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Multimodal Synchrotron Approach: Research Needs and Scientific Vision

Introduction

This report summarizes the outcome of a workshop, "Multimodal Synchrotron Approach—Research Needs and Scientific Vision," held during the National Synchrotron Light Source–II (NSLS-II)/Center for Functional Nanomaterials (CFN) 2019 Users' Meeting at Brookhaven National Laboratory (BNL) on May 22, 2019. Multimodal approaches are defined by the convergence of multiple measurement probes to tackle a single scientific problem. In a synchrotron light source context, this may manifest as the usage of multiple synchrotron beamlines or multiple detection techniques on the same beamline to probe a single sample or system. The synchrotron multimodal approach may be achieved by incorporating ancillary probes into synchrotron beamlines, by exploiting other measurement modalities—such as the electron-based and optical imaging methods—to augment synchrotron datasets, or even by exploiting theory and modeling to complement measurements.

Multimodal approach as a holistic approach offers deeper understanding in complex, heterogeneous systems, critical for increased scientific impact and technological applications. As a facility, NSLS-II, a U.S. Department of Energy (DOE) Office of Science User Facility located at BNL, recognizes both the challenges and opportunities, and thus identifies multimodality as one of the priorities in its strategic plan, launching a Multimodal Issues Task Force in October 2016. Moreover, NSLS-II offers users the possibility to submit one beamtime proposal to include time requests at multiple beamlines (via multimodal proposals), starting in the third cycle of 2019. Such efforts would not reach their potential without outreach to the broader scientific research and technical development community. This workshop intended to address the opportunity and challenges of multimodal analysis at synchrotron light sources.

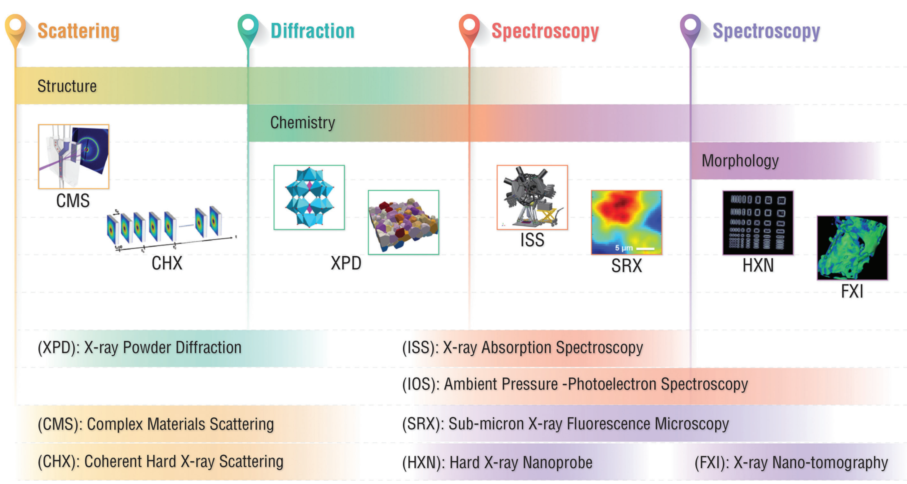
Scientific needs and vision of multimodal approach

Spectroscopic multimodal research—applications to catalysis: Professor Bruce Gates, University of California, Davis, presented "Atomically Dispersed Supported Metal Catalysts: Synthesis, Structural Characterization, and Catalyst Performance," in which he discussed the importance of multimodal research in heterogeneous catalysis. Gates investigated atomically precise metal catalysts dispersed on uniform crystalline supports. Various experimental techniques were used to characterize these materials to reveal complementary information. For example, aberration-corrected scanning transmission electron microscopy (STEM) shows that the metals in well-made samples are atomically dispersed and infrared (IR) spectroscopy shows the uniformity of the metal sites. Synchrotron techniques like extended X-ray absorption fine structure (EXAFS) and X-ray absorption near edge structure (XANES) spectroscopy provide structural and chemical information such as evidence of metal oxidation state and metal-ligand bonding, respectively. Challenges in this field include improving the performance of catalysts and understanding the nature of metal-ligand bonding. Opportunities exist in applying other synchrotron techniques, such as ambient-pressure X-ray photoelectron spectroscopy, high-energy-resolution fluorescence detection—XANES, quick-EXAFS, as well as operando characterization methods.

Multimodal X-ray, neutron, and electron-based characterizations combined with simulations—application to ferroelectric materials: Dr. Igor Levin, National Institute of Standards & Technology (NIST), elaborated on the most recent improvements of the reverse Monte-Carlo (RMC) methods for refining the local (nanoscale) structure of complex materials. The accurate description of the nanoscale ordering is key, as the local structure is known to drive the properties in an extensive range of materials. The power of the combined-technique RMC approach was illustrated by Levin through the study of the classical relaxor ferroelectric $PbMg_{1/3}Nb_{2/3}O_3$ (PMN) perovskite. This case study involved simultaneous fitting of 3D X-ray diffuse scattering from a single crystal of PMN with both X-ray and neutron total scattering measured on a PMN powder. X-ray absorption fine structure (XAFS) spectroscopy characterizing Pb and Nb was also included in the fitting process to improve chemical resolution.

Correlative microscopy and tomography—application in materials science: Dr. Yiyang Li, Sandia National Laboratory, presented work on the subject of "Visualizing Electrochemistry through Multimodal Microscopy for Batteries and Neuromorphic Computing." Li presented the results of studies showing how multimodal synchrotron microscopy enabled detailed visualization and understanding of electrochemistry for batteries: combining soft X-ray scanning transmission X-ray microscopy (STXM), hard X-ray transmission X-ray microscopy (TXM), X-ray diffraction (XRD), STEM (including correlative electron microscopy), Auger electron spectroscopy, and ptychography. Li explained how coupling between electrochemistry and imaging at multiple length-scales with various contrasts could drive the development and understanding in materials science for neuromorphic computing. Li highlighted the scientific motivation in the field of energy storage as a pressing need for society, also discussing exciting new research directions in merging the field of chemistry with electronics, with new paradigms on neuromorphic, analog, and quantum research.

Combining X-ray scattering, diffraction with electron microscopy and simulation—study in additively manufactured stainless steels: Professor Jason Trelewicz, Stony Brook University, showcased a study whereby the microstructure, processing defects, and their connection to the



(Top) Photo of attendees at the "Multimodal Synchrotron Approach: Research Needs and Scientific Vision" workshop. (Bottom) Multimodal synchrotron approach of combining different X-ray techniques across beamlines, as well as with other techniques, with NSLS-II beamlines as examples.

underlying chemical distribution could best be examined by a multimodal approach relying heavily on synchrotron X-ray techniques. Stainless steels produced by additive manufacturing (AM-laser powder-bed fusion) exhibit a complex hierarchical microstructure. Macroscale X-ray diffraction, nanoscale imaging, and fluorescent spectroscopy measurements were performed at the NSLS-II. Results connect the materials printing conditions with the corrosion performance. Trelewicz also summarized other opportunities for multimodal synchrotron studies of nanostructured alloy development with a focus on plasma facing materials for fusion energy platforms.

Discussion for scientific needs and vision of multimodal approach

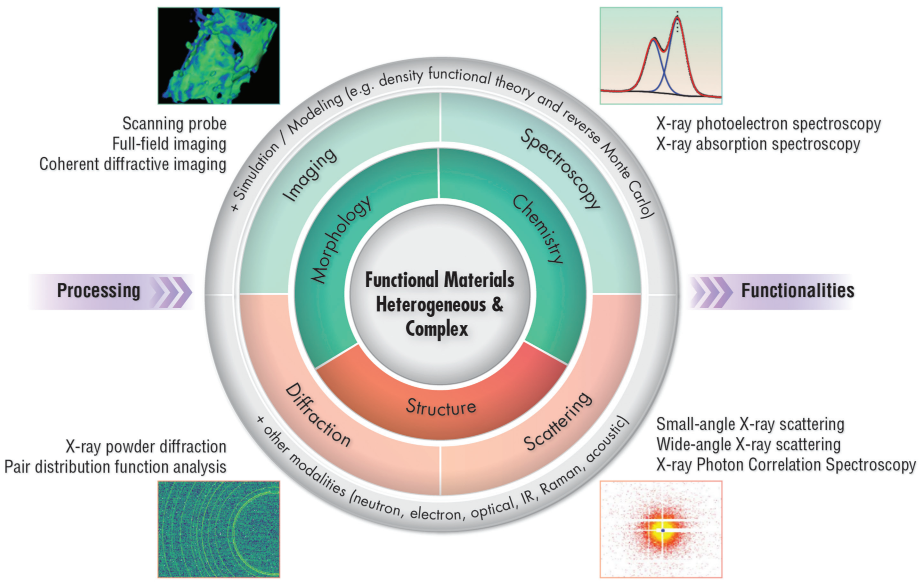
The session-one discussions, led by Dr. Lin Yang, NSLS-II, focused on how scientific research can truly benefit from multimodal approaches, especially considering the newly available multi-beamline General User Proposal at NSLS-II. Yang raised examples using different modes of imaging, scattering, and techniques beyond synchrotron techniques, such as modeling, simulation, and electron microscopy. The various modes of scattering include small-angle X-ray scattering (SAXS), wide-angle X-ray scattering (WAXS), X-ray photon correlation spectroscopy (XPCS), macromolecular crystallography (MX), and XRD. Two types of experiments were discussed:

The first case was in-situ and operando experiments whereby the same system, but not the same sample, is investigated. The needs include leveraging capabilities at multiple beamlines and standardization across beamlines. The key questions are: How to compare data from two beamlines? How to define standard samples to calibrate across beamlines and techniques? In addition, as contamination of surface-sensitive samples is a concern, robust sample handling methods and a trackable history through established protocols are needed when transferring samples between beamlines.

Second, for unique samples being studied across beamlines, several key considerations are raised; fiducials either as part of the sample preparation or as sample holder design scheme to allow cross-beamline alignment would be beneficial. As radiation damage could be a concern for some types of samples, tracking the dose and potentially starting with the less-damaging technique, such as moving from hard X-ray to soft X-ray characterizations, should be considered. When appropriate, having multimodal techniques combined within the same beamline would be beneficial to minimize multiple exposure, and thus reduce the potential radiation damage.

Technical challenges and solutions to multimodality: Correlative and integrating data analysis, visualization, and interpretation

Opportunities with synchrotron multimodal approach: Dr. Mark Rivers, University of Chicago, presented a talk entitled "Mul-



Summary of the multimodal approach discussed during the workshop.

timodal Analysis at GeoSoilEnviroCARS." In the work of the GSECARS program at the Advanced Photon Source (APS), a suite of analysis techniques has grown to provide users with a quick and easy way to align their samples in a beamline, prescreen their samples, and use multiple measurement techniques at the beamline. Users can pre-locate the regions of interest in samples via both optical and scanning electron microscopy (SEM). Moreover, data collected simultaneously with multiple modalities can provide a wealth of information, including fluorescence, transmission, diffraction, and spectroscopy data. For example, researchers studying defects in diamonds conducted simultaneous tomography and diffraction to obtain a 3D representation of the sample along with the crystallographic information.

Open-source software on research across platforms: Dr. Marcus Hanwell, Kitware Inc., gave a presentation entitled "Open Visualization of Multimodal data: from Tomviz to Jupyter." Openness by giving access to data, reproducible workflows, processing code, and visualization tools improves science. An open-source Tomviz package (www.tomviz.org) is easily extendable by allowing the user to integrate data sources and operators written in Python, and also generates reproducible pipelines for the data. This enable pipelines to be run in a batch mode or in other interfaces, such as a Jupyter or IPython. The source code is hosted on GitHub to make use of its features for pull requests, code reviews, issues, and other functions. Kitware currently collaborates with NSLS-II to integrate Tomviz into the bluesky data acquisition system and databroker data management system developed by NSLS-II.

Artificial intelligence to drive multimodal experiments by machine learning: Dr. Kevin G. Yager, BNL, presented "Autonomous X-ray Scattering," sharing the latest developments in autonomous experiments at NSLS-II and their potential to be applied in multimodal analysis. Deep learning methods can be used to "understand" the results of X-ray experiments and then guide experimental execution, including multi-channel machine learning. This level of automation increases experimental efficiency, liberates the scientists from experimental micro-management, and allows them to focus on the high-level scientific meaning of the data. The autonomous method has been applied successfully at the Complex Materials Scattering (CMS) beamline of NSLS-II, including characterization of nanoparticle superlattices, block copolymer assembly using combinatorial samples, and polymer-nanoparticle composites. Envisioned improvements for future autonomous experiments were discussed, including real-time synthesis/processing platforms allowing exploration of the corresponding materials spaces, the inclusion of known materials physics via theory/simulation, and development of general simulations to handle a variety of pertinent systems. This approach would naturally scale to the analysis of multimodal datasets when separate measurement techniques return complementary data characterizing a single sample/material.

Discussion of technical challenges and solutions to multimodality: Correlative and integrating data analysis, visualization, and interpretation

The discussion was led by Dr. Stuart Campbell, NSLS-II. Multimodal experiments naturally generate greater amounts of data, and users need to have all data analyzed quickly to help guide the experimentation. Automation of experiments, aided by machine leaning and informative abstraction of the data and visualization of the sample, is important to find interesting features on highly heterogeneous samples, applicable for multi-scale imaging. On the topic of protocols, data sharing/visualization and sample coordination between beamlines, users also raised the point of the importance of sharing data between beamlines as a crucial step to enable multimodality.

On data processing, the complexity in data processing in a multimodal approach is recognized and can be the limiting factor. This challenge arises largely because the analysis is often research-domain specific, rather than universal. Providing users with adequate access to the software and computing tools, either at the beamline or via remote access, could help alleviate the issue. These discussions point to the importance of the analysis pipeline. In general, having a faster and more automated pipeline is beneficial, so more advanced analysis can be carried out in real-time or near-real-time during the experiment to guide the next steps more optimally.

Conclusions

The workshop provided a snapshot of many exciting emerging opportunities). As NSLS-II continues to grow, developing tools such as those for the prescreening and pre-alignment of samples, adequate data analysis pipelines, simulation/modeling with multimodal inputs, and autonomous experiments becomes increasingly important. The community will continue to develop new strategies for handling rich data from complex, scientifically important systems via multimodal approaches.

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