keller, L.E. Levine et al. Application of finite element, phase-field, and CALPHAD-based methods to additive manufacturing of Ni-based superalloys. Acta Materialia 139, 244-253 (2017).

[9] L.E. Levine et al. 'Full elastic strain and stress tensor measurements from individual dislocation cells in copper through-Si vias'. IUCrJ (2015). 2, 635–642 http://dx.doi.org/10.1107/S2052252515015031.

[10] G. Ice. 'Are X-rays the key to integrated computational materials engineering?' IUCrJ (2015) 2, 605–606 http://dx.doi.org/10.1107/S205225251501951X

memory alloys depend on the evolution of many crystallographic twins. The development of advanced piezoelectric materials depends similarly on understanding their microstructure. The problem is particularly challenging in materials that have been synthesized by additive manufacturing techniques because additive processes have complex thermal histories and create microstructures with large variations over micron-scale distances. A part of the workshop's focus is well reflected in a recent review by G. Ice "Although atomic resolution is still in the future, synchrotron-based X-ray microscopy is rapidly emerging as the standard for nondestructive three-dimensional characterization of materials structure at the macro-, meso- and nanoscales."[15]

The further development of high-photon-energy x-ray diffraction and microscopy techniques and associated data curation and analysis methods is a priority research direction in engineering materials. These methods together provide microstructural data that can be directly employed in improving models of behavior of materials.[11] X-ray microscopy techniques can employ full-field imaging or scanned nanobeam diffraction.[12,13] Near-field 3D x-ray diffraction microscopy provides sub-grain measurements of the crystallographic orientation, the full elastic strain tensor, and the distribution and concentration of extended defects, as in Fig. 5. Complementary full-field techniques track the distribution of grain and macroscopic strain. The microscopy measurements rely on high x-ray photon energies of tens of keV to penetrate macroscopic objects, permitting imaging studies without mechanical sectioning and allowing detailed *in situ* measurements. Future development can potentially provide insight into the structure of surfaces and interfaces within complex microstructures.

Priority Research Direction #2: Exploiting X-ray Coherence to Probe the Structure and Functionality of Materials

A second priority research area in x-ray science is to take advantage of the optical coherence of hard x-rays to provide new insight into materials processes. Coherent x-ray probes can be employed in coherent diffraction imaging (CDI) and ptychography to provide images of structural features at scales far smaller than the overall size of the x-ray beam. Additionally, x-ray photon correlation spectroscopy (XPCS) and related dynamical techniques provide insight directly into kinetic processes.

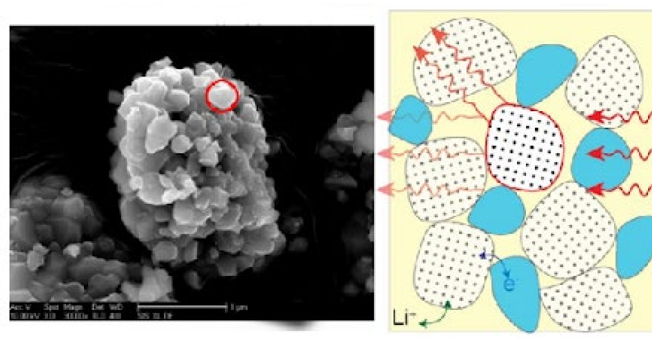


Figure 6: Bragg coherent diffractive imaging in lithium- and sodium-ion battery materials. Courtesy of A. Singer.

CDI techniques are powerful probes of the *in situ* evolution of individual grains of heterogeneous materials. For example, *in situ* CDI techniques allow isolating the nanoscale structural response of an individual cathode crystallite during the lithiation and delithiation process in a fully operational lithium ion battery, as shown in Fig. 6. The spatial and structural precision of CDI allow defects, strain, and the formation of competing phases to be tracked and fed back to materials and process design.[14] Further development of CDI and related coherent imaging techniques is a priority in addressing more complex systems with improved structural and chemical precision and providing

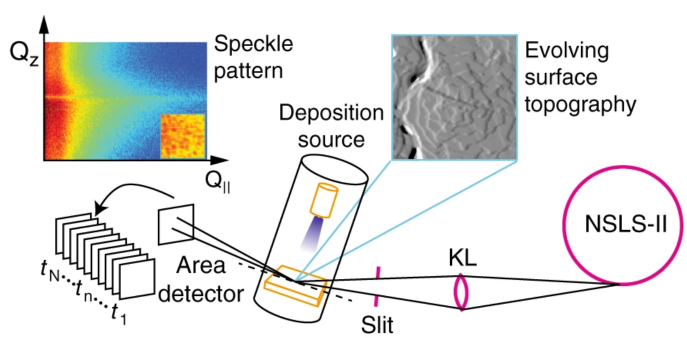


Figure 7: Coherent scattering characterization of C_{60} thin-film deposition. (111)-oriented crystalline mounds form via nucleation events captured by the intensity distribution of scattered x-rays.

[11] K. Kapoor, M. D. Sangid, Mat. Sci. & Eng. A, 729, 53-63 (2018)

[12] Levine, et al., Nature Materials, 5, 619-622 (2006).

[13] G. E. Ice, et al., J. Synchr. Rad. 12, 155-162 (2005).

[14] Singer et al. Nat Energy, https://doi.org/10.1038/ s41560-018-0184-2

time resolution matching the natural timescales of fast materials processes, for example the charging of batteries within minutes.

XPCS provides statistical insight into materials processes involving events that change the structure, phase, or configuration of materials. In epitaxial growth, for example, XPCS can capture the detailed nucleation and dynamics of islands and step edges, processes which in many important cases are critical to epitaxy but detailed information about them is lost in conventional "non-coherent" experiments which average over the surface structure. An example of the use of XPCS in understanding nucleation processes in polycrystalline C_{60} crystal growth is shown in Fig. 7.[15] The formation and growth of terraced mounds by a process of local step flow leads to changes in the speckle pattern that are analyzed to obtain insight into kinetic processes. Traditional x-ray diffraction has usually relied on idealized high quality crystals in order to extract detailed structural information. However, this work shows that for "real-world" surfaces which are not perfect single crystals, the coherence of the x-ray beam can be used to effectively substitute for the coherence of the crystal structure. The further development of XPCS and similar coherent techniques is a priority in extending the impact of this kinetic insight to other *in situ* systems and faster processes.

Priority Research Direction #3: Experimental Methods incorporating AI, Machine Learning, and Automation

MSE is increasingly employing approaches that allow the systematic exploration of large ranges of compounds and processing conditions. These approaches include combinatorial studies on compositional ranges, the use of AI and machine learning in experimental design, and rapid systematic data analysis to speed the process of gaining information from experiments. High-throughput experiments call for rapid measurements (*i.e.* number of samples or number of data captures per unit time) and equally fast (quasi on-line) reduction, visualization and assessment of the data. Synchrotron light sources including NSLS-II are deploying methods that can rapidly probe large arrays of samples, for example using rastered x-ray beams to study compositional spreads. The first synchrotron analysis of a combinatorial library dates back to 1998[16], and since then has gained widespread interest and discussion in the materials community.[17] Experiments can be dramatically sped up by incorporating AI and ML techniques in their workflow, particularly when the execution of the experiment can be guided by machine learning techniques.[18],[19] Sampling a complex, multidimensional parameter space (*e.g.* tuning temperature and pressure and chemistry simultaneously) using AI can be far more effective than an exhaustive grid-scan of that hyper-space. An example of how machine learning can be applied to materials discovery is shown in Fig. 8.[20]

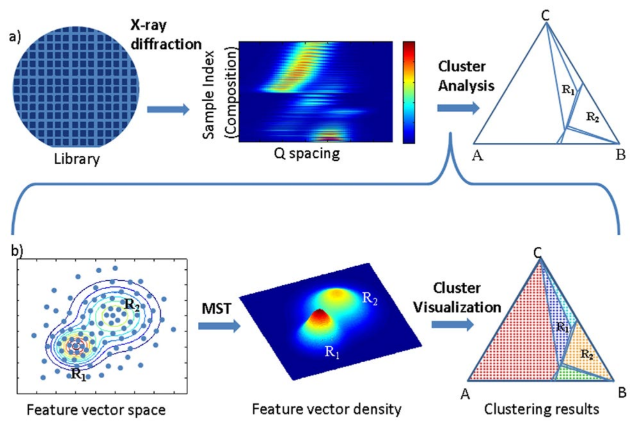


Figure 8: Machine learning strategies in high-throughput experiments searching for rare-earth-free permanent magnets.

Statistically meaningful sets of measurements can be rapidly extracted, a significant advance with respect to basing conclusions intuitively on the analysis of a small number of samples. *In situ* measurements more and more fall

[15] Headrick, R.L., Ulbrandt, J.G., Myint, P. et al. Coherent X-ray measurement of step-flow propagation during growth on polycrystalline thin film surfaces. Nat Commun 10, 2638 (2019). https://doi.org/10.1038/s41467-019-10629-8

[16] E. D. Isaacs and M. Marcus. 'Synchrotron x-ray microbeam diagnostics of combinatorial synthesis'. Appl. Phys. Lett. 73, 1820 (1998); https://doi.org/10.1063/1.122293

[17] https://science.osti.gov/-/media/bes/pdf/reports/2020/AIML_Roundtable_Brochure.pdf (October 2019)

[18] Kusne, A., Gao, T., Mehta, A. et al. On-the-fly machine-learning for high-throughput experiments: search for rare-earth-free permanent magnets. Sci Rep 4, 6367 (2015). https://doi.org/10.1038/srep06367

[19] https://www.nature.com/articles/s41598-019-48114-3

[20] Kusne, A., Gao, T., Mehta, A. et al. On-the-fly machine-learning for high-throughput experiments: search for rare-earth-free permanent magnets. Sci Rep 4, 6367 (2015). https://doi.org/10.1038/srep06367

into this category, where a large series of measurements can be conducted over time as a function of a process variable (T, p, E, gases, …). Combinatorial measurements also contribute to (and benefit from) materials libraries are now at hand (luminescent materials, semiconductor gas sensors, magnetic shape memory alloys, etc). Tagged to these data come strings of metadata, or descriptors, that log and keep track of all relevant information regarding the sample provenance, its history, the experimental conditions, the assumption for theoretical modeling and the authorship.

NSLS-II Capabilities

NSLS-II provides an impressive array of materials research capabilities through techniques that provide spectroscopic and structural data on real and heterogeneous systems over multiple length and time scales. The beamlines at NSLS-II that already have extensive use in materials research include *in situ* studies (ISR) at 4-ID, inner-shell spectroscopy (ISS) at 8-ID, quick absorption and scattering (QAS) at 7-BM, coherent hard x-ray scattering (CHX) at 11-ID, *in situ* and *operando* soft x-ray spectroscopy (IOS) at 23-ID-2, x-ray powder diffraction (XPD) at 28-ID-2, pair distribution function (PDF) at 28-ID-1, NIST partner beamlines soft & tender spectroscopy (SST-1 and SST-2) at 7-ID, and materials measurements (BMM) at 6-BM.

In addition to the existing capabilities at NSLS-II, the workshop identified gaps that fall into two broad categories. The first comprises those facilities that represent the core needs of materials researchers and often involve highly innovative use of what could be considered now-standard techniques. They can be developed as essential and complementary assets with high impact and high productivity, that will make existing high-end capabilities of NSLS-II more focused, efficient and impactful. They rely on: i) large and experienced user communities; and ii) mature technologies and established data analysis protocols. They also are a good match to the multi-modal strategy that NSLS-II is promoting.

The second category are cutting-edge capabilities with a high potential for new user communities and new science focuses. The impact of the research enabled by these capabilities can be measured in both publications and in the enabling of important applications.

A. Core Capabilities for Basic Research Needs

Scientific Capability #1: High-Resolution Diffraction. Detailed knowledge of the atomic structure and its relationship to the properties and functions of the material of interest, underlies all of MSE. Many strategies for resolving the structure of materials rely on diffraction methods. Most materials with a particular functional or industrial relevance are polycrystalline and the exploitation of the x-ray diffraction pattern is hampered by a significant broadening and overlap of the diffraction signal. High resolution diffraction (Fig. 9) is a prerequisite to overcome these limitations. Features in symmetry and atomic structure can be solved or modeled, using direct methods and global optimization methods.

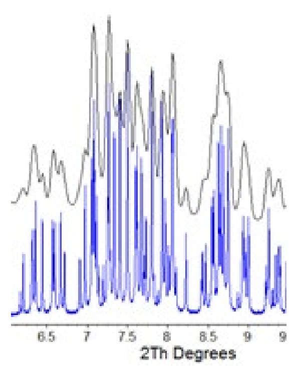


Figure 9: High resolution diffraction patterns of Gadolinium and Uranium based Salt Inclusion Materials (SIMs) for use as nuclear waste forms. Top: NSLS-II 28ID data. Bottom: high resolution APS 11-BM data. Courtesy of S. Gill (BNL) and G. Morrison (U. South Carolina).

There is presently a lack of high-resolution powder diffraction capabilities in the US as a whole. The low-divergence and high flux of an undulator beam in the 10 - 25 keV energy range combined with high angular performance (*e.g.* using analyzer crystal optics) can form the basis of a high-performance instrument that is i) complementary to existing NSLS-II diffraction capabilities, and ii) competitive with state-of-the-art synchrotron capabilities around the globe. This technique is central to MSE, but also extends to physics (dielectrics, thermo- and ferro-electrics, magneto-resistive materials, superconductors), chemical science and engineering (solid oxide fuel cell and battery electrodes, photo-catalyst, clathrates, metal-organic frameworks, metal hydrides), environment and health (carbon capture and gas storage, solar cells, cements, high performance alloys, refractory ceramics, pharmaceuticals and drug engineering, minerals, bio-engineered materials).

<u>*Scientific Capability #2: High-speed, High-Spatial Resolution Imaging*</u>. The workshop revealed a widespread need to provide rapid and reliable 3D imaging of materials under a range of processing conditions. In addition to the specific cases highlighted above, imaging techniques provide insight into fracture of engineering materials under fatigue and corrosive environments, solidification with controlled thermal and chemical conditions (as in Fig. 10), and hierarchical structures of biological biological, biomimetic, and bioinspired materials.[21] The study of complex and heterogeneous systems requires a large-volume, high-throughput, and high-resolution morphological characterization capability. Key parameters are single-to-few-micron spatial resolution, cm-scale field of view, and adequate data collection and transfer rates to accommodate fast scanning. In terms of temporal resolution, x-ray micro computed tomography can already collect more than 200 tomograms per second,[22] and further improvements are expected as detectors and x-ray optics improve.

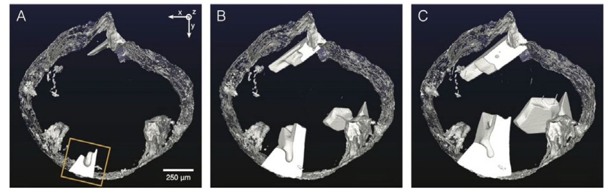


Figure 10: 3-D reconstructions of Si particle growth. White regions are the primary Si particles, translucent light gray region is the oxide skin and the dark blue background is the Cu-enriched liquid. Three images represent (A) 2079 s (875 °C), (B) 4261 s (839 °C) and (C) 8031 s (776 °C).

It is important to combine 3D imaging approaches with complementary x-ray diffraction techniques. The workshop noted that novel beamline designs are available to combine full-field imaging and micro-diffraction. An example of this approach is the "Crack-tip Microscopy" afforded by the DIAD beamline at the Diamond Light Source, as in Fig. 11.[23] The full-field imaging examines the crack initiation and propagation, as well as morphological change of materials around the crack *(e.g.* plastic deformation), while the micro-diffraction technique measures the strain of the crystalline phases near the crack tip. By stitching the real-space and reciprocal-space information together, one can gain a comprehensive understanding of the fracture behaviors of materials and design novel microstructures to increase the toughness of technological significant materials without compromising their strength.

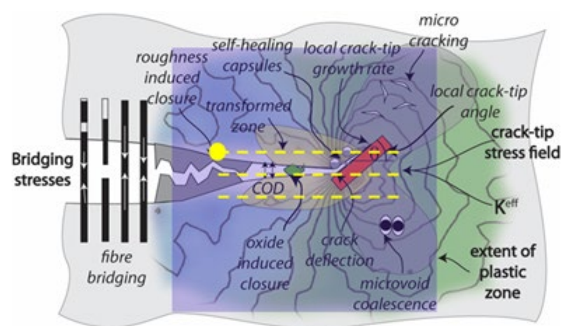


Figure 11: Simultaneous micro-diffraction (yellow) and full-field imaging (blue) for crack growths dynamics.

In addition to integrating multiple x-ray techniques, an imaging facility should include complementary optical, thermal, and chemical characterization capabilities, as well as diverse sample environments. These infrastructures enable *in situ*/*operando* measurements of materials processes driven by changes in the temperature, pressure, and gas atmosphere.

B. Enabling New Characterization Capabilities

<u>*Scientific Capability #3: Sub-Micron Beam Diffraction*</u>. Many of the processes and devices highlighted in the workshop involve understanding and controlling the structure and properties of materials at a length scale of 100 - 500 nm. Key processes and samples in this size range include i) structure of individual particles, defects and inhomogeneities during synthesis reactions, transitions, or device operation (*e.g.* as in the ferroelectric structure

[21] Shahani, A., Gulsoy, E., Poulsen, S. et al. Twin-mediated crystal growth: an enigma resolved. Sci Rep 6, 28651 (2016). https://doi.org/10.1038/srep28651

[22] García-Moreno, F., Kamm, P. H., Neu, T. R., Bülk, F., Mokso, R., Schlepütz, C. M., Stampanoni, M. & Banhart, J. (2019). Using X-ray tomoscopy to explore the dynamics of foaming metal. Nature communications, 10(1), 1-9.

[23] Withers, P.J. (2011), Adv. Eng. Mater., 13: 1096-1100. doi:10.1002/adem.201100092

in Fig12[24]) and ii) sub-micron mapping of the structural changes under external excitation (such as pressure, temperature, photoexcitation or gas exchange). The 100 nm scale naturally matches lithographically patterned semiconductor epitaxial thin films, quantum wells, wires, and dots, and individual nanostructures. NSLS-II currently has a gap in beam size in this range, between the nano-scale and the macro-scale or bulk.

An instrument meeting this need could consist of a high-flux tunable wavelength undulator beamline with in-line focusing optics. The spot size can be changed without moving the sample and optics, enabling a choice of coherent and incoherent sample illumination. The experimental techniques available would combine diffraction mapping capabilities with excellent spatial and temporal resolutions and a large working distance to accommodate a variety of sample environments. The high coherent flux of NSLS-II would allow scanning diffraction measurements to be combined with analysis using ptychography and CDI.

<u>*Scientific Capability #4: Interface/Surface Spectroscopy*</u>. The workshop highlighted challenging materials problems in systems that fundamentally involve multiple oxidation states, including aqueous and solid-phase electrochemical systems that are relevant to batteries, fuel cells, and sensors. This area of MSE also has significant overlap with the needs of the catalysis communities in which interfacial catalysis, corrosion, and electrochemical devices involve similar processes. . The MSE community has developed a variety of tools over the years to establish bulk oxidation states, which in turn provides insight into bulk point defect concentrations. In many instances, the nature of the bulk is used as a proxy to predict surface characteristics. Without verification, however such assumptions are risky, and direct measurement of surface properties are essential.

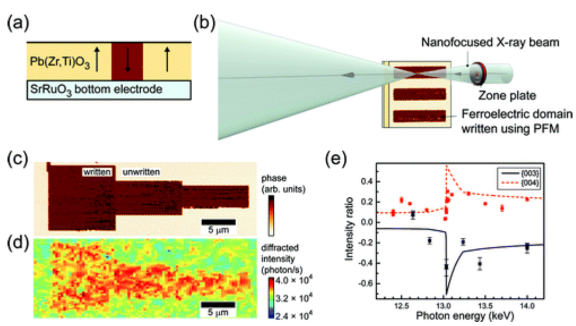


Figure 12: Reversed polarization domain in a $Pb(Zr,Ti)O_3$ ferroelectric thin film. (b) X-ray domain imaging based on a scanning 50 nm diameter probe beam. (c) piezoresponse force microscopy image and (d) map of diffracted X-ray intensity using {002} reflections of a domain pattern.

The challenges associated with understanding the chemical state of multivalent ions at interfaces and surfaces and within small volumes are particularly difficult. To address this issue, high-resolution multimodal chemical (valence- and element-specific) imaging capability would be ideal to map the concentration, valence and coordination state of active species near interfaces. The instrument would be using an undulator beam of tunable 5 µm spot size for Microfocus Spectroscopy (XAS, XES, XEOL).

<u>*Scientific Capability #5: Micron-Resolution Laue Diffraction*</u>. Complementary insight into the three-dimensional distribution of strain, shape (size, orientation) and composition of micro and nano materials and grain boundaries can be obtained using polychromatic diffraction techniques employing a tightly focused beam, as illustrated in Fig. 13. The insight obtained from these measurements is important in plasticity, crystallization, and the variety of nanomaterials problems discussed above. A further key use of micro- or nano-beam Laue diffraction is in surveying the orientation of individual nanocrystals before systematic study by CDI or other monochromatic x-ray beam methods.

An instrument proposed for NSLS-II can meet these needs. The proposed instrument delivers a 0.3 µm focused beam in the 5-23 keV photon energy range. The upper photon energy cutoff can be changed during experiments to allow the photon energy of individual reflections to be determined and thus to disentangle Laue Patterns coming from many grains.

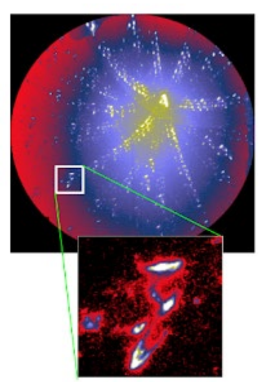


Figure 13. 3D X-ray micro-diffraction using either the triangulation or the differrential aperture X-ray microscopy techniques.[1]

[24] Ji Young Jo et al. Nano Lett. 2011, 11, 8, 3080–3084. https://doi.org/10.1021/nl2009873

Scientific Capability #6: Materials under Stress. The workshop also discussed the need to extend the scope of materials conditions combining high pressure, temperature, and stresses for which x-ray techniques can be applied. These approaches are potentially relevant in materials synthesis at high pressure, including diamond, boron nitride, or oxynitrides as well as in natural environments for predicting earthquakes or developing fracking techniques. The samples for these measurements are multi-grained, multi-phased materials with and without the presence of fluids and melts. A possible approach is to develop an instrument based on a wiggler insertion device combined with a dedicated high-pressure sample environment. The environment employs anvils and gaskets for isotropic pressure (from 50 MPa to 35 GPa), high deviatoric stress, high strain rate, and high temperatures (up to 2,000 K). X-ray techniques available in this approach include full-field 3D imaging, high-energy diffraction microscopy (in both near and far field modes), dark field diffraction imaging techniques, and energy dispersive diffraction.

Scientific Capability #7. Complementary Multimodal Methods and Sample Environments. Challenging materials problems require characterization employing multiple complementary methods. Through appropriate experimental design, advanced synchrotron radiation techniques can simultaneously incorporate these techniques to ensure that the information from multiple probes is obtained under identical conditions. The workshop participants specifically noted that optical, thermal, electronic, and environmental monitoring techniques are essential. Optical techniques of particular interest include optical imaging, dilatometry, and Raman spectroscopy. *In situ* thermal analysis by differential scanning calorimetry and gravimetric analysis are crucial in multiphase systems. Electronic probes include *in situ* resistivity and magnetotransport measurements. Chemical probes of the sample environment using mass spectroscopy are important in chemical synthesis experiments. Further potential complementary techniques include *in situ* scanning-probe or ultrafast optical excitation methods.

In addition to the mechanical, electrochemical, and processing environments discussed above, there are also several sample environments that will greatly increase the impact of materials research experiments at NSLS-II. These include high temperatures, *in situ* electrochemical current-voltage, capacitance, and impedance measurement infrastructure for experiments involving molten salts and a high-power laser for additive manufacturing research.

Finally, there was interest among workshop participants in broadening the scope of materials that can be studied at NSLS-II to include a broader range of radioactive materials. In particular, radioactive samples in difficult sample environments, *e.g.* those required for radioactive molten salts, are not possible with the present suite of instruments at NSLS-II.

C. Research and Characterization Infrastructure

Scientific Capability #8. Advanced Data Acquisition, Analysis, and Curation to Optimize Productivity. The discussion also emphasized the need for information technology resources in data acquisition, analysis, and curation to optimize the productivity of experiments. Key areas include reliable and consistent data acquisition, data management and archiving, and immediate data analysis capabilities to support feedback to experimental design in complex multi-dimensional parameter spaces. The participants emphasized the need for standardized data and metadata/lab record formats, seamless access to data and to computational resources commensurate with the size of the data sets, both during and post experiments. Similarly, all appropriate beamlines should incorporate the capability to employ AI and machine learning for advanced experimental design and for subsequent data analysis. An important consideration was that data confidentiality, security, and the possibility to accommodate intellectual property considerations are all required. All of these digital directions are consistent with goals set out in the current NSLS-II strategic plan.[25]

Scientific Capability #9. Enhanced Experimental Instrumentation and Detectors. The workshop participants also expressed widespread support for key issues in experimental instrumentation. Experiments in imaging and spectroscopy require consistent support for x-ray detection capabilities with some common requirements in area, total number of pixels, dynamic range, energy resolution, energy range, and data rate. The collaborative

[25] "National Synchrotron Light Source II 2020 Strategic Plan,": https://www.bnl.gov/ps/docs/pdf/NSLS2-Strategic-Plan.pdf.

development and support of these detectors was viewed as a priority. Similarly, the simplification and standardization of mechanical, technical, and controls interfaces to experiments would significantly benefit the development of multimodal experiments.

Impact of the New Capabilities

NSLS-II can offer capabilities and a complementary research infrastructure that can contribute to significant advances in materials research. In particular, the field is rapidly advancing towards an understanding of the next generation of materials over a comprehensive range of length and time scales. The role of NSLS-II includes (i) developing key aspects of advanced x-ray techniques in coherence, microscopy, and imaging, (ii) supporting a diverse and rapidly adaptable set of materials and their associated environments, and (iii) providing the engineering, data management, and connectivity with other BNL resources that enables experiments.

Catalysis and Chemical Sciences

Catalysis is central to chemical sciences and chemical engineering. Catalysts increase the reaction rate by lowering the reaction activation energy and increase the yield of the desired product(s) by controlling the relative rates of competing reactions. Catalysts enable many important chemical transformations in energy conversion processes, fossil fuel production, drug synthesis, chemicals production and manufacturing of fertilizers. In fact, catalysts are used in at least one of the processing steps in over 80% of all chemical products and carbon-based energy carriers. The global catalyst market is already expected to reach 35-40 billion dollars by 2025[1] and these materials are crucial to provide for the future energy needs, health and welfare of the entire world population.

The overall feedstock for fuels, and chemicals to a larger extent, has been changing rapidly in the last two decades, from oil to shale gas and more renewable sources such as biomass[2]. Additionally, other alternative energy and carbon sources are being explored to lower the dependence on fossil fuels. For example, a much-desired transition from fossil to solar fuel is needed[3], which necessitates new electrocatalytic processes for water splitting, as one of the most practical ways to transform sunlight to chemical energy. Additionally, new efficient catalysts are needed to convert recycled carbon dioxide into hydrocarbons. Therefore, there is a need for new catalysts and catalytic processes to enable efficient chemical transformations from the different resources to meet our societal needs in fuels and chemicals production[2]. Identifying the catalyst structure and how it adapts to the reaction environment is one crucial step in catalyst design. DOE recognizes the importance of the field with basic research needs summarized in the 2017 report[2].

Catalysts are often very complex, multi-component materials with relevant structural features on multiple length scales. As such, they present a considerable challenge to be characterized and fully understood, especially under working conditions that often include high pressures and/or temperatures. Synchrotron based X-ray techniques, have provided crucial insight into the catalyst behavior. However, new capabilities shall be developed at NSLS-II in order to enable the fundamental understanding of the critical electronic and structural properties and establish their relation to the pathways of catalytic reactions on all relevant spatial and temporal scales. We will address the need to target *catalysis dynamics, heterogeneity on multiple scales* and *dynamics on catalytic interfaces* with the development of new capabilities, including *in situ/operando catalyst characterization with high resolution hard X-ray spectroscopy* (X-ray emission spectroscopy, high resolution fluorescence detection, resonant inelastic scattering), *closing the energy and pressure gap* (through integration across soft, tender and hard X-ray ranges and reaction conditions), and *coupling the X-ray characterization* with additional techniques, data informatics and theory.

Science Questions/Challenges

<u>*Science Challenge #1: Measuring catalyst dynamics and interfaces*</u>. The catalyst structure changes over several time scales, from the synthesis to the initial structure after pretreatment to the active phase during reaction (which can change under different reaction conditions), as well as the changes that proceed during the lifetime of the catalyst (deactivation). Observing these structural changes and interaction of reactants and products with the catalyst (e.g. changes in electronic properties) is key to controlling catalytic transformations.

<u>Example 1:</u> Catalysts are not rigid structures, they adapt and change dynamically during the reaction cycle. Paolucci et al. used *in situ* and *operando* x-ray absorption spectroscopy to study the dynamical nature of zeolite supported isolated Cu ions for the selective catalytic reduction (SCR) of nitrogen oxides (NOx) with ammonia (NH_3). They showed that during the SCR catalytic cycle, isolated Cu sites diffuse in zeolite windows to dynamically and reversibly form Cu-Cu dimers that are more favorable for O_2 activation as shown in Figure 14 (right panel)[4]. Consistent with the dual site requirement, the oxidation of Cu^I can be seen to follow a second order dependence on Cu^I in Figure 14 (left panel).

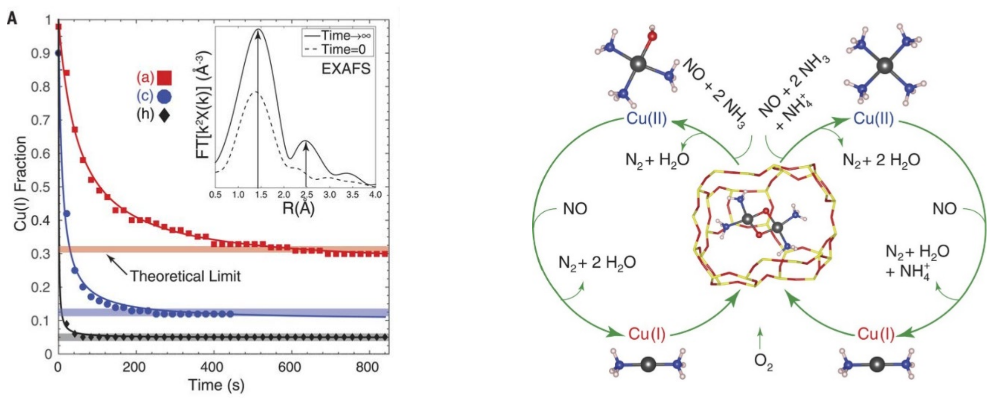


Figure 14: Adapted from ref. 4 <u>Left:</u> Temporal evolution of the XANES-measured Cu^I fraction is plotted for the Cu-CHA-29 (a, red), Cu-CHA-20 (c, blue), and Cu-CHA-15 (h, black) samples during transient oxidation in 10% O_2 at 473 K. The line represent least-squares fit to a second order rate law in Cu^I where the predicted recalcitrant Cu^I fractions are shown as horizontal bars. The Cu^I fractions reported contain an absolute 5% error from linear combination XANES fits. Inset, the Fourier transform of the k^2-weighted EXAFS signal in R-space (R) of Cu-CHA-15 collected before O_2 exposure and after the transient experiment. <u>Right:</u> Reduction steps proceed on site-isolated Cu^{II} ions residing near one (left-hand cycle) or two (right-hand cycle) framework Al centers with constrained diffusion of Cu^I ions into single cages and oxidation by O_2 (inner step). NH_4^+ is formed and consumed in the right-hand cycle to maintain stoichiometry and charge balance. Gray, Cu; yellow, Si; red, O; blue, N; and white, H.

<u>Example 2:</u> Identifying the adsorbates on the catalyst surface (gas/solid interface) and their dynamics during the reaction is crucial to understanding the reaction mechanism. Isolated metal single atoms on a support is a class of catalysts that has recently gained tremendous attention due to its unique properties. Supported metal single atoms maximize the utilization of noble metals and provide an ideal system for understanding the reaction mechanism and how adsorbates change during reaction[5-6]. Lu et al. used HERFD-XANES complemented by FTIR and DFT-calculated XANES (Figure 15) to differentiate between adsorbates and identify the active complex and most stable intermediate (resting state of the catalyst) during CO oxidation on Ir single atoms supported on $MgAl_2O_4$ (Figure 2)[6]. Ir(CO), and Ir(CO)(O) were identified as the active complex and resting state, respectively, showing that a CO ligand is present during the entire catalytic cycle. Despite the strong CO binding to Ir single atoms (−2.1 eV on Ir_1 vs. −1.4 eV on nanoparticles), they are more active than their nanoparticle counterparts. The reaction follows an Eley-Rideal mechanism where a gas phase CO reacts with O* in Ir(CO)(O) to regenerate the site required for O_2 activation. The results reveal the importance of understanding the changes in the local coordination of the Ir single atoms during the reaction cycle to understand the reaction mechanism.

The above examples show the importance of following the catalyst dynamics and understanding the interaction of adsorbates with the catalyst surface. However, characterization of gas/solid or liquid solid interfaces and their dynamics is more challenging due to the often small percentage of atoms at the interface (e.g. surface vs. bulk for metal oxides or larger nanoparticles for metal supported catalysts) requiring more sensitivity. Additionally, faster dynamics (structural or of adsorbates) are more challenging to follow and require faster time resolution.

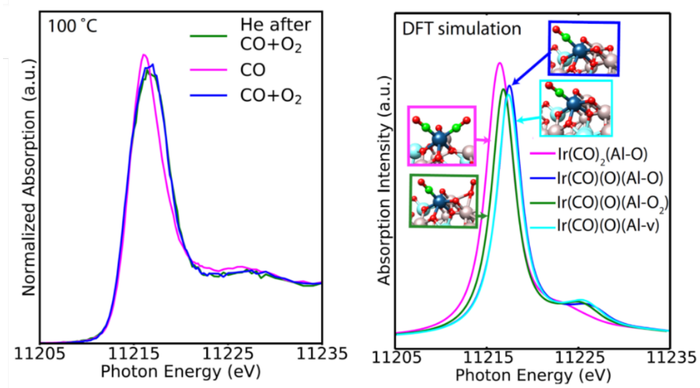


Figure 15: Left: Ir L_3-edge HERFD-XANES spectra of Ir single atoms supported on $MgAl_2O_4$ under He after $CO+O_2$, followed by dosing CO then $CO+O_2$. Right: DFT calculated spectra of the Ir single atoms with two CO ligands and an oxygen vacancy (magenta, $Ir(CO)_2$-(Al-v)), and with a (CO)(O) (blue, Ir(CO)(O)), and with (CO)(O) and adsorbed O_2 on the vacancy on nearest two Al atoms (green, Ir(CO)(O)-(Al-O_2)), from ref. 6.

Science Challenge #2: Understanding catalyst heterogeneity. Catalysts can have heterogeneities at multiple length scales. For example, differences in temperature and reagent concentration on the reactor length scale (µm, mm to cm) can affect the catalyst structure. More intrinsic variations in the structure are due to differences in the local coordination of the catalytic site (nm to Å). The differences can be due to one or more of the following: different coordination of a single isolated site on the support, different sites on a single nanoparticle (mono- or bi-metallic) or due to a distribution of size, shape and/or composition of nanoparticles. Small differences in local coordination/structure can have significant effects on the reactivity of those sites and identifying the reaction pathways associated with different types of sites is a major challenge. Therefore, the ability to distinguish surface sites and identify active vs. spectator sites is necessary for understanding the interaction of adsorbates with the active sites and building structure-activity relations.

Example 1: Not all surface sites on a supported metal nanoparticle are equal. Corner and step sites can have very different reactivity than terrace sites due to being undercoordinated. An extreme example is the case of ammonia synthesis (and the reverse reaction, i.e. decomposition) which is catalyzed by Ru exclusively on B_5 step sites on Ru(0001), while the terrace site are several orders of magnitudes less active (see Figure 3)[7-8]. Therefore, the activity of supported nanoparticles scales with the percentage of surface B_5-type sites which is maximized at a size of ~1.8-3.0 nm for spherical shape nanoparticles[9-11]. Using multiple characterization techniques including microscopy and x-ray absorption spectroscopy, Karim et al. showed that highly defected flake-like shaped nanoparticles maximize the number of B_5 sites and the activity of the catalyst (Figure 16).[12]

Example 2: Despite reaching atomic dispersion, supported metal single atoms can exhibit heterogeneity if the metal atoms are anchored on different sites on the support, or if the metal atoms co-exist with small clusters. This heterogeneity can be originally present after catalyst synthesis or develop under reaction conditions making it difficult to develop structure-activity relationships.

The above examples show that the active sites can be a small fraction of the total available sites making it a major challenge to identify the active sites and to differentiate them from all other spectator sites *in situ* or during reaction without prior knowledge on which type of sites are active (e.g. Ru B_5 sites for NH_3 synthesis/decomposition).

Priority Research Directions

Priority Research Direction #1: Precise identification of the active site. With chemical and structural heterogeneities in the catalysts, especially under reaction conditions, the nature of the catalytically active sites remains elusive. Improving precision, in terms of chemical states and spatial distribution is instrumental in understanding catalysis at the atomic scale and building structure-activity relations.

Priority Research Direction #2: Follow catalyst dynamics. Catalysis is inherently dynamic process where transform-ations occur consciously on various time and spatial scales, from the assembly of precursors into a catalytically active state to interaction of the active sites with of reactants, intermediates, and products. In many cases, the active sites are formed only under reaction conditions and involve a small fraction of the material. Transient measurements coupled with advanced spectroscopy and imaging tools will provide the critical link between dynamic structure and catalytic activity and selectivity.

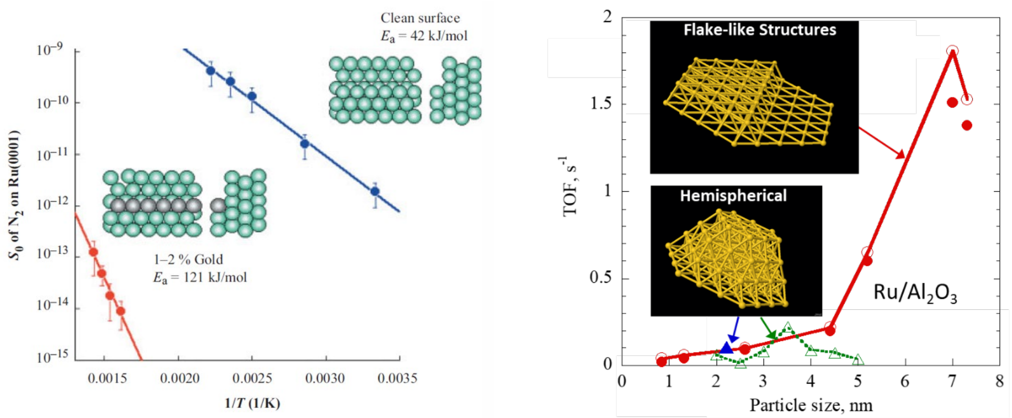


Figure 16: Left: Adapted from ref. 7. Dissociative sticking of N_2 on clean and gold-modified Ru(0001). The sticking coefficient of N_2 drops by several orders of magnitude when the clean surface is modified by gold, which is known to decorate the defect sites. Right: Adapted from ref. 12.Turnover frequency (TOF) in (moles NH_3 reacted/gcat/s)/(exposed moles Ru/gcat), for NH_3 decomposition on Ru/Al_2O_3 at 350 °C and 1 atm as a function of Ru particle size and shape. Experimental data in filled circles and microkinetic model in open circles. TOF using the fraction of B_5 step sites predicted by Gavnholt and Schiøtz for hemispherical nanoparticles (ref. 11) are shown in green.

Priority Research Direction #3: Understand processes at interfaces. Heterogeneous catalysis is an inherently interfacial phenomenon, where transformations occur at the phase boundaries. X-ray characterization of the interfacial regions is challenging as they include a small percentage of the atoms in the catalyst (e.g. surface vs. bulk for metal oxides or larger nanoparticles for metal supported catalysts). Particularly difficult to study are buried interfaces, e.g., surface of metal nanoparticles covered with carbon deposits during the reaction. Developing sensitive multimodal techniques, including extended energy ranges (soft-medium-hard X-rays) and multiple detection channels (electrons, photons) will be critical to identify, and thereby control structural and electronic transformations in the catalytically active interfacial structures. Understanding these phenomena will ultimately lead to new developments for improved catalytic performance.

Priority Research Direction #4: Push temperature and pressure limits. The atomic structure and chemical state of solid catalysts can be significantly influenced by the conditions under which they are probed. Therefore, catalysis researchers strive as much as realistically possible to study catalysts under conditions that mimic those used in standard operations. Industrial processes utilize high pressures to overcome equilibrium limitations, improve reaction rates, and maintain liquid phase conditions in reactors. Two prominent examples include ammonia synthesis reaction at which is carried out at hundreds of bar pressure and catalytic reforming and hydrodeoxy-genation of biomass-derived molecules to produce fuels and chemicals. Also many catalytic reactions require exceptionally high temperatures to achieve measurable reaction rates. The ability to reproduce these reaction conditions in a synchrotron experiment is critical to understand and optimize industrially relevant processes.

Priority research direction #5: Discover correlations and reactivity descriptors in high-throughput data. With the improved synchrotron instrumentation, including the ability to measure the catalysts in-situ and operando, it is possible to explore a much broader parameter space in term of the catalyst composition and reaction conditions. Large volume datasets obtained from multiple characterization techniques could be combined to extract new information and use it to guide material discovery and optimization. The data streams from different sources, together with all relevant metadata, have to be collected, corelated and curated in order to identify functional descriptors. New tools, based on quantum theory, data informatics and machine learning are needed to leverage the high throughput data.

New NSLS-II Capabilities

Workshop Catalysis section participants identified new opportunities to accelerate transformative research in the field, in the context of currently NSLS-II capabilities as well as other DOE synchrotron facilities. Specific capabilities listed below cover the broad range of research needs, and include new methods which require of new beamlines at NSLS-II, developing new endstations for the existing beamlines to address the gap in the energy range and sample environment, incorporation of new data analytics and theoretical tools into the data analysis pipelines as well as improvement of experimental support infrastructure.

The community has also identified the need to increase the capacity for the experiments, which are not flux- or coherence-limited specifically, for spectroscopy and diffraction. Currently all NSLS-II beamlines serving the research in catalysis are highly oversubscribed. The overall capacity of these experiments throughout the DOE complex will be severely limited in the next 5-7 years as other two major synchrotron facilities will go through accelerator upgrades, and as such there is a threat to the ability to aptly support the catalysis field.

Scientific Capability #1: Improving energy resolution. In a traditional XAS experiment, an electron from a core level is excited by the of a photon of the incoming X-rays. This results in the absorption of the incoming photon and emission of the fluorescence photons at lower energy. The energy resolution of the fluorescence detection is defined by a detector (no better than ~125 eV) and ultimately by the core lifetime broadening. Dedicated spectrometers which coupled dispersive crystal optics and area detectors are required to remove the resolution limitations which stem from the finite core-hole lifetime. Using high energy resolution fluorescence detection (HERFD), the broadening of the XANES spectra can be reduced allowing to detect subtle spectral features which are characteristic to active states. This is especially relevant for 5d elements, which L-edge spectra are intrinsically structureless, improving the ability to interpret the data (see Figure 2 for an example). Moreover, in a catalytic process, active component can be present in a number of chemical states and configurations with varying catalytic activity. A regular XAS measurement will deliver an ensemble averaged information, whereas with energy selective detection one can realize selective XAS by tuning the fluorescence energy of the detector to the energy of a fluorescence line of a particular valence.

Currently there is no dedicated instrument at NSLS-II dedicated to emission spectroscopy. To realize a world class experiment, the requirements for the photon beam delivered to the sample is high flux (>10^{13} ph/s) and focal spot of under 50 µm. Undulator based beamline which allows synchronized scanning of the insertion device gap and the monochromator with sufficient resolution will be prime candidate for the photon delivery for this experiment.

Scientific Capability #2: Closing energy and pressure gap. Coupling soft AP-XAS and AP-XPS with additional techniques, such as XES and RIXS will allow higher sensitivity towards valence states, providing information such as oxidation and chemical state, adsorbate-substrate bonding interactions, and charge transfer processes. With the integration of surface-sensitive infrared spectroscopy, surface-adsorbate interactions can be better resolved compared to XPS alone. Through this combined approach an ultimate picture of the catalytic process can be obtained. Decreasing the beam size will improve time resolution and allow higher pressure levels, making possible time-resolved measurements to obtain kinetic information.

The energy range in what constitutes a gap between traditional soft and hard X-ray instruments include K-edges of catalytically relevant sulfur, phosphorus and chlorine as well as L-edges of 4d elements. Only a few beamlines in North America provide access to tender X-rays, however the overall capacity does not come close to matching the research needs. It is therefore important to allow for catalysis related experiments, through developing reaction environments compatible with tender X-ray, as well as providing relevant reaction conditions control such as temperature and gas flow.

AP-XPS will also benefit from the addition of the tender range. Photon energies above >2000 eV improves the technique's ability to interrogate the dynamics of liquid/solid interfaces and conduct higher-pressure studies of gas/solid interfaces. Only one beamline in the US currently allows for this experiment. Adding infrared reflection absorption spectroscopy (IRRAS) for surface characterization to the sample environment chamber would produce a unique beamline with unmatched capabilities while adding a second characterization method that has been shown to give complementary surface information. In this case, there is also a need to develop reaction cells which enable probing, e.g., liquid/solid interfaces in electrocatalytic reactions and critical buried interfaces in carbon nanotubes growth processes.

Scientific Capability #3: Time resolved techniques. Under reaction conditions, multiple transformations in catalyst materials occur simultaneously, leading to crowded operando spectra that are difficult to interpret. Modulation excitation spectroscopy (MES) relies on the periodic perturbation of a given system, either by temperature or reactant composition, in combination with phase-sensitive detection. As a result, this approach significantly reduces background, allows distinguishing between active and spectator species, and enables extraction of kinetic information.

To allow for fast and reliable system perturbations, operando infrastructure (gas delivery, heaters) shall be developed. Equally important, new data acquisition strategy has to be devised to collect the results at the rate comparable with the relevant processes in the sample.

Equally important is the ability to collect comprehensive information about the sample during the process. For example, bimetallic catalysts often contain both 3d and 4d elements, and the instruments have to be reconfigured. Minimizing the time to switch between, e.g., multiple absorption edges allows for new measurements which provide complete data on the catalytic system. Improved detectors allowing for speed and multiple photon ranges have to be deployed to allow for the required time resolution and to suppress beam-induced effects.

Scientific Capability #4: Operando spectral imaging at the nanoscale. The length scales relevant to catalysis span several orders of magnitude. Synchrotron XAS and XRD are bulk techniques, delivering an ensemble averaged information. To resolve the structural and electronics features on the relevant length scale, down to nanometers, the combination of imaging and spectroscopic/scattering methods have to be deployed. Most successfully, transmission X-ray microscopy (TXM) and scanning transmission X-ray microscopy (STXM), were employed to visualize the changes in the structure, aggregate size, and distribution of catalytic active phase and support. Soft X-ray ptychography has allowed to map the distribution and chemical state of metals in a real industrial catalyst particle with a sub-5 nm resolution.

With the exceptional brightness of NSLS-II there is an opportunity to become the world leader in operando spectroscopic imaging in both soft and hard X-ray ranges. Particular attention should be paid to the development of sample environments capable of controlling the catalytic reaction parameters, with short working distance of soft X-ray microscopes being a serious challenge. When properly engineered the reactors can be made compatible with TEM-EELS experiments, adding an important dimension to the information obtained on dynamic catalytic systems.

Scientific Capability #5: Developing theory and data analytics tools. With the improvement of the temporal, spatial and energy resolution of X-ray techniques, higher flux of the third-generation light sources and improved instrumentation for high throughput measurements, the amount of data generated during a synchrotron experiment increased by one-to-two order of magnitude. Traditional analysis methods such as EXAFS spectra fitting do not keep up with the pace: the process requires human supervision and thus is not easily scalable. XANES data analysis is often limited to fingerprinting and basic linear fitting which is often inconclusive. Recent advances in machine learning approaches allow extraction of structural information,[13-14] showing promise for automated analysis of XAS spectra. In order to accelerate material science and catalysis research, there is a need to develop capabilities for data analysis and interpretation that are capable of matching the generation rate of experimental data . Additionally,

While chemometrics, a suite of mathematical and statistical techniques to extract chemical information from complex data, is widely used in traditional spectroscopic data analysis. It is necessary to include these methods into the synchrotron analysis pipeline. Additionally, theoretical tools, such as molecular dynamics and density functional theory are critical to model and interpret the data generated in high-resolution spectroscopic experiments (e.g. XES, HERFD-XANES, RIXS). To improve the understanding of the data generated by these novel methods, a consolidated effort shall be made to provide easily accessible theoretical tools to the users. With integrated data analytics, the researcher will be able to immediately devise pure spectra and reactivity trends from the large datasets. The ability to pinpoint important spectroscopic descriptors during the process, coupled with online analysis of the catalytic products, will not only accelerate catalysis discovery and optimization but will also present an opportunity for autonomous experiments.

Scientific Capability #6: Developing in situ and operando reaction environments. Soft X-ray XPS and XAS are exceptionally informative techniques probing the core levels of the elements, probing chemical and oxidation states of the catalyst, changes in the surface composition of the catalyst, the adsorption and activation of reactant molecules, and the formation of reaction intermediates. With the development of ambient pressure soft X-ray methods, operando characterization became possible. The major impediment of wider application of AP-XPS and AP-XAS is the limited availability of the suitable reaction environments. It is therefore critical to develop in-house capabilities and expertise where NSLS provides and operates facilities which are necessary to develop and construct new operando reaction cells. One example is the graphene membrane-based cell (where graphene on supported on silicon nitride grids or on proton exchange membranes), which enables the application of XPS operando investigations of electrode materials in electrochemical processes such as the oxygen evolution reaction. The fabrication of this kind of cells requires a considerable know-how and equipment related to graphene growth, graphene transfer and fabrication of silicon nitride membranes.

In order to develop and optimize new catalysts, the synthesis pathways have to be carefully characterized. One of the synthetic strategies is atomic layer deposition (ALD), a technique that relies on self-limiting half reactions to deposit a metalloorganic precursor with subsequent ligand removal. With repeated cycles of metal precursor and oxidation, metal oxides and sulfides can be grown in a highly controlled manner. The flexibility of the technique allows growing complex structure with targeted stoichiometry and structure. There is a need to develop a reaction environment which is compatible with X-ray characterization while maintaining mass and heat transfer characteristics of conventional ALD reactors.

In addition to the in-situ/operando sample environments, there is a need to develop high throughput in-situ data collection capability. With the high flux of NSLSII, sample pretreatment is often the limiting factor, making it an opportunity for high throughput data collection to improve beamtime utilization. This is especially important for samples needing routine XANES/EXAFS. The high throughput experiments require the ability to pretreat multiple samples, automated sample alignment and data collection.

The development of the above reaction cells and associated infrastructure requires staff support and dedicated development beamtime.

Scientific Capability #7: Development of support infrastructure. Cells and reactors which are typically used in a bench-top catalytic performance measurements are often not compatible with the requirements of X-ray characterization. Upon arrival to the synchrotron facility, the scientists have to adapt their experiment to the sample environment geometry available at the beamline which often has different mass and heat characteristics compared to the regular reactors. Given the availability of synchrotron beamtime, in situ and operando catalytic experiments are often performed just once per reaction condition, possibly affecting the reliability of the measurements.

In order to support the community better, the is a need of a staging laboratory dedicated to catalysis research. In this laboratory, the user should be able to benchmark their reactions in the sample environment, which will be used for the synchrotron measurements, prior to their actual beamtime. The laboratory should be equipped with the delivery system for relevant gases and liquids as well as the product analysis, including mass-spectrometry and gas chromatography. The sample environment should be easily accessible to the community. With the access of benchtop characterization, e.g., infrared, Raman and UV-Vis spectroscopy, the samples could be screened prior to the synchrotron experiment, improving efficiency. Tighter integration with the transmission electron microscopy capabilities at CFN will also benefit catalysis research.

Many relevant catalytic samples are air sensitive and the capabilities to handle and load these materials into reactors should be made available. These will include dedicated gloveboxes, sample suitcase transport means and possible "glovebox-at-the-endstation" solutions.

References:

1. Markets, F. Catalyst Market by Material (Metal, Chemical, Zeolites, Organometallic Materials), Type, Application, Regions, Global Industry Analysis, Market Size, Share, Growth, Trends, and Forecast 2018 to 2025. https://www.fiormarkets.com/report/catalyst-market-by-material-metal-chemical-zeolites-organometallic-385918.html.
2. Report of the Basic Energy Sciences Workshop on Basic Research Needs for Catalysis Science to Transform Energy Technologies. https://science.osti.gov/bes/Community-Resources/Reports, https://science.osti.gov/-/media/bes/pdf/reports/2017/BRN-Catalysis_factual_ doc.pdf?la=en&hash=07467FDA59C31461A79CBA8AA6224C329AE08ED7.
3. Gray, H. B., Powering the planet with solar fuel. *Nature Chemistry* **2009,** *1* (1), 7-7.
4. Paolucci, C.; Khurana, I.; Parekh, A. A.; Li, S.; Shih, A. J.; Li, H.; Di Iorio, J. R.; Albarracin-Caballero, J. D.; Yezerets, A.; Miller, J. T.; Delgass, W. N.; Ribeiro, F. H.; Schneider, W. F.; Gounder, R., Dynamic multinuclear sites formed by mobilized copper ions in NO_x selective catalytic reduction. *Science* **2017,** *357* (6354), 898.
5. Hoffman, A. S.; Sokaras, D.; Zhang, S.; Debefve, L. M.; Fang, C.-Y.; Gallo, A.; Kroll, T.; Dixon, D. A.; Bare, S. R.; Gates, B. C., High-Energy-Resolution X-ray Absorption Spectroscopy for Identification of Reactive Surface Species on Supported Single-Site Iridium Catalysts. *Chemistry – A European Journal* **2017,** *23* (59), 14760-14768.
6. Lu, Y.; Wang, J.; Yu, L.; Kovarik, L.; Zhang, X.; Hoffman, A. S.; Gallo, A.; Bare, S. R.; Sokaras, D.; Kroll, T.; Dagle, V.; Xin, H.; Karim, A. M., Identification of the active complex for CO oxidation over single-atom Ir-on-$MgAl_2O_4$ catalysts. *Nature Catalysis* **2019,** *2* (2), 149-156.
7. Dahl, S.; Logadottir, A.; Egeberg, R. C.; Larsen, J. H.; Chorkendorff, I.; Törnqvist, E.; Nørskov, J. K., Role of Steps in N_2 Activation on Ru(0001). *Phys. Rev. Lett.* **1999,** *83* (9), 1814-1817.
8. Honkala, K.; Hellman, A.; Remediakis, I. N.; Logadottir, A.; Carlsson, A.; Dahl, S.; Christensen, C. H.; Nørskov, J. K., Ammonia Synthesis from First-Principles Calculations. *Science* **2005,** *307* (5709), 555.
9. Raróg-Pilecka, W.; Miśkiewicz, E.; Szmigiel, D.; Kowalczyk, Z., Structure sensitivity of ammonia synthesis over promoted ruthenium catalysts supported on graphitised carbon. *J. Catal.* **2005,** *231* (1), 11-19.
10. Jacobsen, C. J. H.; Dahl, S.; Hansen, P. L.; Törnqvist, E.; Jensen, L.; Topsøe, H.; Prip, D. V.; Møenshaug, P. B.; Chorkendorff, I., Structure sensitivity of supported ruthenium catalysts for ammonia synthesis. *J. Mol. Catal. A: Chem.* **2000,** *163* (1), 19-26.
11. Gavnholt, J.; Schiotz, J., Structure and reactivity of ruthenium nanoparticles. *Physical Review B* **2008,** *77* (3).
12. Karim, A. M.; Prasad, V.; Mpourmpakis, G.; Lonergan, W. W.; Frenkel, A. I.; Chen, J. G. G.; Vlachos, D. G., Correlating Particle Size and Shape of Supported Ru/gamma-Al_2O_3 Catalysts with NH_3 Decomposition Activity. *J. Am. Chem. Soc.* **2009,** *131* (34), 12230-12239.
13. Timoshenko, J.; Lu, D.; Lin, Y.; Frenkel, A. I., Supervised Machine-Learning-Based Determination of Three-Dimensional Structure of Metallic Nanoparticles. *The Journal of Physical Chemistry Letters* **2017,** *8* (20), 5091-5098.
14. Timoshenko, J.; Wrasman, C. J.; Luneau, M.; Shirman, T.; Cargnello, M.; Bare, S. R.; Aizenberg, J.; Friend, C. M.; Frenkel, A. I., Probing Atomic Distributions in Mono- and Bimetallic Nanoparticles by Supervised Machine Learning. *Nano Lett.* **2019,** *19* (1), 520-529.

Molecular and Cell Biology

The workshop was planned to cover two and half days with the first day covering a broad sweep of synchrotron science undertaken at the NSLS-II, as well as status reports for the science programs. These presentations were held at the Wang Center, Stony Brook University. For structural biology, Wayne Hendrickson (Columbia University) demonstrated the interplay between x-ray diffraction methods and electron microscopy, he discussed the opportunity offered by careful examination of the scientific needs.
The second day focussed on the role of structure in cellular and molecular biology. To achieve dynamic interplay in discussion of biological questions we attempted to provide a program that covered macromolecular crystallography, Small Angle scattering and cryo-EM, imaging and the computing approaches needed to take advantage of these techniques.

The full agenda for both days is presented in the appendix, but briefly, Through the day several themes emerged from the presentations and following discussions. The importance of establishing new methods for study of dynamic processes in biological systems. The importance of solution studies of macromolecules, the continuing need to develop crystallographic methods. But most prominently the value of seizing the opportunity from advances in imaging and the associated improvements in software for analysis and partitioning of images. Each of these themes are touched upon in this report. The question of synchrotron capacity for the Macromolecular Crystallography community during the upgrades of sister facilities is addressed in an appendix.

To make most of the enormous quantities of data that will be generated, it is necessary to work with our funders such that the needs for data processing, analysis, management and curation meet agreed norms. Standardization of data management and analysis will contribute and ease the transition from acquisition through interpretation to publication.

As a community we aim to train the next generation of users by strengthening the workforce, widening the community of users fostering both academia, regulatory and industry. As well as pushing the outreach to make an economic impact in the local community by recruiting, retaining local staff who can contribute to these activities.

Science Questions/Challenges

Based on genome sequence analysis, there are ~200-300 essential biological processes in cells, all carried out by assemblies of molecules, or molecular machines. X-ray crystallography and cryo-EM have been successful in delineating atomic structures of these molecules and machines. *The next grand challenge is to understand how these molecules are organized in the cellular context to conduct specific cellular processes*, and how they are reorganized in different physiological or pathological states. In achieving progress toward this goal we need to know, for example, the structure of proteins in plasma membranes *in situ* and how it enables them to act as cellular gatekeepers. Given the diversity of the cellular function, the opportunities for break through knowledge are almost boundless, some examples:

- What is special about the structural organization of those molecules that function within membrane-bound organelles enabling them to perform specific functions?
- How are molecules organized in the cytosol as they transmit signals from the cell surface to internal components?
- What is now the structural basis of those molecules that form distinct aggregates, or membrane-less organelles?
- Disease state of cells may be morphologically different, if not more sophisticated imaging techniques are needed to allow for an understanding of the impact of disease on the cell function?
- Ultimately we need a four dimensional (3D over time) understanding of the structure, function and dynamics of molecules in a cellular context.

Thus we contend that our challenge is to bring together all the necessary techniques to allow us to develop a multi-scale continuum of dynamic cell structures from molecules to tissues in order to understand the interplay of individual proteins, their organization to the impact on the organism. In achieving this goal, we need to emphasize the importance of understanding molecules – their atomic structure, biological function and their dynamics, and furthermore, to place these molecules in a cellular context through the correlative data available through tomography, light microscopy and advanced data analysis.

The application of a suite of instruments would enable us to imagine a synthesis of more than thirty years of work in biological research areas as diverse as biomedical research and the development of bio-energy crops. As is apparent the societal impact of establishing the ideas discussed here will be profound, as we establish working models of cells and then organisms. An obvious, but important example, is that when the tools and methods described in this report are brought together the ensemble will enable new routes to drug discovery – by allowing the design of cellular assays for molecular treatments to be designed through knowledge of the target molecule's structure and a profound knowledge of the cells morphology.

The challenges to be overcome in this aspect of the research are manifold: data volume, data standardization, access to these data, provenance and versioning history, repeatability and the algorithms to recreate the jigsaw.

The scale of this endeavor is beyond the capacity of an individual PI's laboratory to support and fits well the ability of the National Labs to respond to grand challenges.

Priority Research Directions

We identified the need for the following Priority Research Directions in the area of molecular and cell biology that are relevant to synchrotron light sources.

Priority Research Direction #1: Molecular basis of cellular functions. Multiscale, multimodal, 3D visualizations of sub-cellular organelle structures in intact biological cells, with identifications and incorporations of biological macromolecular structures, in order to understand the molecular basis of cellular functions.

Priority Research Direction #2: Protein dynamics and functions. Visualizations of protein dynamics at the atomic level upon an initiating trigger of a biochemical reaction is crucial to the understanding of the protein function in a cellular environment.

Priority Research Direction #3: Artifact-free biological sample preparation. Sample preparation is vital to artifact-free measurements in cellular imaging. Further investments will be required to allow for cryogenic preparation of bio-samples, particularly for biological specimens as large as biological cells.

Priority Research Direction #4: Multimodal, multiscale data integration, visualization, and analysis. Data interpretation and analysis is the biggest bottleneck in the transition from "nice pretty images" to quantitative scientific interpretation. Multiscale data integration, data handling, and storage access are enormous challenges that need to be addressed.

New NSLS-II Capabilities

To stimulate further thoughts, follow-up discussions and sharing of ideas, Workshop participants identified the following new capabilities that NSLS-II should consider to pursue in the area of Molecular and Cell Biology.

Capability #1: Biological soft X-ray tomography:

Even simple images contain information equivalent to many days or weeks of biochemical and biophysical measurements. For example, you could characterize your pet cat's phenotype by making measurement, gathering experimental and genetic information, or you could simply take a photo of the cat. In this example, an image is an instantaneous, low-cost way to describe the cat's size, hair color, markings, and other phenotypic traits. This principle of using imaging to pull and transmit enormous amounts of information from a specimen also holds in cell biology. With the right microscope, researchers can instantly gain detailed insights on complex biological systems and do so with relatively little effort.

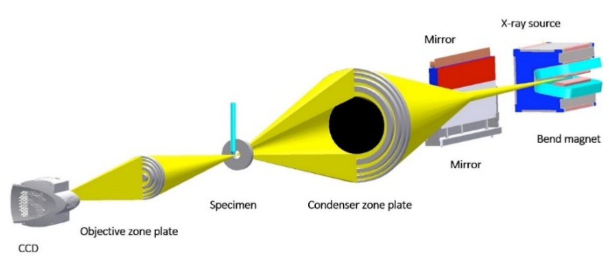


Figure 17. CAD showing XM-2 source, mirrors, optics and the beam path from the bend magnet source, through the specimen, and onto the detector.

Soft X-ray Tomography (SXT) is a relatively new modality for imaging cells. In the U.S., SXT was developed by the National Center for X-ray Tomography (NCXT) at the Advanced Light Source, Berkeley.SXT is a non-invasive, 3D imaging technique capable of measuring volumes, surfaces, interfaces, membranes, and organelle connectivity within an intact cell [1]. SXT data are collected on a transmission soft x-ray microscope, in our case, XM-2 at the ALS [2]. In biological soft x-ray microscopy, the specimen is illuminated with photons at energies within the '*water window*' (i.e., photon energies between the K-absorption edge of carbon at 282 eV and oxygen, 533 eV) [3]. The optical layout of XM-2 and path taken by the illumination through the specimen and onto the detector are shown Fig. 17.

As photons pass through the specimen, they are attenuated. Photons in the water window are absorbed an order of magnitude more strongly by carbon- and nitrogen-containing organic materials than by water (see Fig. 18). Image contrast is obtained directly from the measured attenuation of soft x-rays by the specimen [4, 5]. Consequently, subtle differences in biochemical composition produce measurable contrast in soft x-ray images. Image contrast is, therefore, a quantitative measure of the density and species of biomolecules in each voxel in the tomographic reconstruction. Each voxel in the reconstruction has an associated Linear Absorption Coefficient (LAC), shown in Fig.19. As can be seen in Fig. 19, it is easy to identify subcellular structures and features in a tomogram [6-11].

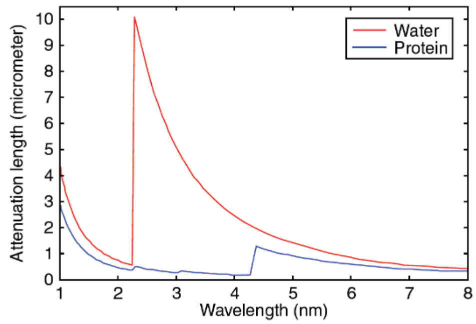


Figure 18. Attenuation of 'water window' soft x-rays by protein and water.

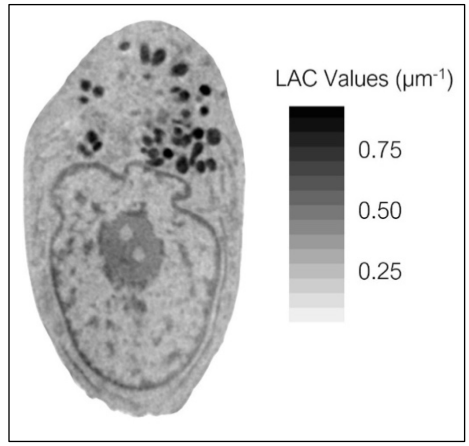


Figure 19. Single voxel thick 'orthoslice' through an SXT reconstruction of a cell. The grey scale corresponding to the measured LAC values shows the contrast range between dark carbon-dense objects and high-water content, low carbon regions.

Once SXT data – which consist of 90 or 180 projection images collected at angular increments around a central rotation axis – have been reconstructed, the tomogram can be segmented, based on the measured LAC values [12]. Segmentation is the process of isolating a specific region from the other cell contents. This can be done readily for all of the major subcellular organelles and structures, or a subset of organelles as shown in Fig. 20.

In summary, the most significant imaging characteristics of SXT are:

- High spatial resolution: Full-field imaging at a spatial resolution of 35 nm or better.
- Wide applicability: Hydrated cells up to 12 μm thick can be imaged intact.
- Quantitative image contrast mechanism: Each voxel in a calculated reconstruction has an associated Linear Absorption Coefficient (LAC).
- High Data Completeness: Specimens are mounted in thin-walled glass capillaries that can be imaged from any position around a central axis of rotation. Results in high completeness tomographic data, i.e., no 'missing wedge' as is the case with flat specimen holders and limited tilt (rather than full rotation) series.
- High specimen throughput: Acquisition of a full tomographic series (180 projection images) from a field-of-view takes a few minutes. A subsequent field-of-view can be imaged by simply translating the specimen-containing capillary in the Z direction. A field-of-view can contain tens or even hundreds of bacteria, five to 10 yeast cells, or one to three mammalian cells.
- Currently, the NCXT is the only facility in the US where biomedical researchers can image cells by SXT.

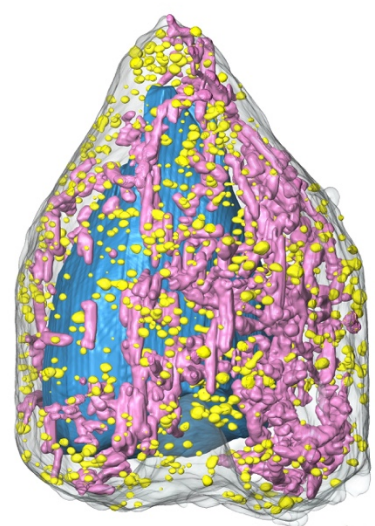


Figure 20. SXT of a pancreatic beta cell, segmented and color-coded to show the nucleus (blue), mitochondria (pink) and insulin granules (yellow), enabling quantitative measurements. There are 926 insulin granules, which occupy 2% of the cell volume; the nucleus occupies 30% and mitochondria 8% of the cell volume.

The success of SXT at ALS has spurred the growth of the technology, with similar microscopes being built in synchrotrons world-wide. Moreover, the huge demand for access to this type of imaging has resulted in several companies building 'table top' soft x-ray sources. America now has significant competition in this area and must keep pushing the boundaries of the technology.

Capability #2: Micro-MX for exploring protein functional states and dynamics.

Protein structures provide atomic-level 3-D details for understanding biological function and form the physical foundation for bio-designing and biomedical applications. By contributing to 90% of protein structures (160,000 structures as of writing) in Protein Data Bank (PDB)(RCSB 2019), X-ray crystallography has been the dominant method in protein structure determination. In typical protein crystals, 30-70% of the volume is water that supports the 3D structure of protein molecules in their crystalline states resembling their physiological structures. Therefore, exploring protein structures in their crystalline states is a powerful tool to understand protein function.

While the time signature (pulse length and instantaneous intensity) of free-electron laser x-ray sources is beyond the reach of current (and likely future) synchrotron sources, it is critical to realize that useful, impactful, fundamental time-resolved structural biology very likely will neither require picosecond/sub-picosecond time resolution nor sub-micron nanocrystals (as accessible to XFELs). The ability of MX beamlines at NSLS-II (AMX, and especially FMX) to determine structures of micron-dimensioned crystals on millisecond/sub-second time-scales opens up new experimental capabilities.

For the study of protein dynamics and enzyme kinetics, the high brightness of the NSLS-II MX beamlines translates to high photon flux in a ~1 μm focal spot, and thereby to time resolutions in the microsecond range - covering essentially the full range of enzyme kinetics [Bar-Even 2011]. The high data collection speeds also enable us to efficiently collect data at various temperatures up to room temperature, essential for the study of physiologically relevant processes.

The NSLS-II MX beamlines' experimental stations were developed to offer a high degree of flexibility and automation - a prerequisite to support a variety of serial sample delivery and reaction trigger methods. This has been demonstrated for different serial crystallography data collection protocols for fixed target sample supports [Guo 2018, Gao 2018, Guo 2019]. Together with our partner users from Arizona State University we offer viscous jet serial crystallography to our user base. Both methods are amenable to a variety of reaction initiations, to address different protein dynamics and enzyme kinetics questions. In addition to time-resolved MX, time-resolved SAXS (on the 16-ID LiX beamline) is also an exciting future direction, with triggering methods developed for MX studies likely to be directly adaptable for SAXS measurements.

Historically, time-resolved crystallography studies were limited by available beam properties, and by challenges due to the large required crystal sizes. This narrowed the available methods to the freeze-trapping of reaction intermediates with associated slow time-scales, and to light-excited repeatable reactions and the very limited number of chromophore driven systems amenable to this type of investigation. With micron-crystal serial crystallography methods now dramatically lowering the required crystal sizes, new reaction initiation methods – triggers – become feasible, thereby greatly widening the scope of dynamic systems that can be investigated at our beamlines. Extension to reaction initiation using the natural substrate molecules in small droplets ejected onto the crystals in a timed sequence was demonstrated by Mehrabi et al. [Nature Methods 2019]. Other reaction triggers beyond light and substrate molecules include temperature jumps, pH, oxygen and photo electrons. Further successful sample delivery methods beyond fixed grids are membranes, microfluidic structures, high viscosity and mixing jets, and capillary or droplet loaded conveyor belt systems. All these sample delivery and reaction triggering methods should be explored at NSLS-II to enable robust and automated time-resolved micro-MX data collection and subsequent heterogenous data and structure analysis.

Capability #3: X-ray footprinting to study proteins in solution.

Synchrotron X-ray footprinting (XF) at the NSLS-II XFP beamline allows structural biologists to, among other things, probe interactions of biomolecules with other biomolecules or small molecule targets, understand structural changes occurring upon complex assembly, and provide structural restraints for intrinsically disordered proteins. The XF method closely aligns with the global structural data obtainable by solution scattering at LiX, as well NSLS-II MX and Cryo-EM capabilities. Time-resolved XF is particularly well suited to address biomolecule dynamics problems by probing changes in solvent/surface accessibility during assembly of a macromolecule complex or binding of a target. This was nicely illustrated by recent work published in *Cell* using time-resolved XF to show a selectivity filter for GPCR-G protein assembly that could not be seen by MX or Cryo-EM. Careful attention to instrumentation design can ensure common solutions for time-resolved dynamics approaches are available at both LiX and XFP as we develop this capability.

However, the requirement for downstream sample processing and mass spectrometry data analysis represents a barrier to uptake of the XF method by the NSLS-II Structural Biology community. Development of biological mass spectrometry (MS) resources at NSLS-II (or nearby on the BNL site) will provide a crucially needed capability for screening of XF exposure conditions and a resource to couple beamline XF with training of the user community in MS sample processing and data analysis methods. Such an MS resource may also benefit other components of the NSLS-II Structural Biology community. Similarly, comprehensive wet-lab facilities for sample preparation and characterization prior to beamline experiments will ensure more effective use of beamtime. This could, for instance, take the form of capabilities for protein purification and characterization in a nearby user laboratory, allowing the community to appropriately poise unstable biomolecules right before beamline experiments.

<u>Capability #4: De novo structure determination for native macromolecules</u>.

The Protein Data Bank (PDB) currently holds nearly 160,000 macromolecular structures and over 10,000 are added each year, of which over 80% are X-ray crystal structures. At present, the vast majority of the new crystal structures derive from known PDB structures, either as isomorphous variants or by the method of molecular replacement. Completely novel structures, those without any known analogs, require *de novo* methods for crystallographic phase evaluation. Single-wavelength anomalous diffraction (SAD) is being used for the great preponderance of *de novo* structure determinations, and selenomethionyl proteins predominate for these SAD phasing evaluation (Se-SAD). Although selenomethionyl proteins can be produced quite conveniently in many cases, substitution levels are often limited for eukaryotic systems and this in turn limits effectiveness in structure analysis.

Fortunately, native SAD phasing has become feasible, relying on the intrinsic sulfur atoms in proteins and phosphorus atoms in nucleic acids, as well as metal atoms and other anomalous scattering ions that are often naturally present or introduced during crystallization. Native SAD experiments can succeed well as conducted on conventional macromolecular crystallography (MX) beamlines when using a relatively low X-ray energy (~ 7 keV), particularly if signal-to-noise ratios are enhanced by averaging from multiple crystals or multiple orientations (Liu, Dahmane et al. 2012, Weinert, Olieric et al. 2015).

Such experiments can be much markedly improved, however, through the use of lower X-ray energies (longer wavelength) and microcrystals to cope with attendant increased absorption (Guo, Zhu et al. 2019). A recent challenging example from NSLS-II is that of the native-SAD structure of a guanine nucleotide exchange factor Ric-8A (Zeng, Mou et al. 2019). By further technical research and development of a beamline optimized for routine operation at X-ray energies of 3-7 keV, we project that native-SAD procedures will replace Se-SAD to dominate *de novo* structure determination and analysis. Similar beamlines are already in operation at Diamond (ID-23) and Photon Factory (BL-1A), and a structure of the Sen1 helicase is another recent challenging example (Basu, Olieric et al. 2019). Since native SAD eliminates the need for heavy-atom derivatization or selenomethionine incorporation, structure determination is virtually assured when the macromolecule is suitably crystallized.

<u>Capability #5: Improved automation for structural biology</u>.

Robotic sample handling and other automation has become, over the last 15 years, the expected norm at synchrotron beamlines for macromolecular crystallography. This expectation is even embodied in the name of one of the NSLS-II MX beamlines, "Highly Automated Macromolecular Crystallography", although equivalent automation is also available at the FMX beamline. Automation is also an important facet of LIX for scanning and mapping of sample properties. The goal of all automation is to deliver equivalent or higher quality data in a shorter period of time.

Automation offers several advantages over manual data collection. It is significantly faster. Data from current synchrotrons indicates that a human can examine between 15 and 20 samples per hour, on average. Once automation is optimized, the NSLS-II beamlines can exceed 50 crystals per hour. This is a particularly important attribute, in order to maintain productivity in the field during the upgrades of the Advanced Photon and Advanced Light Sources between 2022 and 2026 (See Appendix). The speed of dataset collection at the NSLS-II MX beamlines is so much shorter than robotic sample handling and placement that traditional screening should disappear at NSLS-II during the next several years.

Automation also delivers, for most users, higher quality data. Analysis through software of diffraction is more accurate than the human eye. In addition, the human bias is to prefer samples that provide clean scattering patterns from single lattices or domains. There is ample evidence from the last 15 years, however, that many samples whose appearance is suspect can provide excellent data and structures. As was discussed at the workshop, effort needs to be made to improve the statistical relevance of the metrics employed in characterizing crystals and datasets. One example presented compared the quality of metrics used to determine the effectiveness of a collection strategy on the obtained dataset. While a statistical analysis of the raw data demonstrated that two datasets, one with a strategy, one without, were virtually identical, all standard metrics (on scaled data) favored the dataset with a strategy. More work in this area is required in order to have robust collection protocols and statistical accuracy.

The staff of the NSLS-II beamlines recognizes the efforts required to improve automation. The current time to exchange samples on the MX beamlines is ~35 seconds. The state of the art is below 20 seconds. This difference has a major impact, when integrated over the tens of thousands of samples. For microcrystals, the time to raster to locate crystals remains slow, and needs to be at least halved from its current level. Optimization of automation also requires that as many actions be done in parallel. NSLS-II has sufficient expertise in the development of multi-threaded software to achieve this goal. Additional capabilities, such as automatic introduction of ligands into crystals, similar to the XChem facility at Diamond Light Source, and data collection at room temperature from trays are in current development or being considered for the facility. The workshop participants encourage the creation of such capabilities in advance of their need.

For a major subset of experiments, significant time savings can be achieved through the use of automatic mount placement (loop centering), which is the first step in an automatic raster. Currently one third of the academic crystallography proposals are for drug (or inhibitor) development. These types of projects generally yield larger crystals for which rastering is not necessary. Other projects from the university community have similar needs. When a raster is required, such as for microcrystals encased in lipidic cubic phase, there remains significant suspicion of the automatic implementation in even expert parts of the community. NSLS-II should take the lead in demonstrating to users that automation does not compromise quality.

Biosystems, Earth and Environmental Science

Scientists utilizing synchrotron resources for Biosystems, Earth and Environmental Science (BEES) research seek to understand the physics and chemistry of natural systems where controlling reactions and components are often poorly constrained, *a priori,* and heterogeneity dominates. These scientists study planetary bodies, including Earth, from crust-to-core. Their studies target minerals, fluids and organisms that influence metal transformations and mobility within planetary bodies, in the environment and within biological systems. Biological interactions with Earth materials can modify reaction pathways and play a critical role in the processes that drive the flow of mass and energy within geologic media and ecosystems. The nature of BEEs research requires the study of small samples, which are often heterogeneous on the nm-scale. Yet the observations made are used in understanding phenomena from molecular to planetary length scales. Increasingly, our ability to successfully model and understand chemically and physically heterogeneous global systems relies on access to methods that allow us to bridge these length scales analytically and conceptually.

The breadth of research conducted by the BEEs user community utilizing synchrotron techniques is too broad to be adequately summarized in a brief summary such as this. Synchrotron-based techniques utilized by this community include imaging, scattering, and spectroscopy, utilizing both focused-beam and full-field methods, applied at both ambient and extreme conditions, across the entire energy range provided by facilities world-wide, from the softest to the hardest energies. While recognizing that the summary presented here only captures a small segment of the research being conducted by this user community, the NSLS-II New Science Frontiers workshop participants identified six primary focus areas where they believe NSLS-II resources can make a significant contribution:

- *Understanding the physics and chemistry of materials that comprise Earth, solar and extrasolar planets, often under very extreme conditions*.
- *Modeling the petrogenetic evolution of planetary and asteroidal systems*.
- *Understanding the intrinsic controls over heterogeneous reactivity of elements at mineral interfaces*.
- *Understanding the long term health and environmental impacts of environmental contaminants*.
- *Understanding how changing climates affect fluxes of contaminant and nutrient chemical species in soils*.
- *Unraveling controls on ocean productivity by trace metal micronutrients*.

Workshop participants also identified facility technical advances they believe would facilitate research in these focus areas, including:

- Increasing needs for next generation full-field analysis including tomography and bulk-spectroscopy,
- Next-generation spatially-resolved synchrotron beamlines (at both the micrometer to nanometer scale) with multi-modal capabilities,
- Additional resources for conducting experiments at relevant geologic conditions, including at extreme conditions and within experimental cells.

A pressing need exists for additional capacity and for dramatic increases in sample throughput to provide more robust statistical characterization of Earth materials. This last point highlights the increasing need for experiments that require large numbers of analysis, for example to allow scientists to generate more quantitative models of how element cycling in the atmosphere and oceans couples to climatic variations. This includes not only improvements in instrumentation for high throughput analysis, but also computational advances such as multivariate modeling for quickly interpreting large amounts of complex data that will be generated.

Science Questions and Research Directions

Science Question and Research Direction #1: Understanding the physics and chemistry of materials that comprise Earth, solar and extrasolar planets, even under very extreme conditions.

Synchrotron techniques have proven invaluable in allowing Earth scientists to understand the fundamental physics and chemistry of the materials that comprise Earth, solar and extrasolar planets, even under very extreme conditions. This research is fundamentally important in placing mineralogical context to observations made from remote sensing and seismology. Synchrotron measurements of materials under extreme conditions have become critical for testing models for planetary formation and planetary evolution.

The Mineral Physics research being conducted by BEEs researchers often requires the brightest possible X-ray beams with relatively high incident beam energies. They also require the need to measure changes in the morphology, crystallography, scattering behavior, chemistry and seismic properties of samples at extreme pressures and temperatures. Combined, these types of data help Earth scientists constraint the physical and mechanical properties (i.e., elasticity, strength, etc.) of possible mineral phases at the extreme conditions at which they form within planetary bodies. Currently utilized MultiAnvil technologies allow for in-situ diffraction and radiographic imaging at pressures exceeding ~60 GPa and at temperatures of ~2000°C in volumes that vary between ~0.01-1 mm^3. In-situ deformation can also be studied at pressures up to ~20GPa in current devices at strains up to 60%. However, in order to study rheological properties of (Earth) materials under these extreme conditions, new experimental apparatus are needed that will overcome several experimental limitations that currently exist. For many current studies, X-ray diffraction is a rate limiting step, requiring beamlines with higher flux and improved detectors. New studies also increasingly seek to move from measuring static to dynamic processes, including mineral phase transitions, melting, acoustoelastic effects, etc. as P and T are varied in real time. There is also a need to analyze larger samples than is currently feasible under relevant conditions and tomography capabilities at high pressure are highly desirable.

NSLS-II provides new opportunities for synchrotron X-ray high-pressure studies in Large Volume Multi-Anvil Apparatus that will overcome many of the current limitations. For example, the proposed High Energy Engineering X-ray (HEX) beamline will utilize a superconducting wiggler source with potential for both white beam and mono beam studies. This will provide a significant increase in flux with beam sizes significantly larger than currently available. This will enable new scientific programs for studying stress – strain – time relationships in planetary bodies, elastic, anelastic, and plastic properties, kinetics of phase transitions, viscous properties and equation of state for materials, to only name a few pressing questions. Practical applications of new knowledge may include understanding metallurgical and ceramic phase relations at extreme conditions for developing new applied materials, improving our understanding of the physics of fracking / fracture propagation for enhanced energy production, and for developing new carbon sequestration technologies.

Science Question and Research Direction #2: Modeling the petrogenetic evolution of planetary and asteroidal systems.

Crystalline mineral grains and included amorphous magmatic glasses that comprise terrestrial igneous rocks and igneous materials found in meteorites, comets, asteroids, and other planetary bodies contain chemical records of the conditions under which they formed. Deciphering these records can lead to improved understanding of the origin and evolution of the Earth and our Solar System. The oxygen fugacity (fO2) of a magmatic system in particular exerts a first order effect on the crystallization path of magmas and a measure of fO2 at crystallization can provide insights into magma formation and processes that operated on the system before crystallization. Changing fO2 exerts a primary control on the valences of multivalent elements (Ti, V, Cr, Fe, and Eu) that are preserved in magmatic mineral phases and glasses, such that a measure of elemental valence state can then be used as proxies for the oxidation states of the environments from which these mineral grains formed.

This approach has been used on a variety of extraterrestrial materials at relatively coarse spatial scales, but many of the most interesting and relevant phases in understanding early solar system evolution are micrometer scale and smaller. Thus there is an increasing need to have spectroscopy beamlines that can provide not only excellent spectroscopic data for measuring the valence of these multivalent elements, but also for focused beam X-ray absorption spectroscopy of these phases at nanometer spatial scales, at very low elemental abundance and at the extreme conditions relevant to those active at the time of crystallization. For example, beamlines capable of conductive these spectroscopic analyses within hydrothermal and diamond anvil cells at elevated P and T and with varying solution chemistry.

There are also opportunities for developing and applying high energy resolution fluorescence detection XAFS (HERFD-XAFS, ≤ 1 eV) at the micrometer spatial scale for these studies. HERFD-XAFS is achieved by using crystal analyzer spectrometers for measuring the fluorescence XAFS rather than solid state charge discrimination, and can allow peak overlaps to be minimized since only a narrow region of the fluorescence line of interest is integrated. Consequently, backgrounds can be reduced and/or eliminated, potentially allowing XAFS analysis of the multivalent elements of interest at much lower concentration. Another impactful characteristic of HERFD-XAFS is that it reduces core-hole lifetime broadening of XAFS spectra measured in fluorescence mode so that spectroscopic features are dramatically sharper, facilitating interpretation of subtle differences in valence state and speciation.

Science Question and Research Direction #3: Understanding the intrinsic controls over heterogeneous reactivity of elements at mineral interfaces.

Chemical interactions that occur at mineral surfaces often play a crucial role in controlling the catalytic and geochemical behavior of reactive elements in terrestrial surface, subsurface and marine environments. Such chemically reactive mineral interfaces are also important in material science, particularly in regard to emerging energy applications. X-ray interface and surface scattering methods in particular have emerged as powerful techniques for investigating the hierarchical and chemical complexity of mineral interfaces. Surface and interface science techniques, including Crystal truncation rod (CTR) diffraction, Resonant Anomalous X-ray Reflectivity (RAXR), Long Period Standing Wave Fluorescence Yield (LPSW-FY) Spectroscopy, and Grazing Incidence XAFS measurements, provide valuable insights into structures of complex interface systems. For example, these methods have provided new insights in to the sorption behavior of onto solid surfaces from aqueous solutions under conditions relevant to their waste storage and environmental transport.

Opportunities were identified by workshop participants for expanding the breadth and applicability of these techniques in support BEEs research. This included broadening the range of mineralogy to which the techniques can be applied by developing beamlines and instrumentation that can support in situ electrochemistry, microcrystal surface measurements, conducting measurements under geochemical confinement. Upgrades in instrumentation and optics have the potential to enable rapid energy scanning for faster spectroscopy, improvements in spatial resolution and coherence of these techniques expands the range of minerals which can be studies and the provides a potential for coupling these surface sensitive techniques with coherent diffraction imaging.

Science Question and Research Direction #4: Understanding the long term health and environmental impacts of environmental contaminants.

Synchrotron techniques have had a profound impact on the study of contaminant chemical species in the environment. Once deposited to the environment many contaminants, including heavy metal species, actinides, high abundance anthropogenic metals, organic compounds, etc., can undertake transformations to different chemical forms depending on the reactivity and biogeochemical properties of the biogeochemical environment. Properly evaluating a contaminant's bioavailability or bioaccess-ibility requires BEEs researchers to determine both the distribution and speciation of the contaminants and their relationship to the minerals, inorganic/organic components and organisms with which they interact. Synchrotron techniques such as XAFS spectroscopy, X-ray diffraction and scattering, and soft and hard X-ray imaging at nanometer to micrometer resolutions are used by these researchers to analyze the chemical species and crystalline or amorphous phases with which they interact, typically as complex mixtures within media that is heterogeneous at the nanometer scale. For example, synchrotron micro-spectroscopy and diffraction allow these researchers the ability to characterize of both the speciation of the contaminants and the mineralogy of the solid phases to which they are sorbed or incorporated. Constraining these properties is critical for understanding mechanism that may result in the potential release of contaminant to water and results of these types of studies have been used in shaping official government policy to protect our environment.

Opportunities were identified by workshop participants for expanding the breadth and applicability of these techniques in support BEEs research. Particularly emphasized was the need for multi-modal, combination methodologies. The ability to seamlessly transition from, for example, microXRF (element mapping), microXRD (crystal structure identification) and microXAFS (oxidation state, bonding environment) provides the optimal suite of information for fully characterizing contaminant behavior. Workshop participants also highlighted how improvements in beamline brightness, detectors, automation and computing are now driving the development of user research projects that were not previously feasible incorporating long-term, multi-year research projects where large numbers of sample analyses are required, potentially with re-analysis of samples over long periods of time.

For environmental research, such multi-year studies incorporating hundreds or thousands of analyses are needed for developing robust reference databases and statistical models for predicting contaminant behavior over relevant time frames in the environment. There is thus a pressing need for further increases in sample throughput through unattended automation of sample analysis, further improvements in detector performance. The large datasets generated by such studies also require developments in computational analysis and modeling since it becomes difficult for users to evaluate such large datasets efficiently. This includes advances that combine automated data analysis, modeling and data mining. Software development using multivariate modeling for quickly interpreting the large amounts of complex data that will be generated would be useful.

Science Question and Research Direction #5: Understanding how changing climates affect fluxes of contaminant and nutrient chemical species in soils.

Worldwide, economic growth has brought up approximately 80,000 synthetic chemicals over the last 50 years. Although being beneficial for human society, e.g. as nutrients supporting soil quality and plant growth, they also are responsible for a large number and quantity of contaminants dispensed into the environment. Several thousand new chemicals are reviewed by the EPA every year and many are identified as hazardous. The fate and exposure of contaminants in soils depends on their chemical properties, speciation, and on soil properties such as temperature, organic matter content, or microbial activities. These properties are sensitive to aspects of climate change such as rising temperatures, extreme rainfall, extended dry periods, soil erosion, and a rise in sea level. Processes involving loss in soil organic carbon, changes in redox state, and alterations in the microbial community could increase human exposure to soil contaminants.

The availability and mobility of both, nutrients and pollutants, depend on their adsorption and desorption in soils, which are strongly related to soil organic matter. Nitrogen, for example, is a very important nutrient in soil. Soils contain most of the nitrogen pool of terrestrial ecosystems, and regulate its export to the hydrosphere and the atmosphere. Climate warming and continuing high nitrogen inputs will reduce the retention capacity and the overall retention in soils, thus increasing soil nitrogen losses and nitrogen eutrophication of aquatic ecosystems. The lability and mobility of nitrogen in soils differ for different species; therefore, a full characterization requires a detailed knowledge of the nitrogen speciation of soils, stages of soil development, and degrees of nitrogen saturation. Current techniques of nitrogen speciation (e.g. wet-chemical methods, ^{15}N NMR) are unsatisfactory, whereas synchrotron radiation-based spectroscopy and microscopy is a promising tool for a direct, unbiased speciation of soil nitrogen.

Two important groups of contaminants and pollutants in soils are heavy metals and organic compounds.
Heavy metals and metalloids are a group of pollutants which can be dangerous to human health. Many of these metals are vital in very small amounts, but become toxic in larger amounts. They can accumulate in soils, water and the sediments of lakes and rivers, because they do not break down in the environment. it is important to evaluate the contribution of natural emission sources in the environment. Therefore, soils and sediments become reservoirs for a potential secondary release of these metals. Such a release naturally depends on climate conditions and change accordingly. For instance, rising temperatures cause the soil water content to decrease while evapotranspiration increases. As a consequence, soil dust particles carrying heavy metals are resuspended in larger amounts. Extreme rainfalls change the soil chemistry, potentially washing out formerly bound metals.

Persistent organic pollutants represent one group of organic compounds comprising toxic substances produced by industry and released to the environment by human activities. They include PCBs, pesticides, and chemicals such as dioxins and furans. They are distributed globally due to their volatility and persistence in the environment. Long-term data generally displayed decreasing levels but with rising temperatures, these pollutants might return to the atmosphere or being mobilized into aquatic systems. Within the environment, these pollutants are resistant to degradation, and can accumulate over long periods of time. A temperature gradient resulting in a cold-trap has been proposed as a leading mechanism to enhance this accumulation, especially in permafrost, glacial, or arctic environments. Climate change may alter the stability of reservoirs created in that way over time. A release under climate warming may pose risks to the environment and a new challenge to governments and scientists.

While being effective already at very low concentrations down to trace elemental levels, these very small amounts of pollutants and contaminants are a major problem in investigating their fate. Here, using synchrotron radiation-based techniques has a distinct advantage over many other methods in that element-specific analysis can be carried out at close to in-situ conditions (e.g. pH, temperature, or redox state), and at low concentrations in complicated environmental matrices (e.g. soils or sediments). Understanding and possibly predicting the reactivity of trace elements in the environment is key to a true comprehension of processes in the environment.

New state-of-the-art beamlines allow for a combination of very high spatial and temporal resolution. Due to the relatively small number of these beamlines, access to this instrumentation can become a serious bottleneck. In addition, data management, analysis and conclusions are affected by limited computational capacity as well as by the lack of solid data analysis software packages.

Science Question and Research Direction #6: Unraveling controls on ocean productivity by trace metal micronutrients.

The cycling of metals between geologic environments, the ocean and atmosphere on Earth are complexly interrelated on a global scale. Understanding controls on this cycling with respect to mineral phases, marine organisms, micronutrients, and ocean waters aqueous solutions is critical to developing a mechanistic understanding of how competition for metals alters marine productivity and the ocean's role in the global carbon cycle. Trace metal micronutrients such as iron control primary production in approximately 30% of the ocean, yet it remains unclear what primary fluxes for these metals are, how their cycling changes as a result of changing anthropogenic input, volcanic activity and changing global climate.

For example, researchers are actively utilizing synchrotron methods to constrain how large the flux of "hydrothermal" iron to the world's oceans is from deep sea volcanism and how readily Fe can travel from the vent system, the Fe transport potential. This is largely a factor of a number of biotic, abiotic and inorganic reactions in the marine environment that can be studied using synchrotron techniques, including varying hard and soft X-ray spectroscopies, imaging and scattering methods. Constraining these reactions and their efficiency is challenging using traditional elemental analysis techniques due to the low metal contents and the small size of relevant marine particulates and micro-organisms (ca. 500 nanometers). High brilliance bulk- and micro-focused X-ray spectroscopies provide a powerful set of methodologies to measure metal quotas of samples at relevant particle sizes. Workshop participants emphasized that these studies benefit significantly from multi-modal approaches, using instruments that can interrogate samples using soft, tender hard X-ray energies, at multiple spatial scales, and with high throughput.

Science Question and Research Direction #7: Understanding the chemical and physical heterogeneities of contaminants and pollutants and their impact on our environment.

An in-depth examination of single particles in the colloidal size range is a common and important need for research in environmental science. Due to the chemical and physical heterogeneity and the impact of contaminants and pollutants even on trace elemental levels, the necessity to study single particles becomes obvious. The chemical heterogeneity is reflected in the composition of these particles, comprising very often organic and inorganic components at the same time. Related areas of science such as soil science, hydrogeology, geoscience and planetary science very often encounter the same challenge of heterogeneity down to the single particle level. In environmental sciences alone, biogeochemical processes, including sorption and desorption, complexation, dissolution and precipitation, as well as uptake or release by biota, control the mobility of nutrients, pollutants and contaminants, and thus the size, shape, and composition of single particles.

Considering the variety of sample composition and size, versatility is a key factor for the design of a scanning transmission X-ray microscope (STXM) beneficial for environmental research. Generally, a STXM is a microscope working in an X-ray energy range of approx. 200 eV to 2500 eV, delivering routinely a spatial resolution below 100 nm. The accessible energy range allows for X-ray spectroscopy measurements at the K-absorption edges of carbon, nitrogen, and oxygen, as well as at the L-edges of transition metals such as chromium, manganese, iron, nickel or copper. While the former are important for the study of organic constituents of complex agglomerates from the environment, the latter are relevant for many of the inorganic components. The importance of a STXM for such investigations is completed by its capability to study significant elements such as silicon, phosphorus or sulfur at the high-energy end of the accessible X-ray range. Specific research directions may include the following:

Aerosols. Aerosols are particulates found in the lower and upper atmosphere, sub-µm to 10s of µm in size and comprising an extremely heterogenous composition. Many aerosols are built up of an inorganic core and an organic coating around it. The inorganic core contains very often NaCl, originating from sea spray off the oceans, and sulfur, which is anthropogenic in its origin. The core-shell chemistry of such a particulate depends strongly on the overall morphology. This morphology has a strong impact on our climate, e.g. cloud formation, but as well on human health, e.g. through uptake in lungs. A STXM has the necessary spatial resolution to study the morphology, it can be used to study the organic chemistry within single particles, and it can address the inorganic core if made of low-Z elements such as chlorine or sulfur.

Climate history. Airborne dust particles found in deep ice cores provide the best information on global environmental conditions going back many thousands of years. Typically, only very small quantities of dust particles, comprising quite often organic and metallic components, can be liberated from the ice, which furthermore show µm to sub-µm size and a very high complexity chemically and physically.

Geomicrobiology. Microorganisms in aqueous environments are capable of taking up and concentrating large quantities of metals, directly influencing the distribution, mobility and bioavailability of trace and contaminant metals alike. In nature, they often congregate on surfaces to form biofilms, which are complex assemblies of multiple microbial species and extracellular polymeric substances, the latter forming the actual film adhering e.g. to mineral surfaces. Due to the size of single bacterial cells, and due to the necessity to identify organic compounds and the chemical state of the metals, a STXM is extremely well suited for the examination of such biofilms.

Marine chemistry. Iron originating from hydrothermal sources is a major source of this metal spreading into the world's oceans. The transport is largely a result of a number of biotic, abiotic and inorganic reactions in the marine environment, creating organometallic particulates in the colloidal size range. The high spatial resolution and the ability to perform spectroscopy on the organic and inorganic components makes a STXM an ideal instrument for these studies.

Nuclear waste. The management of nuclear waste is one of the most important environmental issues in the coming years. For a comprehensive understanding of the impact to the environment, research is necessary on radionuclide interactions with environmental materials such as minerals, microorganisms and natural waters to predict their mobility and speciation in the environment.

Nutrient cycling in soils. Nitrogen is a very important nutrient in soils, which contain most of the nitrogen pool of terrestrial ecosystems. The lability and mobility of nitrogen in soils differ significantly for different species, thus a full characterization requires a detailed knowledge of the its speciation. Using the accessible X-ray energy range and factoring in the size and shape of soil particulates, a STXM is very well suited for an unbiased speciation of soil nitrogen.

Planetary science. The identification and characterization of particles originating from planetary bodies and of interplanetary dust particles is a critical component of NASA's effort to understand better the origin and the geochemistry of our solar system. Interplanetary dust particles, which are ~5 to 50 micron dust from asteroids and comets, are aggregates of >10,000 individual, generally sub-micron size minerals and organic matter. The small size of the individual organic units requires analyses at the sub-50 nm size scale.

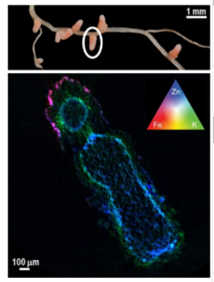
Waste in mining and industrial production. Worldwide and as a result of ore and mineral mining, oil drilling as well as industrial production in general, society is left with large and heterogeneous areas of contaminated land. The nature of pollutants and contaminants and their environmental behavior is very specifically linked to the production processes or the mined materials. Bulk chemical analysis might deliver information about the presence, number, and amount of contaminants, but more specific data are needed to determine the environmental fate, the bioavailability and potential transport risks. Not only metals or metalloids are involved in these processes but organic materials as well. The latter originate from the production or mining processes or constitute the microbial life interacting with these pollutants and contaminants.

New NSLS-II Capabilities and Impacts

BEEs user community members and workshop participants provided input on their utilization of existing capabilities across the DOE synchrotron complex and also highlighted facility technical advances for NSLS-II that they believe would facilitate novel research in the BEEs focus areas and research arena. The table below provides a qualitative list of synchrotron beamlines across the DOE complex that the community has identified as the most highly utilized for BEEs research currently.

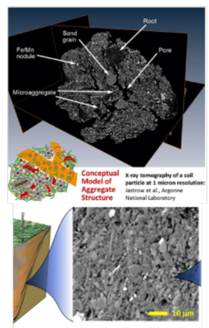
NSLS-II capabilities generally cover as broad a range of techniques relevant to BEEs researchers as is found in the other three DOE synchrotron facilities. NSLS-II hosts a family of spatially-resolved beamlines (XFM, TES, SRX, HXN) for X-ray micro/nano-analysis across a broad range of energy and spatial scales. Many of these instruments provide spectroscopic capabilities and some bulk XAFS capabilities are now becoming available that are suitable for BEEs research focus on three-pole wiggler sources (BMM and QAS), but the community perceives a need for additional resources for these types of studies. In particular, the community strongly recommends development of high energy resolution fluorescence detected XAFS capabilities (HERFD-XAFS) and expresses an urgent need for spectroscopy and imaging capabilities in the soft X-ray range throughout the DOE complex. Soft X-ray spectroscopy is required for constraining carbon speciation in natural materials and NSLS-II and ALS are in the best position to add resources to cover this need. The XPD beamline provides excellent capabilities for

Tender X-ray Nanoprobe



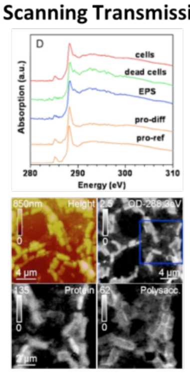
Planned Experimental Capabilities:	
X-ray fluorescence imaging	
Fluorescence microtomography	
X-ray phase contrast imaging	
XANES imaging and spectroscopy	
Specifications:	
Energy	2 – 12 keV
Spatial resolution	30 nm; 100 nm
Flux	10^{10} ph/s
Sample Environment	Helium or vac (Cryo capable.)
Energy selection	Si(111)
Source	IVU
Competitors	APS Bionanoprobe (hard X-ray)

X-ray Microtomography (XCT)



Planned Experimental Capabilities:	
Computed Microtomography	
Absorption-edge Tomography	
Contrast Radiography	
Custom Environment Chambers	
Specifications:	
Energy	10 – 30 keV
Spatial resolution	Micrometer
Flux	10^{11} -10^{13} ph/s
Sample Environment	In-situ capable
Energy selection	Si(111) or multilayer
Source	Broadband
Competitors	APS 13-BM & 2-BM

Scanning Transmission X-ray Microscope (STXM)



Planned Experimental Capabilities:	
Imaging C biomolecules and metals	
C 1s edge nanospectroscopy	
Transition metal L-edge nano spectroscopy	
Cryogenic sample handling	
Specifications:	
Energy	0.2 – 2.4 keV
Spatial resolution	30 nm
Flux	10^{11} ph/s
Sample Environment	Helium or vac (Cryo capable.)
Energy selection	Grating
Source	IVU
Competitors	ALS 11.0.2

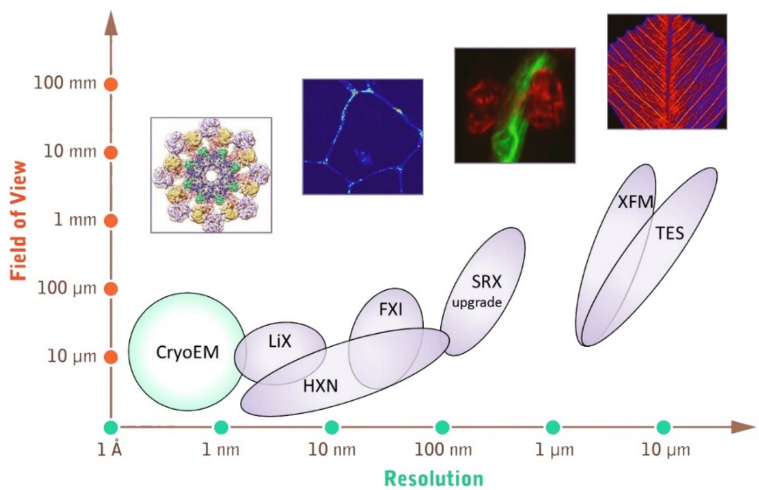
	NSLS-II	APS	ALS	SSRL
Hard X-ray nanoprobe	HXN (3-ID) & SRX (5-ID)	2-ID, 9-ID		
Tender X-ray nanoprobe				
Hard X-ray microprobe	XFM (4-BM)	8-BM, 13-ID & 20-ID	10.3.2 & 10.3.1	2-3, 6-2 & 10-2
Tender X-ray microprobe	TES (8-BM)		10.3.2	14-3
Hard X-ray absorption spectroscopy	BMM (6-BM) & QAS (7-BM)	9-BM, 10-BM, 10-ID & 20-BM		2-2, 4-1, 7-3, 9-3 & 11-2
Soft or Tender X-ray spectroscopy	SST 1&2 (7-ID)	9-BM	9.3.1 & 9.3.2	4-3, 8-2 & 10-1
Microdiffraction	XFM (4-BM)	13-ID & 34-ID	12.3.2	
X-ray diffraction & scattering	XPD (28-ID) & QAS (7-BM)	5-BM, 11-BM & 16-BM		2-1 & 11-3
High-pressure XRD/ Extreme conditions	XPD (28-ID)	3-ID, 6-BM, 13-ID & 16-ID	7.3.1 & 12.2.2	
Micro-CT		2-BM & 13-BM	8.3.2	2-2 & 11-2
Transmission X-ray microscopy	FXI (18-ID)	32-ID	2.1 & 6.1.2	6-2
Scanning transmission X-ray microscopy			11.0.2 & 5.3.2	
sFTIR spectromicroscopy	FIS (22-IR)		1.4, 2.4 & 5.4	

powder diffraction studies relevant for BEEs studies and multimodal X-ray microdiffraction that are being developed on NSLS-II spatially resolved beamlines will provide an excellent set of resources for characterization of the heterogeneous mineralogies observed in BEES research materials.

However, the community did identify opportunities for providing more resources than are currently available at DOE synchrotron facilities for characterizing BEEs-relevant materials using surface scattering and diffraction techniques. They also identified needs to build increased capacity for beamlines that can apply diffraction, spectroscopy, and imaging at extreme pressure and temperature conditions, as the current suite of beamlines that focus on these types of studies at the APS and ALS are all highly oversubscribed.

In fact, one central theme from this exercise is the need for "additional capacity", particularly for high-throughput type experiments that are not necessarily flux-limited. The community in particular identifies a need for additional capacity for bulk and spatially-resolved synchrotron beamlines with multi-modal capabilities. Throughout the DOE complex, the beamlines most highly utilized by the community are commonly 2-3x oversubscribed. It would be highly beneficial to have increased capacity for these types of studies at NSLS-II, particularly given that the APS facility will be dark in 2022-2023 and the ALS facility will be dark in 2024-2025.

With respect to spatially-resolved probes, considering the SRX upgrade from 1 micron to 0.1 micron focusing, a potential resolution gap was identified at the ~ 1 micrometer scale at NSLS-II, a spatial resolution that is highly desirable for BEEs research since it couples well to resolutions currently routinely provided by other instrumentation methods highly utilized by the community (wavelength dispersive electron microprobe, benchtop FTIR, laser ablation ICPMS, SIMS). This gap could be filled through development of an additional undulator-based hard X-ray microprobe at NSLS-II. Much of the infrastructure (front end, hutches, utilities) for such instrument exists currently at sector 5. Facility investments in source and photon delivery would be needed to realize this undulator-based microprobe at NSLS-II. In addition to adding capacity, this instrument will be essential to identify experiments that truly require <100 nm (SRX) or <10 nm (HXN) resolution. An undulator-based hard X-ray microprobe would have broad application in all BEEs focus areas and beyond. Addition of cryo-XRF capability to the endstation would further increase its utility and could be funded by BER contributions to the NSLS-II Center for Biomolecular Structure's Bio-Imaging Core.



In addition to increased capacity, *new capabilities* needed for BEEs research at NSLS-II include:

- Tender-energy nanoprobe,
- X-ray microtomography, and
- Scanning transmission X-ray microscope.

NSLS-II source characteristics are well suited for development of nanofocusing beamlines and the medium energy storage ring is ideal for the tender energy range.

New Capability #1: Tender X-ray nano-probe with cryogenic capabilities

- Interrogate element distributions and speciation at bio/mineral interfaces
- Nano-mm scale chemical and mineralogic imaging and characterization
- High energy resolution spectroscopies.

New Capability #2: Computed microtomography (XCT) with in-situ cell infrastructure

- Image fine-scale structure and distribution of fluids (aqueous, organic)

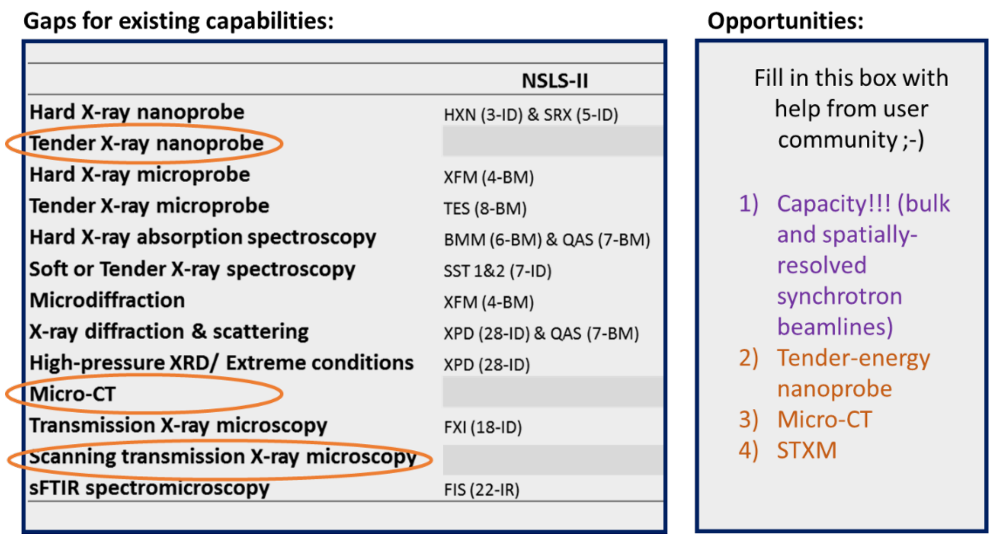
- Study alterations due to perturbations (hydration cycling, dissolution)
- Identify distributions and properties of nano-porous phases.

New Capability #3: STXM with cryogenic capabilities and fluorescence detection

- Understanding the role of organic matter in biogeochemical cycling
- Evaluating carbon speciation at the nanometer length scales.

Other opportunities identified by BEEs include additional resources for conducting experiments at relevant conditions, including at extreme conditions and within experimental cells and requirement for dramatic increases in sample throughput to provide more robust statistical characterization of Earth materials. This would involve technological and computational advances for statistically responsible sampling and analysis.

Both the additional capacity and the new capabilities at NSLS-II, if realized, will have significant impact in all BEES research areas as discussed in the previous sections. The additional capacity at NSLS-II will increase community productivity, and will be essential to maintain a current level of productivity by the BEES community during the APS and ALS facility upgrades (2022-2025).



New capabilities at NSLS-II will have long-term impacts in BEES research. Tender X-ray nanoprobe with full cryogenic sample handing capabilities is required for interrogating trace element distributions and speciation at bio/mineral interfaces and for spatially-resolved chemical and mineralogic characterization at nano-mm scales. X-ray Computed Microtomogarphy (XCT) is needed to image fine-scale structure and distribution of fluids (aqueous, organic) and study alterations due to perturbations (hydration cycling, dissolution). A Scanning Transmission X-ray Microscope (STXM) for 'soft matter and environment' is needed to address many questions related to biogeochemical cycling of nutrients and contaminants in the marine and near-surface terrestrial environments. These new capabilities will have broad impact in all BEES focus areas and beyond.

Appendices

Workshop Charge and Welcome Letter

Dear NSLS-II Workshop Participants,

Thank you for your participation in our upcoming NSLS-II Strategic Planning Workshop *Exploring New Science Frontiers at NSLS-II*, Monday through Wednesday, October 21-23, 2019. The workshop will start on Monday at Stony Brook University's (SBU) Wang Center with plenary presentations and will move to Brookhaven National Laboratory on Tuesday and Wednesday with parallel sessions and a close-out plenary. The agenda is now finalized and updated on the workshop website (https://indico.bnl.gov/event/6840/, and an Agenda-at-a-Glance is attached for your information.

As you may be aware, next week is the five-year anniversary of our first light on October 23, 2014. A lot has happened in the past five years, and our facility is now running at 400 mA, with 28 beamlines in operations and 1 beamline under construction. At the same time, science has advanced, and in some areas has evolved rapidly. Thus, we feel that this workshop is very timely in looking forward to the future, and to ascertain how NSLS-II should further develop to meet the research needs in the evolving scientific fields.

Given this context, we have designed the agenda of this workshop around six science areas: quantum materials, materials and engineering science, catalysis and chemistry, soft matter, molecular and cell biology, and biosystems and earth/environmental sciences. Each area is led by two discussion leaders – one from NSLS-II and one from the external community. These co-leaders have done a tremendous job in organizing the sessions for the science areas, and we are truly grateful for their effort to make this workshop happen.

All speakers and all participants are encouraged to keep in mind the following discussion points:

- **Big Science Questions**: What are the current grand scientific and technological challenges in each of the science areas and related applications?
- **Research Needs:** What research and research capabilities are needed to address the big science questions?
- **NSLS-II Capabilities**: What new scientific capabilities (instrumentation, techniques, analysis, capacity, etc.) should be developed at NSLS-II to meet the research needs in this area?
- **Impact**: What is the expected impact of these new capabilities on the grand challenges in the specific scientific field and/or in other fields?

The outcome of these discussions will be captured in a workshop report that we hope to complete shortly after the workshop. More details about the report will be discussed at the workshop.

A few notes on the logistics:

Identification

All visitors to BNL are required to present proper identification to access the site. Please remember to bring your original passport, visa, supporting documentation (I-20, I94, DS2019, etc.) and driver's license. For more information about identification, please visit the following webpage: http://www.bnl.gov/guv/ID.asp

SBU Parking

A parking garage is available immediately after the SBU entrance and the Charles B. Wang Center is directly adjacent to it. To find the campus via GPS, enter the address "100 Nicolls Road, Stony Brook, NY 11794." Be sure to bring your parking ticket to the registration desk to get a reduced rate. Alternatively, *free parking* space is available as indicated on the attached campus map.

SBU WiFi Network

All registered attendees have received a username and password for access onto the SBU network. Please be sure to print and retain this information if you are planning to access the network.

NSLS-II Tour Requirements

Participants touring the NSLS-II are required to wear long pants or a skirt/dress that extends to the ankle, short-sleeve shirts, and fully enclosed shoes. Anyone arriving at the NSLS-II without the proper attire will not be permitted to tour the facility.

Contact Information:

For urgent purposes only, please contact the Conference Coordinator at (631) 708-4778.

We look forward to seeing you next week! Safe travels.

Best Regards,

Qun Shen
Ignace Jarrige
Juergen Thieme
Mercy Baez

On behalf of all organizing committee members and co-discussion leaders

National Synchrotron Light Source II
Brookhaven National Laboratory

Workshop Agenda at a Glance

Exploring New Science Frontiers at NSLS-II

October 21-23, 2019
www.bnl.gov/newscience2019

Brookhaven National Laboratory
Stony Brook University, Wang Center

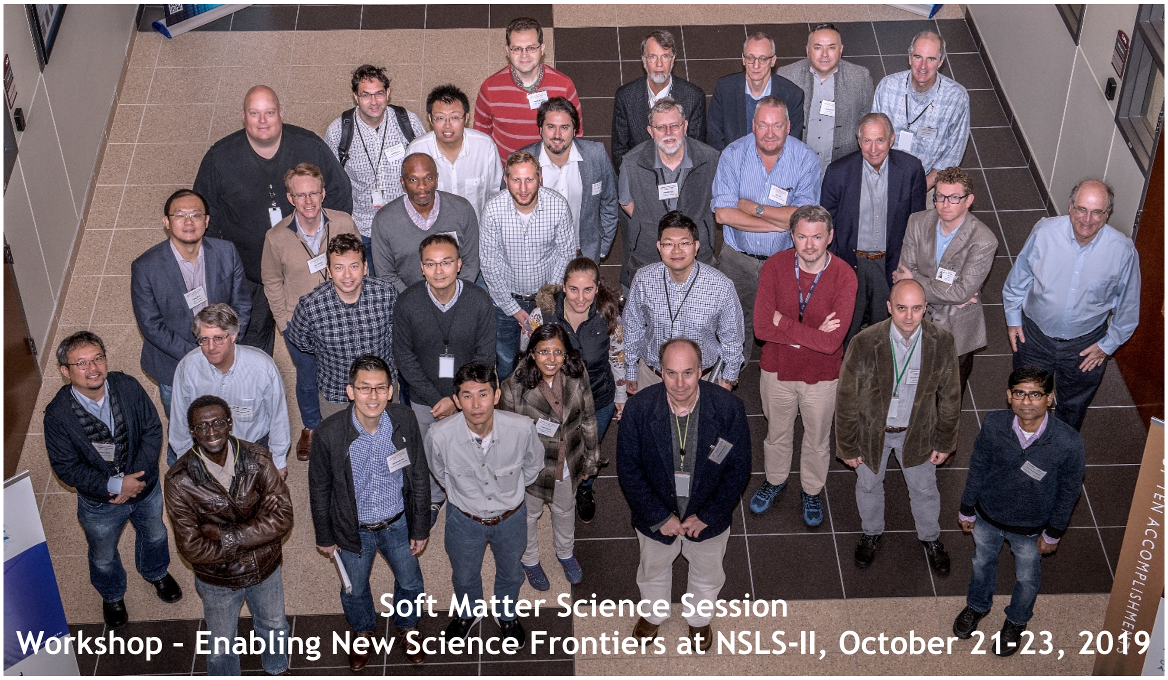
	Monday Morning, October 21, Plenary Session, Theater at Wang Center, Stony Brook University
Welcome and Introduction: ***Chair – S. Gill (NSLS-II UEC Chair, BNL)***	
8:25	**Gather in Theater – Welcome and logistics briefing**
8:30	**R. Reeder** (Vice Provost for Research, Stony Brook): Welcome Remarks [5']
8:35	**J. Hill** (Director, NSLS-II, BNL): NSLS-II Overview and Current Status [20']
8:55	**Q. Shen** (Deputy Director for Science, NSLS-II, BNL): Workshop Charge [5']
Plenary Presentations 1: ***Chair – M. Dean (BNL)***	
9:00	**A. Yacoby** (Harvard): Quantum sensing of quantum materials for QIS applications [45']
9:45	**J. Dailiant** (SOLEIL): Some ideas about soft matter and how synchrotron facilities could make a stronger impact in the field [45']
10:30	***Group Photo***
10:35	**Coffee Break** [25']
Plenary Presentations 2: ***Chair – M. Whitaker (Stony Brook)***	
11:00	**D. Ginley** (NREL): Enabling terawatt renewables – through the development of new functional materials for generation and storage [45']
11:45	**M. Tromp** (Groningen): Characterization of materials - Dynamic functionality using X-rays at synchrotrons: Now and in the future [45']
12:30	**Lunch at Wang Center** [1h]
	Monday Afternoon, October 21, Plenary Session, Theater at Wang Center, Stony Brook University
Plenary Presentations 3: ***Chair – J. Thieme (BNL)***	
13:30	**W. Hendrickson** (Columbia): Challenges and opportunities for synchrotrons in the life sciences [45']
14:15	**B. Toner** (Minnesota): Multi-element, multi-modal synchrotron solutions for Earth and environmental science [45']
15:00	**Coffee Break** [30']
NSLS-II Capability Presentations 1: ***Chair – P. Zschack (BNL)***	
15:30	**C. Mazzoli** (BNL): NSLS-II capabilities for quantum materials research [20']
15:50	**R. Pindak** (BNL): NSLS-II capabilities for soft matter research [20']
16:10	**E. Stavitski** (BNL): NSLS-II capabilities for catalysis and chemical science [20']
NSLS-II Capability Presentations 2: ***Chair – Y. Chu (BNL)***	
16:30	**E. Dooryhee** (BNL): NSLS-II capabilities for materials and engineering science [20']
16:50	**S. McSweeney** (BNL): NSLS-II capabilities for molecular and cell biology [20']
17:10	**R. Tappero** (BNL): NSLS-II capabilities for biosystems and earth & environmental sciences [20']
17:30	**Q. Shen** (BNL): Summary of Day 1 and Plan for Day 2 [10']
17:40	***Adjourn for the Day***
18:00	**Dinner at Hilton Garden Inn (no host)** [1h30']

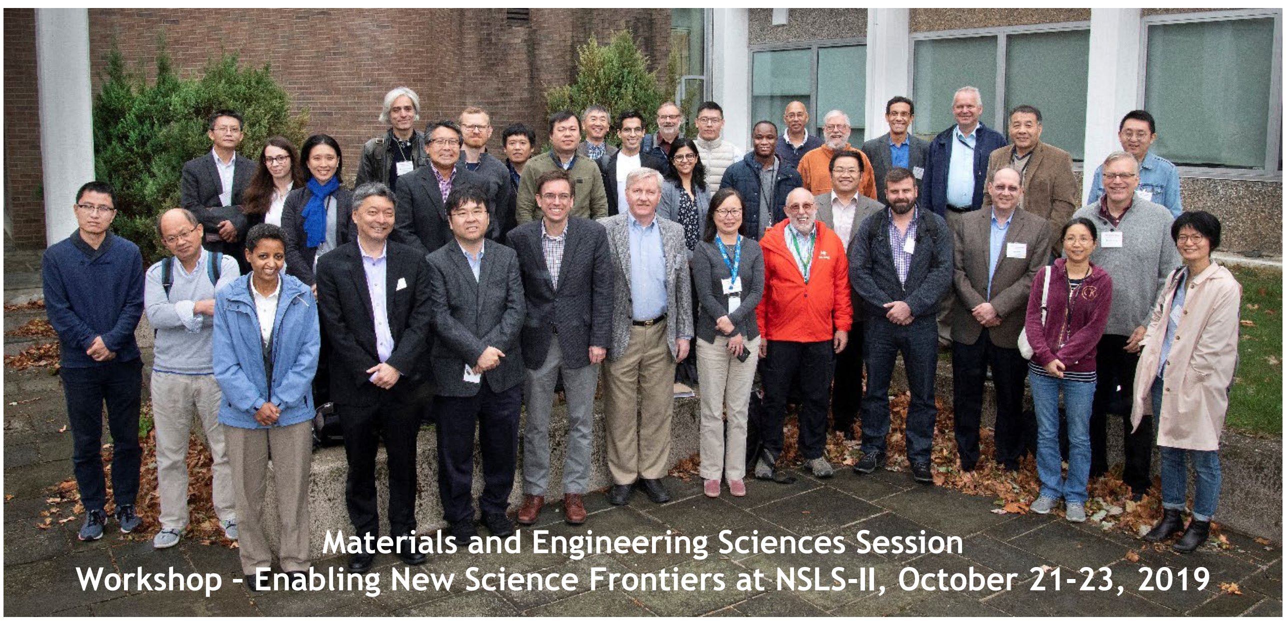
Tuesday Morning, October 22, Parallel Sessions, Brookhaven National Laboratory						
	Quantum Materials Research *Chairs: I. Jarrige (BNL)* *A. Wray (NYU)* ***Large Seminar Room Bldg.463***	**Materials & Engineering Science** *Chairs: E. Dooryhee (BNL)* *P. Evans (Wisconsin)* ***Hamilton Seminar Room Bldg.555***	**Catalysis & Chemical Sciences** *Chairs: E. Stavitski (BNL)* *A. Karim (Virginia Tech)* ***CSI Conference Room Bldg.725***	**Soft Matter Research** *Chairs: R. Pindak (BNL)* *D. DeLongchamp (NIST)* ***CFN Conference Room Bldg.735***	**Molecular & Cell Biology** *Chairs: S. McSweeney (BNL)* *S. Gabelli (Johns Hopkins)* ***Seminar Room 156 Bldg.744***	**Biosystems, Earth & Environment** *Chairs: R. Tappero (BNL)* *A. Lanzirotti (Chicago)* ***Seminar Room 156 Bldg.743***
8:30	**I. Jarrige** (BNL) and **A. Wray** (NYU): Introduction remarks [10'] **P. Abbamonte** (UIUC): Collective excitations with electrons and x-rays [35'] **A. Scholl** (LBNL): Quantum materials research at ALS [25'] **Y-J. Kim** (Toronto): RIXS of Kitaev materials [25'] **Discussion** [10'] *Group Photo*	**E. Dooryhee** (BNL) and **P. Evans** (Wisconsin): Introduction remarks [10'] **L. Levine** (NIST): Maximize impact using multi-modal, in-situ & operando [45'] **A. Bucsek** (Michigan): Deformation across length scales by 3D X-ray microscopy [30'] **K. Ludwig** (BU): Coherent scattering studies of surface processes [30']	**E. Stavitski** (BNL) and **A. Karim** (Virginia Tech): Introduction remarks [10'] **A. Knop-Gericke** (Fritz-Haber Inst): Ambient Pressure-XPS - status and outlook [45'] **M. Dinca** (MIT): Molecule-like reactivity in MOFs [25'] **B. Koel** (Princeton): Catalysis at interfaces [25']	**R. Pindak** (BNL) and **D. DeLongchamp** (NIST): Introduction remarks [5'] **R. Leheny** (John Hopkins): Emerging areas in soft matter research [35'] **R. Headrick** (Vermont): Local step-flow propagation [20'] **M. Schlossmann** (UIC): Liquid-liquid interfaces [20'] **Discussion** [15'] *Group Photo*	[8:45 start] **S. Gabelli** (John Hopkins) and **S. McSweeney** (BNL): Welcome & outline [15'] **M. Wiener** (Virginia): Atomic pairwise distance determination in biological macromolecules [45'] **Q. Liu** (BNL): Protein crystallography with long wavelengths and microcrystals [30']	[8:45 start] **R. Tappero** (BNL) and **A. Lanzirotti** (Chicago): Introduction remarks [15'] **H. Jamieson** (Queen's U): Microanalysis & imaging in environmental chemistry [45'] **J. Stubbs** (Chicago): Chemical complexity at mineral-fluid interfaces [30']
10:15	Coffee Break [30']	Coffee Break [20']	Coffee Break [30']	Coffee Break [30']	Coffee Break [30']	Coffee Break [30']
10:45	**C. Ahn** (Yale): Complex oxide interfaces [35'] **N. Butch** (NIST): Correlations & topology in actinide materials [25'] **Discussion** [15']	**S. Haile** (Northwestern): Surface defect chemistry of ceria and its derivatives [45'] **I. Takeuchi** (Maryland): Combinatorial and machine learning for discovery [30'] **A. Singer** (Cornell): Operando BCDI in Li- & Na-ion battery materials [30'] *Group Photo*	**S. Scott** (UCSB): Operando & time-resolved studies, and modeling of catalysts [45'] **M. Liu** (BNL): Chemical dynamics of atomic layer deposition (ALD) [25'] **Discussion** [35'] *Group Photo*	**C. Osuji** (U Penn): Controlling nanostructure and structure-property relationships [35'] **B. Vogt** (Penn State): Supramolecular interaction mechanisms [20'] **W. Bras** (ORNL): Transient life of chain molecules far out of equilibrium [20'] **Discussion** [15']	**S. Wasserman** (APS): An MX Survival Strategy for 2022-2023 [30'] **X. Kong** (NYU): Fast epitope mapping and vaccine design [30'] **M. Fuchs** (BNL): Protein dynamics [15'] **L. Yang** (BNL): Scanning imaging [15'] **E. Farquar** (Case Western): X-ray footprinting [15']	**M. Whitaker** (Stony Brook): Studying planets from inside out [30'] **S. Myneni** (Princeton): Metal chemistry in natural systems [30'] **M. Keiluweit** (U Mass): Mg-dependence of soil carbon storage [30'] **Discussion** [15']
12:00	Lunch (*Berkner*) [1h]			Lunch (*Berkner*) [1h]		

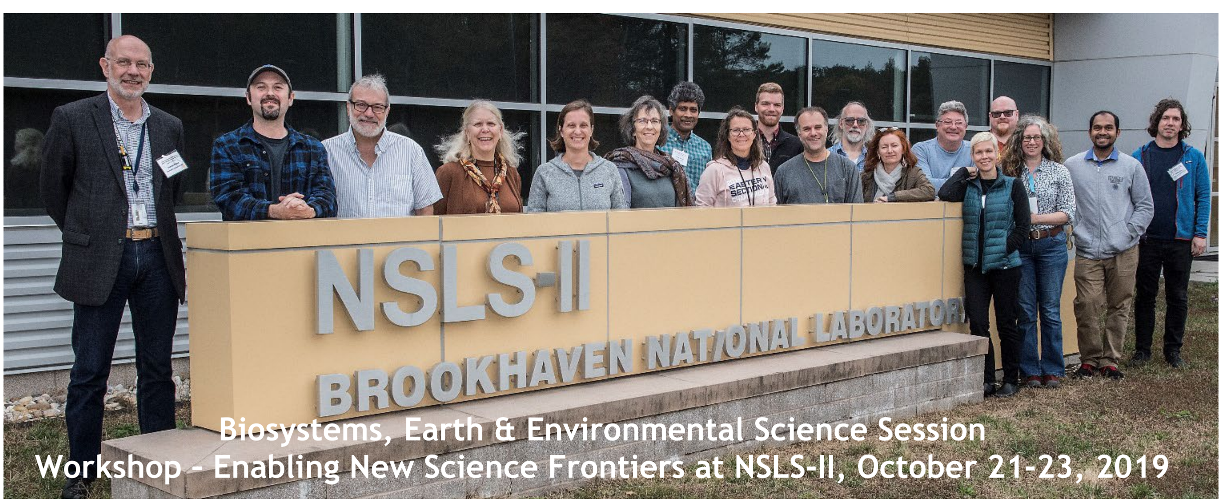
Tuesday Afternoon, October 22, Parallel Sessions, Brookhaven National Laboratory						
	Quantum Materials Research	**Materials & Engineering Science**	**Catalysis & Chemical Sciences**	**Soft Matter Research**	**Molecular & Cell Biology**	**Biosystems, Earth & Environment**
12:30		Lunch (*Berkner*) [1h]	Lunch (*Berkner*) [1h]		Lunch (*Berkner*) [1h]	Lunch (*Berkner*) [1h]
13:00	Beamline Tour			Beamline Tour		
13:30	**K. Plumb** (Brown): 'Multi-messenger' spectroscopies of quantum magnets [25']	Beamline Tour	Beamline Tour		Beamline Tour	Beamline Tour
14:00	**L. Piper** (Binghamton): Scalable neuristors for neuromorphic computing [25'] **M-K. Liu** (Stony Brook): Nano-IR for low energy excitations in quantum materials [25'] **Discussion** [15']	[14:30 start] **C. Lavoie** (IBM): Materials for future computing applications [30'] **T. Sun** (Virginia): Fast micro-imaging for materials and engineering studies [30']	[14:30 start] **R. Davis** (Virginia): Complex heterogeneous catalysts [25'] **A. Frenkel** (Stony Brook): If you want to understand function, study spectrum [25'] **Discussion** [15']	**A. Amassian** (NC State): Intelligent materials synthesis and discovery [20'] **L. Richter** (NIST): Polymer processing from both solution and melt [20'] **C. McNeill** (Monash): STXM for probing molecular orientation and chemical composition [20']	**C. Larabell** (UCSF): TXM imaging of biological cells [45'] **G. Johnson** (Allen Inst.): Prototyping Multiscale Cellular Visualization & Modeling Techniques [30'] *Group Photo*	[14:30 start] **D. Dyar** (Mount Holyoke): Multivariate analysis techniques to quantitative interpretation of valence states [45'] **B. Toner** (Minnesota): What's in your pool? Biogiochemical research at synchrotrons [30'] *Group Photo*
15:00	Coffee Break [30']			Coffee Break [30']	Coffee Break [30']	
15:30	*Soundbite Talks:* *A. Taleb (SOLEIL)* *E. Kotta (NYU)* *K. Kaznatcheev (BNL)* **Discussion and Report Planning**	Coffee Break (30') [16:00 start] ***Soundbite Talks:*** *F. Wang (BNL)* *J. Wishart (BNL)* *D. Weidner (SBU)* **Discussion and Report Planning**	Coffee Break [30'] [16:05 start] **J. Lockard** (Rutgers): XES studies of MOFs [25'] ***Soundbite Talks*** **Discussion and Report Planning**	*Soundbite Talks:* *A. Cunsolo (BNL)* *B. Ocko (BNL)* *L. Wiegart (SBU)* *K. Kaznatcheev (BNL)* *S. Maldonado (Michigan)* *D. Bagchi (U Baroda)* *B. Collins (WSU)* **Discussion and Report Planning**	[15:45 start] **D. Keedy** (CUNY): Illuminating dynamic proteins with multi-dataset X-ray crystallography [30'] ***Soundbite Talks:*** *R. Jain (CWRU)* **Discussion and Report Planning**	Coffee Break [30'] [16:15 start] **A. Lanzirotti** (Chicago): New earth science directions: a GSECARS perspective [30'] ***Soundbite Talks:*** *B. Bostick (Columbia)* *P. Northrup (SBU)* **Discussion and Report Planning**
17:30	***Adjourn for the Day***					

Wednesday Morning, October 23, Parallel Sessions and Close-out Plenary, Brookhaven National Laboratory						
	Quantum Materials Research *Chairs: I. Jarrige (BNL)* *A. Wray (NYU)* ***Physics Conference Room Bldg.510***	**Materials & Engineering Science** *Chairs: E. Dooryhee (BNL)* *P. Evans (Wisconsin)* ***Hamilton Seminar Room Bldg.555***	**Catalysis & Chemical Sciences** *Chairs: E. Stavitski (BNL)* *A. Karim (Virginia Tech)* ***CSI Conference Room Bldg.725***	**Soft Matter Research** *Chairs: R. Pindak (BNL)* *D. DeLongchamp (NIST)* ***CFN Conference Room Bldg.735***	**Molecular & Cell Biology** *Chairs: S. McSweeney (BNL)* *S. Gabelli (John Hopkin)* ***Seminar Room 156 Bldg.744***	**Biosystems, Earth & Environment** *Chairs: R. Tappero (BNL)* *A. Lanzirotti (Chicago)* ***Seminar Room 156 Bldg.743***
8:30	**Report writing and preparation for close-out presentation** [1h30']	**Report writing and preparation for close-out presentation** [1h30']	**Report writing and preparation for close-out presentation** [1h30']	**Report writing and preparation for close-out presentation** [1h30']	**Report writing and preparation for close-out presentation** [1h30']	**Report writing and preparation for close-out presentation** [1h30']
10:00	**Coffee Break (*Physics Conference Room Lobby, Bldg. 510*)**					
10:30	**Close-out Summary Presentations (*Physics Conference Room, Bldg. 510*)** ***Chair: Q. Shen (BNL)*** Quantum materials research [15'] Materials & engineering science [15'] Catalysis & chemical sciences [15'] Soft matter research [15'] Molecular & cell biology [15'] Biosystems, earth & environment [15']					
12:00	***Workshop Adjourns***					

Workshop Photographs


Soft Matter Science Session
Workshop – Enabling New Science Frontiers at NSLS-II, October 21-23, 2019


Materials and Engineering Sciences Session
Workshop – Enabling New Science Frontiers at NSLS-II, October 21-23, 2019


NSLS-II
BROOKHAVEN NATIONAL LABORATORY
Biosystems, Earth & Environmental Science Session
Workshop – Enabling New Science Frontiers at NSLS-II, October 21-23, 2019


Catalysis and Chemical Science Session
Workshop – Enabling New Science Frontiers at NSLS-II, October 21-23, 2019


NSLS-II
BROOKHAVEN NATIONAL LABORATORY
Molecular and Cell Biology Session
Workshop – Enabling New Science Frontiers at NSLS-II, October 21-23, 2019