BNL-52351-2018-FORE

LDRD 2017 Annual Report: Laboratory Directed Research and Development Program Activities

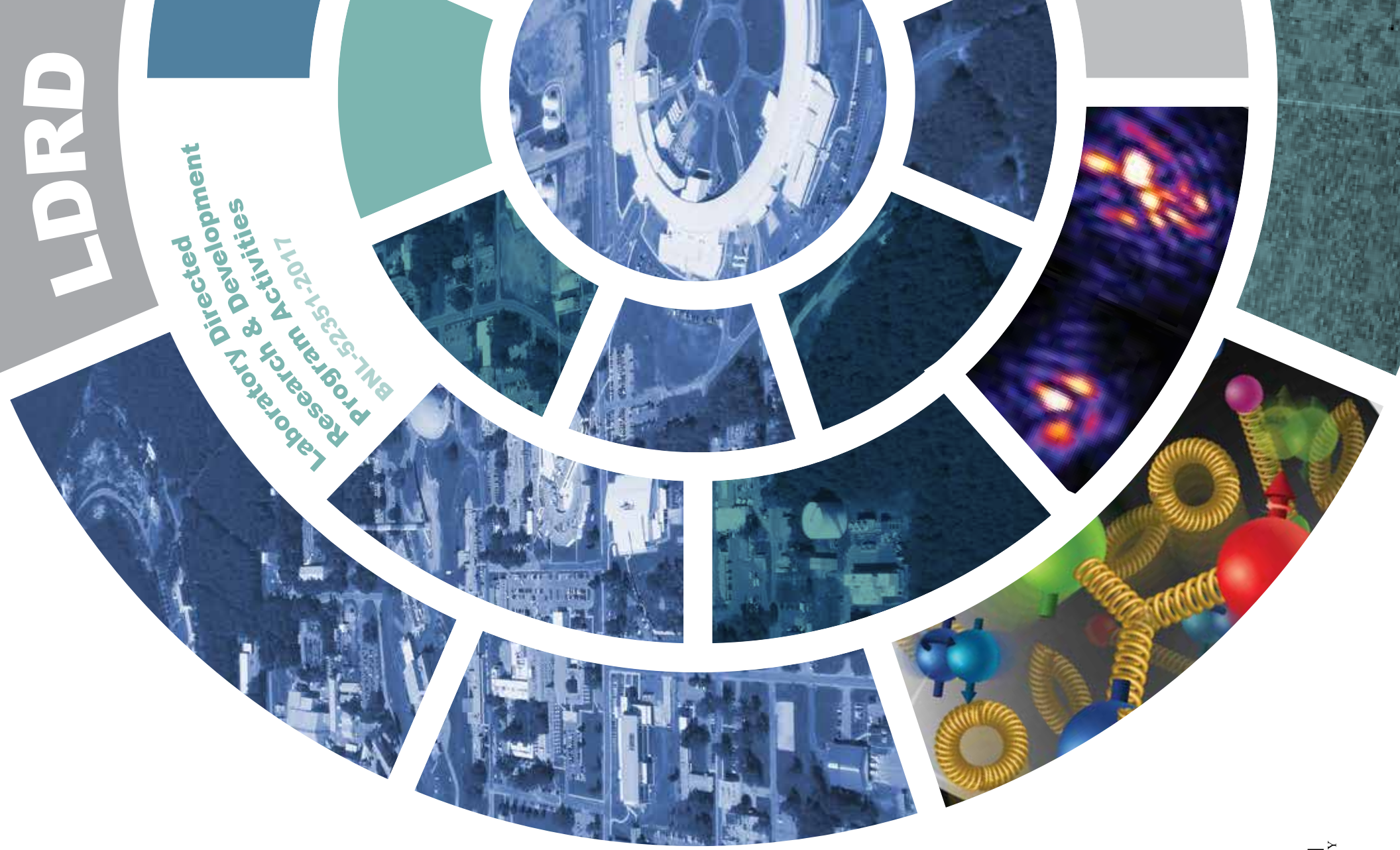
J. Anderson, L. Flynn

March 2018

Director's Office
Brookhaven National Laboratory

U.S. Department of Energy
USDOE Office of Science (SC)

DISCLAIMER



2017 Annual Report

BROOKHAVEN NATIONAL LABORATORY · BROOKHAVEN SCIENCE ASSOCIATES
UPTON, NEW YORK 11973-5000 · UNDER CONTRACT NO. DE-SC0012704
UNITED STATES DEPARTMENT OF ENERGY · March 2018

BROOKHAVEN NATIONAL LABORATORY

Acknowledgments

The Laboratory Directed Research and Development (LDRD) Program is managed by Jack Anderson, who serves as the Interim Director of the Planning, Performance, and Quality Management Office. Contributions to the FY 2017 report were coordinated by Liz Flynn and edited by Kathi Barkigia. Jack wishes to thank Kathi Barkigia, Brian Boyle and Liz Flynn for their assistance in preparing the document. A special thank you is also extended to the Production Services Group for their help in publishing. Of course, a very special acknowledgement is extended to all of the authors of the annual project reports.

Table of Contents

Table of Contents

Table of Contents

Introduction

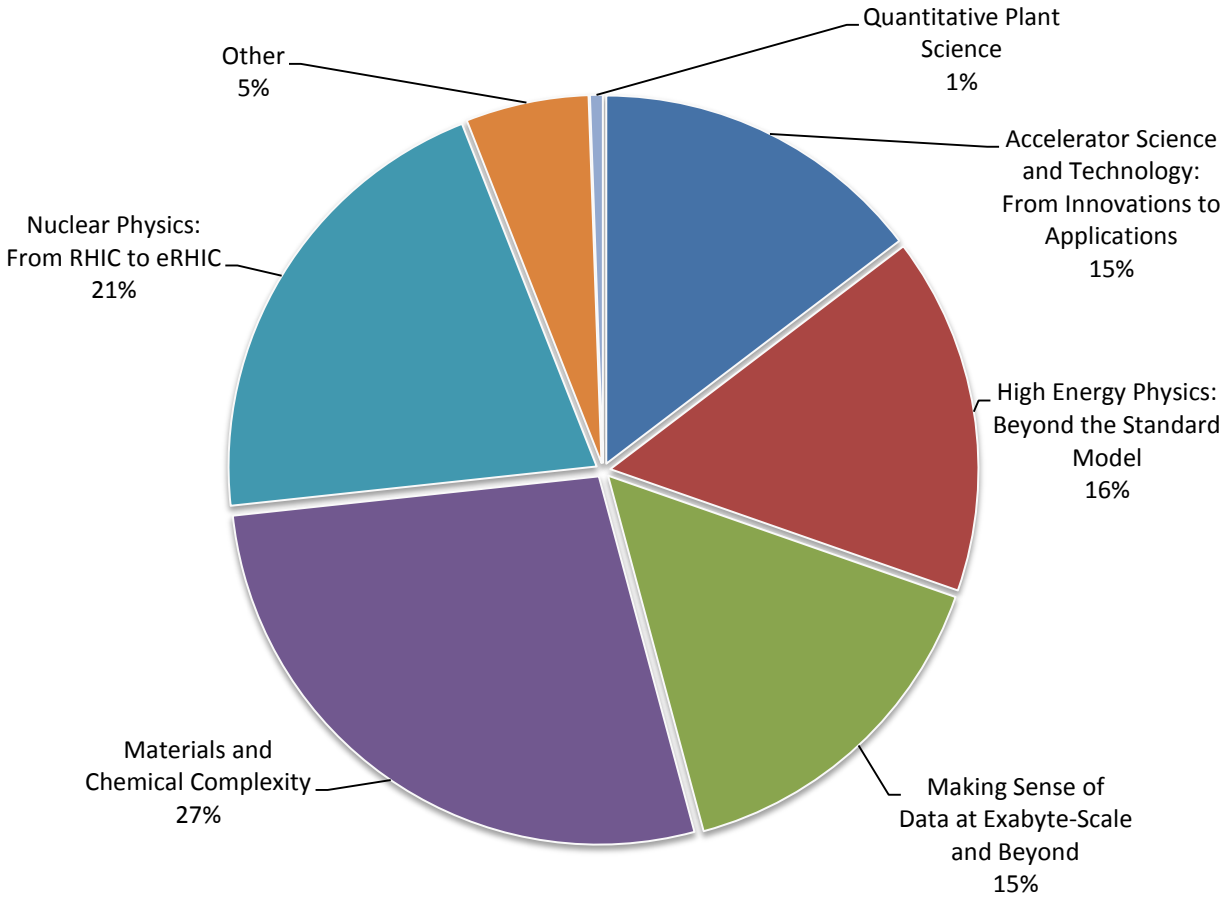
Each year, Brookhaven National Laboratory (BNL) is required to provide a program description and overview of its Laboratory Directed Research and Development Program (LDRD) to the Department of Energy (DOE) in accordance with DOE Order 413.2C dated October 22, 2015. This report provides a detailed look at the scientific and technical activities for each of the LDRD projects funded by BNL in FY 2017, as required. In FY 2017, the BNL LDRD Program funded 46 projects, 13 of which were new starts, at a total cost of $10.4M.

The investments that BNL makes in its LDRD program support the Laboratory's strategic goals. BNL has identified six scientific initiatives that define the Laboratory's scientific future and that will enable it to realize its overall vision. Two operational mission goals address essential operational support for that future: renewal of the BNL campus; and safe, efficient laboratory operations. The six scientific initiatives are:

- Nuclear Physics: From RHIC to eRHIC
- Materials and Chemical Complexity
- Making Sense of Data at the Exabyte Scale and Beyond
- High Energy Physics: Beyond the Standard Model
- Accelerator Science and Technology: From Isotopes to Applications
- Quantitative Plant Science Initiative.

Of the funded LDRD projects, 95% supported the six scientific initiatives (Figure 1). In total, these LDRD investments supported 67 postdoctoral researchers and graduates students in whole or in part and resulted in 87 publications and 8 awards.

Figure 1 - Scientific Initiatives



This Program Activities Report represents the future of BNL science; it is an impressive body of exploratory work that investigates many scientific and technical directions in support of the DOE and BNL Missions.

LABORATORY DIRECTED RESEARCH AND DEVELOPMENT

2017 PROJECT SUMMARIES

Time resolved Imaging of X-rays and Charged Particles

LDRD Project # 13-006
A. Nomerotski

PURPOSE:

Resolving the time evolution of fast processes, measuring the time-of-flight of particles, and looking at time correlations in spatially resolved events are the main drivers for the development of sensors with the best possible time resolution. In this project, we design, characterize and apply fast cameras with 10 ns resolution to time resolved X-ray imaging at National Synchrotron Light Source II and to imaging mass spectrometry in the BNL Chemistry Division. Time resolved X-ray registration is required in X-ray photon correlation spectroscopy, which is an important tool in studies of nanoscale dynamics of materials, while the time-of-flight mass spectrometry is an important analytical tool used widely in chemistry, biology, and medicine.

APPROACH:

We combine features of several established technologies in order to produce a novel device with as-yet unachieved capabilities. We have designed a new silicon pixel sensor, which in combination with the Timepix chip, provides 10 ns time resolution and high quantum efficiency for photons with wavelength between 350 and 1050nm. This resulted in an imager with characteristics far superior to cameras currently available commercially. We employed a commercially available readout system to characterize the new device and tested the new camera with several imaging mass spectrometry groups. An improved version of the readout chip, Timepix3, is fully compatible with the sensor and a Timepix3 based camera will be produced and tested as well.

TECHNICAL PROGRESS AND RESULTS:

We list below the main accomplishments in FY 2017, which amounts to three months of work as the project was funded until 1 January 2017. The main activity was writing up the remaining experiments and submitting papers for publication. The list of published manuscripts during 2017 is provided below:

1. M. Fisher-Levine, R. Boll, F. Ziaee, C. Bomme, B. Erk, D. Rompotis, T. Marchenko, A. Nomerotski and D. Rolles: *"Time-Resolved Ion Imaging at Free-Electron Lasers Using TimepixCam."* Accepted by Journal of Synchrotron Radiation. Submitted in December 2017.
2. A. Zhao, M. van Beuzekom, B. Bouwens, D. Byelov, I. Chakaberia, Ch. Cheng, E. Maddox, A. Nomerotski, P. Svihra, J. Visser, V. Vrba and T. Weinacht: *"Coincidence velocity map imaging using Tpx3Cam, a time stamping optical camera with 1.5 ns timing resolution."*"Rev Sci Instrum. 88(11), 10.1063/1.4996888 (2017).
3. I. Chakaberia, M. Cotlet, M. Fisher-Levine, DR Hodges, J. Nguen and A.Nomerotski: *"Time stamping of single optical photons with 10 ns resolution."* SPIE Commercial+ Scientific Sensing and Imaging, 102120Q-102120Q-7.
4. L. M. Hirvonen, M. Fisher-Levine, K. Suhling, and A. Nomerotski: *"Photon counting phosphorescence lifetime imaging with TimepixCam.*" Rev. Sci. Instrum. 88, 013104 (2017).

5. Nomerotski, Z. Janoska, I. Chakaberia, M. Fisher-Levine, P. Takacs, and T. Tsang: *"Characterization of TimepixCam, a fast imager for time stamping of optical photons*." J. Instrum. 12(1), C01017 (2017).

In summary, we completed all planned milestones for FY 2017 and for the project in general.

1st Light: Elucidating Solid-Solid Interfaces in Energy Storage Systems

LDRD Project # 14-005
E. Takeuchi, K. Takeuchi, A. Marschilok

PURPOSE:
The numerous solid-solid interfaces within an energy storage system can significantly impact the function of energy storage systems. The proposed research will develop a methodology to characterize bimetallic cathodes and anode surfaces. This LDRD will contribute to the broader goal of highlighting National Synchrotron Light Source II (NSLS-II) for energy storage related research.

Polyanion compounds, metal oxides and phosphorous oxides, are of significant interest as energy storage materials, as they provide theoretically high voltage and high capacity. We have recently demonstrated a class of bimetallic electroactive cathodes, which significantly increase in conductivity on reduction. In contrast at the anode interface, solid-solid interfaces (also called solid electrolyte interphase) can lead to significant reduction in battery performance. Surface deposits can form on the anode leading to cell impedance rise, resulting in premature device failure. The proposed research will develop the methodology and then characterize the solid-solid interfaces of bimetallic cathodes and of anode surfaces.

APPROACH:
This LDRD project will establish an important and unique infrastructure for investigations related to energy storage, allowing probing of mechanistic information under application-relevant working conditions. Under this LDRD, cell designs will be fabricated, which will be used to characterize active materials in a cell environment, *operando*. The cells will be compatible with the Sub-micron Resolution X-ray spectroscopy (SRX) and Inner Shell Spectroscopy (ISS) beamlines at NSLS II. SRX provides a focused beam with a broad, tunable and scanable range of photon energy appropriate for elemental imaging and sub-micron spectroscopy accommodating a range of focal spot sizes and geometries. ISS is a high flux damping wiggler spectroscopy beamline, became available in FY 2017 and makes state-of-the-art spectroscopic tools available with *in situ* and *operando* instrumentation for energy research.

TECHNICAL PROGRESS AND RESULTS:
The initial work will focus on the development and demonstration of suitable test cells for sample evaluation under electrochemical use conditions. The test cells will be validated electrochemically to ensure reasonable agreement with performance data collected in previously demonstrated cell configurations. The test cells will then be used at NSLS II *in situ* (not under operation) as appropriate to demonstrate the viability of material analysis at the anode and cathode of the cell. The eventual target is to employ the cells under *operando* conditions at NSLS-II.

Development of the operando environment

With the ISS beamline coming on line in 2017, an appropriate test cell was designed and fabricated, consistent with the space requirements as a prospective approach for an *operando* cell. To compare the results of this X-ray optimized cell with conventional batteries, we performed numerous baseline experiments to characterize and compare the electrical properties of the new cell. Composite electrodes were used for evaluation, as they are fully representative of

functioning batteries. The results of the newly designed cell were acceptable and provided electrochemical data comparable to conventional cell designs.

A significant concern for data collection at a beamline is damage to the material and the cell. Damage was explored in a cell *in situ* first and was found manageable. In 2017, we were able to successfully position a fully functioning electrochemical cell in the ISS beamline. A potentiostat controlling the current of the cell was implemented into the beamline. We were able to collect data during material reduction as well as oxidation. Correlations of the number of coulombs under potentiostatic control were compared with the change in oxidation state of the active material within the battery.

Implementation of a cell for use with SRX demanded different considerations due to collection by fluorescence and the use of a mapping beamline. Sample cells were sized consistent with the target set of experiments for integration into the SRX infrastructure. Also, the thickness of the cell window housing material needed to be carefully selected and adjusted to provide appropriate beam throughput while retaining a sealed environment. The data were collected using X-ray fluorescence imaging and step-scan X-ray Absorption Near Edge Structure spectroscopy (with sub-100 nm spatial resolution in 2D, a unique capability of SRX). The spot sizes of SRX enabled the ability to probe individual aggregates to determine homogeneity of reduction as a function of applied current, Fig. 1. The initial studies were focused on as synthesized and partially discharged systems, *in situ* but not in *operando*. Follow on studies using *operando* techniques were also demonstrated as feasible.

Finally, *in situ* results were obtained on the Hard X-ray Nanoprobe beamline. A custom electrochemical cell design was developed that enabled the collection of the data.

As a result of this project, data collection methodologies appropriate for *operando* interrogation of energy storage materials were demonstrated.

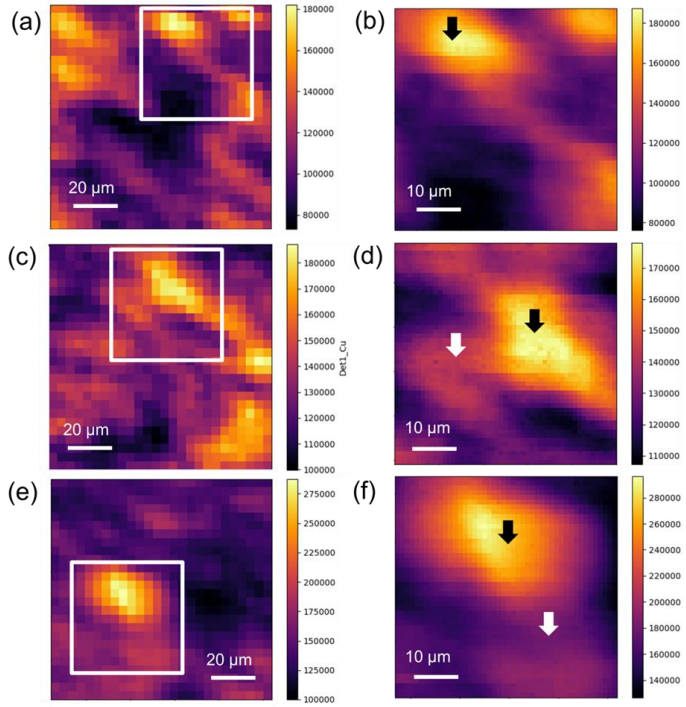


Figure 1: (a, c, e) Coarse resolution and (b, d, f) fine resolution copper elemental maps of copper ferrite (CFO) nanorod-carbon nanotube binder-free electrodes which were (a, b) undischarged, (c, d) discharged to 0.3 V at 1600 mA/g and (e, f) discharged and recharged to 3.0 V at 1600 mA/g. The white box insets in the coarse maps indicate the area selected for the fine resolution map. Arrows signify 0.5 µm x 0.5 µm spots where µ-X-ray absorption spectroscopy measurements were collected; black arrows indicate selected areas at the center of the high intensity (high concentration of CFO) regions within the binder-free electrode, white arrows indicate measurements collected at the surface of higher intensity regions.

Correlative Microscopy, Spectroscopy and Diffraction with a Micro-Reactor

LDRD Project # 14-036
E. Stach, A. Frenkel

PURPOSE:

The purpose of this effort is to first develop and then utilize a novel correlative characterization approach based on microfabricated catalytic reactors. This approach allows the acquisition of data from heterogeneous catalysis systems at atmospheric pressure, using both advanced analytical electron microscopes at the Center for Functional Nanomaterials (CFN) as well as X-ray spectroscopy, diffraction and imaging at the National Synchrotron Light Source (NSLS/NSLS-II), and at other synchrotrons during the transition period from NSLS to NSLS-II. The measurement of reaction products during the data acquisition allows "*operando*" characterization, *i.e.* characterization of the catalysts as they are actively working to catalyze the chemical reaction. By obtaining the data in an *operando* condition, it becomes possible to directly correlate the information about the system obtained from the measurements using each technique. The capabilities being developed through this effort are intimately linked to the proposed efforts of the Energy Frontier Research Center on Site-Selective Nanocatalysis, the core research programs at the CFN and Chemistry Division, as well as the Synchrotron Catalysis Consortium: in short, the methods being developed within this LDRD are well-aligned with the overall Laboratory strategy.

APPROACH:

Supported metal nanoparticles are commonly used in heterogeneous catalysis, and as such are critically important to myriad industrial processes. They generally possess a large variety of three-dimensional structures, which are known to directly affect their catalytic function. Effects due to features such as cluster size, state of atomic order, bond strain, facet orientation, the support, and bimetallic composition have been shown to affect catalytic activity, selectivity and stability. Importantly – even when considering model catalysts – there may be a coexistence of different particle sizes, shapes, compositions and degrees of crystalline order within the catalyst population. This heterogeneity poses a formidable challenge to commonly used ensemble averaging characterization techniques, such as X-ray absorption structure (XAS) spectroscopy, which collapses this structural heterogeneity into a single average measurement. Techniques such as electron microscopy can be used to directly measure the structural heterogeneity in a sample, but can suffer from limited statistics and experimental artifacts associated with beam damage and insufficient resolution. Thus, it is increasingly common to utilize a combination of multiple experimental and theoretical methods to more accurately describe the structural complexities that are inherent in these nanoscale systems. Further complications arise when considering the structure of supported nanoparticles subjected to reaction conditions. It has become increasingly apparent that they can exhibit substantial structural changes when they are in a working (*operando*) state and that these structural changes may no longer be apparent when the sample is returned to a non-working state or examined *ex situ*. A variety of characterization techniques have been developed in recent years that allow these structural changes to be either inferred or directly observed *in situ* (*i.e.* when subjected to approximate working conditions through the application of temperature and pressure) or under *operando* conditions. We are confronting this significant challenge through the use of microfabricated catalytic reactors that are compatible with a broad range of photon and electron based characterization methods based on imaging, spectroscopy and diffraction. These reactors allow provision of reactant streams at temperatures

of up to 800°C and pressure in excess of 1 atmosphere, and which can measure reactant streams to confirm that the system is in a working condition. This allows us to make direct comparison between the measurements in an unprecedented fashion.

TECHNICAL PROGRESS AND RESULTS:

During the past year, we completed a study on a reversed water gas shift reaction by $NiPt/SiO_2$ heterogeneous catalysts. Alloy nanoparticles have been reported to possess unique catalytic activities that are different from their parent metals, but the understanding of the structure of supported bimetallic catalysts under reaction conditions is generally lacking. Notably unclear are the details of the varying oxidation states and atomic arrangements of the catalytic components during chemical reactions. In this work, we quantitatively analyzed the variations in the oxidation state of several components of the silica supported Pt-Ni bimetallic catalyst for the reverse water gas shift reaction by a correlated use of the *in situ* XAS, Diffuse Reflectance Infrared Fourier Transform Spectroscopy (DRIFTS) and *ex situ* aberration corrected Scanning Transmission Electron Microscopy (STEM). The metal-silica interaction prevented formation of phases containing Ni-Ni bonds and thus suppressed unwanted CH_4 formation. This helped the structure evolve to the active state of the catalyst: bimetallic nanoparticles that possessed an intermetallic structure, and a Ni-rich phase. This work has been recently submitted for publication to the Journal of the American Chemical Society.

Design, Fabrication and Test of Superconducting RadioFrequency Cavity Prototype for eRHIC Energy Recovery Linac

LDRD Project # 15-006

W. Xu

PURPOSE:

The objective of this project was to design, prototype and test a 422 MHz high current superconducting radio frequency (SRF) cavity for the eRHIC SRF linac for the Linac-Ring design option. This application required the cavity to operate at a 16 MV/m continuous wave (CW) accelerating gradient, with a quality factor of $2 \cdot 10^{10}$, representing a state-of-the-art performance challenge. Following a recommendation from the Collider-Accelerator Department (C-AD) Machine Advisory Committee in 2015, C-AD management directed a change of the cavity frequency to 647 MHz. Anticipating a possible beam test at the Cornell BNL Energy Recovery Linac (ERL) Test Accelerator, the project was redirected to design, prototype and test a 5-cell 650 MHz SRF cavity – such frequency being compatible with Cornell's high current electron injector.

While the focus of eRHIC R&D has since shifted to the Ring-Ring design option, the 650 MHz niobium (Nb) prototype cavity remains a valuable cavity for use in exploring and validating state-of-the-art fabrication, processing and handling procedures for high performance, high current SRF cavities. For other frequencies and geometries, such cavities remain a crucial element of the eRHIC Ring-Ring design option, *e.g.* in the electron storage ring, the hadron storage ring and a possible high current ERL for electron cooling of hadrons. The ultimate goal was to carry out an SRF performance study of the cavity, with various post-processing procedures, in order to develop and validate production and processing procedures for eRHIC.

APPROACH:

The cavity design involves the physics design, mechanical analysis and design, and multiphysics simulations. The cavity design includes optimization of the cavity's fundamental SRF performance, as well as its very strong Higher Order Mode (HOM) damping capability. As the average HOM power is linear with the cavity loss factor, the loss factor is also an important optimization factor during the cavity design. The mechanical design has to consider various testing and operational scenarios: cryogenic vertical tests, horizontal cryomodule tests, required tuning range, fundamental power coupler requirements and stresses, HOM couplers, and so on. The multiphysics simulations are used to analyze complex cavity behaviors, *e.g.* multipacting and Lorentz detuning.

The cavity fabrication was contracted to RI Research Instruments GmbH in Germany, a company which has successfully delivered hundreds of SRF cavities to many customers, including very high performance cavities for the X-ray Free Electron Laser and the upgrade to the Linear Coherent Light Source. Through weekly telephone conferences and visits to the vendor facility for critical inspections and tests, we closely monitored every fabrication step from design of the dies, to deep drawing or hydroforming, machining, chemistry and electron-beam welding and tuning. As part of the R&D goals, we had specified tight mechanical deformation tolerances for the cavity fabrication. Because this 5-cell 650 MHz Nb cavity was the largest multi-cell cavity ever fabricated by RI, RI encountered more difficulty than anticipated in achieving the tolerance requirements, leading to seven months of delay in completion of the cavity. However, with the

strong, cooperative effort between BNL and RI, RI successfully fabricated a cavity that meets all of our specifications. The cavity was delivered in October 2017.

With the cavity delivered, it will next be post-processed through the most advanced treatments of chemical polishing, vacuum furnace firing and high-pressure rinsing that are applied to SRF cavities. Following such post-processing, cryogenic vertical tests to study the performance of the cavity, including measurements of its quality factor as a function of accelerating field, the residual resistance, helium bath pressure sensitivity and Lorentz detuning, will be performed.

TECHNICAL PROGRESS AND RESULTS:

- Cavity design - the physics and engineering design of the cavity was completed in February 2016. A 650 MHz 5-cell eRHIC cavity was designed, including cavity geometry optimization, analysis of the HOM spectrum, analysis of multipacting in the cavity, and optimization of the Lorentz detuning factor. The engineering design was completed, which included mechanical analysis of the cavity and tuning plate design. If a horizontal test is desired, a helium vessel and cryomodule can be added onto this cavity for an eRHIC cryomodule test in the future.

- Cavity fabrication - the cavity was contracted to RI in March 2016 and delivered to BNL in October 2017. Figure 1 shows the Nb cavity, which is tuned to 99% of the field flatness.

Figure 1: Nb 650 MHz 5-cell cavity

- Final LDRD Status - with the completion of the cavity fabrication and delivery to BNL, the project is complete. The cavity will continue to serve as a useful tool for the study of high performance SRF cavities.

Nanoconfined Polymer Electrolytes for Rechargeable Lithium-Metal Batteries

LDRD Project # 15-009
C. Black

PURPOSE:
Lithium (Li) metal is widely viewed as an ideal battery anode material, with a theoretical energy storage capacity 10 times higher than state-of-art commercial anodes. Despite successful commercialization in single-use batteries, Li is unusable for rechargeable batteries because growth of dendrites short-circuits the device after a small number of charge/discharge cycles. Solid polymer electrolytes with sufficient mechanical strength to suppress Li dendrite formation and/or disconnection have not yet been realized. We are exploring a new battery design that confines polymer electrolytes within nanoscale structural templates, which we believe will frustrate crystallization and provide high ionic conductivity with sufficient rigidity to suppress dendrite formation/growth. The goal of this research is to develop the fundamental understanding necessary to realize this concept.

APPROACH:
Rechargeable Li-ion batteries are the best vehicle for supplying mobile electrical power, storing electrochemical energy with graphite anodes by reversibly shuttling Li ions between cathode and graphite interstices. Although using Li metal as the anode improves storage capacity over graphite, this technology cannot be used for *rechargeable* batteries due to formation of dendrites during recharging, which short-circuit the device.

A long-proposed, but never-realized solution uses a solid polymer electrolyte with sufficient mechanical strength to suppress Li dendrite formation during battery cycling. Lithium metal batteries with polymer electrolytes, such as polyethylene oxide (PEO), have shown promise. However, the crystallinity of PEO poses a dilemma: Crystallization increases PEO mechanical modulus but hinders Li ion conduction. Melting PEO at an elevated temperature restores ionic conductivity, but decreases its mechanical strength.

We have undertaken an ambitious project to understand the role of nanoconfinement on the structural, electrical, and mechanical properties of polymer battery electrolytes, with the goal of realizing a new, rechargeable Li-metal battery architecture with high cyclability, capacity, and power delivery density. The project relies on unique BNL facilities and experimental techniques, foremost of which are X-ray scattering capabilities at the Coherent Hard X-ray Scattering (CHX) beamline at National Synchrotron Light Source II. We are measuring the crystallinity and orientation of polymer electrolytes in different degrees of nanoconfinement and at different temperatures, Li-ion doping ratios, and polymer molecular weights using Grazing-Incidence Wide-Angle Scattering. Simultaneously, we are measuring the polymer viscoelastic moduli through X-ray Photon Correlation Spectroscopy by incorporating nanoparticle tracers into the material. We have fabricated confining templates at the Center for Functional Nanomaterials using state-of-the-art lithography and etching methods.

TECHNICAL PROGRESS AND RESULTS:
Following foundational work on nanofabrication and initial measurements completed during FY 2016, this year we have completed characterization of the structure and properties of nano-confined polymer electrolytes —with results described in two different manuscripts in preparation.

Mechanical characterization: We have exploited the unique "tracer-based" viscosity measurement capabilities of the CHX beamline to establish that the nano-confined ion-conducting polymers exhibit a viscosity dramatically boosted compared to their bulk value. By measuring polymers filled

in both nanogratings (a1-a2) and nanopores (b1-b2), we conclude that the increase in viscosity is positively correlated with the degree of confinement (Figure 1).

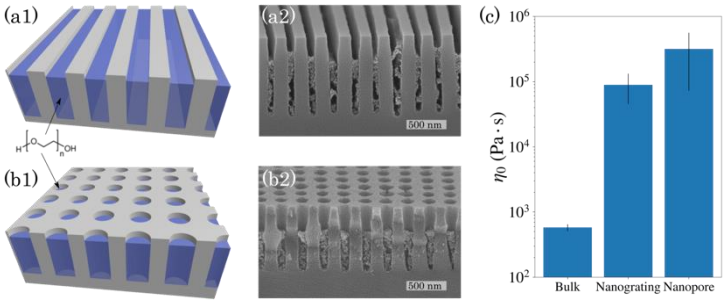


Figure 1: Schematic illustrations and scanning electron microscope images of ion-conducting polymers filled in (a1-a2) line gratings and (b1-b2) nanopores. (c) Measured zero-shear viscosities for polymers in bulk, nanograting, and nanopore geometries, showing an increase in viscosity of more than 500 times.

Structural characterization: We have completed a series of experiments to understand how the crystallinity of PEO-based electrolytes is influenced by nanoconfinement and salt additives. Most significantly, we observed that the nanoconfinements, when acting together with salt complexing, suppress crystallization of PEO (evidenced by the disappearance of crystalline peaks in Figure 2(d)), even though addition of salt alone does not.

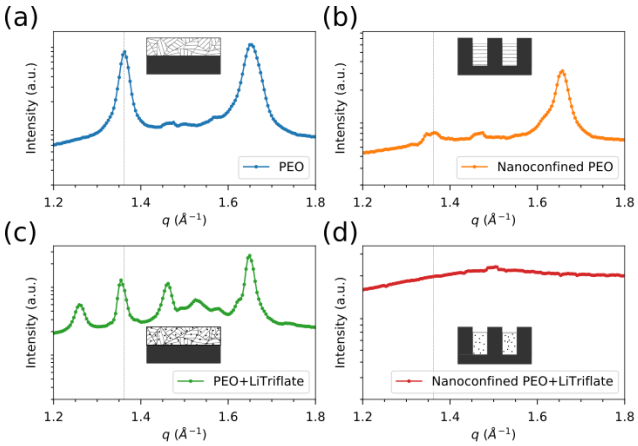


Figure 2: X-ray scattering patterns of (a) PEO (b) Nanoconfined PEO (c) PEO+LiTriflate salt (d) Nanoconfined PEO+LiTriflate salt.

Milestones:
Forthcoming experiments will nanoconfine polymer electrolytes in prototype devices (shown in Figure 3 below) and measure battery conductivity, capacity, and cyclability.

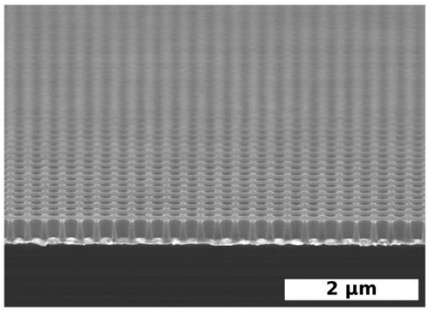


Figure 3: Scanning electron microscope image of wide-area nanopatterned battery architecture, including a metal bottom-contact for electrochemical cycling.

Hydrocarbon Chemistry on Zeolite Model Systems: Towards a Detailed Understanding of Energy-relevant Chemical Transformations Using In-Situ Techniques at National Synchrotron Light Source II, Center for Functional Nanomaterials and Chemistry Department

LDRD Project # 15-010
J. Boscoboinik

PURPOSE:

The purpose of this work is to elucidate fundamental aspects of energy-related hydrocarbon transformations using zeolite model systems that take advantage of advanced surface science tools available at BNL (National Synchrotron Light Source II [NSLS-II], Center for Functional Nanomaterials [CFN] and Chemistry Department) and tools to be developed as part of this project. While zeolites themselves are the most widely used industrial catalysts, in processes such as the cracking of crude oil and methanol to gasoline conversion, the mechanisms by which these processes occur are still far from well-understood. Detailed knowledge on other catalysts has been obtained using surface science methods, and we will use a related strategy to study zeolite chemistry, using a 2D-zeolite model system developed by the Principal Investigator (PI). Given the complexity of hydrocarbon chemistry in zeolites, it is necessary to take full advantage of the *in situ* methods available at BNL and to develop new experimental capabilities.

APPROACH:

Zeolites are the most important industrial catalysts. They are used for many energy related chemical transformations, including cracking of hydrocarbons and conversion of methanol to hydrocarbons. The active site for these processes is an extremely acidic bridging hydroxyl group within the crystalline porous framework of the zeolite. Despite extensive research carried out around the globe, we are still far from understanding the mechanistic aspects of these processes. On the other hand, the surface science community has successfully used simplified versions (models) of industrial catalysts to provide insights on structure–reactivity relationships. Using "model" catalysts, the complexity can be reduced in order to disentangle structural, chemical and electronic effects in the reactions. Zeolites remained a challenge to the surface science community due to the lack of a suitable zeolite model, until one was developed by the PI. In parallel, the PI is participating in developing surface science instrumentation for studies at elevated pressures, as opposed to traditional ultra-high vacuum studies. This includes instrumentation at the CFN and NSLS-II: ambient pressure photoelectron spectroscopy (AP-PES), polarization modulation infrared reflection absorption spectroscopy (PM-IRRAS) and reactor scanning tunneling microscopy (r-STM). The combination of a model system for zeolites and state-of-the-art instrumentation to study these processes at realistic conditions puts BNL at the forefront of research that will allow the elucidation of reaction mechanisms for some of the most important energy-related industrial chemical processes.

The AP-PES endstation and the CFN PM-IRRAS system count with mass spectrometers fed by capillary tubes adjacent to the sample surface to follow the conversion from reactants to products in the vicinity of the catalysts' surface. Some experiments were carried out with the IRRAS instrument in collaboration with Dr. Dario Stacchiola, then in the Chemistry Department. The reaction pathways hypothesized based on this data are being studied using Density Functional Theory (DFT), in collaboration with Dr. Deyu Lu at the CFN.

TECHNICAL PROGRESS AND RESULTS:
This research started in the second half FY 2015, with the hiring of postdoctoral researcher Dr. JianQiang Zhong to carry out experimental work. In June, Stony Brook University student Mengen Wang joined to carry out DFT calculations to complement the experimental work.

Dr. Zhong prepared zeolite model system samples at the r-STM at the CFN. These samples were then transferred to the AP-PES endstation at the Coherent Soft X-ray Scattering and Spectroscopy (CSX-2) beamline of NSLS-II, where they were used for technical commissioning of the endstation and beamline. In order to gain knowledge to later implement PM-IRRAS at the AP-PES endstation, a PM-IRRAS system was built at the CFN by a postdoc (John Kestell) supervised by the PI.

In FY 2016, these first experiments at CSX-2 related to the project resulted in the first publication from the CSX-2 beamline, including authors from NSLS-II and the CFN (J. Phys. Chem. C 2016, DOI: 10.1021/acs.jpcc.6b02851.). Dr. JianQiang Zhong won the best poster award at the BNL Young Researcher Symposium for this work. This was a groundwork study where the zeolite model systems (2D silicates and aluminosilicates), prepared on a Ru(0001) support, were exposed to O_2 and H_2 molecules at elevated pressures and temperatures. DFT calculations were carried out by Mengen Wang to understand core level shifts observed in the framework elements of the zeolite model. This work is important as the basis for mechanistic studies, since the catalytic activity is expected to be affected by the electronic structure of the system. This has also resulted in a second publication including theory and experiments (Top. Catal., 2016, DOI: 10.1007/s11244-016-0704-x). DFT calculations related to these experiments were carried out and another manuscript was prepared and is ready for submission. We also started exploring methanol adsorption within zeolite nanosheets using AP-PES at CSX-2 and the newly built PM-IRRAS system at the CFN, which resulted in a third publication in FY 2016 (Catal. Today, 2016, DOI: 10.1016/j.cattod.2016.07.015).

In FY 2017, work carried out using the AP-PES CSX-2 endstation on trapping small inert molecules in the pores of the zeolite model, was published in Nature Communications (Nat. Commun. 2017, DOI: 10.1038/ncomms16118), highlighted by DOE, and chosen as one of the top-10 achievements at BNL in 2017. We also managed to stabilize Cu^{2+} clusters on the zeolite model (https://doi.org/10.1007/s11244-017-0879-9). A technical paper on the design and construction of the PM-IRRAS system was accepted for publication in FY 2017 (https://doi.org/10.1063/1.5007024). We continued experiments to study cracking of small hydrocarbons on the zeolite model and carried out calculations to aid in the analysis and interpretation of the data. We have continued the design for the AP-PES+PM-IRRAS endstation and procured a new lab-based AP-PES system at the CFN scheduled for installation in 2018.

Milestones:
Analyze and publish results. Obtain further funding to continue after LDRD. Start C-C bond formation experiments using alcohols at the combined AP-PES/PM-IRRAS system. Carry out calculations to support these experiments. Summarize results of LDRD and plan future directions.

Revealing the Structure and Dynamics of Discrete Meso-Architectures

LDRD Project # 15-011

O. Gang, K. Yager, K. Kaznatcheev, A. Fluerasu, L. Wiegart, D. Yu

PURPOSE:

These studies aim to develop methods for assembly of non-periodic mesoscale structures and to develop coherent scattering and photon-correlation methods as new tools for probing finite-size mesoscale objects. Another goal of this research is the development of experimental and data analysis methods for resolving the structures of meso-assemblies using *in situ* X-ray methods.

APPROACH:

We developed a method for assembly of complex mesoclusters using nanoparticles imbedded in DNA frames to direct their interactions in a programmable manner. Our approach provides the potential for the formation of linear nanoparticles architectures of designed configurations and compositions. We exploited correlation analysis of Coherent Scattering (CS) for X-ray analysis. A rapid succession of coherent X-ray scattering images, obtained for a construct tumbling in solution, encodes meso-scale order. The correlations (*e.g.* angular) in a CS image can be accumulated to uncover local order (*e.g.* coordination number).

TECHNICAL PROGRESS AND RESULTS:

Our study tackles an important question in nanoscale self-assembly: how to fabricate low-dimensional architectures with targeted mesoscale morphology and internal organization of nano-components. The method developed is inspired by supramolecular systems, where intermolecular stacking is a key factor for the formation of the commonly observed linear morphology, for example for peptides and small organic molecules. We have developed a general nano-assembly strategy using DNA-programmable interactions that mimic stacking interactions for nanoscale objects. As result, linear architectures with complex nanoparticle arrangements can be rationally fabricated. In our method, programmable stacking allows for the highly specific assembly of the designed planar nanoparticle clusters into linear structures, so called "pillars." These multilayered pillar architectures offer a well-defined nanoparticle organization, with ability to incorporate particles of different sizes and types, as has been shown in our study. We have conducted the detailed structural characterization using electron microscopy and X-ray scattering methods to reveal the 3D structure of the formed pillar architectures and to relate them to the system parameters. We also demonstrated that the assembled pillars can be used to regulate a collective plasmon resonance for metal nanostructures.

Another key goal and accomplishment of this project was the extension of conventional X-ray scattering methods to measure meso-clusters (*e.g.* engineered assemblies of small numbers of nanoparticles). The measurement of these structures is extremely challenging since individual meso-clusters scatter extremely weakly, whereas measurements of ensembles of such structures average out the relevant structural information (local packing motifs).

We developed variance scattering methods that probe correlations within X-ray scattering datasets in order to extract otherwise hidden structural information. In particular, we demonstrated how angular correlation methods can be used to measure the structure of finite-sized meso-clusters. We nano-fabricated test samples to validate our approach and used the Coherent Hard X-ray beamline at National Synchrotron Light Source II to perform experiments

at the frontier of X-ray scattering. We demonstrated the ability to experimentally measure as few as 20 nanoparticles, thus demonstrating that it is possible to resolve individual meso-clusters using synchrotron methods. We identified the key roadblocks to implementing angular correlation methods in a realistic experimental context. The result of this work is a new, robust, data analysis pipeline, including a method for handling experimental artifacts (background, masking, noise, etc.). This work was published in the Journal of Applied Crystallography.

This research project also developed a new, ambitious measurement mode for studying weakly-scattering samples (Figure 1). The technique consists of bringing a strongly-scattering "amplifier" into proximity with a (weakly-scattering) sample of interest. Through coherent interference, the designed structure amplifies the weak scattering of the sample, allowing the measurement of the structure in previously-impossible regimes.

Combined with angular correlation analysis, this allows the structure of an otherwise invisible (below background) sample to be measured; importantly, this method scales to dynamic samples *in situ* (*e.g*. meso-structures tumbling in solution). This novel "XAmp" method was published in the International Union of Crystallography Journal (the premier journal in X-ray science).

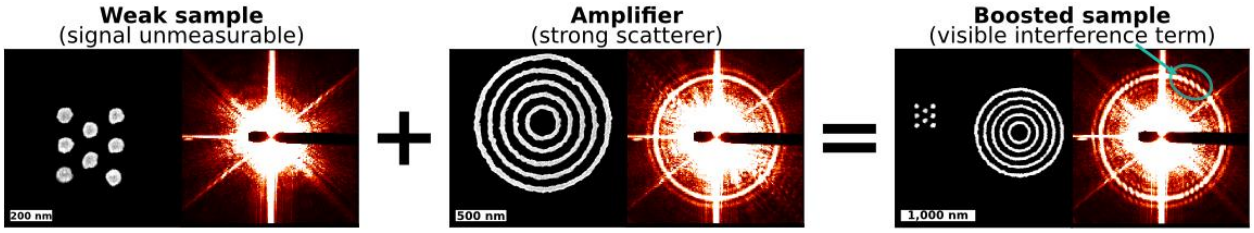


Figure 1: A new, ambitious measurement mode for studying weakly-scattering samples.

Publications:

Lhermitte, J.R.; Tian, C.; Stein, A.; Rahman, A.; Zhang, Y.; Wiegart, L.; Fluerasu, A.; Gang, O.; Yager, K.G. "Robust X-ray angular correlations for the study of meso-structures" Journal of Applied Crystallography 2017, 50, 805–819.

Lhermitte, J.R.; Stein, A.; Tian, C.; Zhang, Y.; Wiegart, L.; Fluerasu, A.; Gang, O.; Yager, K.G. "Coherent amplification of X-ray scattering from meso-structures" IUCrJ 2017, 4, 604–613.

Tian, C., Cordeiro, M. A. L., Lhermitte, J., Xin, H. L. L., Shani, L., Liu, M. Z., Ma, C. L., Yeshurun, Y., DiMarzio, D. & Gang, O. "Supra-Nanoparticle Functional Assemblies through Programmable Stacking" ACS Nano 11, 7036-7048, doi:10.1021/acsnano.7b02671 (2017).

A New Frontier for Improving Processes for Regional and Global Climate Modeling

LDRD Project # 15-020
W. Lin, S. Endo, Y. Liu, A. Vogelmann, M. Jensen

PURPOSE

This project aims to develop a novel modeling system to advance regional and global climate simulations, particularly climate change projections of extreme weather events. Deficiencies in physical process representation are long known to be responsible for uncertainty in climate change projection, more so for the projection of extreme precipitation. The new development has the potential to provide a platform to study and improve process representation in climate models and a modeling framework for regional climate simulations. Such unique combined capability aligns well with the Environmental and Climate Sciences Department's long term strategic vision to be a leader in regional and local climate modeling for the Northeastern U.S. This project also addresses the DOE Climate and Environmental Sciences Division strategic goals to "develop, test, and simulate process-level understanding of atmospheric systems" and to "develop core capability to target the research on key earth system processes that represent significant uncertainty and currently limit the predictive understanding of climate."

APPROACH:

The fidelity of climate projections depends on the representations of a wide range of cloud and turbulence processes. These processes operate at scales that are smaller than climate models and can explicitly resolve and, thus, have to rely on overly simplified representations, or so-called physical parameterizations to realize their collective influences in the climate system. Deficiencies in such parameterizations are known to be responsible for large uncertainties in climate projections that heavily rely on global climate models.

Even more challenging is the simulation of extreme weather events under climate change scenarios. This is because key physical processes such as clouds, turbulence and convection influence the mean state and variability of the large-scale atmospheric environment that spawns the extremes and dictate the extent of individual weather extremes when they occur.

The modeling system this project aims for is motivated by these challenges to develop an integrated capability to deconstruct the parameterizations of entangled atmospheric physical processes in order to improve their representations in climate models, while providing a venue to more accurately simulate weather extremes at local and regional scales in the context of global climate projections. This is achieved by integrating the regional Weather Research and Forecasting (WRF) model into the framework of the Community Earth System Model (CESM) to enable the inline coupling of the high-resolution process model (WRF) with the global climate model CAM (Community Atmosphere Model, the atmospheric model component of the CESM). In tandem, extensive analysis of standalone CAM simulations are performed to identify major climate model biases attributable to the deficiencies in physical parameterizations, followed by contrasting process level investigation, using explicit simulations of the same problem with the WRF model. In FY 2017, a new emphasis was placed on employing machine learning techniques to develop a predictive framework that can be used to characterize extreme weather conditions, and atmospheric process level relationships in order to explore novel approaches to improving process representation in climate models. The development of such a machine learning based framework is led by our new postdoctoral research associate Dr. Tao Zhang.

TECHNICAL PROGRESS AND RESULTS:

The project focused on utilizing the strength of high-resolution atmospheric modeling to identify the deficiency and improve the representation of cloud processes in global climate models. In previous years, we contributed to the improvement of the WRF model's capability for modeling at large-eddy scale with the incorporation of a new moisture treatment in the dynamical core; developed building blocks for the one-way integrated WRF-CAM system through establishing physical connections between the explicitly resolved and parameterized representation of common cloud processes; analyzed the potential of a new unified scheme for boundary turbulence and shallow convection for improving the simulations of climatically important low-level clouds; and investigated the challenges facing the climate change projection of extreme precipitations over the northeastern U.S. During FY 2017, our main progress was made on 1) developing a predictive framework based on machine learning techniques in order to explore novel approaches for improving convective parameterizations in climate models and forecasting of extreme weathers such as tropical cyclones; 2) diagnostic study of the simulation of tropical diurnal convection in the DOE's Energy Exascale Earth System Model to characterize the existing model behavior and identify key elements for model improvement.

The machine learning framework was developed with an initial application toward the prediction of tropical cyclogenesis. Interfaces were developed to enable the use of a rich set of machine learning algorithms for predicting the occurrence of tropical cyclones (TCs) from early stages of mesoscale convective systems (MCSs) that are detected by weather satellites. The machine learning algorithms extract essential properties associated with MCSs and their environment and predict the possibility of becoming TCs at specified lead times. The predictive capability is achieved by establishing relationships among the properties along the track of individual MCSs through exhaustive machine learning.

Utility software was developed to automatically extract environmental data associated with the MCSs from major data centers in order to derive the needed properties to feed the machine learning algorithms. Fig. 1 illustrates the structure of the developed machine learning framework as applied to historical MCS data over global tropical oceans to train and develop the predictive classifier to determine whether an MCS can develop into a TC.

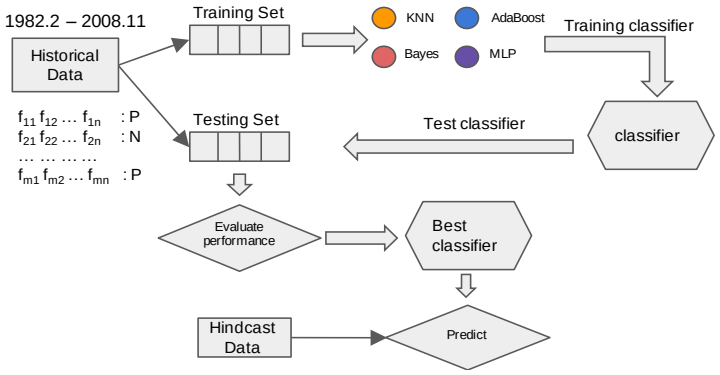


Figure 1: Flowchart for the machine learning framework.

Nine algorithms (horizontal labels in Fig. 2) were tested and very promising results were obtained particularly with the ada boost method. In the figure, *Precision* reflects the accuracy that the predicted TC actually occurs, and *Recall* reflects the reliability that all actual TCs are predicted. The F1 score is the harmonic mean of the two metrics. The adaboost method achieved a near perfect F1 score of 96%.

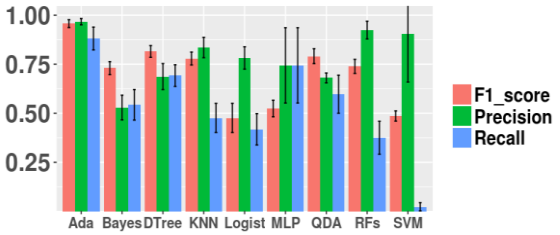


Figure 2: Skills in predicting TC Genesis develop a predictive classifier to replace the convective trigger in climate models that will have broad implication including improving the simulations of diurnal convections.

Growth of Self-activated Scintillators for Dual Gamma- and Neutron-Detection

LDRD Project # 15-025
U. Roy, G. Camarda

PURPOSE:

The main purpose of this project is to establish BNL as a major R&D contributor in the field of scintillator detectors for national security applications. There is an urgent need for large-volume, economical, uniform scintillators for detecting and imaging gamma-rays and neutrons for several nonproliferation applications, such as emergency response, counter-terrorism, treaty verification, and nuclear safeguards, and for medical imaging. Our objective is to develop intrinsic scintillating compounds for simultaneously detecting gamma-rays and neutrons, which will resolve the problems present in doped scintillators, such as Ce-doped Cs_2LiYCl_6 (CLYC). The results and outcomes of the project are expected to lead to substantial new funding from various agencies, such as the National Nuclear Security Administration, the Domestic Nuclear Detection Office, and the Defense Threat Reduction Agency.

APPROACH:

Cs_2LiYCl_6 doped with Ce is perhaps the most efficient scintillator for a dual gamma- and neutron-detector. Detectors fabricated using doped scintillators undergo significant degradation with increasing volume, due to the segregation/striations of the dopant (*i.e.*, the activator). The resulting non-uniformity of response reduces the yield of large detectors and increases their cost. In contrast, intrinsic compounds will ensure the homogeneity of the material throughout the grown ingot volume, and thus, assure a uniform response from the detector regardless of its volume.

We are developing $Cs_2LiCeCl_6$ (CLCC), an intrinsic scintillator for the dual detection of gamma-rays and neutrons. CLCC crystals have a cubic structure, and were grown by the vertical Bridgman growth technique. The crystals were characterized for different scintillator properties and device performance for both gammas and neutrons. Collaborators are Y. Cui, G. Yang, A. Hossain, P. Vanier and R. James. We have also established a collaboration with Steve Payne's group at Lawrence Livermore National Laboratory (LLNL).

TECHNICAL PROGRESS AND RESULTS:

CLCC crystals of different diameters, up to 22 mm, were grown by the vertical Bridgman growth technique. Figure 1 shows the emission of CLCC. A double peak was observed at ~384 and 402 nm. The appearance of a double peak is typical for the Ce^{3+} state. The decay time which is one of the most important parameters for scintillators was estimated using 662 keV gamma excitation. Figure 2 shows the decay time of CLCC, which comprises three components. The fastest decay time constant for ~80% of the total light yield is ~90 ns, which is much faster than CLYC. Figure 3 shows the gamma response from a ^{137}Cs source; the energy resolution, measured at LLNL, is 5.1% at 662 keV. It is worth noting that, our first crystal showed 7% resolution at 662 keV. The resolution can be further improved by purifying the

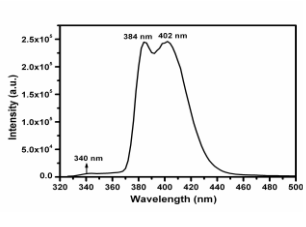


Figure 1: Emission spectrum of CLCC.

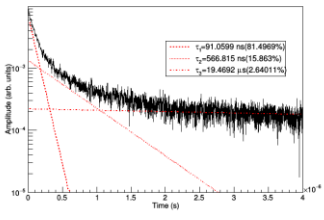


Figure 2: Fluorescence decay obtained from CLCC using 662-keV excitation from a ^{137}Cs source.

starting material by zone refining. The light output estimated by comparing to Bismuth Germanate (BGO) was ~20,000 ph/MeV, while 34,000 ph/MeV light output was obtained at LLNL comparing to NaI. The reason for the lower light yield obtained at BNL might be due to the highly hygroscopic nature of the sample. We performed the sample preparation and measurements under ambient conditions, immersed in oil/optical grease. The surface degradation might have caused lower light yield. At LLNL, all the processes were done in a controlled atmosphere. The results show that the light output is higher than CLYC. The non-proportionality for CLCC was estimated by Steve Payne's group using the Scintillator Light-Yield Non-proportionality Characterization Instrument (SLYNCI)[1] (Figure 4). It was observed that carrier trapping in CLCC is much smaller than CLYC or CLLB ($Cs_2LiLaBr_6$):Ce, while for both CLYC and CLCC, the percent of free electrons/holes created is similar, ~ 22% and 23% respectively.

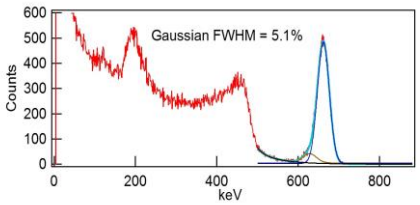


Figure 3: Pulse height spectra of CLCC crystal for ^{137}Cs gamma source.

The response to dual gammas and neutrons was evaluated by measuring the pulse-height spectra using Am-Be and ^{137}Cs sources together. Figure 5 shows the dual gamma and thermal neutron spectrum for the device using CLCC. The measurement was carried out at BNL. The well-resolved 662 keV gamma line and the thermal neutron line are marked with arrows in Figure 5. The inset shows the CLCC sample irradiated with Ultraviolet (UV) light, depicting the emission from the sample under the UV excitation. The gamma-equivalent peak position of the neutron line was estimated to be at 1.46 MeVee with a resolution of ~4%, while the resolution obtained at LLNL was ~3%, with the gamma-equivalent peak position of the neutron at 1.4 MeVee. The pulse shape discrimination for gamma and neutron of the CLCC sample (Figure 6) was measured at LLNL using a moderated Cf-252 source. The gamma and neutron events are well-separated.

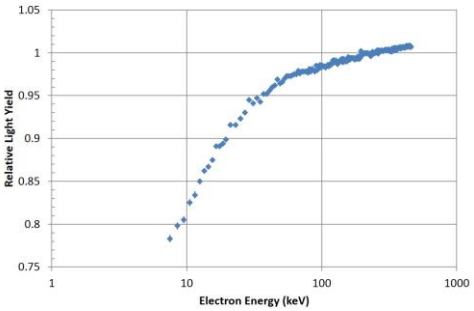


Figure 4: Non-proportionality plot for CLCC, measured at SLYNCI.

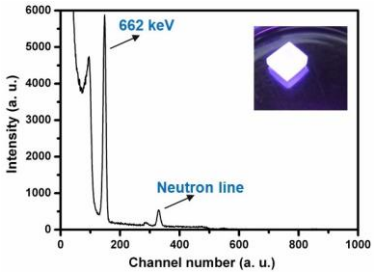


Figure 5: Pulse height spectra of $Cs_2LiCeCl_6$ crystal from Am-Be thermal-neutron and ^{137}Cs gamma source. Inset shows the sample excited by UV light.

Summary: We have successfully grown intrinsic $Cs_2LiCeCl_6$ for dual gamma-ray and neutron detection. The CLCC showed several advantages compared to the well-known crystal CLYC for dual gamma-neutron radiation detectors. CLCC is faster and brighter than CLYC. However, the gamma equivalent peak position of the thermal neutron line is lower than for CLYC.

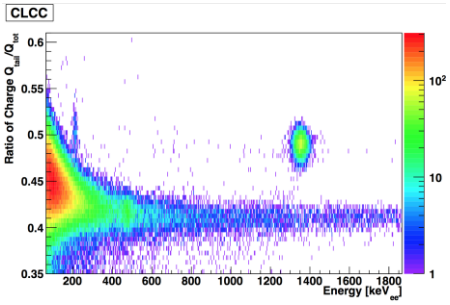


Figure 6: Pulse shape discrimination for CLCC using Cf-252 source.

[1]Payne *et al.*, IEEE Trans. Nucl. Sci., 58 (6), December 2011.

Inelastic X-ray Scattering Determination of the Inter- and Intra-Particle Dynamics of Nanoparticle Superlattices: Key to the Development of THz Phononic Crystals

LDRD Project # 15-031
A. Cunsolo, Y. Cai, O. Gang

PURPOSE:
The goal of this project is to investigate phonon propagation in programmable assemblies of nanoparticles, with the aim of searching for metamaterials that hold the potential of new functionalities. It is aligned with BNL's strategic vision on complex nanostructured materials from both an applied and fundamental point of view. The main purposes are: 1) exploration of a new science frontier - characterization of the inter- and intra-particle high frequency elastic (acoustic) properties of nanoparticle superlattice assemblies synthesized at the Center for Functional Nanomaterials (CFN); 2) development of THz phononic structures; 3) high-impact first experiments using the unique capabilities of the Inelastic X-ray Scattering (ISX) beamline at National Synchrotron Light Source II (NSLS-II).

APPROACH:
The goal of THz acoustic manipulation is aggressively pursued through the synthesis of nanoparticles of various shapes, sizes and materials, arranged in nm-spaced superlattices and linked by ligands with tunable strength. The aim is to form new structures functioning as THz phononic crystals, *i.e.* enabling manipulation/programming of phonon transport and, consequently, of heat management based upon structural engineering. The use of the new high-resolution/high-contrast IXS spectrometer at NSLS-II is combined with the most advanced nanostructure engineering developed at the CFN.

TECHNICAL PROGRESS AND RESULTS:
The project terminated in July 2017, when Dima Bolmatov, the postdoctoral research associate hired for the project, left BNL for a position at Oak Ridge National Laboratory. In the past year, several measurements were performed using allocated beamtime at the 10ID beamline of NSLS-II. Surita Bhatia of Stony Brook University collaborated with us on the project.

Most importantly, the major achievement was the first scientific publication entirely focused on measurements performed on this beamline. Proposals for beamtime were submitted and related measurements were performed.

In FY 2017, our effort was mostly focused on IXS measurements on phononic properties on Soft Matter systems. The following proposals were submitted and were allocated beamtime:

- Proposal title: "Phononic gaps in liquid crystals as fingerprints of order-disorder transitions at THz scale"- measurements performed at 10ID IXS beamline using beamtime allocated in the 2017 - 1st cycle
- Proposal title: "Block-copolymers as a new generation of phononic and optomechanical metamaterials"- measurements performed at 10ID IXS beamline using beamtime allocated in the 2017 - 2nd cycle

- Proposal title: “Phonon propagation in super-lattices of Au nanospheres” - measurements performed at 10ID IXS using beamtime allocated in the 2017 - 2nd cycle
- Proposal title: "Search for optical phononic modes in mesogenic systems”- measurements performed at 10ID IXS beamline using beamtime allocated in the 2017 - 2nd cycle.

The results of these measurements are still to be analyzed and expectedly will be the focus of high-profile publications.

As mentioned above, the most important result of this intensive research effort is the first publication focused on scientific results obtained at the 10ID beamline in a high profile (Impact Factor 14.2) research journal: D. Bolmatov, M. Zhernenkov, L. Sharpnack, D. M. Agra-Kooijman, S. Kumar, A. Suvorov, R. Pindak, Y. Q. Cai, A. Cunsolo, “*Emergent optical phononic modes upon nanoscale mesogenic phase transitions*”, Nano Letters 17, 3870–3876 (2017).

This publication has also been selected as a DOE Science Highlight.

Searching and Sorting Haystacks

LDRD Project # 15-034
S. McSweeney, Q. Liu, W. Hendrickson

PURPOSE:

This research has successfully employed the tools (developed through the first years of the project) to assemble complete datasets from macromolecular crystals presenting many thousands of fragmentary sets. Traditional macromolecular crystallography (MX) experiments require one or at most two (a low and a high resolution) sweep data collection plans (Figure 1a); more complex strategies are possible based upon the radiation robustness of the crystal (Figures 1b, c). However, National Synchrotron Light Source II (NSLS-II) will bring scientists into completely new regimes where data sets will be generated from thousands of crystals (Figure 1d).

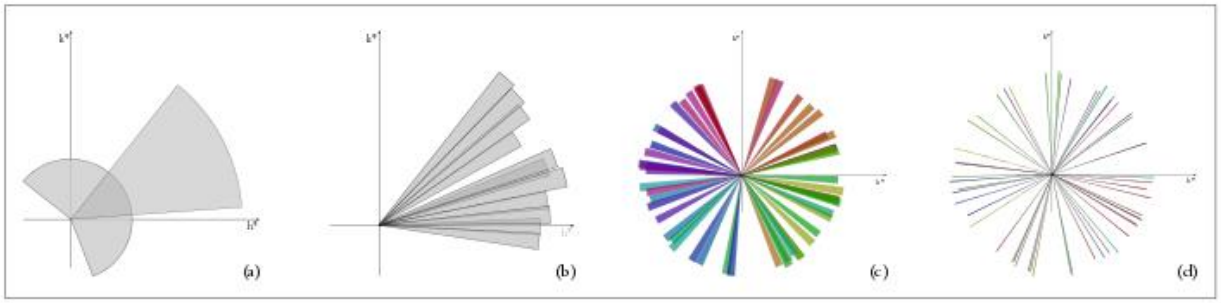


Figure 1: Data collection strategies for MX data, each wedge represents an experiment sweep revealing part of the data for a crystal structure. (a) Low and high resolution sweeps (b) "complicated" multi-orientation strategies (c) optimistic multi-crystal strategies where several degrees of data per sample are possible, many samples lead to a data set (d) at NSLS-II narrow fragments of data sets will be possible for each crystal 0.2 degrees per-sample; hundreds or thousands of crystals will be necessary for each data set. In (c) and (d) the color represents the order in which the samples contributed in the simulation; changes in length represent different diffraction limits for the samples.

APPROACH:

Our approach to this problem was first to develop the infrastructure and methods for simulating the diffraction experiments expected at our new beamlines – this has been done and we are now able to sustain data rates of up to 750Hz.

In parallel, tools for examining these data were created and benchmarked. We have a number of data collection and analysis “pipelines” established to allow for smooth interpretation of these data. In addition, to supplement these computational investigations, we will be developing crystal samples to provide experimental data for further refinement of the technique.

Several experiments representing beamline usage scenarios were explored and adjustments made to account for the subtle differences between these measurements. These software tools will be made more robust to allow for routine use at NSLS-II. The results will also be published as peer-reviewed manuscripts (two manuscripts submitted, more in preparation). This work will form the basis for continued support by an application through competitive funding routes – we have developed a proposal.

TECHNICAL PROGRESS AND RESULTS:

We have been able to demonstrate the value of the approach to both the determination of metabolites binding and to *ab initio* phasing of macromolecules. Figure 2, taken from our

recently submitted manuscript, shows the statistics that allow us to differentiate between partial datasets determined from 95 individual crystals. These results and their application should allow us to continue to explore the possibilities that the macromolecular crystallography beamlines offer.

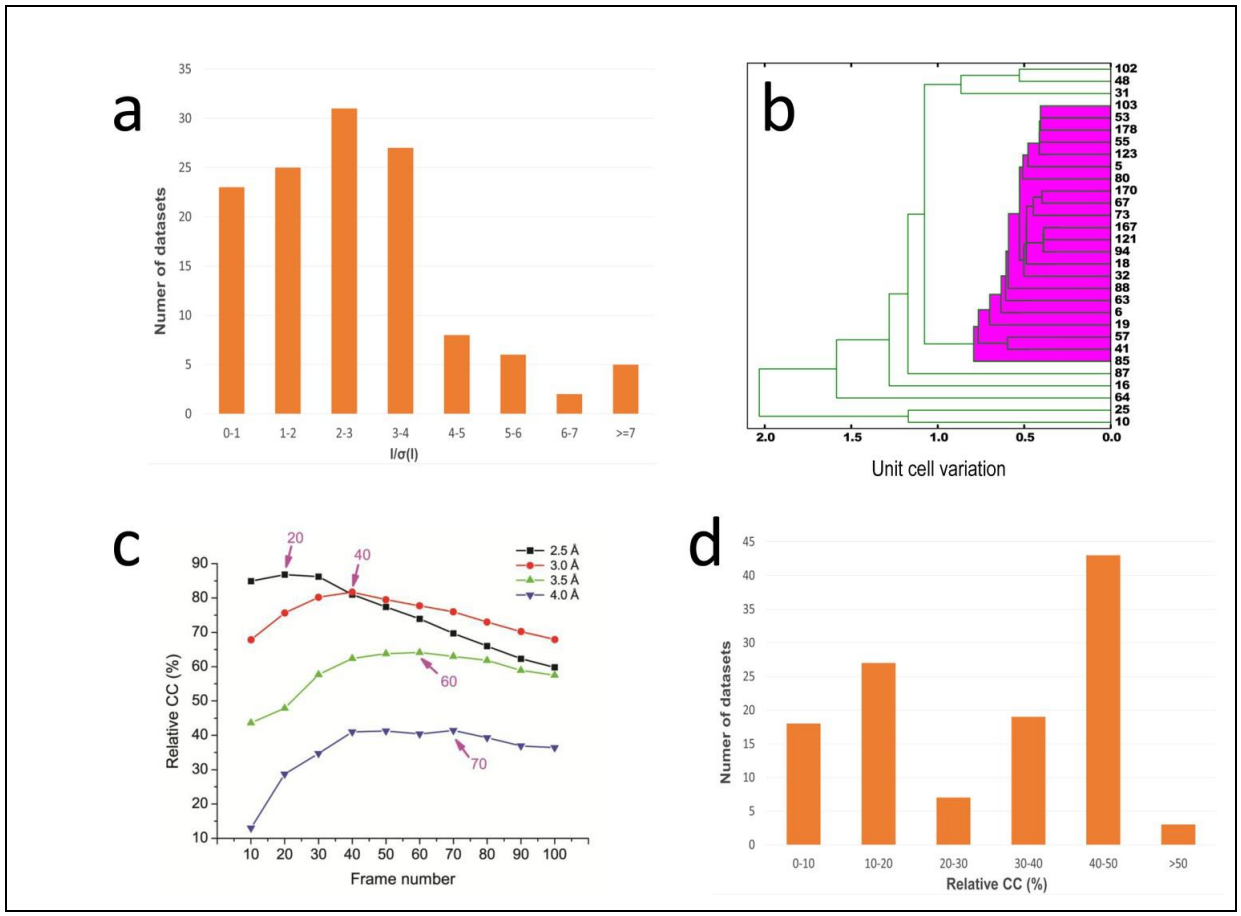


Figure 2: Data analysis of individual microcrystals. (a) Histogram distribution of I/σ(I) values for single-crystal datasets used to obtain the reference dataset. (b) Unit cell variation analysis for classification of single-crystal datasets in the reference dataset. The eight crystals in the magenta-colored cluster are representative of the 95 crystals that co-clustered in the dendrogram. (c) Relative correlation coefficient (RCC) of a typical single-crystal dataset to the reference dataset at four different diffraction limits. (d) Histogram distribution of RCC values for the 117 selected single-crystal datasets.

In-situ Microscopy Investigation of Complex Manganese Oxides for Energy Storage

LDRD Project # 15-037
Y. Chu

PURPOSE

Advanced battery materials have complex hierarchical structures, ranging from the atomic scale to the millimeter scale. Development of more efficient and safe batteries requires a comprehensive understanding of how these complex interfacial structures are transforming during the charge/discharge reactions. One of the major challenges is a lack of suitable imaging methods that can provide comprehensive structural and chemical information with sufficient resolution and sensitivity. The scientific purpose of this research is to develop multi-scale and multi-modality imaging methods in an *in situ* sample environment. A key fundamental scientific goal of the project is to understand the interplay between the nanoscale electrode structure and electrochemical reversibility, by leveraging the world-leading microscopy capabilities of the Hard X-ray Nanoprobe (HXN) beamline at National Synchrotron Light Source II (NSLS-II) and electron microscopes at the Center for Functional Nanomaterials (CFN). Successful implementation of the project will tremendously strengthen BNL's scientific leadership in energy research, and the developed capabilities will be offered to the general users of these facilities.

APPROACH

In order to investigate battery materials from the mesoscale down to the atomic scale, we employ the HXN beamline and transmission electron microscopes (TEM). In addition, we collaborate with Prof. Esther Takeuchi of BNL/Stony Brook University, who has world-leading materials' knowledge on a wide range of battery materials. There are two specific aims of this research. Our first aim is to develop liquid electrochemical cells that allow HXN and TEM investigation under *in situ* conditions. We developed three different *in situ* cells with different purposes. An *in situ* cell developed through a commercial vendor provides the capability to perform TEM and X-ray microscopy measurements. This cell was developed, commissioned, and used for TEM and X-ray experiments. The second cell is an optimized X-ray cell, which allows both X-ray fluorescence and diffraction measurements. It uses a much larger X-ray window and has better electrical contacts to the sample. The third cell is a modified coin-cell that offers the highest familiarity to battery researchers. All the cells are used at the HXN beamline for this research as well as for the general user science experiments at the HXN beamline.

Our second aim is to use these cells to conduct *in situ* microscopy experiments in order to image how the nanostructure at the battery interface transforms under *in situ* controls. In the course of our research, we realized that we had to develop methods to analyze the collected image data. We began tackling a way to quantify the amount of different chemical species in a sample. We also began exploring a method to solve the self-absorption problem for fluorescence nanotomography. Initial attempts demonstrated highly encouraging results. We are now applying these methods to the user experiments at the HXN beamline.

TECHNICAL PROGRESS AND RESULTS

The developed cells enabled us to perform *in situ* microscopy experiments on a broad range of material systems. We used the TEM/Nanoprobe *in situ* cell that we developed for investigating the electrochemical deposition of silver (Ag) nanoparticles using a scanning transmission electron microscope (STEM). In this work, we observed a highly unusual behavior of Ag

nanoparticle growth from solution. We attributed this unusual growth mode to the interaction of the electron beam in solution plus the catalytic behavior of metallic platinum A summary of this work was published (Mingyuan Ge, Ming Lu, Yong S. Chu, and Huolin Xin, "Anomalous Growth Rate of Ag Nanocrystals Revealed by in-situ STEM", Sci. Reports, **7**, 16420 (2017)). Fig. 1a shows the schematics of our optimized X-ray *in situ* cell. This cell offers significantly greater flexibility and robustness. It has a dedicated sub-chamber for lithium (Li), so that Li does not get depleted during the *in situ* reaction. It allows different materials (Kapton, Mylar, or Carbon sheet) as X-ray windows, so that the type of window can be chosen optimally for each experiment. Fig. 1b shows *in situ* discharging and charging of an iron oxide battery material. Fig 1c shows the oxidation state image of iron and iron oxide species when discharged (A) and charged (B). The images show their relative concentrations during the *in situ* reaction. Both cells are offered to the general users of the HXN Beamline.

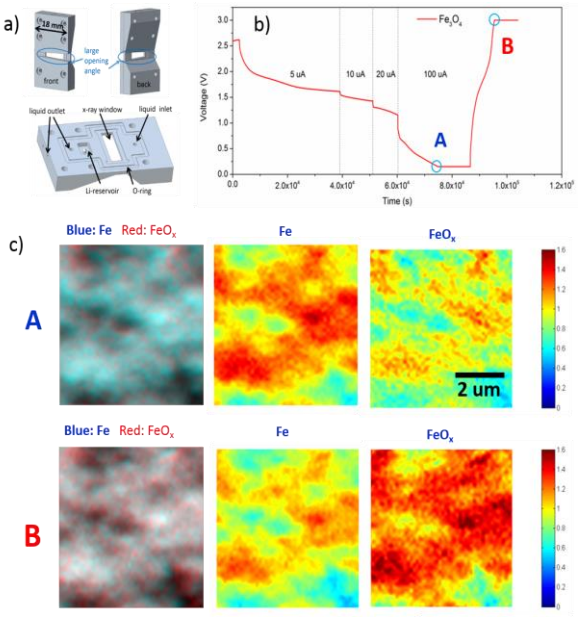


Figure 1: (a) Optimized X-ray *in situ* cell; (b) *In situ* electrochemical control of an FeO battery material; (c) Chemical map of the sample at fully discharged (A) and fully charged (B). The image in the first column shows the relative fractions of metallic Fe (blue) and Fe oxides (red).

We also developed a robust tomography analysis method for analyzing 3D elemental distribution. Until now, such analysis was not possible due to a well-known problem called self-absorption. A significant fraction of emitted fluorescence X-rays are absorbed in the sample, producing significant errors for 3D tomographic reconstruction of the detected signals. For the first time, we demonstrated that self-absorption can be corrected without relying on pre-knowledge or using model-based calculation. This is a significant achievement in the field of X-ray microscopy, which will significantly broaden the application of 3D fluorescence tomography. A manuscript summarizing this work is in preparation.

In summary, this research resulted in significant scientific capability at the HXN beamline, allowing general users to perform *in situ* imaging experiments using both electrons and X-rays. The *in situ* cells that were developed are actively used by the HXN users. In addition, we developed a robust 3D fluorescence imaging method, solving the self-absorption problem.

Chiral Magnetic Effect (CME): from Quark-gluon Plasma at the Relativistic Heavy Ion Collider to Dirac Semimetals at National Synchrotron Light Source II

LDRD Project # 16-004
Q. Li, D. Kharzeev

PURPOSE:
This cross-disciplinary project aims at establishing the existence of quantum phenomena specific to relativistic quasi-particles in condensed matter experiments and exploring their potential applications. The project will combine experimental and theoretical studies to understand the nature of the chiral magnetic effect (CME) - generation of electric current induced by the chirality imbalance between left- and right-handed fermions in a magnetic field, a dramatic consequence of chiral anomaly in the theory of Dirac fermions.

APPROACH:
The relativistic theory of charged chiral fermions (massless spin 1/2 particles with a definite projection of spin on momentum) in three spatial dimensions possesses so-called chiral anomaly – non-conservation of chiral charge induced by the external gauge fields with non-trivial topology. One of the anomalies is the chiral magnetic effect. This phenomenon is currently under intense study in relativistic heavy ion collisions at the Relativistic Heavy Ion Collider (RHIC) at BNL and at the Large Hadron Collider at CERN.

Interactions of quasiparticles in Dirac and Weyl semimetals, having linear energy dispersions, lead to chiral fermions that opened unprecedented opportunities to study the quantum dynamics of relativistic field theory in condensed matter experiments, with the potential for important practical applications, such as non-dissipative charge transport [1,2]. Currently, topological physics in 3D Dirac systems is a very active field. Data are rapidly being produced on electronic structures and transport properties. However, the pieces of information are not well connected, leaving important questions unanswered, for example, how does the chiral magnetic effect respond to the change of electronic structure and topology in Dirac/Weyl semimetals? A coordinated investigation that brings together theory, materials synthesis, and characterization of crystal structure, electronic structure, and transport properties is desirable. This coordinated investigation is our approach used in this LDRD.

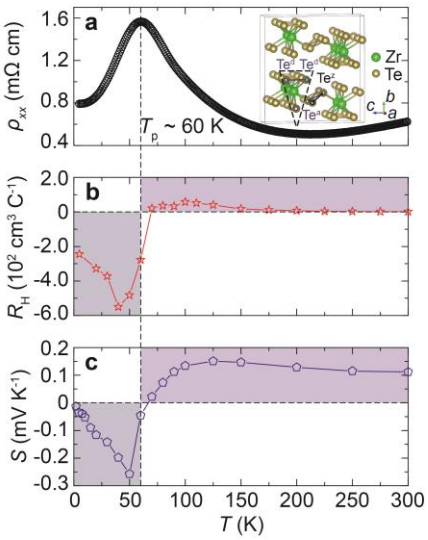


Figure 1: The temperature-dependent (**a**) electrical resistivity ρ_{xx}, (**b**) Hall coefficient R_H, and (**c**) Seebeck coefficient S. The dashed line, at the ρ_{xx} peak temperature T_p (approximately 60 K), indicates the resistivity "anomaly" occurs essentially at the temperature when both R_H and S change sign. The crystal structure of $ZrTe_5$ is illustrated in the inset of (**a**).

TECHNICAL PROGRESS AND RESULTS:
Zirconium pentatelluride $ZrTe_5$ hosting chiral fermions exhibits a strong chiral magnetic effect [1]. However, the origin underlying its anomalous resistivity peak (Fig. 1) has been under debate for decades. We provided transport evidence substantiating the anomaly to be a direct

manifestation of a Lifshitz transition in the Dirac band with an ultrahigh carrier mobility. We demonstrated that the Lifshitz transition was readily controllable by means of carrier doping, which sets the anomaly peak temperature T_p. T_p is found to scale approximately as $n_H^{0.27}$, where the Hall carrier concentration n_H is linked with the Fermi level by $\varepsilon_F \propto n_H^{1/3}$ in a linearly dispersed Dirac band. This relationship indicates T_p monotonically increases with ε_F, which serves as an effective knob for fine tuning transport properties in pentatelluride-based Dirac semimetals [3].

In a strong magnetic field, the longitudinal magnetoconductivity in 3D chiral materials is shown to exhibit a new type of quantum oscillation, arising from the chiral magnetic effect. These quantum CME oscillations are predicted to dominate over the Shubnikov-de Haas (SdH) ones in chiral materials with an approximately conserved chirality of quasiparticles at strong magnetic fields. The phase of quantum CME oscillations differs from the phase of the conventional SdH oscillations by $\pi/2$ [4].

Experimental verifications of the theoretically predicted topological insulators are essential steps toward the applications of the topological quantum phenomena. In the past, theoretically predicted topological insulators were mostly verified by measurements of the topological surface states. However, as another key feature of the nontrivial topology in topological insulators, an inversion between the bulk bands has rarely been observed by experiments. Here, by studying the optical transitions between the bulk Landau levels of $ZrTe_5$, we not only offer spectroscopic evidence for the bulk-band inversion — the crossing of the two zeroth Landau levels in a magnetic field, but also quantitatively demonstrate three-dimensional massive Dirac fermions with nearly linear band dispersions in $ZrTe_5$. Our investigation provides a paradigm for identifying topological insulator states in candidate materials. This is a collaborative study led by the Institute of Physics (Beijing) and Peking University in China [5].

REFERENCES:

[1] Q. Li, D. E. Kharzeev, C. Zhang, Y. Huang, I. Pletikosic, A. V. Fedorov, R. D. Zhong, J. A. Schneeloch, G. D. Gu and T. Valla. "Chiral magnetic effect in $ZrTe_5$" arXive:1412.6542, *Nature Physics* **12** 550 (2016) doi:10.1038/nphys3648

[2] Q. Li and D. E. Kharzeev "Chiral magnetic effect in condensed matter systems" *Nuclear Physics* A **956** 107–111110 (2016)

[3] H. Chi, C. Zhang, G. Gu, D. E. Kharzeev, X. Dai, Q. Li "Lifshitz transition mediated electronic transport anomaly in bulk $ZrTe_5$" New Journal of Physics **19** 015005 (2017)

[4] S. Kaushik and D. E. Kharzeev "Quantum oscillations in the chiral magnetic conductivity" Phys. Rev. B **95**, 235136 (2017)

[5] Z. G. Chen, R. Y. Chen, R. D. Zhong, J. Schneeloch, C. Zhang, Y. Huang, F. Qu, R. Yu, Q. Li, G. D. Gu, N. L. Wang "Spectroscopic evidence for bulk-band inversion and three-dimensional massive Dirac fermions in $ZrTe_5$" Proceedings of the National Academy of Sciences **114**, 816 (2017).

Serial Micro Crystallography at Full Flux

LDRD Project # 16-006
M. Fuchs, E. Nazaretski

PURPOSE:

The Frontier Micro-focus Macromolecular Crystallography (FMX) beamline at National Synchrotron Light Source II (NSLS-II) with its 1 micron beam size and photon flux of 3×10^{12} ph/s at the current operating current has reached new and unprecedented dose rates for the structure determination of biological macromolecules. The goal of this project is to develop a new ultra-high-speed high-precision scanning goniometer to support fixed target serial micro crystallography at the full flux of this new beamline. The high dose rate presents a great advantage for serial micro crystallography in cutting measurement times from hours to minutes, and for time-resolved serial crystallography measurements in providing ms time resolution. In serial crystallography, a complete crystallographic dataset for determining the structure of a biological molecule is assembled from hundreds of partial datasets obtained from micron-sized, and therefore short-lived, crystals. To provide the instrumentation basis for such measurements, we have designed and constructed an ultra-high-speed, high precision goniometer and complementary specialized sample supports. This new instrument enables data collection from previously intractably small crystal sizes, often encountered in challenging structural biology projects, such as membrane proteins. The results bring the data collection speed of scanning serial crystallography from several hours into the few minutes range, and thereby establish it as a new standard tool to tackle crystallography with the most challenging microcrystals.

APPROACH:

Serial crystallography methods for room-temperature crystallography were recently adapted for data collection from microcrystals at Free Electron Laser (FEL) facilities. With microfocus synchrotron beamlines now able to collect useable data from ever smaller crystals, these methods have been re-appropriated by the synchrotron crystallographic community for their ability to compile full datasets from partial microcrystal-datasets. With the FMX beamline's full-flux crystal half-life in the 10 ms range, to realize the speed advantage, a nanometer-precision high-speed piezo scanner optimized for fast rotation is needed for the required sample positioning performance.

The scanner unit designed and constructed in FY 2016 required the integration of piezo positioners on an infinite rotation rotary spindle, which to our knowledge has not been done with the repeatability requirements needed for our application. A suitable metrology setup was implemented using capacitive and optical micrometer distance sensors to calibrate the device during commissioning. The instrument is being embedded in the beamlines controls and data processing environment. Successful structure solutions of challenging test microcrystal samples have proven the validity of the scanning approach up to the maximal detector speeds of the FMX beamline at full flux. This establishes the method of rapid scanning serial micro crystallography as a unique tool for structure determination of previously intractable molecular targets.

Yuan Gao, the postdoctoral research associate on the team since May 2016, works full-time on the project. W. Xu from the NSLS-II Experiment Group provided metrology support. M. Lu from the Center for Functional Nanomaterials (CFN) helped with sample holder microfabrication. S. Myers, B. Seiva-Martins and J. Skinner from the BNL Controls Group supported control system integration. W. Shi from Case Western University and J. Jakoncic from BNL provided support with crystallographic data processing, and A. Soares (BNL) provided test samples.

TECHNICAL PROGRESS AND RESULTS:
The core objective for FY 2017 was to fully commission the ultra-high-speed high-accuracy scanning goniometer and perform first experiments. After final load tuning at the manufacturer (PI Physik Instrumente), the new crystal goniometer, the Nanocube piezo scanner, was returned to BNL in February 2017. It was extensively tested and calibrated in BNL's nanometrology laboratory, to establish the necessary calibration routines and optimize the driver settings for the high scanning speed performance. A dedicated LabVIEW driver was developed to upload the optimized trajectories to the controller and provide an interface to the FMX beamline control system. To integrate the new device into the beamline control system, the scanner translations needed to be synchronized with the crystallographic rotation axis and with the detector readout. This was achieved by integrating it with the NSLS-II standard scanning controller unit. The new scanning logic of the scanner with its combination of coarse and fine stages was then integrated with the graphical user interface and controls server LSDC.

In the first experimental run in May, the delay settings of all devices were tuned in scanning experiments over the full speed range using thin crystal needles to ensure the precision of the combined systems. Using the first standard test datasets, the scanner's performance was compared with the established slow goniometer hardware with a focus on achieving equally high data quality, as measured by the R-factors achieved. In September and December, we collected full flux serial microcrystal datasets on proteinase K. The collections were run at a set of different scanning speeds up to the maximum framerate of 750 Hertz of the beamline's Eiger 16M detector, and at raster precisions down to 1 µm, surpassing current macromolecular crystallography goniometer systems by an order of magnitude (to the best of our knowledge). To measure and calibrate high-speed movements of the device, a fast camera system was installed and tested.

To enable efficient data processing at the newly achieved collection speeds, Yuan is developing and evaluating a Python-based pipeline for automated data processing that sequences established crystallographic codes for the required processing steps. The challenge was to optimize a workflow for rotation data collection from the smallest crystals, as opposed to the FEL-like still image collection, which requires statistical averaging and uses significantly more crystals for a complete dataset.

To complement the new goniometer, a first batch of new silicon wafer-based sample holders was microfabricated at the CFN. In the first tests, we verified their performance in sample loading and their compatibility with the sample cooling system and established an accessible rotation range well past 50°. Their main advantage is high stiffness and the ability to remove excess buffer solution to minimize background scattering and thereby optimize signal to noise. An inspection microscope and a high speed linear motor will be integrated in a cryo-plunger system to support loading of the sample holders and freezing without ice-crystal formation.

Milestones:
In FY 2018, we will extend the method to other sample systems. We have arranged experimental runs with the first external user groups, Q. Liu of BNL and S. Gabelli of Johns Hopkins University. The controls system work will focus on automation and on integrating the user interface in order to turn the system into a standard user operation device.

3D Ptychography Imaging without Rotation using Highly Convergent X-ray Beam

LDRD Project # 16-007
X. Huang

PURPOSE:

The objective of this project is to develop a novel microscopy method for obtaining high-resolution three-dimensional (3D) images without sample rotation. This technique will revolutionize the way of extracting 3D structural information, since the performance and resolving capability of existing microscopy tools are inherently limited by the detection geometry. The successful implementation of this method will find wide application in quantitative characterization of specimens in an *in situ* environment. Elimination of the rotation requirement removes the limitation imposed by the usually bulky sample cell and provides high quality 3D structure with ultimate spatial resolution.

APPROACH:

Multi-slice ptychography was proposed to solve long standing problems in imaging thick samples with diffraction limited spatial resolution. This approach numerically decomposes a thick material into slices and models the interaction between the incident X-ray and sample, slice by slice. The structural information on each slice is encoded into the propagating wavefront independently. Under this framework, multiple planes can be reconstructed simultaneously. They can form either a 3D view of the specimen or provide a projection image with extended depth of field beyond the limitation imposed by the imaging system. This method is capable of providing high resolution images of thick samples or samples embedded in complex *in situ* environments. With further improvement of the depth resolution, ultimately a 3D image can be obtained without sample rotation.

TECHNICAL PROGRESS AND RESULTS:

Before we applied the technique to a true 3D sample system, we verified our experimental approach and algorithms using a two-layered structure, which can be considered as a pseudo and simplified 3D system. We prepared nanoparticles on two sides of a silicon membrane. The membrane thickness matches the depth resolving capability provided by the multilayer Laue lens (MLL) used in our experiment. The normal way to enhance achievable depth resolution is to collect high spatial frequency scattering signals as far as possible. However, as the scattering signal decreases dramatically with the increment of spatial frequency, in practice, it is a challenge to measure those signals with a sufficiently high signal-to-noise ratio. Reported results used 10 seconds of exposure time to record high-q signals. The novelty of our approach is that instead of chasing those weakly scattered signal, we use only the intense holographic area diverged from the focused beam. Due to the high numerical aperture of the MLL optics, the holographic area provides lateral resolution better than 12 nm and depth resolution of around 4 um. The most important point is that it allows us to collect data with 0.05 seconds exposure time, which improves the throughput of the method by three orders, while achieving better resolution. We tested our idea with an experimental measurement, and we successfully reconstructed two layers separated by 10 um with 8.1 nm lateral resolution, which is the best to our knowledge. The manuscript has been submitted [1].

To improve the depth resolution further, we plan to explore how to redistribute scattering power into the high-q region. We will introduce certain high frequency features into the focused beam, which will enhance the high-q signal. This goal can be realized by using a diffuser in front of the nano-focusing optics or by designing new MLL lens with additional zone displacement errors to produce the desired beam profiles.

The performance of a related technique, Bragg coherent diffraction imaging method, was systematically characterized through simulation. The optimum conditions for achieving the best reconstruction image quality were revealed, which will provide a general guideline for rational experimental design. This result is published in New Journal of Physics [2].

To increase the throughput of the ptychography method and make it applicable to more practical cases, we explored the integration of ptychography with a scanning probe microscope operated in on-the-fly scan mode. A strategy was proposed to suppress the periodic artifact without sacrificing achievable resolution, while enabling multi-modality measurements. This result is published in Applied Physics Letters [3].

References:
[1] H. Ozturk, et al, "Multi-slice ptychography with large numerical aperture multilayer Laue lenss", submitted (2017).
[2] H. Ozturk, et al, "Performance evaluation of Bragg coherent diffraction imaging", 19, 103001, New Journal of Physics, (2017).
[3] X. Huang, et al, "Artifact mitigation of ptychography integrated with on-the-fly scanning probe microscopy", 111, 023103, Applied Physics Letters, (2017).

Milestones:

- We've submitted a manuscript that reports achievement of 8 nm lateral resolution with an extended depth of field from a thick sample. (H. Ozturk, et. al., submitted (2018)).

Toward 100fs Single-shot Electron Imaging Using Electron Beam Slicing Technology

LDRD Project # 16-010

L. Yu, Y. Zhu, T. Shaftan, L. Doom

PURPOSE:

The original objective of the project is to realize the following:

- Gain control over the beam size and the divergence angle
- Focus the electron bunch to 30 μm size in the new diagnostics chamber
- Define the specifications and design of an Ultrafast Electron Microscope (UEM)
- Design the compressor for a high charge short pulse (although not currently funded).

The new capability afforded by the Ultrafast Electron Diffraction (UED) set-up will better position us to respond to DOE funding opportunities for ultrafast electron scattering experiments and provide technology breakthroughs in key development areas.

APPROACH:

The immediate objective of Phase I of this project was to design, build and experimentally confirm the focusing of electron bunches to 30 microns in size within the range of a few fC to 50 pC, with a divergence angle of 0.1 to a few mradians. Phase II will develop a design for the UEM system.

In addition to the principal investigators, our collaborators from the Condensed Matter and Materials Science Division, National Synchrotron Light Source II (NSLS-II), and the Accelerator Test Facility (ATF) include J. Tao, J. Li, W. Cheng, V. Smalyuk, X. Yang, A. He, M. Fedurin, M. Babzian, and others on the ATF team.

TECHNICAL PROGRESS AND RESULTS:

In FY 2016, we completed the physics design and development of the beamline lattice. Specifications and procurement documents for the quadrupole magnets and power supplies were finalized.

In FY 2017, we completed the system design, manufacture, installation and initial testing (Mechanical, Electrical, Vacuum, and Diagnostics).

Design and Manufacture: Beamline design and manufacture of the vacuum system was completed, including chamber gages and pumps, a corrector power supply, and a diagnostic system that included the beam position monitors and diagnostic chamber for the measurement of beam size, corrector magnets and a support system. Models, manufacturing, electrical and assembly drawings were developed for all components and systems.

Installation: Assembly of all magnets, diagnostics and vacuum components was completed. All components were surveyed and positioned. The girder assembly was installed, surveyed and grouted in the UED room of Building 912. The completed vacuum system was leak checked, pumped down and baked. New electronics racks were installed in the UED area. All systems were wired to the control system and associated components.

Testing and Commissioning: System testing was completed to prepare the beamline for current user experiments. The next step will include testing the focusing and divergence angle. All subsystem integration tests were completed.

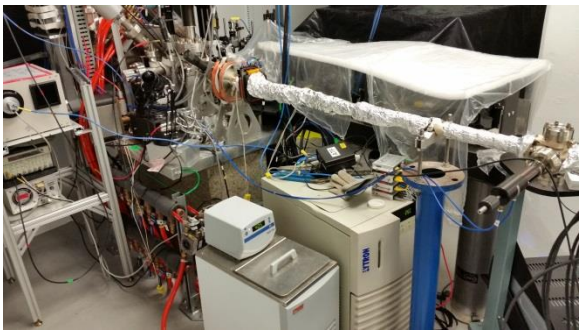


Figure 1: UED room before component installation.

Figure 2: UED room after component installation.

Figure 1 and Figure 2 compare the UED configuration before and after the components developed under this project were installed. The system installation was completed by the end of October. At the beginning of November, commissioning of magnet power supplies, pop-in monitors, and system controls, etc. started.

The NSLS-II team worked closely with the ATF staff and UED users to characterize this new mode of operation with the components from this project installed. The first priority was to commission the system so that the condensed matter physics experiments could continue with the existing set-up with the new components installed.

Initial commissioning identified the system's sensitivity to remnant magnet fields; multiple steps were completed and tested to remove the remnant fields. With these steps accomplished, the user experiments have begun taking data again. We are in the process of making the transition between user experiments and the focusing experiment that is part of this project simple and fast.

In FY 2017, our plan was to commission the beamline with focusing and its diagnostics with sufficient tuning range for the diffraction experiments. The beamline was made sufficiently flexible for us to vary, within several orders of magnitude, the focal point size, divergence angle and bunch charge, and to use the diagnostics system to verify the results.

Milestones:
As the first part of Phase II of our project, we plan to prepare a preliminary design for a UEM system.

In situ Synchrotron Studies of Subsurface Material Interfaces using X-ray Fluorescence Mapping and X-ray Tomography at National Synchrotron Light Source II

LDRD Project # 16-019
S. Gill, M. Elbakhshwan, L. Ecker

PURPOSE:
There is a lack of fundamental understanding of kinetics and reaction mechanisms that occur at solid-fluid and solid-solid interfaces under extreme conditions of temperature, pressure and corrosive environments present in various energy systems, such as subsurface and enhanced geothermal systems. It is critical to study interfacial interactions to understand the degradation mechanisms involved in subsurface materials. Such knowledge will not only help predict economic viability of sequestration and well integrity, but will also help in designing next generation materials with enhanced performance under extreme geothermal conditions. The world-leading intensity and brightness of National Synchrotron Light Source II (NSLS-II, coupled with advanced sample environments, presents an exceptional opportunity to understand the fundamental interfacial processes at solid-fluid (rock/mineral-fluid) and solid-solid (cement-rock/clay and cement-casing) interfaces involved in the degradation of subsurface materials under extreme environments.

This project is aimed at utilizing the high brightness of NSLS-II and high sensitivity of micro-and nanobeams to elucidate the interfacial reaction processes and structure of the reaction products in subsurface materials using *in situ* and *ex situ* synchrotron (X-ray fluorescence [XRF] and X-ray absorption near edge structure spectroscopy, X-Ray Diffraction [XRD] and computed tomography) studies. In addition, it will develop a capability to characterize subsurface materials under a supercritical carbon dioxide (sc-CO_2) environment. Such studies will elucidate the phase, chemical and microstructural changes associated with dissolution and precipitation reactions occurring at subsurface material interfaces.

The research will provide fundamental understanding of subsurface material degradation of advanced cements and cement-casing interfaces from the meso- to nanoscale using synchrotron methods, which will aid in design of next generation self-healing cements. It strategically leverages NSLS-II and positions BNL as a key player in characterizing subsurface materials using high resolution and ultra-fast X-ray methods. This will position BNL to be a strategic key partner in the Subsurface Technology and Engineering (SubTER) R&D team and compete in the Energy Frontier Research Center (EFRC) call on subsurface materials, and future DOE funding.

APPROACH:
Develop a capability to characterize and design subsurface materials under a sc-CO_2 environment: in order to qualify and deploy advanced cements for geothermal well applications, it is critical to evaluate their performance after exposure to extreme environments, such as CO_2 environments. Our goal is to develop a capability where structural and chemical changes in advanced cements can be monitored in real time using high resolution nano and micro-beams at NSLS-II beamlines.

Characterization of microstructure and chemical composition of foamed and polymer-based cements (with B. Kutchko at National Energy Technology Laboratory and C. Fernandez at Pacific Northwest National Laboratory [PNNL]): the storage of CO_2 in deep geologic formations

requires the assurance of wellbore integrity, including the evaluation of the stability of wellbore cements over time. Investigations into well bore cements should include studying the effects of geologically-relevant parameters on the geochemical and geomechanical stability of existing conventional and novel self-healing cements. Our goal is to study microstructure, chemical composition and structure of conventional and advanced, as-synthesized cements and after exposure to CO_2.

TECHNICAL PROGRESS AND RESULTS: In FY 2017, three milestones were achieved:

- Test and commission a sc-CO_2 loop to study subsurface materials under geothermal environments: a test loop for studying interfacial interactions of subsurface materials after exposure to a sc-CO_2 environment was designed and built. The sample environment design includes a capillary sample holder (Figure 1a) to hold powdered samples, a controller to monitor the CO_2 flow and a pump for delivering CO_2 flow (Figure 1b). The test loop was tested prior to testing and commissioning at the X-Ray Powder Diffraction (XPD) beamline at NSLS-II.

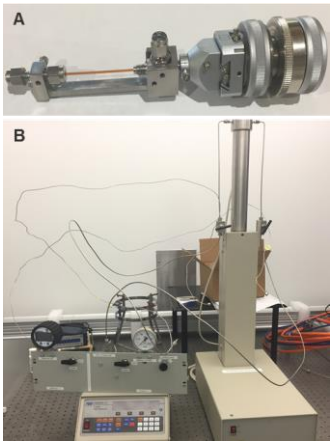


Figure 1: (a) Sample holder for CO_2 exposure (b) Pump and controller for sc-CO_2 flow.

- Synchrotron based *in situ* XRD studies in a sc-CO_2 environment: carbonation of brucite was studied *in situ* at the XPD beamline using the sc-CO_2 test loop. The kapton capillary was filled with the Brucite powder and then CO_2 was introduced to the system. The pressure was increased slowly up to 1100 psi (75.8 bar), then the capillary temperature was increased to 50ºC using N_2 gas from a cryostream. The total exposure time to sc-CO_2 was 6 hours at 1100 psi and 50°C. XRD measurements to monitor structural changes were done with a beam size of 500x500 µm and wavelength of 0.2366Å (52.4025 keV). A clear conversion of brucite ($Mg(OH)_2$) to Nesquehonite [$MgCO_3.3H_2O$] was observed immediately after sc-CO_2 exposure (Figure 2).

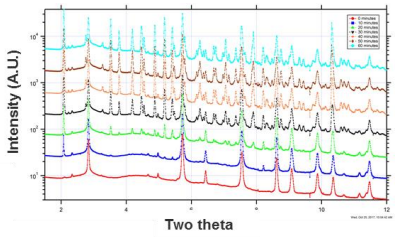


Figure 2: *In situ* XRD patterns showing carbonation of Brucite (Red) to Nesquehonite (Green onwards)

- Synchrotron studies of advanced foamed and self-healing cements after exposure to extreme environments: X-ray Ray Fluourescence (XRF) studies of self-healing cements and foamed cements exposed to CO_2 and CO_2-H_2S environments were performed at the Submicron Resolution X-ray Spectroscopy (SRX) and Tender Energy X-ray Absorption Spectroscopy (TES) beamlines and clear carbonation fronts were observed at the interface with calcium and sulfur rich regions.

Knowledge of chemical and structural changes is crucial for understanding dissolution and precipitation reactions in subsurface materials and to design next generation well cements with superior strength and bonding properties. A manuscript on self-healing cements was submitted with PNNL to the journal of Environmental Science and Technology. Results were leveraged to participate in an EFRC led by Washington University focused on subsurface materials.

Milestones:
Perform synchrotron studies of subsurface materials at NSLS-II and publish results presented here.

Characterization of Gold Photocathode and Photoelectrons in Liquefied Noble Gases

LDRD # 16-021
T. Rao, T. Tsang

PURPOSE:

The purpose of this project is to investigate the possibility of integrating a photocathode as a part of the Time Projection Chambers (TPCs) for *in situ* charge calibration. Many large-scale liquefied noble gas TPCs are being designed and built for neutrino and dark matter experiments such as the Long Baseline Neutrino Facility (LBNF), the Deep Underground Neutrino Experiment (DUNE), and the next Enriched Xenon Observatory (nEXO). A robust and reliable electron charge calibration is crucial for these high purity liquefied noble gas detectors.

APPROACH:

In collaboration with Stony Brook University, a high purity gas xenon (Xe) purification and recirculation system is constructed to recapture the expensive Xe. A small TPC drift stack made of macor and sapphire is used to study the electron transport in noble liquids. Photoelectrons are generated by back illuminating a 22 nm thick semi-transparent gold photocathode on sapphire with time synchronized ultraviolet (UV) laser pulses. Photoelectrons drifted from the photocathode towards the anode is collected and measured. Field shaping rings and a wire grid positioned between the cathode and anode help shape the field lines and improve the charge collection efficiency. We examine the quantum efficiency (QE) of the photocathode and the photoelectron transport properties in liquefied xenon (LXe) as a function of drift field and impurity. The extracted electron lifetime depends strongly on impurity that can be used as a technique for charge calibration in large TPCs.

TECHNICAL PROGRESS AND RESULTS:

We successfully built and operated a small drift cell for the study of electron transport in LXe. To date, we have completed well over 100 cycles of liquefaction. The great repeatability of the system allows us to study in detail the drift electron properties in LXe. We measured the drift velocity and the longitudinal diffusion coefficient D_L in LXe as a function of drift field and temperature, see Figs. 1(a), 1(b), and 2(a). The increase in D_L with decreasing drift fields is in qualitative agreement with theoretical predictions. At the drift field of 0.5 kV/cm, the measured drift velocities and corresponding temperature coefficients are 1.96 mm/μs and -0.4%/K for LXe, and 1.42 mm/μs and +0.11%/K for gaseous xenon (GXe). In addition, we measured longitudinal diffusion coefficients of 17.7 cm^2/s and 131 cm^2/s, for LXe and GXe, respectively. The field dependence of the electron drift velocity in LXe agrees qualitatively with other investigators, while the transport behavior in GXe agrees well with literature values.

The quantum efficiency of the gold photocathode is measured to be $5x10^{-7}$ in LXe, and $4x10^{-7}$ in GXe, at the photon energy of 4.7 eV, see Fig. 2(b). It is important to note that unlike gaseous argon (GAr) and liquid argon (LAr), where the QEs differ by nearly an order of magnitude (QE of GAr greater than LAr), here the QEs of the gold photocathode in GXe and LXe are nearly the same. This finding might hint to the importance of electro-negative affinity of photocathodes in various noble liquid environments. The multiphoton ionization in noble liquids is of interest in LAr and LXe detectors, such as the LBNF and nEXO experiments. Although the ionization

energy of LXe is comparable to LAr, the 2-photon ionization cross section in LXe is expected to be ~4 orders of magnitude greater than in LAr. Here we employ picosecond and nanosecond UV lasers of different peak powers, but otherwise the same energy densities to explore the role of multiphoton ionization in LXe, see Fig. 3. We demonstrated that multiphoton ionization in LXe becomes significant when the laser peak power density increases from kW/cm^2 to MW/cm^2. This finding suggests that a careful choice of laser will be needed for an *in situ* internal charge calibration scheme. The results of these charge transport properties are important in designing future large scale noble liquid detectors.

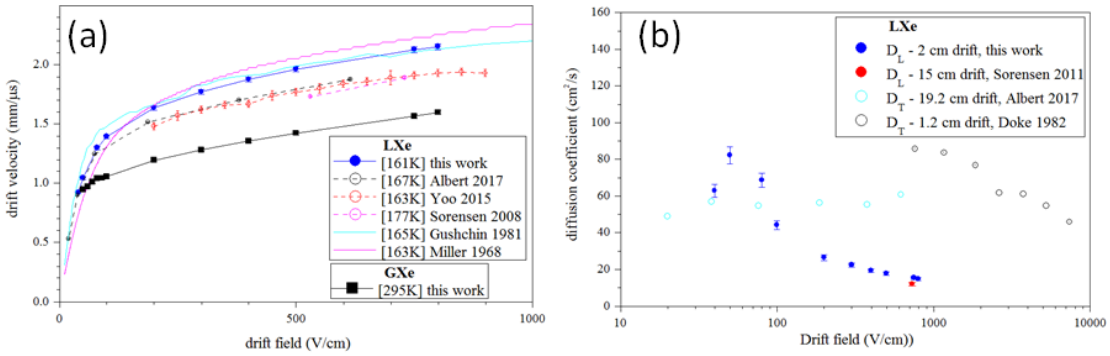


Figure 1. (a) field dependence of electron drift velocity in LXe, for comparison, a set of GXe data is overlayed. (b) field dependence of electron longitudinal diffusion coefficient in LXe, for comparison, a set of transverse diffusion coefficient is also displayed.

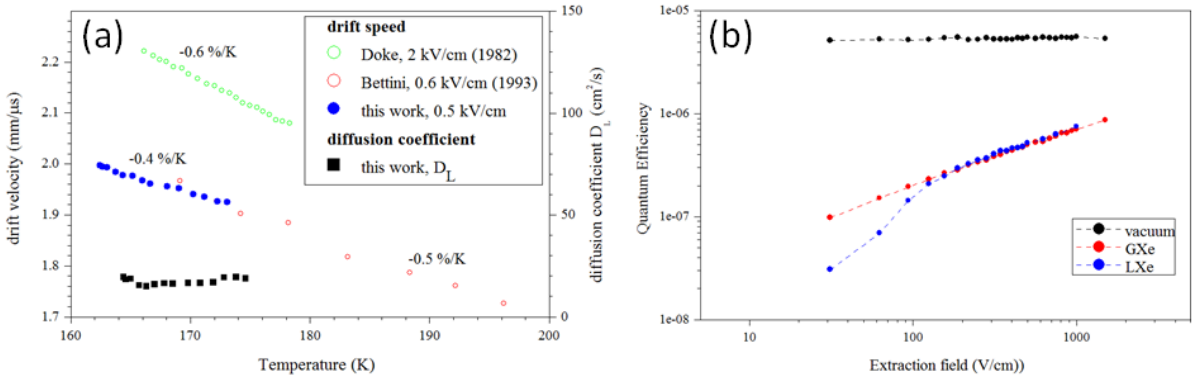


Figure 2. (a) temperature dependence of the electron drift velocity and diffusion coefficient in LXe, (b) field dependence of the quantum efficiency of the gold photocathode in vacuum, gas, and liquid xenon.

Furthermore, we evaluated the electron lifetime by comparing the ratio of electrons arriving at the anode to that of leaving the photocathode at a fixed drift field. A consistent ratio of 0.6 was obtained at 0.5 kV.cm field gradient. All these results highlight the importance of minimizing impurity to extend the electron lifetime in LXe and to establish the reliability of the electron charge calibration scheme based on photocathode technology.

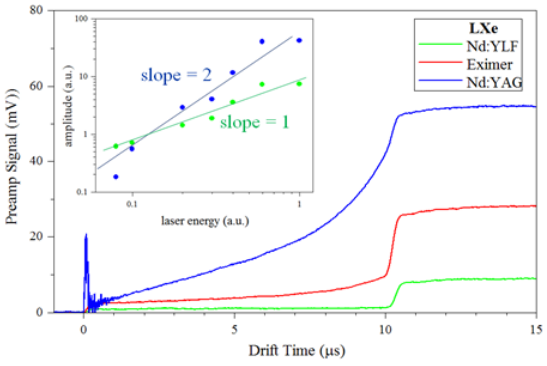


Figure 3. Electron charge signal pulse shape when lasers of different peak powers but otherwise same energy densities are employed. The log-log plot of their signal amplitude highlighting a slope of 2 for multiphoton ionization in LXe and a slope of 1 for linear photoemission from the gold photocathode.

Investigation of the Performance Characteristics of Silicon Photomultiplier (SiPM) Photosensors under Extreme Conditions for Use in Nuclear and Particle Physics Detectors

LDRD Project # 16-022
T. Tsang, C. Woody

PURPOSE:
The purpose of this project is to study the properties of silicon photomultipliers (SiPMs) under extreme conditions (cryogenic temperatures and/or high radiation environments) for use with other detectors in nuclear and particle physics experiments to study the fundamental properties of energy and matter. BNL plays a significant role in many of these experiments, such as PHENIX and STAR at the Relativistic Heavy Ion Collider (RHIC), ATLAS at the Large Hadron Collider, the Deep Underground Neutrino Experiment, and the next Enriched Xenon Observatory (nEXO). Improving their performance would benefit new programs at BNL, such as eRHIC and many others.

APPROACH:
A systematic study on the operation of SiPMs before, during and after irradiation was carried out at BNL. Devices of different pixel sizes are exposed to various doses of neutrons and gammas. Their operating parameters, dark current and single photoelectron spectra were characterized both at room temperature and cryogenic temperature. Recovery from radiation damage is observed after thermal annealing. Cryogenic operation and performance of Vacuum Ultraviolet (VUV) sensitive SiPMs are evaluated.

TECHNICAL PROGRESS AND RESULTS:
Comparison of radiation damage from neutrons and gammas: We observe different effects when SiPMs are exposed to ~1 MeV gammas up to 1 Mrad and 14 MeV neutrons up to 10^{10} n/cm^2 (Figure 1). This difference is indicative of bulk displacement damage in the silicon caused by defects produced by irradiation.

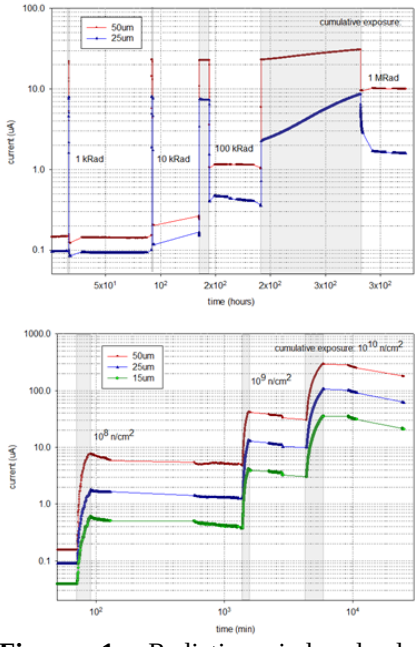


Figure 1: Radiation induced dark current in SiPMs with different pixel sizes for increasing doses of gammas (top) and neutrons (bottom).

Studies of neutron radiation damage and recovery: SiPM generally lost its performance characteristics with elevated dark current when exposed to high dose of neutrons. However, the distinct hallmark of SiPMs' photon-number-resolving capability (PNR) can be recovered by operating them at cryogenic temperature or by high temperature thermal annealing. Figure 2 shows representative I-V plots and their corresponding photoelectron spectrum during all stages of the experiment. It is evident that after thermal annealing, the dark current is lowered and photoelectron resolving capability is restored. The annealing is permanent and the level of recovery scales with radiation dosage and the annealing temperature. However, we found no significant difference if SiPMs are irradiated at room or liquid nitrogen temperatures (Figure 3). But this annealing process is effective on SiPMs

from difference vendors (HPK and FBK), laying the foundation on extending the useful lifetime of large scale SiPM detectors deployed in an ionization radiation environment.

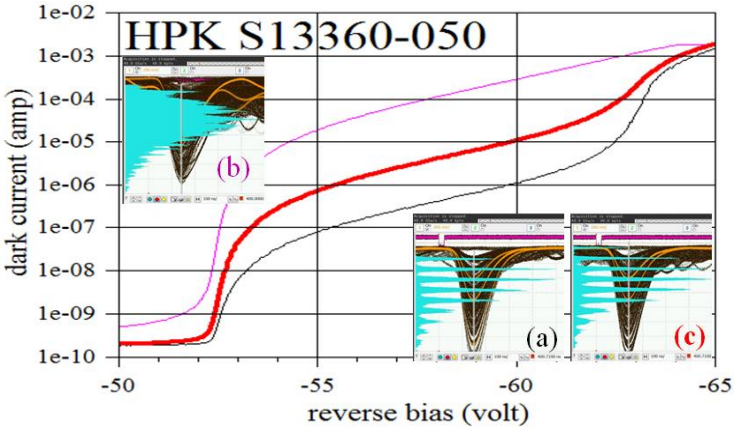


Figure 3: Representative room temperature I-V plots of (a) before 10^9 n/cm^2 irradiation; (b) after irradiation; and (c) after annealing, and their corresponding single photoelectron spectrum.

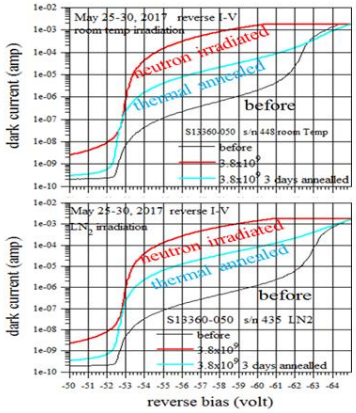


Figure 2: I-V plots when SiPMs are irradiated at room (top) or liquid nitrogen (bottom) temperatures followed by thermal annealing.

Studies at cryogenic temperatures: Scintillating photons of liquid xenon in the nEXO experiment are at ~178 nm VUV wavelength. It is important to identify SiPMs that can meet the specification of the nEXO detector. We generated spectrally pure and precisely triggered 177.3 nm VUV photons from the 3rd harmonic of a frequency doubled Nd:YAG laser using a nonlinear odd-harmonic generation scheme on a piece of ordinary glass slice. We use the VUV photons to perform time-correlated photon counting and to characterize the single-photoelectron spectrum of SiPMs at 177 nm (Figure 4(a)). At 2-volt Over Voltage (OV), the charge gain (Figure 4 (b)), dark-count-rate, crosstalk, and the 1-photoelectron resolution (Figure 4(c)) met the nEXO specifications, While the photo-detection-efficiency (PDE) at 178 nm is ~20%, the higher fill factor of ~0.6 improved the PDE. However, we observed a spectral and temperature dependence of the PDE on the avalanche probability. The overall performance of this SiPM still meets the VUV wavelength requirements for nEXO.

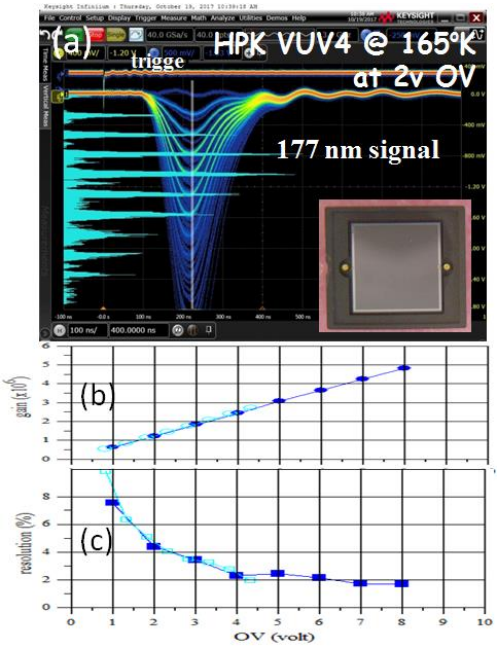


Figure 4: (a) Time-correlated PNR of HPK VUV4 SiPM at 166K, using 177 nm VUV photons (b) charge gain and (c) resolution.

Overall, in FY 2016, we demonstrated the SiPM works well in cold and identified a technique to restore radiation damaged devices. In FY 2017, we: identified that gammas and neutrons produce different radiation effects; established thermal annealing is effective on SiPMs from different vendors; found no significant difference if SiPMs are irradiated at room or liquid nitrogen temperatures; and identified that the HPK VUV4 SiPM meets the nEXO specifications.

Analog-to-Digital Converter and Gbit/s Serializer/Driver in Complementary Metal–oxide–Semiconductor for Large Data Generation and in Operando Analysis

LDRD Project # 16-023
S. Li, E. Vernon

PURPOSE:
Modern radiation detectors are required to be compact with increasing demand for a larger number of channels, better resolution, and faster data throughput. Low noise, low power, and high functionality front-end Application Specific Integrated Circuits (ASICs) are the enabling technology behind advances in modern radiation detector design. The purpose of this work is to develop an analog-to-digital converter (ADC) for signal processing, and an encoder-serializer/driver (ESERD) to transmit the digitized data off chip at Giga-bits/second (Gb/s) speeds. If resources allow, these circuits will be developed to operate at both room and cryogenic temperatures. Compared to conventional systems built from discrete components, if successful, two critical enabling circuits (ADC and ESERD) will be integrated into next generation front-end ASICs as systems on chip (SoC), thus directly impacting on the future of micro-electronics and state-of-the-art detector developments at BNL. The applications will cover various R&D programs in Nuclear and Particle Physics, Synchrotron Light Sources (such as National Synchrotron Light Source II [NSLS-II]), and National Security. There will be an immediate impact on large scale Liquid Argon Time Projection Chambers for the Long Baseline Neutrino Facility, large scale neutrino-less 2β-decay detectors for the next Enriched Xenon Observatory, dark matter detectors, and X- and Gamma-ray imagers needed at NSLS-II.

APPROACH:
The micro-electronics group in the Instrumentation Division at BNL has been developing ASICs to readout gas, liquid, and solid state radiation detectors. Conventional designs incorporate ASICs, which measure the amplitude and timing of an event and multiplex the analog data off-chip for digitization outside the detector with discrete ADCs. As the need for more channels increases, conventional designs are approaching a bottleneck in data rates and the number of feed-throughs required, which also increase the noise from cross-talk.

Alternatively, advances in low noise ASICs and direct interface with the sensor electrodes inside the detector volume have improved the noise performance and reduced the complexity of detector design by requiring fewer feed-throughs. In addition, by implementing ADCs and ESERDs on the ASIC fabric to form SoCs, the data bandwidth will be significantly improved for next generation detectors.

In phase I of the design process, the successive approximation register analog-to-digital converter (SARADC) design is under study. The approach involves: the exploration of various topologies; developing functional models for the signal chain; systematically converting functional blocks to transistor level designs; implementing the physical layout; verifying performance with post layout simulations; fabrication of the first prototype on a multi-project wafer; and bench top testing, with the possibility for a design revision. In phase II, the ESERD will follow the same design trajectory to achieve parallel testing of the two circuits. This work is

being realized through the collaborative efforts of Yuan Mei (a new hire), Krithika Yethiraj (Ph.D student), and Wenbin Hou (Ph.D student).

TECHNICAL PROGRESS AND RESULTS:

During FY 2017, the SARADC topology was finalized, the schematics were developed, and the physical layout was initiated. The ADC was designed for 14 conversion cycles to obtain 12 bits. To compensate for inherent device mismatches and process variations, the two redundant cycles were used to implement a digital foreground calibration scheme. The layout of one of the ADC channels is shown in Figure 1.

Figure 1: Layout of an ADC channel using sub-radix-2 digital-to-analog converter.

A data pipeline and readout logic were also designed and realized in layout. In addition, the registers used to configure the ADC were implemented, with a pseudo serial peripheral interface. Figure 2 shows the preliminary results for the transistor level schematic simulation of one ADC channel. The ADC achieved 12.1 effective number of bits with no missing codes after calibrations at the typical process corner, with 1.2 V supply and a sampling frequency of 5 MS/s.

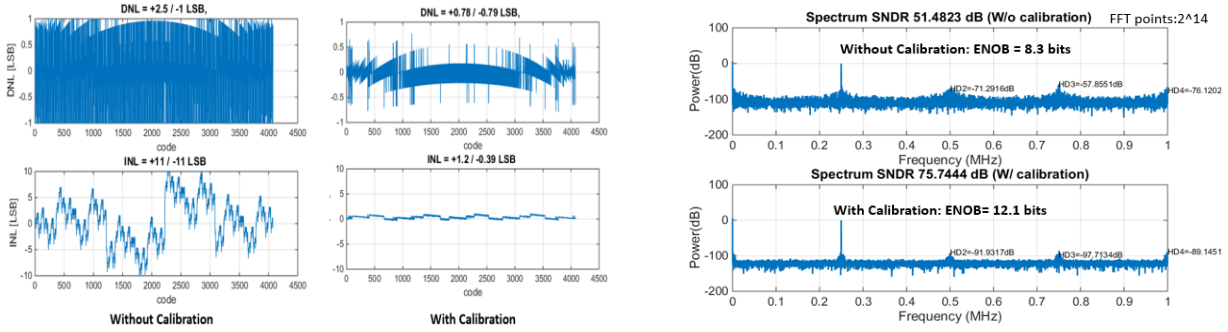


Figure 2: (a) Simulated Integral Non-Linearity and Differential Non-Linearity. (b) Simulated effective number of bits.

Milestones:

- Complete the layout of the functional blocks
- Chip level integration
- Post layout simulations
- Fabrication of the first ASIC prototype (ADC)
- Characterization of the first prototype
- Design revisions and fabrication of the second ASIC prototype
- Characterization of the second ASIC prototype.

Improved X-ray Spectroscopy Detectors

LDRD Project # 16-024
G. Giacomini, D. Elliott

PURPOSE:

High-rate X-ray spectroscopy allows the determination of the elemental composition of a sample by detecting the element's characteristic fluorescence X-rays that are emitted in response to an excitation produced by the synchrotron X-rays. Nowadays, this is done by an array of p-in-n diodes, *e.g.* the BNL-developed detector called Maia, whose energy resolution is essentially limited by the noise coming from the capacitance of each diode (and, in the second order, by the leakage current of each cell). Such resolution allows only the detection of X-rays emitted by elements heavier than aluminum, while lighter elements are beyond detection. To extend the reach of the elemental tagging of the first elements of the periodic table, the detector must feature a better energy resolution, such as the one achieved by Silicon Drift Detectors (SDDs), which, due to the complexity of the device, exist commercially only as single channel devices. To retain the high throughput typical of the original Maia array (tens of megahertz), each diode has to be replaced by a single drift sensor. This translates to an improved energy resolution at shorter shaping times, which in turn, further increases the acquisition rate (or decreases the measurement time).

Improved spectroscopic performance will allow exploring new kinds of samples; a number of beamlines at National Synchrotron Light Source II will benefit from this new development, while generating interest from other beamlines at other synchrotrons world-wide, some of them already equipped with versions of the diode-based Maia.

APPROACH:

Maia is a high-rate X-ray spectroscopic detector, which consists of an array of 384 1x1 mm2 p-in-n diodes. The sensor itself is fabricated in the Silicon Processing Facility of the Instrumentation Division at BNL; the read-out electronics and the boards have been designed at BNL and the mounting is performed here, too. The energy resolution – and thus the capability of detecting X-rays emitted by light elements - is intrinsically limited by the capacitance of the p-in-n diodes, which negatively also affects the measurement times.

As a first step towards the achievement of an enhanced energy resolution and shorter acquisition times, it is fundamental to replace the sensor itself. Each 1x1 mm2 diode must be replaced by an SDD, whose small anode capacitance allows the achievement of both goals.

To carry out the development, the following actions must be undertaken and completed: design and fabrication of SDDs with a low-leakage process flow in the class-100 Silicon Processing Clean Room at BNL (Wei Chen); at the probe station, electrical characterization of fabricated devices to select good devices; laser scribing of wafers to identify the good devices; mounting on printed-circuit boards and connection through wire bonding to the read-out electronics (Donald Pinelli); and in-lab X-ray spectroscopic measurements (Peter Siddons). The boards and electronics are the same as for the original Maia. From the results of the first characterization, usually a few problems arise, which must be tackled and solved with proper modifications. Thus, a second iteration follows (and possibly more in the future), in which all the problems encountered in the first batch are fixed.

TECHNICAL PROGRESS AND RESULTS:

In 2016, the first design was completed, the related photolithographic masks were produced, and the first batch of silicon wafers was processed. The electrical characterization of devices at the probe station allowed the selection of a few suitable sensors to be mounted on boards and connected by wire bonding to the read-out electronics. From these results, it was apparent that a modification of both the design and the process flow was necessary. Thus, in 2017, a new batch consisting only of test structures was designed and several process splittings were adopted during the fabrication. This effort was successful, because it clearly pointed out the limitations inherent in the old process, while demonstrating the advantages of the new process. This new process is now adopted in all the batches fabricated in Instrumentation. A new set of photolithographic masks was designed and produced.

In particular, three types of arrays of MAIA SDDs were considered (see Figure 1). Clearly, each version has pros and cons, which must be individually tested. Scheme a) is particularly difficult, since it requires two metals. A lot of effort was dedicated to this topic, which can be useful in future batches. A new batch of SDDs exploiting the new process and the new mask set was fabricated. At the probe station, the electrical characterization showed very low leakage currents and essentially functional devices. However, due to the complexity of the device, the yield is an issue and must be tested while connected to the read-out electronics. The functional characterization with radioactive sources was done with the type c) array only.

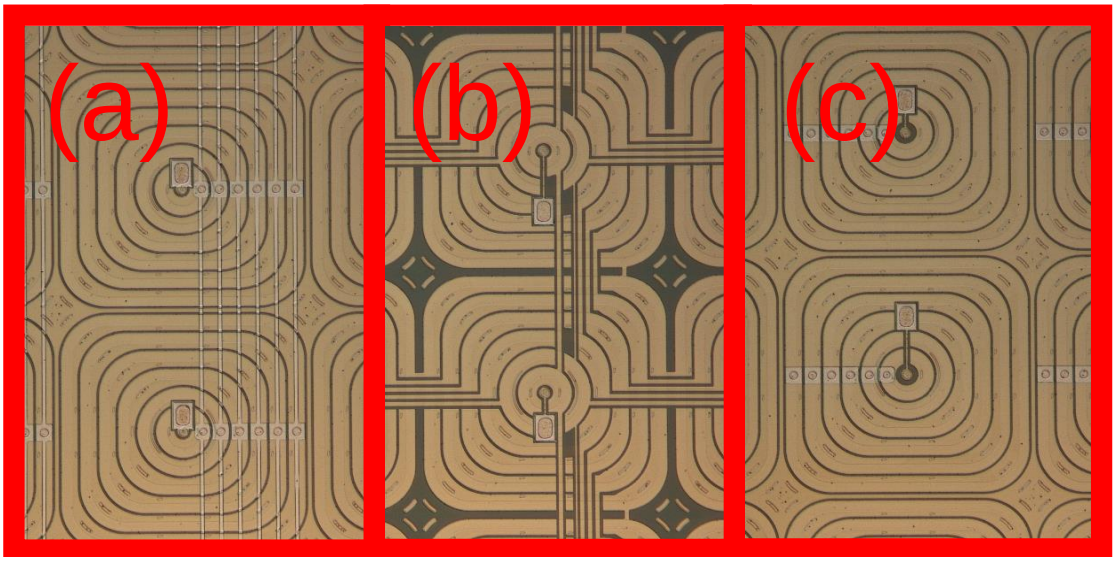


Figure 1: Three types of arrays of Maia SDDs: a) with a voltage distribution scheme through a second metal, running over an insulator; b) with a proper routing (at the first metal level) of bias lines that bias each drift ring of each cell; and c) with floating, self-biased drift rings surrounding every anode.

Microwave Kinetic Inductance Detectors: from Cosmology to National Synchrotron Light Source II

LDRD Project # 16-026

P. O'Connor, A. Nomerotski, A. Slosar

PURPOSE:

This project will develop feasibility demonstrations of next-generation cosmic frontier survey instrumentation, particularly in the radiofrequency domain.

APPROACH:

The DOE Office of High Energy Physics Cosmic Frontier research area has funded Stage 3 and Stage 4 Dark Energy experiments, all of which involve massive surveys in the optical wavelength range. The flagship Stage 4 experiments are the Large Synoptic Survey Telescpe (LSST) (optical imaging) and the Dark Energy Spectroscopic Instrument (DESI) (optical spectroscopy), both using silicon charge-coupled device sensors and each nearing completion of construction. Together with BNL collaborators on related LDRD 16-038, we are working to leverage our collective expertise in detectors and instrumentation, cosmological large-scale structure studies, and data science to study and prototype design ideas for a possible post-LSST/post-DESI Dark Energy experiment.

A promising approach is to map the large-scale distribution of hydrogen, the most abundant element in the Universe by observing its redshifted 21-cm emission. This can now be done with cost-effective technology, thanks to advances in digital radiofrequency signal processing driven by the needs of the wireless telecommunication industry. In addition, for radiotelescopes the required large collecting area can be realized for a fraction of the cost of similar-size optical mirrors. The goal of an eventual large program is to expand the volume of the Universe surveyed by a factor of 3 – 10 over present surveys, using aggregate emission of hundreds-to-thousands of distant galaxies in voxels matched to the cosmological scales of interest, on the order of 10 megaparsecs on a side. The sharp, isolated 21-cm spectral feature allows precise redshifts to be determined far more rapidly than by conventional optical spectroscopy of individual galaxies.

In this project, we will demonstrate key elements of a 21-cm intensity mapping experiment by making laboratory and on-sky measurements using novel reflector and electronics design techniques.

In parallel with our work on 21-cm intensity mapping, we have carried out several investigations of Microwave Kinetic Inductance Detectors (MKIDS), a superconducting detector technology once thought promising for large-scale optical surveys. As a result of modeling and fabrication studies, we now better appreciate the limitations of this detector and do not plan to follow through with additional prototyping.

TECHNICAL PROGRESS AND RESULTS:

The prior year's main accomplishments were:

- Formed loose collaboration ("Baryon Mapping eXperiment", BMX) with individual Oak Ridge National Laboratory, Stony Brook University, Columbia, U. Michigan, and U. Arizona scientists
- Designed front-end electronics with novel gain calibration

- Implemented a high-throughput digital back-end processor
- Prototyped low-cost dish antenna construction and characterization techniques
- Mapped the radiofrequency interference spectrum around the BNL site and obtained approval for construction of a demonstrator experiment in a well-shielded location in the East part of the campus.

During this year, we made exciting progress in the following areas:

- Completed construction of the first full 4 m dish and receiver tower
- Measured dish surface parabolicity to ~0.05%
- Characterized the S-parameter of the orthomode transducer for the first dish
- Designed and constructed the horn and transition elements of the feed
- Demonstrated broadband gain calibration using a pulsed noise diode
- Assembled and characterized the full analog amplifier/filter chain in the laboratory and installed it in on the receiver tower
- Characterized gain and noise performance of the entire signal chain *in situ*, using hot and cold loads
- Developed data acquisition and data reduction software, including automated 24/7 collection and transfer from the remote site to the astro cluster in the RHIC Computing Facility
- Achieved "first light" in July
- Demonstrated detection of the first astronomical targets, including 21-cm emission from the Milky Way and identified several extragalactic source transits
- Developed foreground simulation code, including galactic and extragalactic contaminants.

In the MKIDs area, we used the Instrumentation Division Silicon Detector facility to fabricate a low-frequency MKIDs array, which was delivered to collaborators at Columbia, where it is undergoing testing.

Milestones:

For the remaining year of this project, our goals are to:

- Continue observations and generate a deep co-added dataset of the stripe observed by BMX's single dish
- Develop foreground and radiofrequency interference removal code and use it to process the BMX coadded dataset, and attempt to detect the cosmological 21-cm signal in cross-correlation with Sloan Digital Sky Survey galaxies
- Construct at least one more (duplicate) dish and receiver and operate the pair of telescopes as an interferometer
- Using the results of the BMX measurements, develop a plan forward and proposal towards the next step of our proposed 21-cm roadmap.

Detector Calibration and Material Analysis – Expanding the Capabilities of National Synchroton Ligh Source II

LDRD Project # 16-027
J. Smedley

PURPOSE:
The central goal of this project is to make X-ray Beam Induced Current (XBIC) a technique available at National Synchrotron Light Source-II (NSLS-II). NSLS was a leader in this research area, with two beamlines dedicated to this work, and two more also accepting proposals in this area. The project will also enable X-ray topography, which has a strong synergy with XBIC. Taken together, these capabilities allow study of both the charge motion in semiconductors and the isolation of electrically active defects, which can affect device performance.

APPROACH:
We have identified a beamline to enable topography and spectroscopic X-ray beam induced current measurements at NSLS-II (the inner-shell spectroscopy beamline [ISS]). While this capability was being constructed, we used our initial beamline capabilities at the Cornell High Energy Synchrotron Source (CHESS) to calibrate 16 devices for NSLS-II beamlines, and have assisted four independent user groups with these techniques, who will continue their work when the NSLS-II capability is available. We have since used capabilities at the ISS and X-ray Footprinting (XFP) beamlines at NSLS-II to calibrate a further 12 devices for NSLS-II, including four ultra-thin devices for soft X-rays. We have secured funding for developing medical dosimetry, which is a spinoff of the detector effort and required the new capability at NSLS-II to be successful and used the capabilities at ISS and XFP to understand proton-induced radiation damage in diamond.

TECHNICAL PROGRESS AND RESULTS:
The hardware allowing XBIC mapping at ISS has been purchased and assembled in compliance with NSLS-II requirements, including use of delta-tau motion control systems. The software to allow these motion control systems to interface with the beamline has been written; we will also be able to use the data acquisition capability of the delta-tau to read the detector/sample current. This system can accommodate samples up to 30 cm x 30 cm active area, and up to 10 kg, enabling it to be useful for detectors with large sensitive areas, as well as typical photovoltaic cells. The system has been serving user groups for the last three cycles of NSLS-II operations (not only our work).

While this system was being designed and constructed, we utilized a smaller, stand-alone motor set at the G2 beamline at CHESS to characterize detectors for NSLS-II, including ultra-thin detectors for the Soft Matter Interfaces (SMI) beamline. We have also supported four external user groups – two from industry (one doing SiC detectors and the other diamond detectors), and two from Los Alamos National Laboratory (LANL) (one of which was our LANL collaborator James Distel).

We have obtained funding from the DOE Office of High Energy Physics (Accelerator Stewardship) to develop detectors for proton and carbon ion radiation therapy ($500k over two years, half of which comes to BNL and half to Stony Brook University). This proposal relies

heavily on the XBIC capability being developed, as this is the tool which allows us to understand radiation damage induced by the charged particle beams.

Below are measurements performed with the new XBIC tools on proton irradiated diamonds. Figure 1 shows an edge-on XBIC map of a proton damaged diamond. This method utilizes the bright, small X-ray beams available at NSLS-II and permits localization of the proton-induced defects (which are predominantly on the proton-exit side of the diamond). This also allows the impact of defects on electron and hole transport to be differentiated (by changing the applied bias).

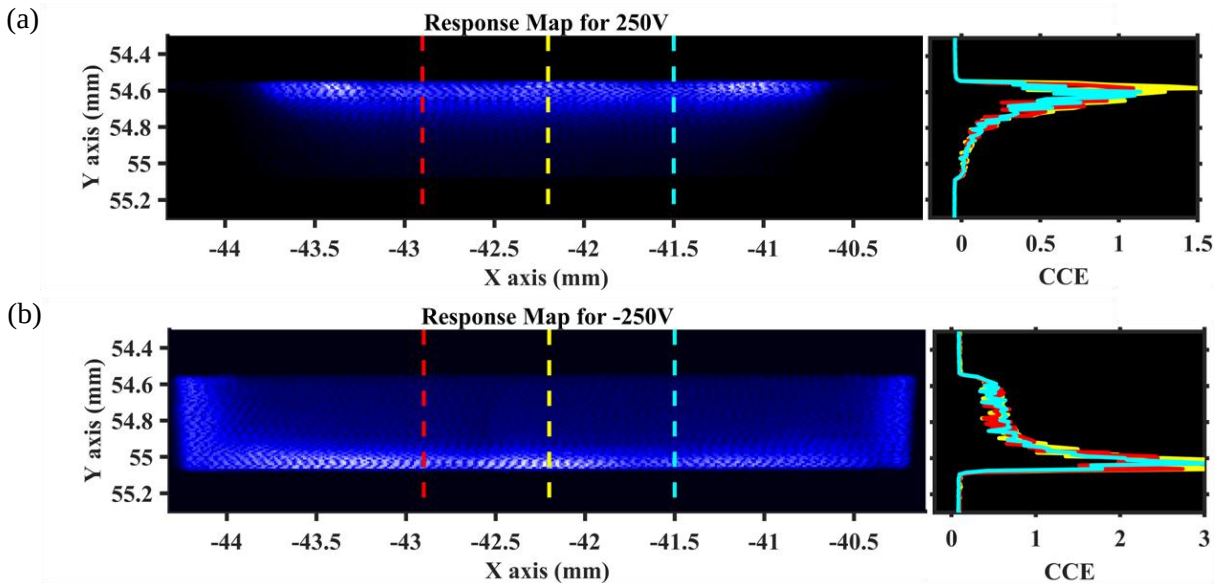


Figure 1: Response map from diamond cross section and corresponding line profiles from different positions under (a) positive (+0.5 V/µm) electric field and (b) negative (-0.5 V/µm) electric field.

Figure 2 shows the lateral XBIC map of the Charge Collection Efficiency (CCE) of the diamond before (2b) and after (2c) proton irradiation. Fig. 2A shows the field dependence of the CCE, for both cases.

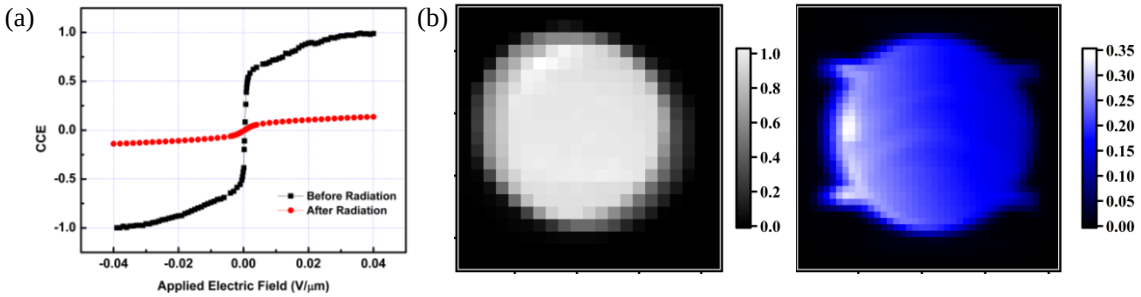


Figure 2: (a) CCE with applied electric field before and after proton radiation; (b) CCE map of the entire active area before (left) and after (right) proton radiation.

These measurements are part of a paper that is being prepared for submission in the coming weeks.

Higher-Order-Mode (HOM) Damping for Full Luminosity of eRHIC

LDRD Project # 16-029
W. Xu

PURPOSE:
The objective of this project is to increase the eRHIC luminosity limit, through demonstrating a high-power, full-spectrum Higher-Order-Mode (HOM) damping scheme for the eRHIC Energy Recovery Linac (ERL). The HOM damping requirement and scheme of the Superconducting Radio Frequency (SRF) linacs (electron and proton) and storage cavity for the Ring-Ring eRHIC design will be studied and verified as well.

APPROACH:
The proposed HOM damping scheme for a high current SRF cavity comprises a combination of waveguide HOM dampers (for low frequency HOMs) and room temperature beam pipe absorber (for high frequency HOMs). The expected results are:

- HOM spectrum study on a detachable copper (Cu) cavity.
- RF design and simulation, which is the starting point, includes design of the ridge waveguide, waveguide absorber, and beampipe absorber
- Engineering design that includes thermal analysis and mechanical design
- A test of the waveguide HOM damper and beampipe absorber prototypes on a Cu cavity to verify the HOM damping results
- Modification of the Niobium SRF cavity to attach the waveguide damper and carry out vertical cold testing.

TECHNICAL PROGRESS AND RESULTS:

- Version 1 design of the HOM damping with six ridge waveguides and beampipe absorbers was completed, including mulitpacting simulation, thermal analysis, HOM load design, HOM power scan, and preliminary mechanical design. Two 3D-printed prototypes (one 3D-printed stereolithography with Cu plating and one 3D printed-aluminum) of the cavity endgroup HOM couplers were fabricated and tested.
- The Cu cavity prototype for the HOM spectrum study was fabricated by *RI Research Instruments GmbH* in Germany and delivered to BNL for HOM studies. Figure 1 shows the 5-cell Cu cavity HOM study setup and measurement results. The measurement results on the HOMs of interest agreed very well with simulation results.

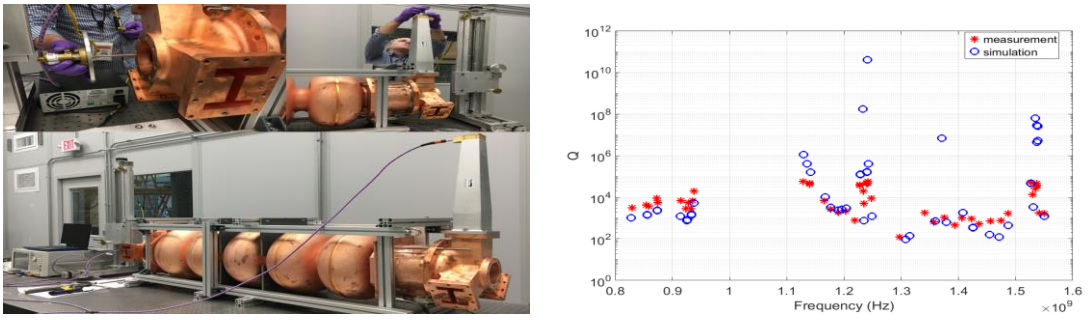


Figure 1: HOM study on 5-cell 650 MHz Cu cavity with Version 1 HOM damper.

- Completed study of different configuration of HOM damper (Version 2). Comparison results are shown in Table 1. The conclusion is that the V2 configuration with 4 B-shape waveguides (Figure 2) will fit our requirement, with reasonable cost/complexity. The RF design and preliminary engineering design were completed.

Table 1. Comparison of the Different Waveguide Configurations

Waveguide Configurations	**Min R_{th}/R_{cav}**	**F (GHz)**	**Normalized R_{th}/R_{cav}**
6 H-shape (Version #1)	6.2	0.856	5.3
6 Eight-shape	6.0	1.6	9.6
4 B-shape (30 ^{o}) (Version #2)	5.9	1.53	9.0
3 B-shape (60 ^{o})	3.6	1.25	4.5
2 B-shape (120 ^{o})	3.5	1.25	4.3

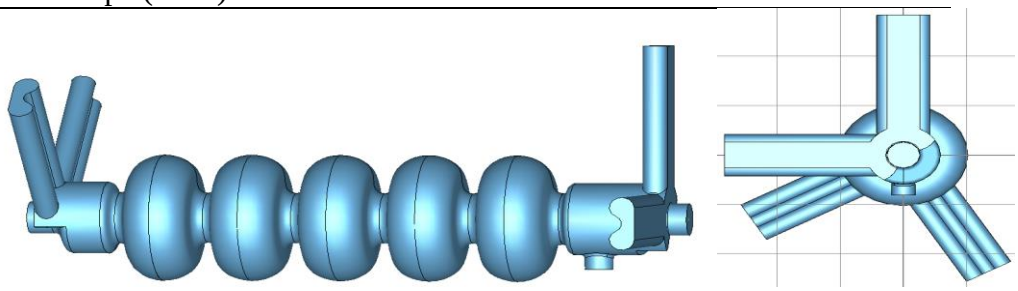


Figure 2: B-Shape HOM damping scheme.

- Completed RF design of the beampipe absorber (Figure 3). This beampipe absorber will damp the high frequency HOMs excited in the cavity. The RF design and engineering design were completed.

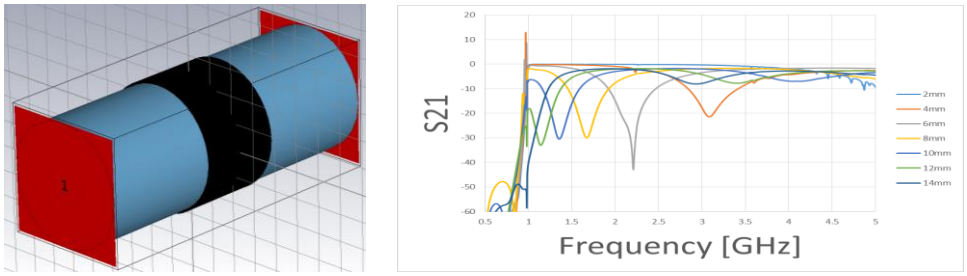


Figure 3: Beampipe absorber design.

Final Project Status:

A tremendous of work was accomplished in this project, benefiting the Ring-Ring eRHIC design in the area of the HOM dampers for storage RF cavities and the ERL for hadron cooling. For example, the work specific to the development of warm SiC beamline HOM absorbers is being leveraged to great advantage for several eRHIC Ring-Ring RF system pre-conceptual designs, including the electron storage ring, hadron storage ring and potentially for an e-cooling ERL. This project is complete.

Advanced Silicon Detectors R&D

LDRD Project # 16-034
F. Lanni, H. Chen, G. Giacomini, H. Takai, A. Tricoli

PURPOSE:
The focus of this project is on the development and characterization of three innovative Silicon-(Si)-based technologies for their potential application in tracking and calorimeter system upgrades of current experiments (*e.g.* ATLAS and CMS at the high luminosity Large Hadron Collider), and of future experiments at the next generation of colliders in nuclear and particle physics (*e.g.* eRHIC and a Future Circular Collider). These are high voltage/high resistivity complementary metal–oxide–semiconductor (CMOS) hybrid sensors (HV/HR-CMOS), ultra-fast Si detectors through Low Gain Amplification Devices (LGAD), and Monolithic Active Pixel sensors based on HV-CMOS (HV-MAPS). These technologies are being evaluated in terms of performance, reliability and radiation tolerance. In the process, we will take advantage of the existing facilities at BNL for understanding properties in detectors and electronics, specifically in the Instrumentation Division. The ultimate objective of the project is the exploration of fully monolithic sensors, which integrate both sensor technologies and readout devices. Advances in these technologies may also impact other fields, such as high precision/fast read-out sensors for dynamic structural analysis for time-resolved crystallography and beam monitoring at photon science facilities, and medical applications, such as precision timing-based Positron Emission Tomography. The project, if successful, will heighten BNL's reputation as a leading organization in Si sensor R&D cutting-edge technology.

APPROACH:
Precision measurements of multi-hadronic final states at high energy colliders will require larger and larger areas of Si detector coverage and readout channels and miniaturization of feature-size. Today's available technology is limiting this capability because of cost implications and the amount of service that these detectors require, which ultimately will compromise the performance in reconstructing charged particles and the underlying events to be measured.

The motivation of this project and the strategy that we have pursued is to advance technology in the following directions:

- Research the development of sensors through industrial fabrication
- Maximize integration of front-end electronics on sensors in small feature size processes
- Enable the discovery of rare events at future very high luminosity colliders, which means that experimental measurements are complicated by the number of primary vertices per bunch collision. Silicon detectors with high precision timing reconstruction of a track, together with the vertex information, enable identification and discrimination of particles emerging from the hard scatter vertex.

TECHNICAL PROGRESS AND RESULTS:
The following are the main achievements for FY 2017. FY 2017 was the second year of this project.

- Manufactured additional LGAD structures, designed by G. Giacomini, using processing at the Si fabrication facility in the Instrumentation Division. Preliminary characterization of those structures is on-going (see Figure 1)

- Continuing our international collaboration for HV-CMOS (U. of Geneva, U. Bern, U. Liverpool, Karlsruhe Institute of Technology) and for LGAD (U. California Santa Cruz, U. Paris VI, Laboratoire de l'Accélérateur Linéaire Orsay, Centro Nacional De Microelectronica (CNM) Barcelona, Jožef Stefan Institute Ljubliana, CERN, Istituto Nazionale di Fisica Nucleare Turin)
- Participated in testbeam campaigns at CERN, as part of the High Timing Detector Proposal in the ATLAS experiment, of which the first author was the proponent and coordinator until December 2016
- Characterized HV-CMOS testbeam for non-irradiated and irradiated large area sensors, achieving excellent minimum ionizing particle efficiency above 99.7%.

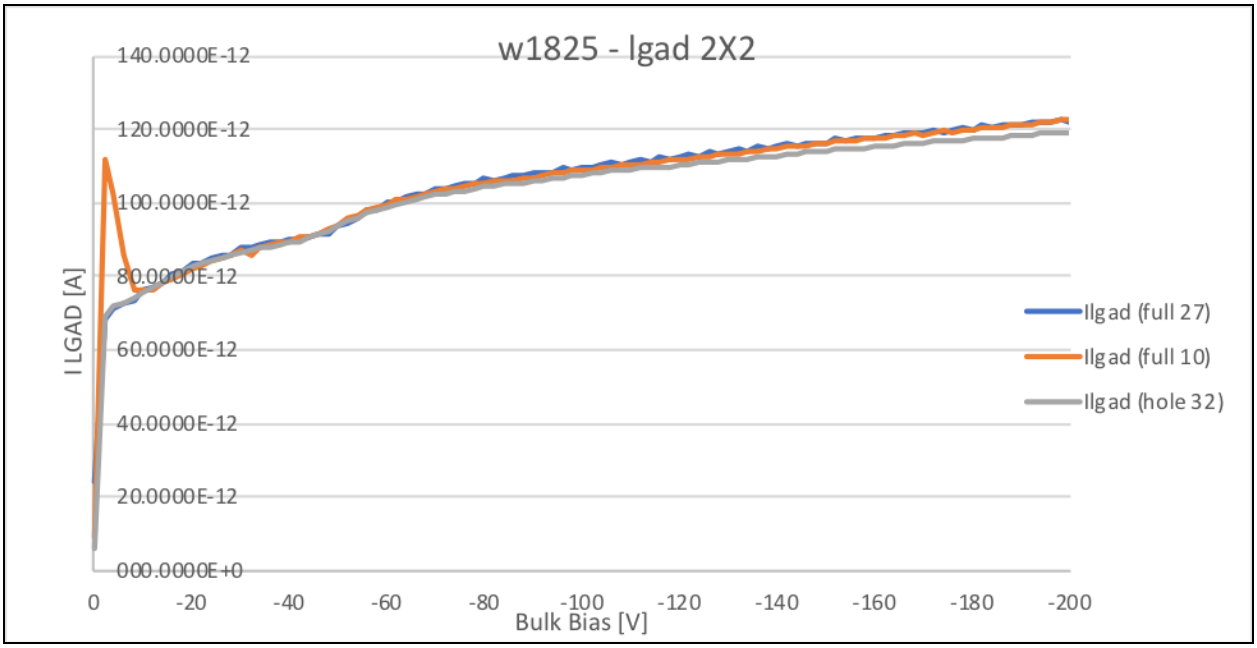


Figure 1: Leakage current from three different LGADs belonging to the latest fabrication batch.

The results allowed us to define priorities and milestones for FY 2018, continuing the program outlined in the original proposal as:

- Laboratory characterization at BNL and through testbeam at Fermilab (FNAL) and CERN (Mar 2018 @ FNAL, April-Jul 2018 @ CERN)
- Irradiation of devices for radiation damage studies with photons and pions
- Completion of irradiation studies. First annealing studies at BNL.

Resolving Technical Issues of a Compact Time Projection Chamber for Use at both the Relativistic Heavy Ion Collider and a Future Electron Ion Collider

LDRD Project # 16-035
E. O'Brien

PURPOSE:
The purpose of the project is to develop a concept for building a compact Time Projection Chamber (TPC) that could be implemented as a detector for both the Relativistic Heavy Ion Collider (RHIC) and a future Electron-Ion Collider (EIC). The challenges are: the mechanical footprint of the detector must be only minimally larger than the active volume of the TPC; the design of the high voltage field cage must minimize field distortions especially at the inner radius; the readout plane must minimize ion feedback and space charge build-up; and the electronics chain must be able to handle both the high multiplicities during RHIC heavy ion operation and the high luminosities during RHIC polarized proton running. At the end of this project, we plan to have completed a conceptual design for the compact TPC that could operate at both RHIC and EIC.

Time Projection Chambers are an excellent technology for RHIC and EIC. They measure the momentum of all charged particle with good precision. They are generally low mass devices meaning that they do not significantly scatter the particles that they are measuring. Their limitations include that their performance degrades at high particle multiplicities and high rates due to field distortions from ion build up, and they have trouble tracking at radii close to the interaction point, at radii less than 60 cm. If the space charge effects could be reduced at low radii and high multiplicity, then their utility at RHIC and EIC would be markedly improved. A TPC with a smaller outer radius than those currently employed at RHIC and LHC would enable the installation of calorimetry at a much smaller radius and result in calorimeters with significantly smaller volumes that translate to significantly lower costs.

APPROACH:
The Principal Investigator (PI) has been involved in the design, construction and operation of tracking detectors for use in both High Energy Physics and Nuclear Physics experiments for over three decades. As a member of the sPHENIX project, the PI has become interested in solving a number of challenges associated with compact TPCs that would allow for their use in both a RHIC and EIC detector. The PI also has a number of colleagues that share this interest and are collaborating with him on this project including: Craig Woody: John Haggerty; Takao Sakaguchi; Jin Huang; and Bob Azmoun at BNL, Tom Hemmick and Klaus Dehmelt at Stony Brook University (SBU), and Alexander Milov at the Weizmann Institute of Science. We will use design studies, simulation studies, bench tests and fabrication of prototypes to test our ideas on how to solve the challenges previously described.

TECHNICAL PROGRESS AND RESULTS:
In 2017, the engineering design of a full-scale prototype of a compact TPC is reaching completion. The design of the prototype field cage, central membrane, end caps and external support structure is advanced. Construction of the prototype is progressing at SBU. Readout module prototype design has begun. Software simulations that model the response of the detector gas to charged particles has made good progress. Approaches to minimize space charge effects look very promising in simulation. Prototype tests to confirm the simulations will take place in

2018. A prototype front-end electronics board is built. We have performed a successful readout of the full electronics chain in prototype versions.

The construction of the prototype field cage (Figure 1) is very advanced and we are preparing a small TPC prototype for tests in the Fermilab test beam in March 2018. This project has allowed us to answer many design and simulations questions related to compact TPCs and given us enough confidence to include a compact TPC in the design of the sPHENIX detector.

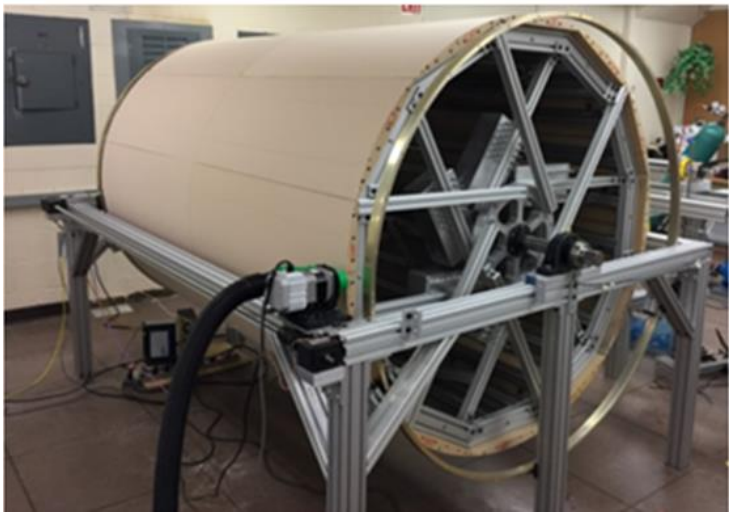


Figure 1: Mandrel that will be used to form the outer cylinder of the field cage for the prototype Time Projection Chamber.

Exploring Hadron Structure with ab initio Lattice Quantum Chromodynamics Calculations and Making Predictions for eRHIC

LDRD Project # 16-037
T. Izubuchi, J. Qiu, P. Petrezcky, R. Venugopalan

PURPOSE:
For reliable theoretical prediction of the motion of quarks and gluons (collectively called partons) inside a nucleon, we compute the Parton Distribution Function (PDF) and transverse momentum dependent (TMD) PDF by solving Quantum Chromo Dynamics (QCD) calculations in discretized space-time using supercomputers (Lattice QCD). This will provide useful guidance for the future Electron-Ion Collier (EIC) experiments. To examine recently proposed formalisms, which enable a direct computation of PDFs from Euclidean space-time, a collaboration of experts in Lattice QCD and perturbative QCD is organized to make *ab initio* predictions for eRHIC.

APPROACH:
Lattice QCD provides a systematic way to study the non-perturbative region of QCD. For example, lattice QCD can be used to compute hadron and hadronic matrix elements, reaching the one percent level or even higher accuracy. In the new lattice formalism for PDF (*e.g.* [1-3]), we compute the nucleon's matrix elements of a non-local QCD object (Wilson line) at a large momentum to obtain the quasi-PDFs, which are related to the real PDF via perturbative QCD factorization. To obtain reliable results, it is essential to use the correct convolution integral between computed function and PDF, a large enough momentum for hadrons, and proper renormalization of the Wilson line. We collaborate with Luchang Jin (RIKEN BNL Research Center [RBRC] and the University of Connecticut) and Sergey Syritsyn (Stony Brook University/RBRC).

TECHNICAL PROGRESS AND RESULTS:
In FY 2017, we addressed operator renormalization, a proper convolution integral between a computed object and a PDF and started the actual lattice calculation for the hadron PDF. For the renormalization procedure, both perturbative and non-perturbative renormalization were studied, and the results were published [4,5].

The large momentum effective theory was studied to establish a clear relationship between the matrix elements accessible from lattice QCD and the light-cone PDF. The relationship was provided by a factorization theorem with a non-trivial matching coefficient [6].

To check the large momentum limit of hadrons, we first examined a pion PDF as a feasibility study. We used a staggered QCD ensemble with very fine lattice spacing (a=0.060 and 0.040 fm) with staggered 2+1 flavor quarks with 300 MeV pions, which allow us to reach a large (~ O(1) GeV) momentum. To enhance the statistical signal for large momentum hadrons, we utilized recently proposed quark smearing for high momentum [7]. We wrote proposals to USQCD and the DOE Innovative and Novel Computational Impact on Theory and Experiment (INCITE) program for computational resources.

Milestones:

FY17/18

- With the establishment of the methodologies and computer resources, we computed the PDF of pions with fine lattice spacing (0.06, 0.04 fm), while keeping an adequate volume size. Complete pion PDF on Domain-Wall Fermions (DWF) on a coarse lattice, Wilson on a fine lattice (48, 64 cubes)
- Perform global analysis of the generated lattice "data" in terms of a QCD factorization formalism to extract the PDFs, as a function of x, to establish in practice a complete example of extracting PDFs from the lattice QCD calculations, based on QCD factorization. Renormalize using fine lattices.
- Quantify the improvement from the first exploratory work. Control large Pz ~ O(1) GeV behavior
- Set up a Next-to-Leading Order (NLO) computation of Deeply Virtual Compton Scattering (DVCS) at small x and perform first computations.

FY18/19

- Perform nucleon PDF on a realistic lattice based calculation using the INCITE allocation for the ratios of PDFs, such as d(x)/u(x) at large x (> 0.1), and compare it with calculations from various models and existing data
- Extend the program to cover polarized PDFs, in particular, the gluon helicity contribution to the proton's spin: DG(x), which is one of the key measurements at eRHIC.
- Consider DWF calculation
- Try to identify new lattice computable hadronic matrix elements, which are also factorizable to PDFs, TMDs, and generalized parton distributions, in order to establish a full program to extract the hadron's quark-gluon correlation functions from lattice QCD calculations, which is complementary to the ongoing worldwide efforts using data from high-energy scattering experiments
- Combine NLO computations with nonperturbative (lattice motivated) initial conditions to make systematic predictions for DVCS at small x. Connect small x approaches to computations in p+p and p+A final states at the Relativistic Heavy Ion Collider/Large Hadron Collider.

[1] X. Ji, Phys. Rev. Lett. 110, 262002 (2013), 1305.1539.

[2] Y.-Q. Ma and J.-W. Qiu, "Extracting Parton Distribution Functions from Lattice QCD Calculations" preprint arxiv 1404.6860 [hep-ph], 10 Sep 2017

[3] X. Xiong, X. Ji, J.-H. Zhang, and Y. Zhao, Phys. Rev. D90, 014051 (2014), 1310.7471.

[4] T. Ishikawa, Y. Q. Ma, J. W. Qiu and S. Yoshida, "On the Renormalizability of Quasiparton Distribution Functions," Phys. Rev. D 96 (2017) 094019 [arXiv:1707.03107 [hep-ph]].

[5] J.-W. Chen, T. Ishikawa, L. Jin, H.-W. Lin, Y.-B. Yang, J.-H. Zhang, and Y. Zhao, "Parton Distribution Function with Non-perturbative Renormalization from Lattice QCD," to be published in Phys. Rev. D [arXiv:1706.01295 [hep-lat]].

[6] Taku Izubuchi, Xiangdong Ji, Luchang Jin, Iain W. Stewart, Yong Zhao. Jan 11, 2018. 19 pp.

"Factorization Theorem Relating Euclidean and Light-Cone Parton Distributions" e-Print: arXiv:1801.03917 [hep-ph]

[7] G. Bali et al, Phys. Rev. D 93, 094515 (2016).

Preconceptual Design Study for Large Scale Structure Experiment post Large Synoptic Survey Telescope/Dark Energy Spectroscopic Instrument

LDRD Project # 16-038
A. Slosar, P. O'Connor, A. Nomerotski

PURPOSE:
The main purpose of this LDRD is to develop know-how for 21-cm cosmology within the DOE Laboratory complex. With its sister LDRD (16-026), we are building a small radio-telescope prototype. This prototype will test several novel techniques for radio astronomy that could be used in an eventual instrument with the aim of developing a systematically very clean experiment. The second goal of the LDRD is to determine the science reach of a possible future DOE experiment and to work towards a design that could become a DOE project in the 2020s.

APPROACH:
The Principal Investigator was member of the Cosmic Visions Dark Energy Committee, which was created by the DOE to find possible directions for a future survey experiment in the DOE cosmic frontier. A report was written which suggests four possible experimental approaches in the post Large Synoptic Survey Telescope/Dark Energy Spectroscopic Instrument era. One of these is the 21-cm radio experiment, which relies on measuring the hydrogen spin-flip transition (with rest-frame wavelength of 21-cm) by galaxies in the universe. With the goal of developing a future DOE-funded experiment in this field, we are: i) building a small in-house prototype to learn relevant techniques in order to be considered a lead lab in this field; ii) studying the synergies between DOE expertise and radio astronomy; and iii) further developing the science case for such an experiment. On the experimental side, we collaborate with Jeff McMahon (Michigan), Daniel Marlow (Princeton) and Phil Mauskopf (Arizona). Paul Stankus, an Oak Ridge National Laboratory employee stationed at BNL, works on this project. Our postdoc Chris Sheehy is also a Goldhaber fellow. In addition, we have an undergraduate and a Ph.D, student from Stony Brook working on it.

TECHNICAL PROGRESS AND RESULTS:
We have continued to make significant progress on our prototype and expect first publishable results soon. In particular:

- We have designed, built and installed a low-cost 4 meter reflector.
- We have designed, built and installed a support tower structure.
- We have designed and built the primary feed and associated front-end electronics, including all the necessary mechanical, electrical and weather protection.
- We have built and tested a 550MHz spectrometer using a custom built PC with a digitizer and a GPU processing unit.

The system was integrated on site in the summer of 2017 and started to observe in a single channel single dish mode (see Fig. 1). We have successfully detected our own galaxy in 21-cm and in broadband emission and also several calibration sources. We have achieved a measured on-sky noise temperature of around 50K. However, many aspects of our system remain poorly understood. We hope to extend the system to two or four dishes over the coming year and the full 4 channels and perhaps detect a signal in cross-correlation with low redshift galaxies.

The goal is to iteratively improve our system performance and our understanding of its properties, while designing a long-term program in 21-cm. To this end, we are also developing a thriving 21-cm program in collaboration with Yale and are attempting to create a seed collaboration for a U.S. cosmology-focused 21-cm experiment.

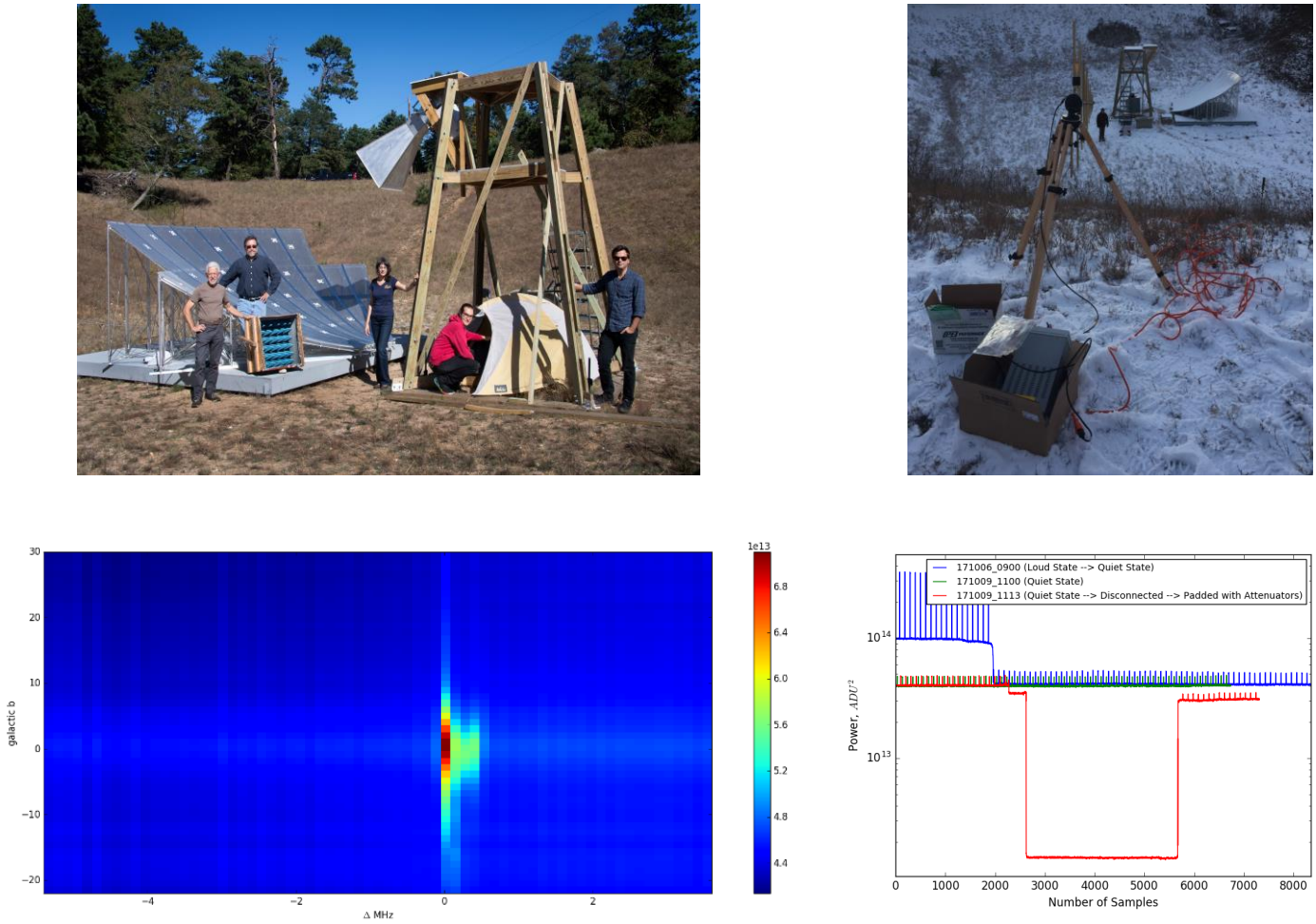


Figure 1: The full system integrated for the first time (upper left; from left to right: Paul O'Connor, Paul Stankus, Justine Haupt, Will Tyndall, Chris Sheehy), measuring side-lobe response (upper right), detection of the Milky Way transiting and emitting in 21-cm (bottom left), the front-end calibration pulses generating comb in the raw data (bottom right).

Milestones:

- Resolve remaining issues with calibration at 1.3 GHz
- Take, analyze and calibrate six months of data
- Measure cross-correlation with low-redshift galaxy survey data and publish papers on results and telescope design
- Continue developing beam calibration methods, including drown-flown artificial source calibration.

Machine Learning Assisted Material Discovery

LDRD Project # 16-039
S. Yoo, D. Lu, D. Zakharov, E. Stach, A. Frenkel

PURPOSE:

X-ray and Transmission Electron Microscopy (TEM)-based imaging and spectroscopies are powerful tools to characterize structures and electronic properties in materials. Under *in situ* and *operando* conditions, information from such measurements is critical input towards a mechanistic understanding of the underlying dynamical processes and functional properties of materials, such as electrodes in energy storage or nanocatalysts in heterogeneous catalysis.

Our original objectives are three fold: 1) develop image analysis methods to allow near real time analysis of TEM images from a 3 Giga Byte (GB)/s image stream, solving a general problem that will soon be seen at multiple beamlines at National Synchrotron Light Source II (NSLS-II); 2) develop a Spectroscopy Genome that allows deep insights into the atomic-scale changes that occur in complex dynamic processes in *operando* conditions; 3) combine the activities in 1) and 2) with first principles calculations in a robust, integrated manner to describe the dynamic structural changes. We dropped the third goal given the budget constraints.

This project provides the quantitative information needed to rationally design functional materials for energy applications. These techniques exploit both *in situ* and *operando* approaches, which generate high rate data streams. Thus, automatic real time analyses and data driven discovery have been identified as critical Laboratory goals, as BNL is developing a diverse array of characterization techniques at both NSLS-II and the Center for Functional Nanomaterials. These will provide the quantitative information needed to rationally design functional materials for energy applications.

APPROACH:

For TEM image analysis, most of the image analyses are done manually or limited image processing tools are applied. We investigate the diverse material (semi-)automated quantitative image analysis difficulties and develop novel near real-time approximated computer vision algorithms customized for TEM to enable *in situ* analysis.

In the Spectroscopy Genome project, we propose to develop and apply a robust first principles approach for inverse modeling of core-level absorption spectra. With the focus on *in situ* and *operando* probes, the correlation between atomistic details of key structural motifs (*e.g.*, atomic position, coordination, and bond length) and core-level spectral features is systematically investigated for well-defined systems, with close cooperation from theory, simulation and machine learning algorithms. The proposed study involves constructing experimental spectra standards, extracting significant spectral signatures, developing corresponding high-quality computational spectra standards, and identifying relevant local structural descriptors. Success will allow us to interpret the changes to key local structural motifs that are due to stochastic or dynamic effects under *operando* conditions using the corresponding experimental spectral features.

TECHNICAL PROGRESS AND RESULTS:

For TEM image analysis, we built a prototype pipeline for automated detection and tracking of nanoparticles. From a 3 GB/s data stream, we developed an algorithm for frame-to-frame gold

nanoparticle detection/segmentation and tracking during a 137 minute heating experiment inside an Environmental Transmission Electron Microscope (ETEM) [1] (Figure 1). We also evaluated the feasibility of deep learning-based particle object detection and tracking and found that the results are more robust to noise around the edge of the images compared to traditional computer vision techniques. These are encouraging results for future development consideration. For our algorithm development effort in streaming analysis to accelerate the high velocity data stream, we prototyped algorithms of streaming approximation of classical multidimensional scaling and produced a nearly identical streaming approximation of classical multidimensional scaling.

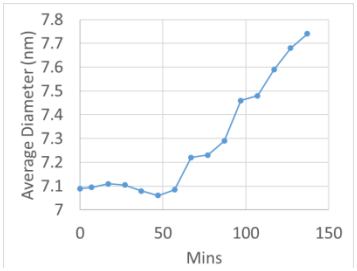


Figure 1: the average particle diameter changes over 137 minutes, which is nearly impossible to be traced by hand.

In the Spectroscopy Genome project, Lu and Frenkel demonstrated the use of an artificial neural network to unravel the hidden relationship between X-ray Absorption Near Edge Structure (XANES) features and the 3D geometry of metal nanoparticles [2], where high-throughput *ab initio* XANES simulations were used to build the training set for machine learning. Our approach allows us for the first time to accurately reconstruct the average size, shape, and morphology of small metal nanoparticles on-the-fly from their XANES spectra (Figure 2). This method can be generalized to other nanoscale systems and opens new avenues for real time tracking of, *e.g.*, nanoparticle growth or the structural changes of catalysts during chemical reactions. This work was highlighted by DOE (Oct.10, 2017) [3] and as one of BNL's 2017's top-10 discoveries and scientific achievements [4].

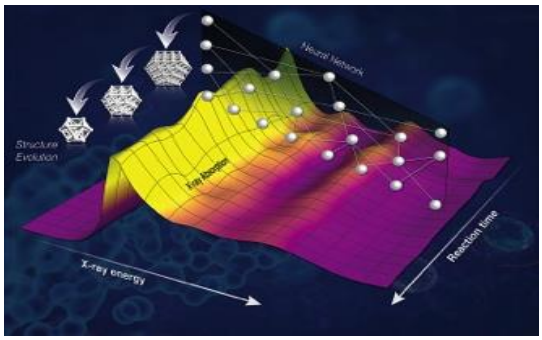


Figure 2 Schematics of machine learning assisted structure determination using XANES.

Milestones:

- XANES analysis tools deployed at the Inner-Shell Spectroscopy beamline at NSLS-II (collaboration with Eli Stavitski and Klaus Attenkofer)
- Manuscript of forward machine learning to predict XANES spectra from molecular structures
- Manuscript of backward machine learning to predict local atomic structures from XANES spectra
- Book chapter on "machine learning-assisted metal nanoparticle structure determination using XANES."

References:

1. Lin, Y., et al., (2017, August). Near real time ETEM streaming video analysis. In *Scientific Data Summit (NYSDS), 2017 New York* (pp. 1-4). IEEE.
2 Timoshenko, J. et al., Supervised Machine-Learning-Based Determination of Three-Dimensional Structure of Metallic Nanoparticles. *The Journal of Physical Chemistry Letters* 2017**,** *8* (20), 5091-5098.
3. https://science.energy.gov/news/featured-articles/2017/?page=2.
4. https://www.bnl.gov/newsroom/news.php?a=112669.

Dynamic Visualization and Visual Analytics for Scientific Data of National Synchrotron Light Source II

LDRD Project # 16-041
W. Xu

PURPOSE:

This project aims to establish a prototype system that visually represents, explores and interacts with extreme-scale multivariate scientific data to empower future science research at National Synchrotron Light Source II (NSLS-II) and the Center for Functional Nanomaterials (CFN). To our knowledge, there are no existing visualization and visual analytics solutions satisfying the unique requirements of NSLS-II i.e., the visual analytic environment for extreme-scale data and complex analyses so that scientists can better integrate, understand and interact with their data and analysis. On the success of this project, we expect to secure funding from the DOE Office of Advanced Scientific Computing Research for programs including scientific data management, analysis and visualization. The research will also foster cross-disciplinary collaborations.

APPROACH:

An online visual analysis tool that can process, manipulate and visualize extreme-scale data is critical for scientists to make the right decision while onsite and adjust their measurement strategies during the experiment. Therefore, we identified three pilot projects that, according to our scientific collaborators, have extreme difficulties in 2D/3D/high-dimensional data visualization, because the capabilities for interactive strategic exploration of raw data, metadata and analyzed results are lacking, thereby impeding the desired scientific insights.

- **Enable visual comparisons and integrations of multi-modal imaging techniques:**

Co-investigators: H. Yan and Y. Chu (NSLS-II), K. Mueller (Stony Brook University)

We devise an interactive visual analytics tool that can combine, correlate and process different image datasets so as to enable scientists to fully understand relationships among the chemical, elemental, structural and physical quantities being imaged.

- **Automatic large-scale image set organization, summarization, and analysis:**

Co-investigators: K. Yager (CFN), M. Fukuto (NSLS-II), Klaus Mueller (Stony Brook)

Optimizing the functionality of material design requires strategic exploration of the vast parameter spaces associated with complex materials. We thus develop visualization tools ideally suited to managing and exploring large collections of X-ray scattering images, providing the tools necessary to efficiently summarize and explore correlations and trends in large scattering datasets.

- **Dynamic 3D visualization and interaction:**

Co-investigators: W-K. Lee, Y. Chu, Dan Allen (NSLS-II), Klaus Mueller (Stony Brook)

For 3D datasets with multiple attributes, we propose a dynamic way to visualize data in different data channels, while the raw data keep screaming in. Assisted plots illustrating the angle coverage of the sample, as well as meta-data display will also be implemented.

TECHNICAL PROGRESS AND RESULTS:

In the first year, we completed the proof of concept work for the first two pilots. In the second year, we further extended that into two software releases. We met all milestones and published good quality papers. Specifically, we made the following progress.

- *ColoMapND*: Extending 2D color-mapping for multivariate dataset in pseudo-3D

Based on the first year's development, we released the software and installed it on the Hard X-ray Nanoprobe beamline. We further extended the color mapping algorithm from a 2D multivariate dataset to stack-based 3D dataset (Fig. 1.) Users can alter the slice number and view perspective to check the whole 3D dataset colorization effects.

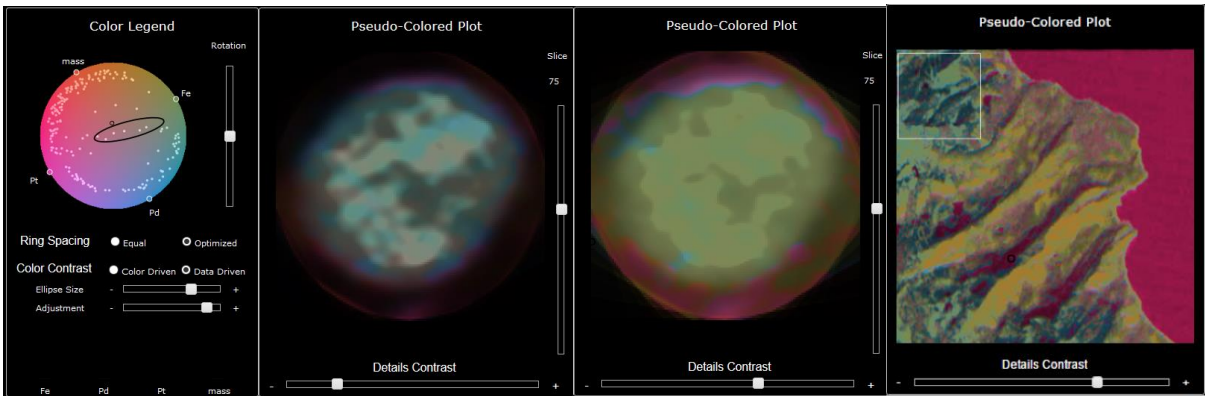


Figure 1: Color mapping for multivariate dataset in pseudo-3D: left: the color space, middle left: equal spacing mode, middle right: optimized spacing mode, and right: a multi-field mountain image.

- *MultiSciView*: Multi-level visual exploration for X-ray scattering images

We further refined the previous framework that provided semantic zooming with multiple levels of details for multivariate X-ray images. There are three major visualizations: multi-level scatterplot, cross filters and attribute projection, altogether serving as a cross-space exploration as shown in Fig. 2. This software is also released and will be installed at the Complex Materials Scattering beamline.

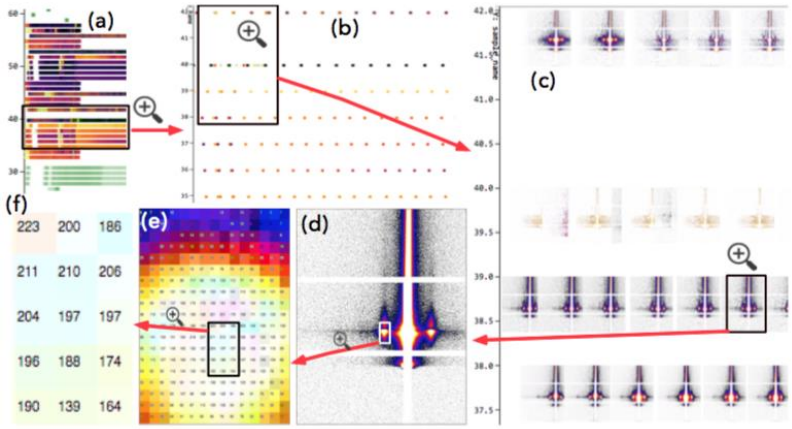
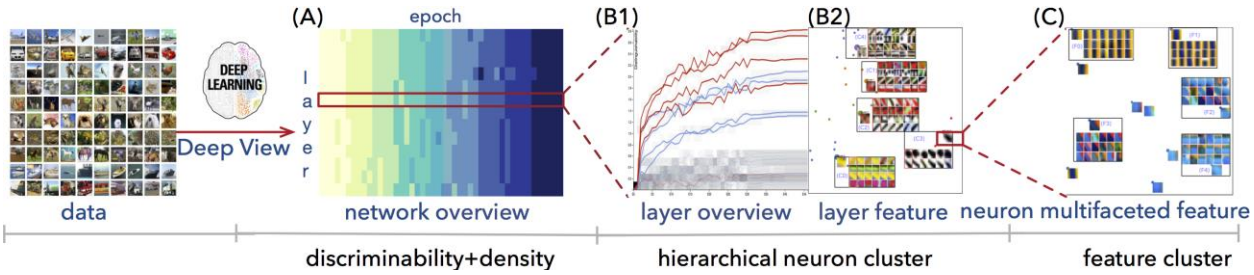


Figure. 2: Multi-level visual exploration for X-ray scattering images: (a) the scatterplot overview, (b) the zoom-in scatterplot, (c) the corresponding image series, (d) the selected image view, (e) its zoom-in view with pixel values plotted, and (f) the further zoom-in with details of the pixel values.

- Evolutionary visualization for deep neural networks

We completed a proof of concept on a framework to visualize the evolution of convolutional neural networks. We provided three levels of scales: macroscale as an overview of the progress of the network training, mesoscale as layer visualization, and microscale for neuron multifaceted visualization.



Milestones:

In FY 2018, we will: 1) finalize color mapping to support real 3D exploration for pilot project 3; 2) enhance attribute space exploration for MultiSciView project; 3) complete corresponding publications.

Deep Structured Analysis for Image Datasets from Center for Functional Nanomaterials and National Synchrotron Light Source II

LDRD Project # 16-043
K. Yager, D. Yu, M. Fukuto

PURPOSE:
We are building towards a transformative new paradigm for scientific research, where experimental acquisition, analysis, and decision-making, are all automated, thereby liberating scientists to focus on high-level scientific questions. Our approach involves exploiting recent advances in machine learning to massively automate data analysis. We are adapting deep learning methods to the problem-space of scientific data and thereby developing "physics-aware deep learning" methods. The ultimate goal of this project is to develop an automated data-analysis pipeline for X-ray scattering experiments and to deploy this software at National Synchrotron Light Source II (NSLS-II) beamlines.

APPROACH:
Modern scientific instruments are now generating data at unprecedented rates. In particular, NSLS-II offers unprecedented X-ray brightness and high-speed detectors. The correspondingly large data-rate is beyond the ability of human experimenters to manually interpret. It is now evident that a crucial complement to high-throughput instruments is automated analysis methods, which can categorize, tag, and analyze scientific data without human intervention. This automation liberates the human scientist to concentrate on high-level scientific questions and focus their attention on the subset of the data most meaningful for a given problem. This extreme automation, in turn, enables more ambitious scientific projects. In particular, these methods enable streamlined materials discovery, where new materials with desired performance (mechanical, light-harvesting, energy storage, etc.) can be efficiently found.

This project is building automated, streaming analysis pipelines for extracting scientifically-meaningful insights from datasets relevant to materials discovery, especially X-ray scattering images (Figure 1). We are developing data-analysis pipelines that serve a dual role: providing experimenters with useful (physically-meaningful) intermediate results, and using these analysis results as inputs to machine learning methods. In particular, we are leveraging the recent successes in "deep learning," where carefully structured neural hierarchies are trained to identify hierarchies of structures in data. By using multiple crafted input channels (raw data, data after physics-based decomposition, analysis outputs) into a deep learning network, the machine learning can be strongly optimized to a particular problem space (collaborations with Minh Hoai Nguyen, Stony Brook University (SBU); Hong Qin, SBU; and Dantong Yu, New Jersey Institute of Technology/BNL). Thus, we are creating both efficient analysis pipelines and new physics-aware machine learning algorithms. These methods can be applied to a variety of "images": the raw detector images generated by instruments, reconstructed maps of sample structures, and the abstract phase spaces of materials science. This deep-learning centric analysis will eventually be exploited to automate the scientific experiment itself, by providing feedback to algorithms that can efficiently explore scientific problems and make decisions about what experiments to conduct next. Overall, this project aims to deliver a data analysis pipeline for X-ray synchrotron instruments, empowering more ambitious materials discovery experiments.

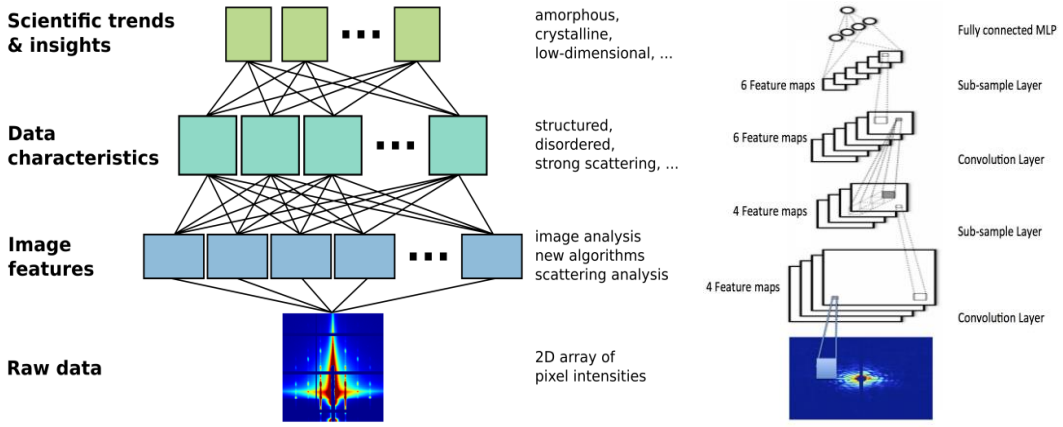


Figure 1: This project is exploiting machine learning methods to analyze X-ray scattering data (bottom). Using structured deep learning methods (right), scientifically-meaningful insights will be automatically extracted from the data (left).

TECHNICAL PROGRESS AND RESULTS:

In FY 2016, we designed the required analysis pipeline, created a preliminary software framework for data-analysis modules, and crafted several domain-specific modules, including a physics-aware in-painting method. We also investigated the use of conventional deep learning methods (convolutional neural networks acting on the image data) to automatically categorize X-ray scattering images. In FY 2017, we have greatly augmented these approaches, especially by crafting domain-specific transformation methods. We have investigated multi-view (or multi-channel) deep learning, where several representations of the data fed into a neural network. By carefully selecting these representations, domain-specific information can be implicitly included in the learning. We developed a Fourier-Bessel decomposition for X-ray scattering images, which highlights symmetry features in the images and developed a "geometry correction" method that can transform complex reflection-mode scattering images into much simpler transmission-mode data (thereby eliminating several complicated data distortion effects). We have demonstrated that the combination of domain-specific data transformation with convolutional networks enables vastly improved data analysis performance.

Simultaneously, we designed and implemented a new pipeline framework targeted towards immediate analysis of data at a synchrotron beamline. This code connects to the instrumental data collection software used at NSLS-II and allows specialized analysis modules (including deep learning code) to be triggered. This pipeline framework is now operating at the Complex Materials Scattering beamline at NSLS-II, allowing us to test our methods in a realistic data collection context.

We will continue our foundational development in machine learning for scientific data and implementation of these methods in a real-world experimental context. We will connect this work to parallel efforts in data visualization and autonomous experimental control, where the deep learning pipeline will feed analysis results to these other codebases.

Milestones:

FY18 milestones are: (1) to demonstrate that physics-informed multi-channel machine learning is a viable and powerful strategy to improve scientific data analysis; (2) to transition the developed analysis pipeline from prototyping into user operations at the beamline; and to use (1) and (2) in a proof-of-principle autonomous experiment, where the analysis pipeline acts as input to a decision-making software module.

Catalysis Program in Sustainable Fuels

LDRD Project # 16-045

J. Chen

PURPOSE:

This LDRD project established a research program led by Jingguang Chen as a Joint Appointee between BNL and Columbia University. The research program addresses the need for improved catalytic pathways for sustainable fuel synthesis. It focuses on chemical routes for the synthesis of fuels by recycling carbon dioxide, including catalytic processes to convert CO_2 into CO using H_2 as a source of energy. At present, about 95% of H_2 is produced from hydrocarbon reforming, which produces CO_2 as a byproduct in the process of H_2 production. Therefore, this LDRD project also explores two alternative ways to convert CO_2: (1) the reduction of CO_2 directly by hydrocarbons without using H_2 and (2) the discovery of active and low-cost electrocatalysts for water electrolysis to produce CO_2-free H_2 that can be subsequently used to convert CO_2.

APPROACH:

Motivation: The primary energy source for transportation is currently the combustion of fossil fuel hydrocarbons. This contributes to rising atmospheric carbon dioxide levels. Chemical routes for the synthesis of fuels by recycling carbon dioxide driven by renewable energy sources would reduce the net emission of carbon. This research program specifically focuses on chemical routes for the synthesis of fuels by recycling carbon dioxide.

This LDRD project established a research program at both BNL and Columbia University. At BNL, the Chen group is collaborating with the existing BNL catalysis and electrocatalysis research groups. The FY 2017 research effort under this LDRD was carried out primarily at BNL. The project strengthens the BNL catalysis science program through linked research thrusts on CO_2 activation. The projects are carried out using a combination of theoretical and experimental methods and *in situ* techniques at the Center for Functional Nanomaterials (CFN) and National Synchrotron Light Source II (NSLS-II).

TECHNICAL PROGRESS AND RESULTS:

During the past year of LDRD funding, the Chen group at both BNL and Columbia University continued to identify design principles of electrocatalysis to expand our efforts in using metal-modified carbides as potentially low-cost catalysts for water electrolysis for the production of produce CO_2-free H_2. Using density functional theory (DFT) calculations, we discovered that the hydrogen binding energy can be used as a descriptor to design metal-modified carbides in both acid and alkaline electrolytes. Furthermore, we applied the same design principle to predict that metal-modified nitrides should also be active and stable for water electrolysis. So far we have confirmed these predictions by experimentally measuring the activity of metal-modified carbides; the synthesis and evaluation of metal-modified nitrides are underway.

We also continued to explore the catalytic conversion of CO_2 with light alkanes, such as ethane that is abundant from shale gas. The reaction of CO_2 with ethane simultaneously converts CO_2 to CO and dehydrogenates ethane to ethylene, which is one of the most highly demanded chemicals. Therefore, this reaction allows both CO_2 mitigation and effective upgrading of under-utilized ethane in shale gas. Although we discovered that Fe-Ni bimetallic catalysts are active for this reaction, we did not understand the active sites and mechanisms.

During the past funding period, we performed extensive DFT calculations using the CFN clusters, carried out electron microscopy characterization with CFN facilities, and utilized synchrotron techniques at NSLS-II to determine the physical and electronic properties of the catalysts. These combined theoretical and experimental studies revealed that the active sites for the reaction of CO_2 and ethane are the interfaces between metallic Ni and oxidized Fe. These discoveries potentially allow us to design more effective and stable catalysts for the simultaneous conversion of CO_2 and ethane.

Milestones:
Future LDRD research efforts will continue to focus on the following two areas:

- Discovery of electrocatalysts to produce CO_2-free H_2 from water electrolysis: Based on DFT predictions from the past funding period, we will evaluate several promising metal-modified nitrides as active and stable electrocatalysts for water electrolysis. The utilization of metal-modified nitrides can potentially mitigate the instability issues that were observed previously for metal-modified carbides. If successful, these results can potentially open up a new frontier in the utilization of nitrides as stable and low-cost electrocatalysts.

- Catalyst design for converting CO_2 using ethane and other light alkanes: Based on the observation of the important roles of the FeO/Ni interface from the past funding period, we will design catalysts that optimize the interfacial area and stability of FeO/Ni. In addition, results from the past funding period also provided guidance on exploring other oxide/metal interfaces for the reaction of CO_2 and ethane. If the initial results are successful, we will also explore the possibility of converting CO_2 with other light alkanes (propane and butane), which, based on our thermodynamic calculations, could reduce the reaction temperature for the conversion of CO_2.

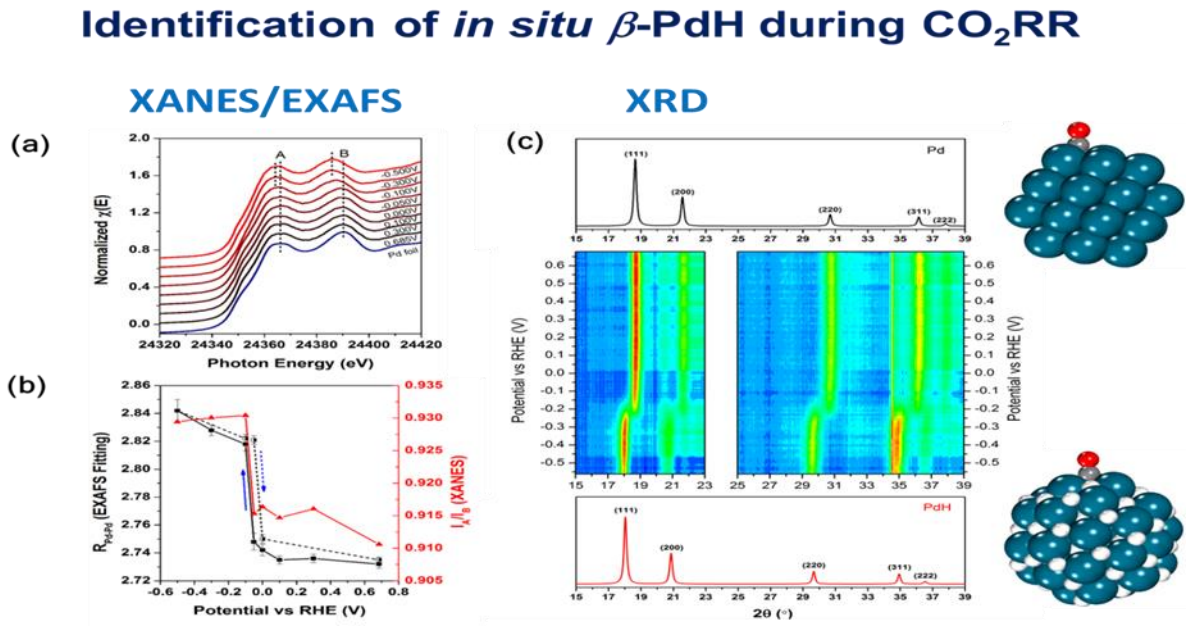


Figure 1: Example of utilization of *in situ* techniques, X-ay Absorption Near Edge Spectroscopy (XANES), Extended X-ray Absorption Fine Structure (EXAFS) and X-ray Diffraction (XRD) for determination of active sites for the CO_2 reduction reaction (CO_2 RR) under reaction conditions.

Strong-Strong Beam-Beam Interaction Studies for a Ring-Ring Based Electron Ion Collider

LDRD Project # 16-046
F. Willeke

PURPOSE:
The interaction between particles of opposite beams colliding in high luminosity storage rings is strongly nonlinear and the densities of the beams in collisions, which provide high luminosity, is limited by a number of coherent and incoherent instabilities and nonlinear dynamic effects. This effect, if it occurs in eRHIC would limit the luminosity below the design values and could prevent stable high luminosity operations. Therefore, it must be considered as a risk, which deserves to be studied thoroughly before a decision on the scheme of the eRHIC electron hadron collider is made. The purpose of the project is to assess the feasibility of high luminosity operations of the eRHIC ring-ring option with computer simulation that will use computer codes based on a strong-strong interaction model, in which the mutual nonlinear forces of the two beams upon each other are assessed.

APPROACH:
Electromagnetic fields of a colliding beam act on the beam particles like a very special electromagnetic lens, which is highly non-linear (forces depend on the particle distribution) and time-dependent (the particle distribution is changed due to the interaction). The beam-beam interaction results in the following effects of particle beam dynamics: intensity-dependent shift of betatron tunes; changing effective beta function at the interaction point (dynamic beta effect); changing transverse emittances of the colliding beams; and coherent beam instabilities. These effects result in deterioration of the collider luminosity.

Study of the beam-beam effects includes modeling of the field-particle interaction, non-linear beam dynamics, and multi-particle dynamics. An analytical approach is limited to a few simplified cases; computer simulation is the main tool.

In the weak-strong simulations, the strong bunch is represented by a rigid Gaussian not affected by the weak beam, and the weak bunch is represented by macro-particles. The weak-strong approach is used to study a single particle's long-term stability; it is time efficient and has negligible numerical noise. However the weak-strong simulation is not self-consistent.

In the strong-strong simulations, both bunches are represented by a large number of macro-particles, the particle-in-cell method is used to solve the 2D Poisson equation. The strong-strong approach is self-consistent, although it is much more time consuming and affected by numerical noise. The strong-strong simulations are used to study coherent beam-beam motion and its stability.

For this project, the BBSS computer code developed by K. Ohmi (from the High Energy Accelerator Research Organization in Japan known as KEK) has been selected for strong-strong beam-beam simulations. This code was previously used to study the performance of crab-cavities at KEKB (the particle accelerator used in the Belle experiment at KEK), and for studies of the luminosity degradation due to incoherent emittance growth and coherent beam-beam instability in the Large Hadron Collider. The BBSS code has been installed with the developer’s help on a

dedicated node of the National Synchrotron Light Source II Accelerator Physics computing cluster. The project work plan includes: simulations for the Hadron-Electron Ring Accelerator (HERA) case and comparison of the results with experimental data to benchmark the code; strong-strong simulations for the eRHIC ring-ring option, analysis of the results; and writing a contribution to the eRHIC design report.

TECHNICAL PROGRESS AND RESULTS:

All the tasks listed in the project work plan for FY 2017, have been completed. The results were presented and discussed with invited experts at the eRHIC Design Choice Validation Review in April 2017 and at the Electron Ion Collider Collaboration Meeting in October 2017.

Optimal simulation parameters have been determined for the strong beam-beam simulations by comprehensive convergence studies of the strong-strong simulation code BBSS.

The BBSS code has been also benchmarked against experimental data from HERA, the only electron-proton collider that was ever in operation. No beam-beam instability was observed at the design beam parameters, this is consistent with 2006 HERA-II experimental runs. The beam-beam effects, such as increase in the beam emittance, have been observed in simulations, if the beam intensities exceed four times the design values.

A 2D map of beam stability as a function of the electron betatron tunes has been calculated. The optimal tune working points have been found: the horizontal tune is 0.31 for the proton ring and 0.08 for the electron ring; the vertical tune is 0.305 for the proton ring and 0.06 for the electron ring.

As has been found using strong-strong simulations, the horizontal coherent instability occurs if the number of protons exceeds the design beam intensity at least by a factor of two.

For the nominal eRHIC parameters, neither significant emittance growth nor coherent instability was observed in the simulation results at the optimal working points.

Additional studies, such as analysis of the effect of numerical noise on the slow proton emittance increase in strong-strong simulations and simulations of beam-beam effects in crab crossing collisions, were also done.

The results of the beam-beam simulations are summarized in a draft of the beam-beam interaction section of the eRHIC Pre-Conceptual Design Report.

High-powered Erbium-doped Fiber Laser for 50 mA Highly Polarized Electron Beam

LDRD Project # 17-002
Z. Zhao

PURPOSE:

The objective of this project is to design high-power laser systems and generate output pulses at 780 nm for a polarized DC gun. An Erbium (Er)-doped fiber oscillator, ultrafast switch, high-power photonic crystal fiber amplifiers, and frequency doubling modules will be built to meet this specific goal. The laser system will be used as a driver laser for the existing polarized DC gun and will be integrated into eRHIC in the future.

APPROACH:

In the past two decades, scaling of the average power in fiber laser sources has been significant, but the focus has been largely put on the development of ytterbium-doped fiber laser systems in which the operation wavelength is around 1040 nm. Recently, development of large-diameter-mode-area Er-doped fibers and high-power single-mode diodes at 1480 nm enable the opportunity to achieve high-power Er-doped fiber amplifiers and produce optical pulses at 1560 nm. The project will take advantage of the state-of-the-art fiber and diode technologies and develop a high-power Er-doped fiber laser system. Furthermore, frequency doubling is carried out to produce optical pulses at 780 nm for the polarized DC gun.

Specifically, the laser system will be designed to meet a few key specifications, including (1) 9.3 MHz repetition rate; (2) ultrafast 1x8 switch; (3) 1 ns Gaussian temporal profile; (4) 10 ps Root Mean Square (RMS) time jitter with an external Radio Frequency (RF) clock; (5) eight 10 W average power at 780 nm; (6) high pulse contrast ratio (10^5) for minimizing the longitudinal halo; (7) low intensity fluctuation (10^{-3}); and (8) high-quality spatial beam profile.

In the oscillator, an active-mode-locking Er-doped fiber laser is designed to produce optical pulses with 9.3 MHz repetition rate and 1 ns duration. The wavelength is centered at 1560 nm. The laser is actively modulated and locked to an external RF clock and the RMS time jitter is less than 10 ps. The gain factor and the modulation depth are balanced to vary the pulse duration. The laser pulse contrast ratio between the laser pulse peak intensity and any pre-pulse or pedestal will be optimized to over 50 dB. The pulse train is separated into eight channels through a three-stage ultrafast fiber electro-optic modulator. In each channel, optical pulses are first amplified in preamp stages, and then coupled into a main Er-doped fiber amplifier to generate an average power of eight 20 W at 1560 nm. Through effective frequency doubling in a periodically poled lithium niobate or a bulk crystal, an average power of 10 W at 780 nm will be produced. Furthermore, a fast-feedback system using an electro-optic modulator (EOM) will be developed to suppress the intensity fluctuation.

TECHNICAL PROGRESS AND RESULTS:

The project started in May 2017; a lot of work was conducted to advance the objective of the first year. Parts including pump diodes, electrical devices, gain fibers, optical elements and frequency doubling crystals were ordered and assembled. A preamp Er-doped fiber amplifier was built to generate an average power of 5 W at 1560 nm. Frequency doubling modules using both a bulk lithium triborate crystal and a periodically poled lithium niobate were also installed. Effort is underway to achieve an active mode locking fiber oscillator (9.3 MHz repetition rate, 1 ns pulse

duration, and 1560 nm central wavelength) and high-power Er-doped fiber amplifier. The developments achieved so far were requested for use in the existing polarized DC gun. We will try to accommodate such developments, while LDRD development schedule is not impacted.

Milestones:
In FY 2018, we will complete three major goals. First, we will build an oscillator delivering pulses with 9.3 MHz repetition rate, 1 ns duration, and 10 ps RMS time jitter. Second, we will build a pulsed laser generating 10 W average power at 780 nm. Third, we will achieve an ultrafast 1x2 switch.

In FY 2019, we will complete three major goals. First, we will build multiple 780nm, 10 W average power lasers or single 780 nm, 40 W average power laser. Second, we will characterize the laser in terms of pulse contrast ratio, spatial beam profile, and intensity noise. A feedback system will be built to suppress the intensity fluctuation to 10-3. Third, a high-power laser system will be built and installed for polarized beam experiment.

Integrated Low-Noise and Low Drop-Out Voltage Regulator for Front-End Application-Specific Integrated Circuits

LDRD Project # 17-003
S. Li

PURPOSE:
Front-End Application-Specific Integrated Circuits (FE-ASICs) have become essential to enable high-resolution, large-scale modern radiation detectors, especially in extreme environments (high radiation, cryogenic operation, long lifetime, etc.). A critical circuit device, currently part of the ancillary discrete components surrounding FE-ASICs, is the low-noise, Low Drop-Out (LDO) voltage regulator. The purpose of this work is to develop integrated low-noise LDO regulators to match our FE-ASICs and to operate both at room temperature and in cryogenic environments with a long lifetime. The regulator will consist of a low-noise Band-Gap Reference (BGR) circuit, an LDO pass device, a low-noise differential amplifier in a high-gain control loop; it will provide a low-noise regulated voltage at 1.2 V or 1.8 V with a drop-out voltage below 300 mV for low power requirements. The applications will extend to several programs in Nuclear and Particle Physics, Synchrotron Light Sources, and National Security. There will be an immediate impact on large scale Liquid Argon Time Projection Chambers (e.g. Deep Underground Neutrino Experiment [DUNE]), large scale detectors for $0\nu\,\beta\beta$ decay (e.g. next-generation Enriched Xenon Observatory), dark matter detectors, and X- and Gamma-ray imagers needed at National Synchrotron Light Source II and in National Security.

APPROACH:
FE-ASICs are characterized by their small physical size, optimized low-noise amplification, low power dissipation, high functionality, extensive programmability, and the possibility of design for radiation tolerance, cryogenic operation, and long lifetime. In the past, commercial voltage regulators were carefully selected and adopted for our FE-ASICs. Among the stringent requirements of such regulators are a very low-noise and a low drop-out voltage (voltage difference between the input and to output), the former critical for the detector resolution, the latter for the power dissipation.

Due to their general-purpose nature, commercial regulators are designed to satisfy many requirements at the same time, including high load and line regulation and fast transient response, some of which are not of primary importance for our FE-ASICs. As a result, commercial regulators are subject to trade-offs on noise, power, and drop-out voltage for requirements not needed for our applications. Our proposal consists of developing low-noise, low drop-out voltage regulators in selected Complementary metal–oxide–semiconductor (CMOS) technologies to match our next generation FE-ASICs. Our proven knowledge on designing low-noise low-power cryogenic-operating ASICs will greatly benefit the design of the proposed application-specific low-noise regulator.

In phase I, the technical requirements of the regulator to fulfill immediate needs of the FE-ASIC for DUNE and the noise parameters of the most critical blocks in CMOS technologies are studied; the first prototype is developed to match the FE-ASIC in DUNE; the noise performance will be characterized in a bench-top test. In phase II, prototypes will be developed and integrated with our state-of-the-art low-noise FE-ASICs.

Our collaborators include: Wenbin Hou (Ph.D. student from Stony Brook University [SBU]), Gianluigi De Geronimo (Adjunct Professor at SBU), Veljko Redeka (Senior Scientist at BNL Instrumentation), and Emre Salman (Professor at SBU).

TECHNICAL PROGRESS AND RESULTS:

During FY 2017, issues with commercial regulators in the µBooNE experiment were investigated; state-of-the-art LDO regulators architectures in the literature and in industry were studied; technical requirements of the low-noise low-dropout integrated regulator to support DUNE FE-ASIC were specified; noise parameters of two target technologies, 180nm and 65nm, were compared and analyzed; and a first prototype of the low-noise low-dropout integrated regulator core was designed and simulated for optimized performance.

In FY 2018, a low-noise Band-Gap Reference circuit for the regulator core was designed and simulated; physical layout of the first prototype is being developed for fabrication in 2018.

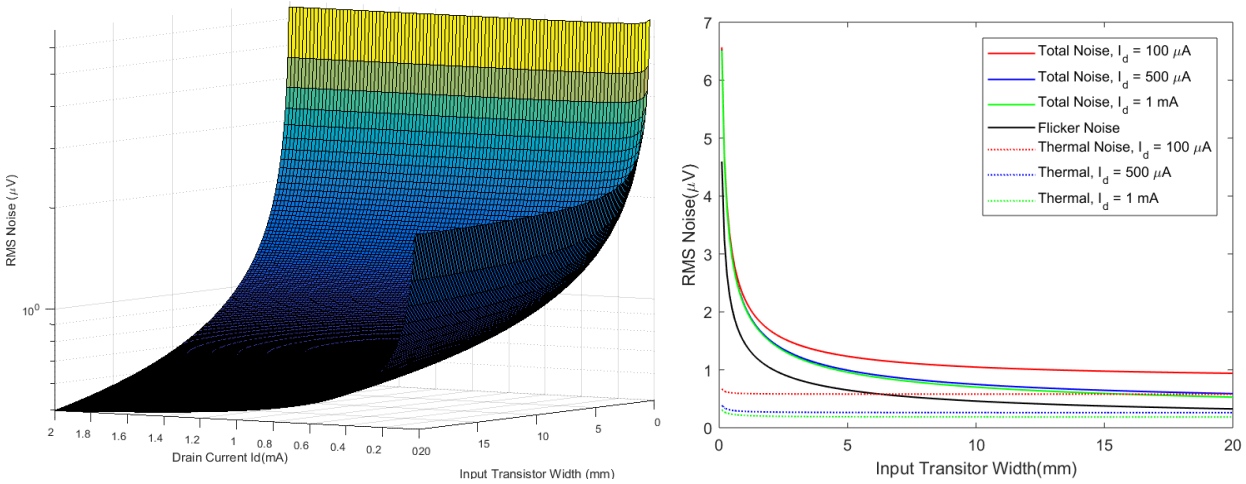


Figure 1: Noise optimization of the key transistor in LDO core.

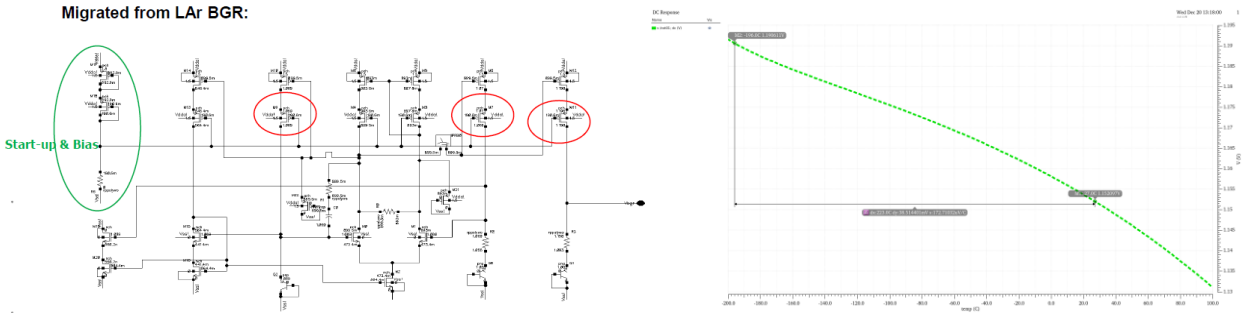


Figure 2: (a) BGR circuit adopted from Liquid Argon (LAr) FE-ASIC, and (b) temperature effect on transistor operation region.

Milestones:

FY 2018

- Fabrication of the first LDO regulator prototype
- Bench-top characterization of the first LDO regulator prototype
- Design revision upon characterization results of the first prototype
- Fabrications of the revised LDO regulator prototype

FY 2019

- Characterization of the revised LDO regulator prototype
- Design LDO regulator prototypes integrated with selected FE-ASICs
- Fabrication of integrated LDO regulator with FE-ASICs
- Characterization of integrated LDO regulator with FE-ASICs

Next Generation Pad Readout for Neutron Detectors

LDRD Project # 17-004
B. Yu. N. Schaknowski

PURPOSE:

The concept of operating ^{3}He-filled neutron detectors in ionization mode (no gas gain), with anode pad readout, has been developed at BNL with significant success. To date only symmetrically shaped 5x5mm sensing pads have been utilized. A number of detectors using this approach have been fabricated for neutron user facilities and for Strategic Partnership Projects (SPPs), with interest and demand from user facilities and agencies progressively increasing over the last few years. The resolution performance of these detectors is adequate for a number of experimental techniques at neutron user facilities, for example Small Angle Neutron Scattering, but generally not good enough for other techniques such as macromolecular neutron diffraction, powder diffraction, and reflectometry. The technical goal of this project is to evaluate the practical performance limits of this innovative pad readout method for ^{3}He based neutron detectors. Exploring the performance of rectangular anode pad geometry will help to determine the practical position resolution that this technology is capable of providing. This work can ultimately result in the development of highly stable neutron detectors which allow researchers to gather more accurate information using shorter lengths of valuable beam time while reducing the need for detector service. This accomplishment will increase the competitiveness of BNL in attracting requests for advanced neutron detectors, especially for DOE User Facilities and SPPs.

APPROACH:

The development of gas-based detectors has for many years been based on anode structures around which it is possible to develop electric fields in excess of 10kV per cm, high enough to initiate electron multiplication. The gas gain achievable can be as much as 10^{4} and hence signal to noise is improved. Unfortunately there are drawbacks associated with high levels of gas gain. Aging effects associated with gas gain can produce localized areas of low efficiency and rate capability can be limited. Our goal is to enable a new generation of highly efficient position sensitive neutron detectors with improved position resolution, capable of recording incident fluxes that are higher than ever before achieved without the limitations associated with other detectors technologies.

The scope of this study is twofold. Improving the resolution capability of a pad detector requires the study of the performance of a detector with reduced pad size. This is coupled with the study of detector performance with different stopping gasses at different partial pressures. Dr. Graham Smith (BNL) and Dr. Gerry Doumas of the U.S. Merchant Marine Academy are significant collaborators on the gas mixture studies.

We designed a test detector with rectangular pad geometry where individual pads are 2.5x10mm. Pad detectors are essentially a printed circuit board (PCB) with the sensing anode pads on one side routed through to the opposite side of the board where the signal is coupled to the input of an Application Specific Integrated Circuit (ASIC). The ASIC and PCB design and fabrication are quite complex and demanding. We are able to use the ASIC we developed for the 5x5mm pad detectors, but the PCB requires a redesign. The design, fabrication and assembly of appropriate pressure vessel and readout electronics and software is also required. Test setups

where the pad board can be tested electrically and with an alpha source are required. Neutron testing with a collimated beam will provide the final test of performance.

TECHNICAL PROGRESS AND RESULTS:

We have designed and fabricated the printed circuit board (PCB) with individual pad size of 2.5mm x 10mm. The board can accommodate 36 ASICs. We have fabricated a full size (24x24cm) pad board so that if successful, we will have a useful working detector. Each ASIC has 64 channels routed to 16 pads (2.5mm) in one dimension and 4 pads (10mm) in the other dimension forming an area of coverage which is 4cm square.

We have populated the central location of this board with four ASICs and other components. This gives an active area of 8x8cm square.

We have fabricated a test fixture for electrical testing of the board. We have adapted LabView based software for the readout. We have adapted a readout board for use in the electronic test fixture.

We successfully completed electronic testing of the new rectangular pad board. All channels are alive and respond appropriately to an input pulse. Tests of baseline noise, crosstalk gain uniformity were completed. This verified that the PCB layout and fabrication are as intended.

We have constructed a gas tight enclosure and an aluminized Mylar entrance window electrode for assembly onto the electronic test enclosure to allow testing with an alpha emitter source.

Work has begun on adapting an existing pressure vessel to accommodate the new pad board for neutron testing

Implications for Future Work

The design and layout of this pad board is complex and was a concern prior to commencing work. Essentially the PCB is the detector, so it was critical to confirm that we could design and build a pad board with greater density capable of producing better position resolution. .The success of our testing proves that the new pad board will provide a suitable step toward achieving improved position resolution.

Milestones:

FY 2018:

- Test pad board in test setup with alpha source to characterize the performance of the complete signal chain from pad through the readout electronics.
- Install detector in pressure vessel with appropriate readout and verify performance with neutrons.

FY 2019:

- Experiment with stopping gasses. Determine if C_3F_8 can provide suitable resolution at lower partial pressure than CF_4.
- Conduct collimated beam measurements of resolution and rate capability studies with neutrons at the High Intensity Flux Reactor at Oak Ridge National Laboratory.

Investigation of Novel Materials for Generating Polarized Electron Beams

LDRD # 17-005

T. Rao, P. Johnson

PURPOSE:

In this program, we plan to investigate the properties of Topological Insulators (TI), such as Bi_2Se_3 and Bi_2Te_3 to determine if TIs rather than GaAs can be a suitable alternative to deliver polarized electrons. We will determine their Quantum Efficiency (QE) and work function of in-vacuum cleaved crystals followed by deposition of alkali metal to the surface. We will examine the electron emission properties of the TI, such as the angular and energy spread of the emitted electrons, the surface charge limit, life time, and their degree of polarization.

APPROACH:

We intend to modify one of our existing ultrahigh vacuum systems to study the QE of several TI materials. We will lower their work function and determine their spectral response after exposing them to a thin layer of alkali metal onto their surface. We will then measure the emitted charge as a function of optical power, repetition rate and extraction field to determine the surface charge limit and life-time. Finally, we will build a Mott's polarimeter and measure the degree of polarization of the emitted electrons.

TECHNICAL PROGRESS AND RESULTS:

We have modified one of our existing ultra high vacuum (UHV) systems. Additional pumping sections, vacuum components, and gauges have been added to improve the vacuum to 10^{-12} torr, a necessary vacuum environment for TI samples to work properly. A new cathode mounting assembly has been designed and fabricated. An optical fiber delivery system with limited motion capability is designed and assembled for photon illumination onto a typical ~3x3 mm^2 area of the TI sample, see Fig. 1.

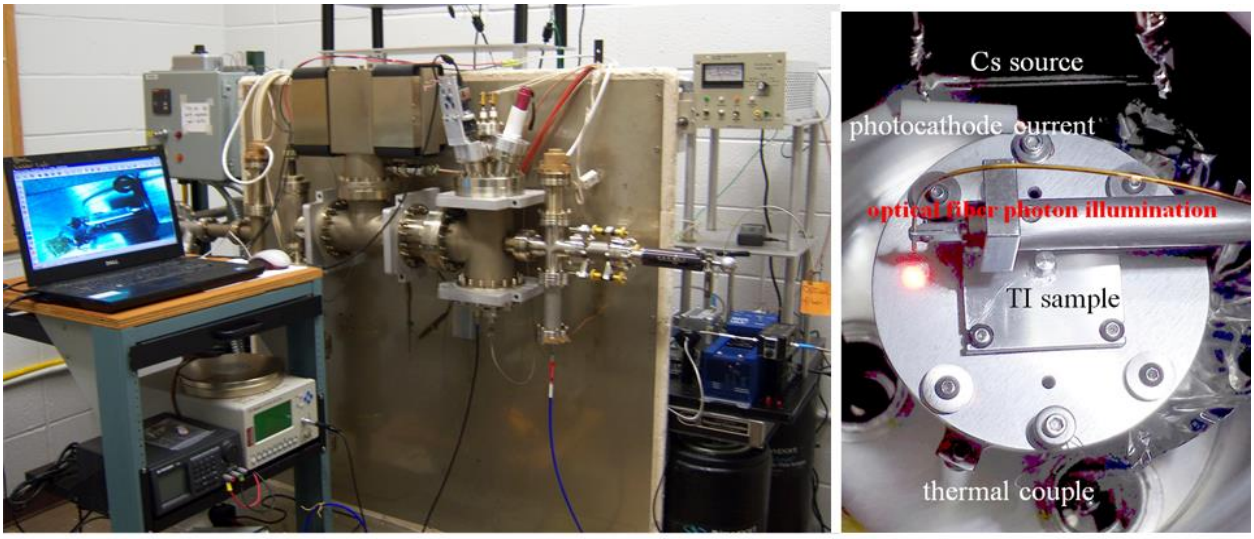


Figure 1. Left: UHV vacuum system, right: TI sample and photon illumination arrangement.

For photon illumination, we employed a laser triggered plasma cw white light source spanning from the vacuum ultraviolet (UV) to the visible range. Photons are fiber-coupled to a

monochromator and fiber delivered in a 10 nm bandwidth to a 600 μm diameter in-vacuum polarization resistance optical feedthrough, followed by a 30 cm long in-vacuum UV fiber, ended directly ~3 mm away from the surface of TI sample. Photocurrent leaving the TI photocathode is measured on a picoammeter and optical power is measured on a calibrated power meter. Quantum Efficiency is defined as:

$$QE = \frac{photocurrent\ (Amp)}{optical\ power\ (Watt)} \times\ photon\ energy\ (eV)$$

At the vacuum level of $2x10^{-11}$ Torr, we performed in-vacuum cleaving of one or several TI samples at once. We then deposited a ~nm thick cesium layer onto the surface of the TI sample to lower the work function, thus enhancing the electron emission process. We monitored the photocurrent of the TI using a fiber-coupled 268 nm UV-Light Emitting Diode before, during, and after cesiation. Fig. 2(a) summarizes the QE of three in-vacuum cleaved TI samples: Bi_2Se_3, BiSb(TeSe), and Bi_2Te_3, and two noble metal reference photocathodes: Au and Ag. They are mounted on the same flat-form substrate and measurements are performed the same day.

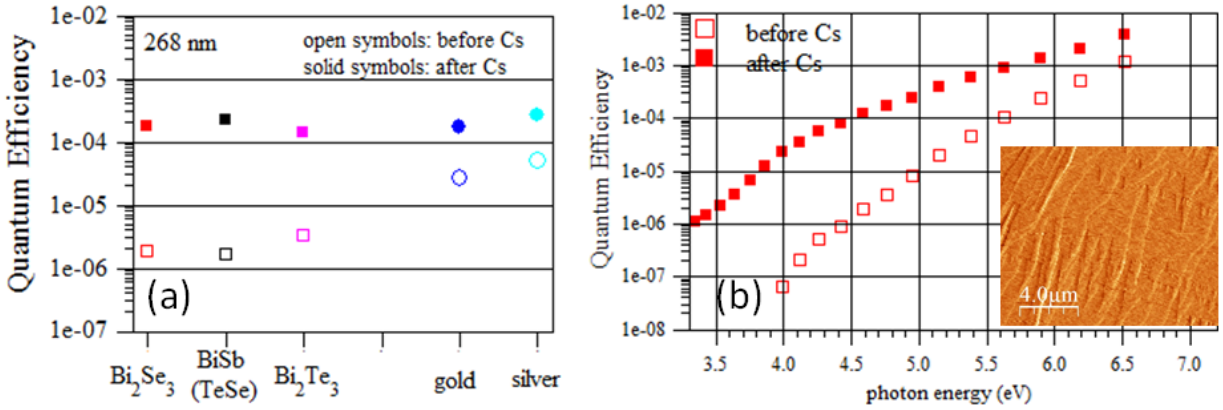


Figure 2. (a) QE of sereral TI samples before and after cesiation, samples of noble metals are also shown for comparison. (b) Spectral response of a typical Bi_2Se_3, inset: AFM image of an as-cleaved Bi_2Se_3 sample indicating the typical smoothness of the TI sample surface.

At a wavelength of 268 nm, the QEs of freshly in-vacuum cleaved TI samples are all in $\sim10^{-6}$; however, the QEs of air-cleaved TI samples are markedly higher (cleaved in air prior to installation in the vacuum chamber, data are not shown here). After cesiation, the QE of all TI samples reached $\sim10^{-4}$. Unlike noble metal photocathodes, where the QE improves only by a factor of ~10 after cesiation, the QE of TI photocathodes evidently improved by a factor of ~100, see Figure 2. The spectral dependence of QE on all TI samples has similar characteristic and is shown representatively in Fig. 2(b). We note that after cesiation, QE improvement is substantially higher at lower (<5eV) photon energy and it plateaus at higher photon energy (>6.5eV); however, the maximum QE is below 1% even at the highest photon energy of 6.5 eV.

Milestones:
FY 2018: Measure the degree of polarization of emitted electrons using a Mott's polarimeter.

Engineered Protein Arrays for Structural and In-Operando Studies

LDRD Project # 17-011

O. Gang, Q. Liu, E. Stach, H Xin, S. McSweeney

PURPOSE:

Revealing the 3D structure of proteins is transforming our understanding of the biological complexity across the entire spectrum of biology and providing the structural basis for discovering and developing strategies for treatment of diseases, including cancers. Single-particle cryo-Electron Microscopy (Cryo-EM) and crystallography are instrumental in such structure characterization; however, difficulties in making suitable single particles or crystalline samples and obtaining adequate signal-to-noise levels are major bottlenecks in revealing the structures of many challenging proteins using traditional approaches. Our overall objective is to develop the DNA-based protein assembly methodology to accelerate structure analyses by cryo-EM and crystallography. The proposed approach makes use of DNA origami nanocages to capture and to orient proteins as single particles for cryo-EM and as 2D and 3D arrays for crystallography.

APPROACH:

To enable DNA-based protein assembly in arrays, we develop two major approaches required for the fabrication of such systems:

- Caging proteins in DNA frames. We establish the methods for conjugating proteins with polyhedral DNA cages using covalent conjugation of proteins with modified DNA staples. We develop a set of origami cages to match protein properties, including their coating, sizes, and shapes.
- DNA-frames and their crystallization. We will innovate the methods of making well diffracting DNA cages (2D and 3D) that will be used to organize proteins. We will develop and optimize methods for individual steps from sample preparation and for characterization and analysis using X-ray/electron beams. We will innovate the methods that will combine X-ray and electron data for correlated studies.

TECHNICAL PROGRESS AND RESULTS:

We have focused our efforts in creating protein arrays on the FtsH family proteins, which are important in repairing the damaged photosystem II during conversion of light into bioenergy. FtsH is a class of membrane protein which has both hydrolysis and ATPase functionalities. Therefore, understanding the structure and function of FtsH is key to engineering plants for improved energy capture and conversion. However, the protein tends to aggregate after production and purification and is recalcitrant to structural determination by cryo-EM. One aim of this project is to develop a novel biotechnology for protein organization in caged particles for structure determination by cryo-EM. The technology will revolutionize the current practice in cryo-EM sample preparation: isolated and shielded by DNA nanocages, proteins are always preserved in their native states during the vitrification processes, thus allowing for sample preparation with minimal distortion and dehydration. We have produced FtsH and incorporated it in DNA origami frames by using an enzyme-catalyzed covalent conjugation reaction. We are currently using the Center for Functional Nanomaterials microscopes to optimize the ratio between DNA and protein for optimized protein incorporation. By encaging FtsH within DNA frames, the contrast of particles will be enhanced, which will facilitate various steps in 3D image reconstruction, including motion correction, particle picking, 2D/3D class averages and high-

resolution refinement. Protein structures enabled by this technology will provide fundamental knowledge, which will lead to the development of structure-inspired biosystems for renewable bioenergy.

Through a combination of DNA origami design and expressed protein ligation, we aim to covalently encapsulate and orient single proteins in individual frames. Our preliminary experiments show the encapsulation of AAA+ protease FtSH (*E. Coli*) by incorporating copper-free click chemistry and expressed protein ligation (EPL) to ensure oriented bioconjugation at the C-terminal protein domain (Figure 1). In principal, this allows one protein in each frame based on the linker length and size of the protein (Mw = 426 kDa, ~15 nm in diameter). This system has been characterized primarily by agarose gel electrophoresis and negatively-stained Transmission Electron Microscopy (TEM) imaging. To achieve single domain, sub-nanometer resolution of individual encapsulated proteins, electron tomography (ET) in a cryogenic stage will be a powerful tool to analyze the system. In particular, we will use the individual-particle ET (IPET) technique, which allows direct determination of high-resolution structure of a single protein using intermediate resolution imaging (<1-2 nm).

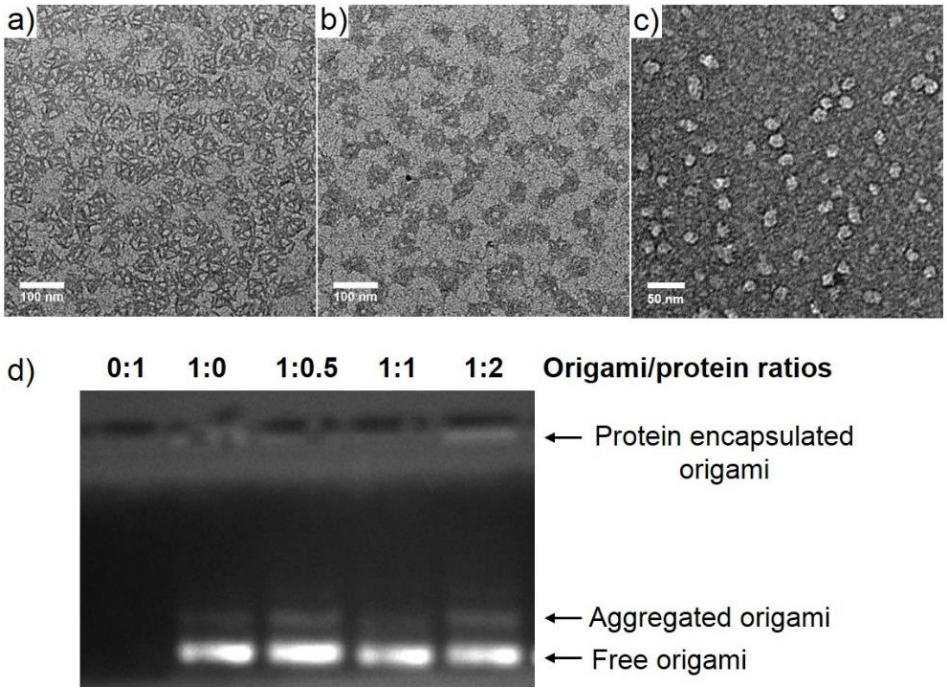


Figure 1: Negatively-stained TEM images showing (a) DNA origami only, (b) DNA origami with AAA+ protease FtSH encapsulation, and (c) protein in solution (obtained by Liu's group). (d) The protein encapsulation is also confirmed by agarose gel electrophoresis. Compared to the free origami frames, the presence of proteins reduces the mobility of the origami. Further optimization of the conjugation and sample purification will be performed prior to cryo-TEM tomography imaging.

Milestones:

- Arranging protein particles into DNA frames and optimizing frame design, conjugation and stabilization for high-yield structure formation
- Assembling 2D and 3D protein arrays and performing characterization by X-ray scattering; developing relevant experimental and data analysis procedures
- Establishing and applying the single-particle cryo-EM capability for systems using the environmental Titan microscope.

National Synchrotron Light Source II High Brightness Upgrade Design Studies

LDRD Project # 17-015
E. Blum

PURPOSE:
The goal of this project is to study how the brightness of the National Synchrotron Light Source II (NSLS-II) synchrotron radiation source can be improved by reducing the horizontal emittance of the NSLS-II storage ring without major modifications to the storage ring components. The project was planned to explore the following avenues to achieve this goal: storage ring lattice modifications, off-energy ring operation, and damping wiggler modifications. Increasing the NSLS-II brightness will allow the facility to remain competitive as a radiation source as fourth generation synchrotron light sources go into operation.

APPROACH:
The original design for NSLS-II was a fairly conventional third generation storage ring with a double bend achromat lattice with damping wigglers. This study will explore changes to the machine lattice and other modifications that improve the brightness of the x-ray beam by reducing the beam emittance. We hope to reduce the emittance up to three-fold without replacing any of the major storage ring magnets.

The storage ring emittance is determined by the formula

$$\epsilon = C_q \gamma^2 \mathcal{H} / J_x$$

where C_q = 3.84x10^{13}, m is a constant, γ is the Lorentz factor for electrons in the storage ring, $\boldsymbol{\mathcal{H}}$ is a function of the storage ring lattice, and J_x is the horizontal damping partition number. For a given electron energy, the emittance can be reduced by reducing (a) or increasing J_x (b). In this project we examine the possibility of doing both.

(a) The function $\boldsymbol{\mathcal{H}}$ is an integral over the dispersion of the electron beam in the ring dipoles. By reducing the dispersion, the emittance can be reduced. The theoretical minimum emittance for a double bend achromat lattice with NSLS-II geometry is 0.92 nm-rad compared to the 2.1 nm-rad emittance of the NSLS-II lattice without damping wigglers. The theoretical minimum emittance is never realized in practice for several reasons, including that the required quadrupole and sextupole strength would be much higher than can be obtained from practical magnets if the dynamic aperture becomes too small. This study will explore the minimum emittance that can be achieved in NSLS-II with the existing storage ring magnets.

(b) The damping partition number $J_x \approx 1$ for a storage ring like NSLS-II built with pure dipole magnets with no gradient in their magnetic field. It can be significantly greater than 1 if there is a gradient but replacing the dipoles can be expensive. Alternatively, a dipole magnetic field with gradient components can be introduced into the lattice by operating the ring off its nominal energy by mismatching the RF frequency, which forces the electrons onto an off-momentum orbit. This will make the electrons pass through the quadrupoles off-center in the dispersive region where they will see a dipole component in the magnetic field. We will optimize the lattice

taking into account the off-axis passage of the beams through the dipoles, quadrupoles, and sextupoles.

We will develop a lattice with the smallest practical value of $\mathcal{H}$ and the largest possible J_x. We will test the lattice in the NSLS-II storage ring to compare its performance with prediction, and to determine that the lattice will be practical in regular operation. Initial testing will be conducted with a small electron beam current. After we demonstrate through ray tracing that the storage ring vacuum chamber will not be harmed by synchrotron radiation from the off-axis electron orbit, we will test the lattice at standard operating electron beam current and demonstrate predicted emittance scaling with current.

Lattice development was carried out in collaboration with Weiming Guo. Lattice testing was performed with Weiming Guo, Joshi Hidaka, Xi Yang, and Victor Smalyuk. A preliminary ray tracing of the synchrotron radiation from the off-axis electron orbit was performed by Sushil Sharma and Cynthia Longo.

TECHNICAL PROGRESS AND RESULTS:

Funding was awarded for this project and work began in January 2017. We first studied the minimum emittance that can be realistically obtained in the NSLS-II storage ring. Although the theoretical minimum emittance of the NSLS-II lattice is 0.92 nm-rad (with $J_x \approx 1$), our analysis shows that in practice the emittance cannot be reduced to much less than 1.5 nm-rad with the existing quadrupole magnets.

We then studied off-energy operation to increase J_x. Decreasing the RF frequency by 1.4% drives the electrons onto an orbit with a maximum displacement of 6 mm from the nominal axis at points of maximum dispersion in the arcs resulting in $J_x = 1.4$. To match the 3.0 GeV injector energy, we increase the storage ring dipole current by 6 A. By optimizing the quadrupole strengths to reduce the $\mathcal{H}$ function, we created a lattice with an emittance of 1 nm-rad, a 50% reduction from normal operating conditions. We tested this lattice in the storage ring with a 5 mA electron beam and measured a 1.1 nm-rad beam emittance. The measured lattice functions do not fully agree with the design and injection efficiency is only 70%, but we expect to correct these problems during FY 2018.

We hope to reduce the emittance as low as 0.65 nm-rad with $J_x = 2.0$ by increasing the orbit displacement to 9 mm. Since we have been able to increase J_x to this value with RF frequency shift alone, we did not study damping wiggler modifications.

All the lattice testing to date has been limited to 5 mA to preclude any chance of damaging the vacuum chamber by synchrotron radiation from the electrons on the displaced orbit.

Milestones:

During FY 2018, we will complete ray tracing of the synchrotron radiation and hope to test the lattice at full operating current. Preliminary analysis shows that the three pole wiggler beamlines must be shifted by 0.5 mrad before the lattice can be used in operation. During FY 2019, we will estimate the cost of realigning these beamlines. We will also analyze the safety of top-off injection with this lattice.

Diffraction Limited and Wavefront Preserving Reflective Optics Development

LDRD Project # 17-016
M. Idir, L. Huang, N. Bouet

PURPOSE:

There is a worldwide interest in upgrading existing and constructing new light sources, such as the National Synchrotron Light Source II (NSLS-II) and the Advanced Photon Source upgrade, where the electron beam emittance approaches the diffraction limit, even for 1 Angstrom X-rays. However, our capacity to utilize these ultrabright sources is still limited by our ability to focus, monochromatize, and manipulate these beams with adequate X-ray optics. Many of the unique scientific opportunities at the nanoscale afforded by diffraction-limited X-ray sources, such as coherent X-ray imaging, nanodiffraction, and nanospectroscopy, require maximum intensity in the focus and/or extreme attention to wavefront distortion. These applications require mirrors with ≤0.5 nm rms figure height errors (ideally ~0.3 nm rms) for surface error wavelengths, ranging from 100 microns to the full optical aperture of the optic, which can approach 1 m for hard X-ray optics. Our project aims to complete the development of ion-directed profiled reactive etching for fabrication of X-ray mirrors that are capable of reaching diffraction-limited focusing and wavefront preservation. This capability is essential for realizing the potential of ultra-high brilliance sources such as NSLS-Il and will foster the development of the next generation of advanced synchrotron instrumentation and science.

APPROACH:

Our project focuses on the development of a complete technological process for fabrication of precise X-ray and vacuum ultraviolet mirrors with high sag (small radius of curvature). The process of surface figuring, based on conventional chemical-mechanical polishing, cannot provide sub nm figure errors, as plastic deformation and brittle fracture due to abrasive-driven deformation lead to unavoidable multiplication of defects and dislocations. Computer-controlled plasma chemical vaporization machining and elastic emission machining were developed within the framework of the Center for Atomistic Fabrication Technology (Japan) and enable the figuring of mirror surfaces to a peak-to-valley accuracy better that 1 nm and a lateral resolution close to 0.1mm. Although state-of-the-art mirrors can be produced by these processes, several interactive steps are required before final 1 nm performance is met because of the slow erosion rate. Such crosschecks are time consuming and costly, and are the source of additional errors, as tolerances of alignment accuracy, environmental stability, and stress relief are very demanding. This project will develop a polishing process with significant higher removal rate, whose industrial impact is large and reduces the number of figuring-metrology iterations.

After analysis of alternative routes, we are developing a mirror fabrication method combining high material removal rates, while keeping surface roughness at the atomic level. The technique merges ion erosion, chemically assisted etching, and surface profiling coupled with state-of-the-art optical metrology.

TECHNICAL PROGRESS AND RESULTS:

We have started the design of a dedicated and optimized Ion Beam Figuring (IBF) plant (in collaboration with Yi Zhu). IBF is a well-known technique, which uses low (<2000eV) energy

argon ions for erosion and mostly relies on the kinetic impact of impinged ions on a surface layer. Fig. 1 shows some images of the future IBF station.

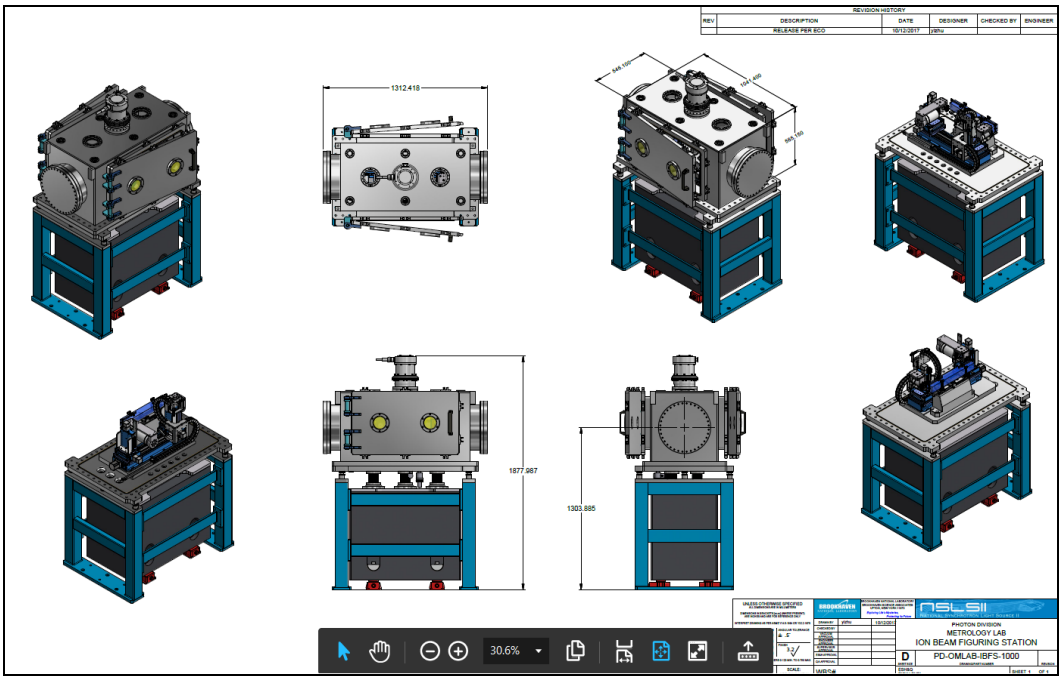


Figure 1: Some images of the future IBF station.

In parallel, we have also worked on dedicated software to perform all the necessary steps from metrology map to dwell time calculation as illustrated in Fig. 2.

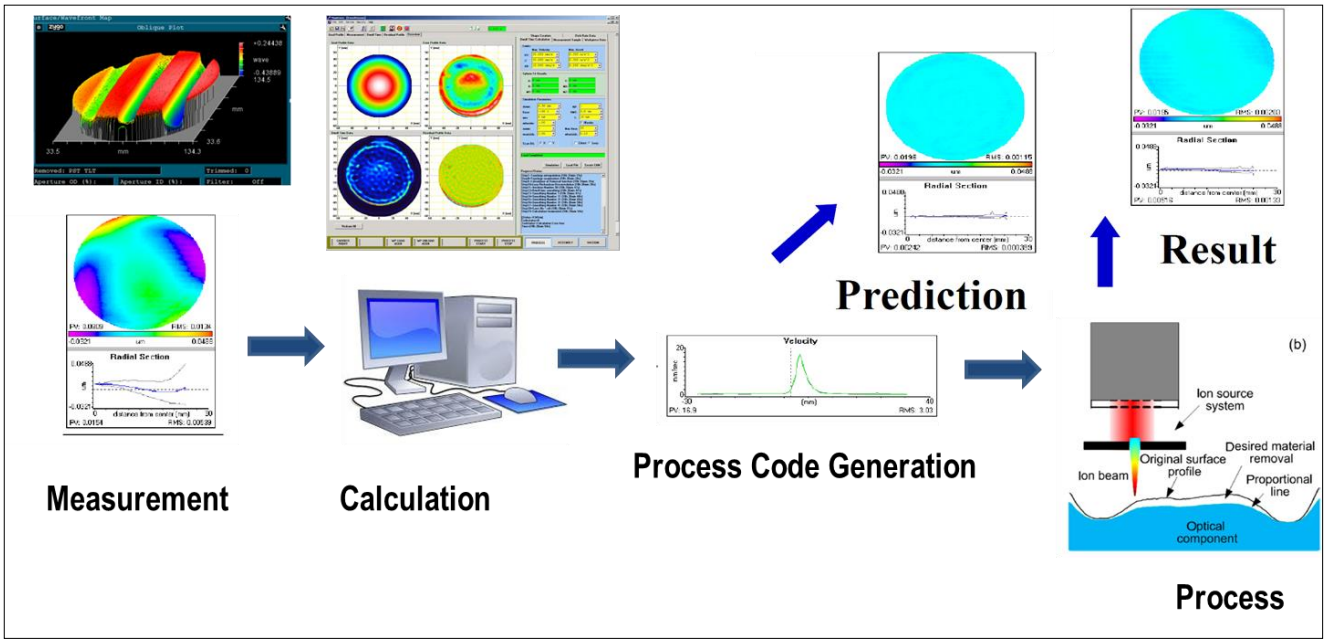


Figure 2: Schematic of Dedicated Software for IBF.

Milestones:
FY 2018: Build the dedicated Ion Beam figuring Plant, install and conduct the first test.

Development of Compact, High Efficiency Nanofocusing Optics for Hard X-ray Nano-imaging

LDRD Project # 17-017
E. Nazaretski

PURPOSE:

X-ray microscopy is a route to achieve real space nanometer-scale imaging with micron-scale penetration depth. To achieve down to 10 nm focal spot size, Multilayer Laue Lenses (MLL) are utilized at the Hard X-ray Nanoprobe (HXN) beamline at the National Synchrotron Light Source II (NSLS-II). Alignment of two linear MLLs is extremely complex (required to achieve point-focus); therefore to push the imaging resolution further and elucidate phenomena with characteristic length scales of 5 nm and below novel approaches are required. As a first step towards direct sub-5 nm imaging, we are developing a novel method suitable for alignment and bonding of two linear MLL optics together through microfabrication of a silicon (Si) alignment template. This approach greatly reduces complexity of an MLL microscope yielding higher stability and ultimately better spatial resolution.

APPROACH:

Non-invasive hard X-ray imaging with resolution of 10 nm and below is critical for many areas of science and technology. To achieve down to 10 nm focal spot size MLLs are deployed at the HXN beamline at the NSLS-II. Alignment of two linear MLLs is extremely complex (requires 8 degrees of motion: five translations and three rotations; all must be stable down to a single nanometer level). In order to push the imaging resolution further and elucidate phenomena with characteristic length scales of 5 nm and below, novel approaches are required i.e. fabrication of monolithic, pre-aligned 2D MLL structures. This approach overcomes limitations posed by conventional piezo-mechanical systems (large form factor impeding alignment of nanofocusing optics, thermal drifts, resolution, and repeatability). It also scales down the size of a microscope itself, providing a route for imaging experiments with spatial resolution of 5 nm and below. Moreover, 2D monolithic MLL optics can be used not only in scanning X-ray microscopy applications, but may also be adopted by full field X-ray imaging systems.

We utilize microfabrication technology to fabricate a Si template to hold two linear MLLs together as a monolithic 2D optics in a pre-aligned configuration. The concept of a Si template is shown in Fig. 1a. It includes an aperture, aligning edges, and Si springs. Two aligning edges and springs are fabricated on both sides of a Si template with aligning edges orthogonal to each other. The mask aligner used in the microfabrication process provides adequate angular accuracy to satisfy 5-nm point focus requirements. During the packaging process, two linear MLL optics are aligned with respect to aligning edges and secured by Si springs. Their effective area (marked as yellow) overlaps at the aperture location. The separation between two lenses is adjusted as a part of the microfabrication process to ensure overlapping of two focal planes. The MLLs packed into a template will be characterized and used for measurements at NSLS-II. Design, fabrication, characterization and measurements of the monolithic 2D MLLs is a joint effort of the NSLS-II microscopy instrumentation group (Evgeny Nazaretski, Wei Xu, Weihe Xu), optics fabrication group (Nathalie Bouet, Abdiel Quetz, Juan Zhou), Center for Functional Nanomaterials (CFN) cleanroom (Ming Lu) and HXN beamline team (Yong S. Chu, Hanfei Yan, Xiaojing Huang).

TECHNICAL PROGRESS AND RESULTS:

In May 2017 Postdoctoral Research Associate, Wei Xu, joined the group. Since he arrived, we have designed Si templates at the R&D labs of NSLS-II (Fig. 1a). The design process included active interaction with the MLLs fabrication group and the HXN team to finalize all dimensions of MLLs and templates. Dimensions of the developed template are less than 1 cm x 1 cm. We have studied displacements/stress applied to the template during the packaging process of MLLs utilizing the Inventor software package. Finite Element Analysis revealed that the stress is less than the yield strength of Si (Fig. 1b), suggesting adequate strength of a template to withstand positioning and holding forces of two linear MLLs. We have derived the microfabrication process, which includes two steps of metal deposition, four steps of photolithography, and four steps of deep reactive ion etching (Fig. 1c). The fabrication process has been tested and executed at the CFN. Fig. 1d demonstrates a nearly complete, unreleased MLL template.

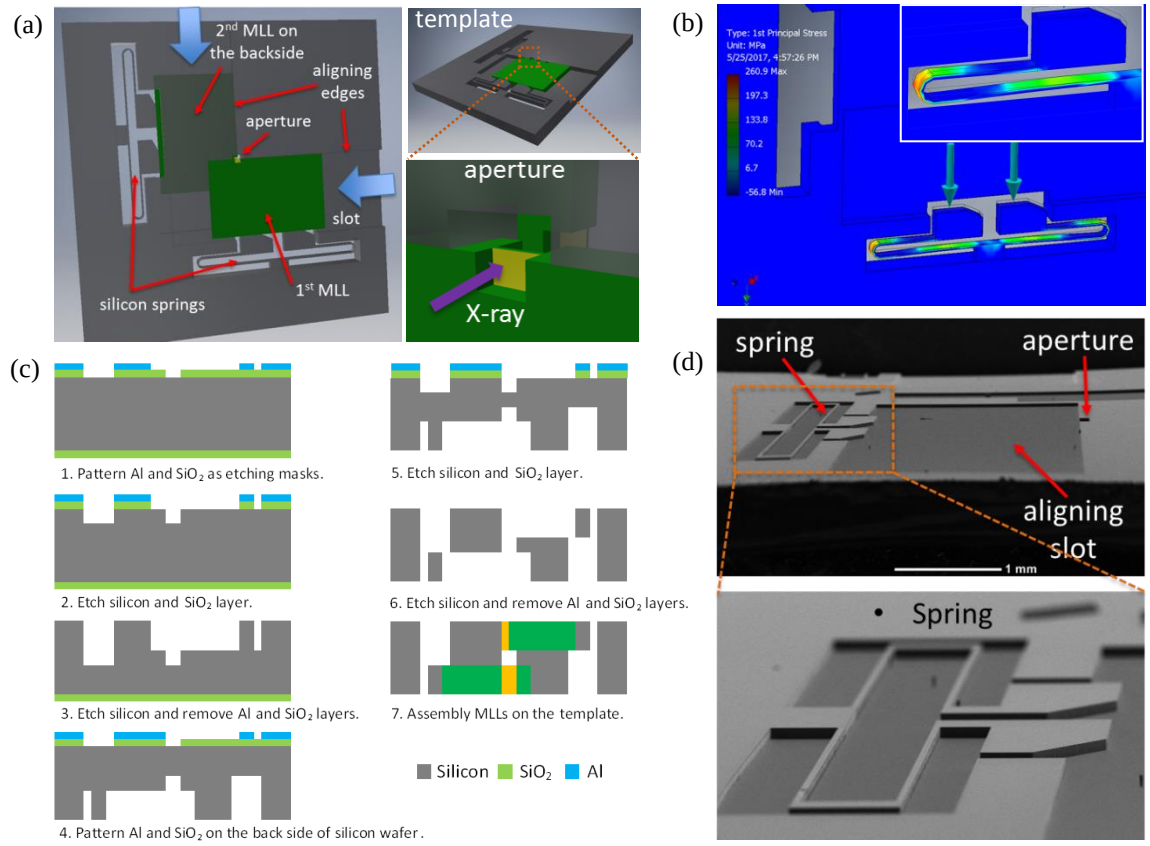


Figure 1: (a) Schematic of a microfabricated Si template used to align two linear MLLs. (b) Displacement and stress analysis of a Si template. (c) Step-by-step microfabrication process of a Si template. (d) Scanning Electron Microscope image of a partially etched Si template.

Milestones:

- In FY 2018, we will complete fabrication of the Si templates. Once fully released, replica-MLLs will be installed, and fabricated devices will be evaluated for strength and stability. The fabrication parameters will be adjusted to ensure best performance of the manufactured templates.
- When available, fabricated devices will be equipped with dedicated MLL lenses and will be tested for their focusing performance at the HXN beamline. A beam time proposal for the nanofocusing optics evaluation has been already submitted and approved.

Genomes to Predictive Biology: Machine Learning for the Integration of Inter-Species Functional Genomics Data

LDRD Project # 17-018
I. Blaby, S. Yoo

PURPOSE:
This project will establish a novel computational platform capable of extracting events (i.e. data, observations, hypotheses, etc.) from the scientific literature and large datasets. While our primary focus is biology, our approaches are foundational and will consequently be of direct relevance to all scientific disciplines. In the instance of biological analyses, the platform will be designed to propagate protein-specific data to conserved proteins within other organisms that may share biological function due to a common evolutionary ancestor. This will lead to a "hypothesis engine" which will enable biological experiments to become increasingly targeted, accelerating progress in all of biology. We will build a community platform and engage users by encouraging open access through publications and a web-based implementation. We foresee this project as providing key leadership in the field of natural language processing (NLP) at BNL.

APPROACH:
To accelerate gene functional discovery, significant paradigm shifts in life science research must take place. Ultimately, computing infrastructures can assist bench scientists in designing experiments that fill knowledge gaps in biological networks. The design and implementation of such an infrastructure is a large task. The data derived from biological experiments can be as complex as the organisms from which the data is collected: datasets and experimental results are multi-faceted, multi-dimensional, and originate from different sources (i.e. organisms), and interpretation often requires understanding and analyzing multiple fields of research.

The key objective of the proposed work is to design and implement a dynamic biological big data paradigm: prospecting literature for protein function evidence across kingdoms. A major input of high value data will be the scientific literature, which is the primary venue in biological research for dissemination of experiments, data, hypotheses, observations, conclusions and speculation. This project aims to extract information associated with specific genes, such as characteristics of their encoded proteins or the behavior (phenotypes) of mutant strains lacking that gene. We will exploit protein evolutionary relationships in order to integrate and contextualize inter- species designations. Although our focus is on biological research, the underlying principles are equally valid to other branches of science. We therefore foresee significant relevance in proposed activities beyond biology.

The scientific literature is a rich source of knowledge that is essential for contextualizing past, present and future data mining. Yet this vast resource, comprising the accumulation of > 2x107 peer reviewed articles exploring all aspects of biology, is difficult to mine in an automated fashion for several reasons. First, there is a need to interpret context in order to differentiate between data, conclusions and speculation. Because articles rely on natural language and not a standardized format, they are not ideally suited to machine learning. Second, with the exception of key words, no standards exist for article metadata/indexing. Present implementations of biological text mining fall short of contextualizing extracted events and instead rely upon string (keyword) searches or ontology (i.e. prediction, rather than fact) based constructs. Additionally, no automated inter-organism system exists for correlating observations made on a protein in one

organism to a related protein in another organism. This is critical, since protein function is often conserved among orthologously-related (genes derived from a common ancestor) proteins, meaning an experiment performed on a protein in one organism is relevant to results gained in another. For example, catalytic activity of a particular protein from a bacterium should be co-extracted with a phenotype associated with the deletion of the corresponding gene in a plant.

The task of mining functional information associated with specific proteins/genes from unstructured data (such as published articles) requires querying question-answering (QA) systems. In recent years, significant technological progress has been made in QA systems. Two implementations of QA systems are available: IBM's Watson and Stanford's DeepDive. The IBM Watson QA system is based on the Apache Unstructured Information Management Applications (UIMA) framework, which provides ecosystems of building blocks for the QA system. Stanford's DeepDive is based on Stanford CoreNLP, an NLP engine, which relies on tokenizer, Part Of Speech (POS) tagging, NER (Named Entity Recognition) tagging, and dependency parsers for NLP requirements. Although both of these systems are open source, an understanding of NLP and machine learning is essential to use such tools. Of note for this proposal is that DeepDive has already processed articles published in BioMed Central and Public Library of Science and has made their knowledge base available with open licenses. While we are starting with biological literature mining, we intend to generalize to encompass other science domains where the Department of Energy and BNL have interests, such as material science and physics. Therefore, we will still need to evaluate available open source options and customize the knowledge base to our needs. A second task is to contextualize multiple evidence extractions of protein function from a protein in one organism to a conserved protein in another organism. Multiple methods exist for this purpose, including phylogenetic relationships (i.e. orthology) and conservation rate. To aid progress with this task, we will leverage existing resources such as the Systems Biology Knowledgebase (KBase) homology service and computed orthologous relations via InParanoid. Additionally, we plan to develop a Bayesian inference model to aid with transfer of extractions between organisms. A third task is to address computational infrastructure needs. For this, we intend to leverage a portion of the BNL KBase site and submit a proposal to use the BNL Institutional Cluster.

TECHNICAL PROGRESS AND RESULTS:

After an extensive search, we made an offer to our top postdoctoral candidate, Dr. Carlos Soto who began work at BNL in January 2018. Prior to this hiring, Co-Principal Investigators Ian Blaby and Shinjae Yoo have worked closely to explore the feasibility of several computational platforms for datamining including DeepDive and Apache UIMA to extract gene-specific events from the biological literature. Since the postdoctoral research associate arrived, the project has proceeded rapidly and we jointly submitted a proposal to use the BNL Institutional Cluster. In parallel, Carlos is ensuring optimal progress towards this project by familiarizing himself and performing preliminary analyses using DeepDive, as we work towards the milestones below.

Milestones:

- 6 month: test applicability of preexisting open source platforms
- 12 month: finalize evaluations of platform
- 24 month: modify Probabilistic Graphical Model as required, start to contact publishers to increase article access
- 36 month: construct web interface, open tool to wider community.

Molecular Mechanisms of Alkane Hydroxylation

LDRD Project # 17-023
Q. Liu

PURPOSE:

Membrane-bound redox-enzyme complexes catalyze a critical step in conversion of hydrocarbon to high-value bioproducts, avoiding the need for expensive and nonspecific chemical conversion. To harness the potential of these highly-specific and energy-efficient biocatalysts, we propose to study the molecular mechanisms of chemically recalcitrant C-H bond activation in alkanes catalyzed by a hydroxylation complex. We propose to determine the atomic structure of the alkane hydroxylation complex and characterize its molecular function in C-H bond activation and alkane bioconversion. The project will reveal the underlying mechanisms guiding substrate selectivity, electron transfer and alkane hydroxylation, thus providing the structural foundation to confidently redesign the hydroxylation complex for improved and new functionalities. The project will significantly enhance the DOE Office of Basic Energy Sciences physical biosciences capability at BNL and contribute to the development of the DOE Office of Biological and Environmental Research structural biology program at BNL.

APPROACH:

Alkanes are saturated hydrocarbons that constitute 20 to 50% of crude oils. Due to the abundance and cost effectiveness, alkanes are very attractive starting materials for producing high-value products. Alkanes contain only carbon and hydrogen atoms with fully occupied electronic orbitals, making alkanes almost chemically inert. To functionalize alkanes, they must first be activated in order to form alcohols, aldehydes, carboxylic acids, or epoxides, which represent a family of high-value products and feedstocks for further syntheses. Direct chemical conversion of alkane is possible; however, the current processes require high-temperature, harsh conditions and tend to produce unpredictable byproducts by expensive and toxic catalysts.

In nature, some microbes, for example the gram-negative bacterium pseudomonas oleovorans thrive in oil-rich environments where they can use alkanes as their sole carbon and energy source through the process of alkane hydroxylation. The alkane ω-hydroxylase, AlkB, from pseudomonas oleovorans, oxidizes the methyl group of alkanes to produce the corresponding alcohols. By forming an electron transport complex with rubredoxin (AlkG) and rubredoxin reductase (AlkT), AlkB catalyzes the hydroxylation reactions of alkanes (Fig. 1A). This AlkBGT hydroxylation complex converts alkanes to alcohols at ambient temperature and pressure with high specificity, thus is attractive for exploitation for real-life applications. However, due to absence of valuable structural data, the exact molecular mechanisms for the hydroxylation complex are largely unknown, which hampered the use of the system for enzymatic C-H bond activation and alkane bioconversion.

In collaboration with J. Shanklin (Biology) on protein biochemistry, S. McSweeney on synchrotron micro-crystallography (National Synchrotron Light Source II), and H. van Dam on molecular and quantum mechanics simulation and design (Computational Science Initiative), we propose to determine structures of the hydroxylation complex and characterize the underlying molecular mechanisms of electron transfer, C-H bond activation and alkane hydroxylation. Specifically, we will produce and characterize individual components as well as their assemblies of the hydroxylation complex. We will determine structures of AlkB alone, AlkB in complex with AlkG, and the entire AlkBGT complex. We will use molecular and quantum mechanics

computation to understand the underlying catalytic principles as well as to design new hydroxylation complexes for improved and new functionalities.

TECHNICAL PROGRESS AND RESULTS:

In FY 2017, we have cloned and expressed three individual proteins of the complex and did functional characterization of these proteins in alkane bioconversion. AlkB is a multi-pass transmembrane protein and has been resistant to stable production. To obtain stable AlkB protein, we made four constructs and identified two that led to large scale production. To stabilize AlkB, we made the AlkBG complex by attaching AlkG to AlkB (Fig. 1B). The complex has a higher molecular weight than AlkB alone, indicating the formation of a complex (Fig. 1C). To assure that the AlkBG complex is functionally active, we measured the activity of AlkBG in biological conversion of alkanes to alkanols (Fig. 1D). We also tried to express the third protein of the complex, AlkT. The expression level of AlkT is high; while the purity was not sufficient for structural and functional work. We plan to optimize our purification protocol of AlkT.

With the purified AlkB and AlkBG, we did crystallization screening by both the vapor diffusion method as well as the lipidic cubic phase method. We obtained diffraction quality crystals of a lipid/detergent complex and determined its crystal structure at high resolution (Fig. 1E). We hypothesize that these lipids bind to AlkB tightly and may have disturbed the crystallization of AlkB. We therefore developed a wash protocol to effectively remove most lipids but still maintain the protein in soluble state. Crystallization screening with reduced lipid content is underway.

In addition to our crystallization work, we prepared and screened AlkBG for suitability of cryo-Electron Microscopy (EM). We found that AlkBG complex is stable in solution. Preliminary analysis using a negative staining electron microscope showed well-separated nanoparticles, promising for single-particle cryo-EM analysis (Fig. 1F). The sizes of these nanoparticles vary. So, we are optimizing the sample preparation conditions to get more homogenous particles desirable for cryo-EM.

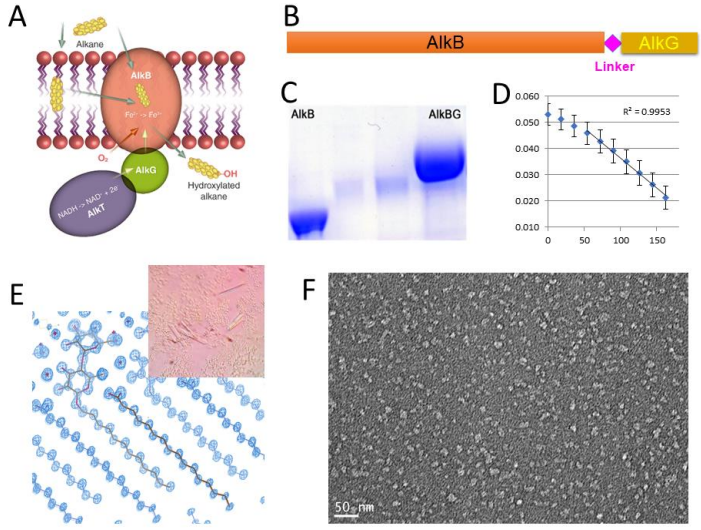


Figure 1: Alkane Hydroxylation Complex. (A) Schematic diagram of the complex consisting of AlkB, AlkG, and AlkT. (B) AlkB and AlkG fusion complex. (C) Coomassie Blue staining sodium dodecyl sulfate polyacrylamide gel electrophoresis (aka SDS-PAGE) analysis for the AlkBG complex. (D) Alkane conversion activity catalyzed by the AlkBG complex. (E) Crystals and structure of a detergent/lipid complex. (F) Negative staining Transmission Electron Microscopy for AlkBG complex imaged at a nominal magnification of 50K.

Milestones:

FY 2018: Determine the structures of AlkBG by cryo-EM and/or X-ray crystallography. Production and characterization of the AlkBGT complex.

FY 2019: Determine the structure of the entire AlkBGT complex with and without substrates; prepare atomic models suitable for biocatalyst design by molecular and quantum mechanics computations.

Development of A New Approach to Remotely Measure Limitations on Plant Growth

LDRD Project # 17-024
A. Rogers

PURPOSE:
This LDRD will develop the scientific foundation and technical ability to measure the balance between plant resource capture and utilization, repeatedly and non-destructively using remote sensing techniques and will provide new sophisticated biological markers for physiological breeding programs. The LDRD contributes to the development of a comprehensive plant science research portfolio at BNL, is aligned with the DOE Office of Biological and Environmental Research and is synergistic with the development of the Quantitative Plant Science Initiative at BNL. The development of this new capability is consistent with the Laboratory Agenda.

APPROACH:
The LDRD will leverage two areas of existing expertise to develop a new capability with a cross cutting impact. Plant growth is a major determinant of yield in food and biofuel feedstock crops. Therefore, increases in productivity will depend on a quantitative understanding of the extent to which sources (leaves) and sinks (e.g. grain or biomass) limit growth. In other words, we need to identify the processes that are bottlenecks limiting growth and yield. Currently identification of key bottlenecks is hindered by our inability to monitor source-sink balance non-destructively throughout plant development. This LDRD will leverage expertise in understanding and measurement of plant source-sink status (A. Rogers), and in remote sensing of plant functional properties (S. Serbin), to develop a new capability to remotely and non-destructively measure biological markers associated with the balance between plant resource capture and utilization. The concept can be broadly applied to scales ranging from high throughput platforms in controlled environments to large physiological breeding programs in field conditions. We will develop links between plant leaf traits and the spectral signatures of their leaves to enable those traits to be sensed remotely and non-destructively. Initial work (now underway in FY 2018) will establish links between leaf spectra and plant traits associated with carbon and nitrogen metabolism, and plant response to drought. Future steps are highlighted in the table below.

TECHNICAL PROGRESS AND RESULTS:
This LDRD started in September 2017 so there is limited FY 2017 progress to report. Essentially we hired a post-post – who started on October 1st 2017 – and began to plan a glass house experiment. That experiment is now underway (December 2017).

Milestones:
- Develop spectral signatures for assessment of source-sink balance
- Develop instrument package
- Complete evaluation of capability – Greenhouse
- Complete evaluation of capability – Field

High Performance X-ray Diffraction Simulation Toolkit Using Graphics Processing Unit and Central Processing Unit Clusters

LDRD Project # 17-029
M. Lin, Wei Xu

PURPOSE:

X-ray diffraction is an essential tool for crystal characterizations that was developed more than 100 years ago. However, high-performance computational tools for general X-ray diffraction forward simulation, data analysis and visualization are still lacking. The objective of this project is to improve the algorithms for the forward simulations based on the dynamical diffraction theory, port and optimize the software on both Central Processing Unit (CPU) and Graphics Processing Unit (GPU)-based High Performance Computing (HPC) platforms, and design a user-friendly graphical interface to help the scientists design their X-ray diffraction experiments, understand their data and verify mechanical models more rapidly. This tool will enable virtual X-ray diffraction experiments that are vital for verifying new concepts, optimizing experimental conditions, and testing new data analysis algorithms. It has applications in nano- and micro-crystal diffraction, nano- and micro-beam diffraction and diffraction topography. While the immediate use cases are for the National Synchrotron Light Source II (NSLS-II) facility at Brookhaven National Laboratory, it can benefit X-ray diffraction beamline scientists and user communities broadly in academia, government and industry.

APPROACH:

X-ray diffraction from crystals is a powerful tool to identify the crystal structure, accurately measure the strain and manipulate X-rays. The theoretical foundation for solving the general dynamical diffraction problems lies in the Takagi-Taupin equations (TTE), which can only be solved numerically except for a few special cases. Finite-difference methods can be employed to solve TTE with certain boundary conditions, but they are not generally applicable for crystals with complex shapes and mixed diffraction geometries. In 2014, one of our Co-Principal Investigators, Dr. Hanfei Yan, proposed [1] a universal modeling approach that can solve TTE for general X-ray diffraction cases based on an iterative procedure. The new approach converts TTE into integral equations for which an integral solution satisfying the boundary condition can be readily obtained. Moreover, this generic diffraction modeling approach is well suited for parallel processing, as the calculations of the wave fields at different positions inside the crystal are independent of each other, making it possible for us to take advantage of the computing powers of modern CPU or GPU-based systems. The toolkit we are developing will also have a user-friendly graphical interface that includes a control panel for the input simulation parameters and a display panel that has 3D visualization capability for the output images. To accomplish these goals, we have hired a student assistant, Ryan Hilbert, who is a computer science undergraduate student at Stony Brook University, and a postdoctoral research associate, Sangsoo Ha, who specializes in visualization and HPC, to work on the project.

TECHNICAL PROGRESS AND RESULTS:

Our prototype toolkit used NVIDIA's CUDA programming model for parallel processing on the GPUs. In order to make the toolkit portable across different architectures, we ported the software using OpenCL (Open Computing Language) that supports many heterogeneous configurations CPU+X, where X can be NVIDIA CPUs, AMD GPUs, Intel GPUs or even the IBM Cell Blade.

The toolkit supports parallel processing of the simulation with a single scan point, as well as asynchronous parallel processing with multiple scan points.

As shown in Fig. 1, a graphical user interface (GUI) consisting of a control panel and a display panel was developed using Qt and QCustomPlot libraries. The display panel will show the input beamline orientation relative to the crystal placement before the calculation and will switch to showing the diffraction image after the calculation is completed.

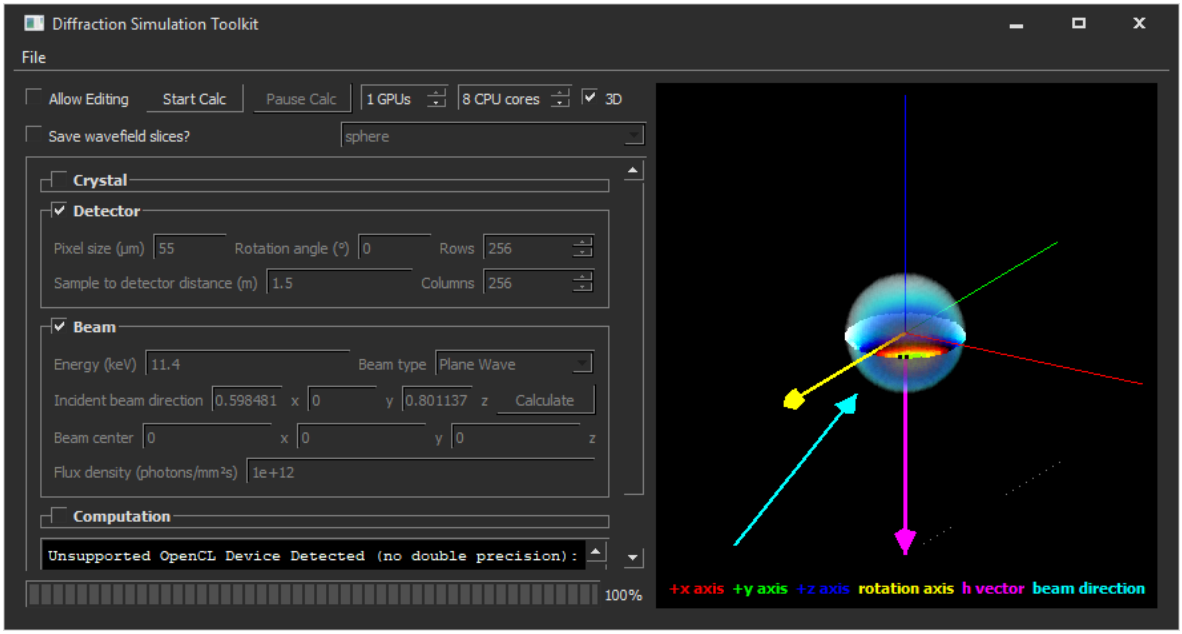


Figure 1: Snapshot of the GUI for the diffraction simulation toolkit at the calculation setup stage.

The first software releases of the toolkit can be obtained from GitHub [2]. The current release supports both Windows and Linux operating systems. In order to support the client-server mode for the cluster-based implementations, and be aligned with the current Python-based data acquisition system of NSLS-II, we will provide another web-based GUI on Jupyter notebook.

Milestones;

For FY 2018 and FY 2019 (half year), we plan to install our software first at the Hard X-ray Nanoprobe (HXN) beamline and perform maintenance. In order to make our project more scientifically impactful, we plan to extend this collaboration module to additional scientific problems and focus on the following tasks:

- Enhance the performance of ptychography reconstruction
- Investigate new research problems, including projection alignment for tomography and new phase finding for fluorescence data
- Leverage onsite visualization work and apply it to HXN modalities.

REFERENCES:

[1] Yan, H. & Li, L. X-ray dynamical diffraction from single crystals with arbitrary shape and strain field: A universal approach to modeling. Physical Review B 89, 014104 (2014)
[2] https://github.com/r-hilbert/diffraction-simulation-toolkit/releases

Transitions in Strongly Correlated Metal Oxides

LDRD Project # 17-035

I. Robinson

PURPOSE:

The goal of this LDRD was to oversee the X-ray Scattering Group (XSG), a group comprised of two scientists, a 50% joint appointment and postdoctoral research associates. It was also expected to jump-start a collaboration with National Synchrotron Light Source II (NSLS-II) to build a new coherent-imaging beamline. The achievements to date are to introduce Bragg Coherent Diffractive Imaging (BCDI) and merge it with the existing Pair Distribution Function (PDF) program within the XSG. A side achievement was to explore Ultrafast Electron Diffraction (UED) and prepare successful applications for laser beamtime. We have already shown that BCDI can deliver contributions to the Division, specifically Iridium Telluride ($IrTe_2$) domain imaging which will appear in a future PDF paper by Emil Bozin, and introduction of the concept of "pinning physics" in another pending publication.

APPROACH:

The main event of the year was the successful renewal of the X-ray Scattering field work proposal in April 2017, which is now fully funded in FY 2018. The senior XSG group members moving forward are Emil Bozin and Simon Billinge (50% joint appointment at Columbia) and two postdoctoral research associates. Specifically, the methods of BCDI have been advanced with X-ray experiments at NSLS-II and the Advanced Photon Source (APS) (Argonne) and the planning of a new beamline for CDI at NSLS-II. We undertook a successful experiment at the Korean X-ray Free Electron Laser facility to explore the structure of gold (Au) on the ultrafast time scale immediately following a laser excitation. Those data are being analyzed using the PDF method.

TECHNICAL PROGRESS AND RESULTS:

We made preliminary studies on the Perovskite manganite $La_{1-x}Ca_xMnO_3$ (LCMO) with x=0.5. Nanocrystalline samples were necessary to meet the oversampling requirement of BCDI. Typical diffraction patterns, which nominally represent the 3D Fourier transform of the crystal shape, show a set of concentric rings resembling the Airy pattern of a compact solid object with streaks attributed to its prominent facets. Our LCMO data, shown in Fig. 1, are more "speckled" with several subsidiary maxima of intensity, indicating a more complicated structure with significant lattice distortions or strain. Once the BCDI diffraction pattern is inverted, the strain is mapped in the resulting complex real-space 3D image as the phase of the complex

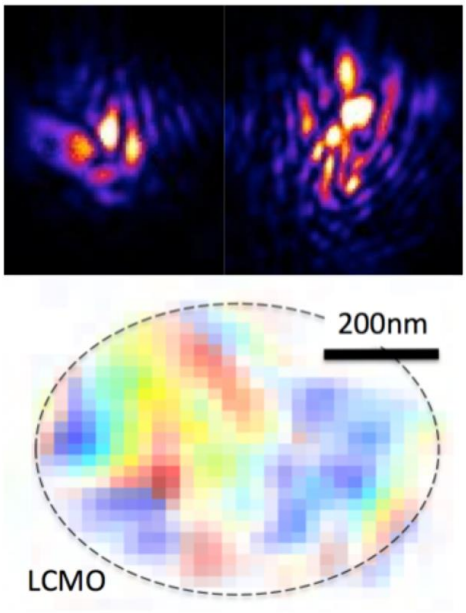


Figure 1: (Top) BCDI diffraction patterns of two single grains of LCMO measured at the 200 reflection at room temperature using APS. (Bottom) Section of a 3D image of the mosaic domains in a crystal of LCMO. The colors indicate the relative phase shifts on the scale (blue-red) of one 200 d-spacing.

number at each voxel location. This phase $\Delta\varphi$ corresponds to a projection of the local crystal displacement vector **u**, according to the simple relation, $\Delta\varphi = \mathbf{Q}\cdot\mathbf{u}$, where **Q** is the momentum transfer vector of the Bragg peak measured.

Preliminary images show massive strain effects in Fig. 1: whole blocks inside the crystal have different (real space) phase. We identify this to be a new form of crystal mosaic, which is fundamental to the crystal growth and subsequent treatment. Nanometer-sized blocks of crystal are laterally shifted with respect to each other and acquire different phases with in-grown planar defects separating these blocks.

Domain formation is critical to the understanding of the rhombohedral phase transitions in $IrTe_2$, where the Ir dimerization direction may drive the pattern of domains seen, as our BCDI experiments have shown. The pinning of Charge Density Wave (CDW) stripes in $La_{1.825}Ba_{0.125}CuO_4$ (LBCO) was studied to explore the connection with domain formation at its Low Temperature Orthorhombic (LTO) to High Temperature Tetragonal (HTT) phase transition in Fig. 2. BCDI experiments on carefully ground samples of LBCO have produced preliminary images of the pattern of LTO domains within the HTT material and, importantly, have determined the grain sizes of those domains.

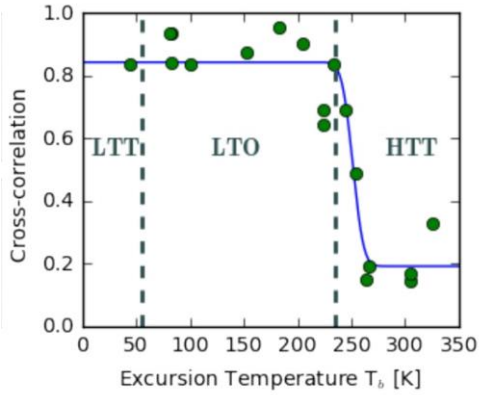


Figure 2: Correlation between coherent diffraction patterns of LBCO before and after temperature excursion to the designated temperature. Data are from the CSX-I beamline of NSLS-II.

In order to avoid beamline drift problems between the illuminating beam and the sample, we have fabricated pinholes in thin sheets of Au and laid them directly on the face of the LBCO samples, cleaved to their c-axis. Since the (0.24,0,1.5) peak on resonance appears at near normal incidence, the shadowing effect of such a sample-pinhole is minimized at the Cu L_3 resonance at the Coherent Soft X-ray Scattering (CSX) beamline. We are making progress towards imaging the individual CDW domains contributing to the speckle pattern. Using the on-sample pinhole to prevent beam-sample drifts, we studied the "depinning" of the CDW domains, to investigate the physics of pinning to relate to the body of theoretical work on this subject. The results of Fig. 2 show that the observed pattern of pinning is extremely robust and survives temperature excursions as high as 250K, well above the melting temperature of the CDW and likely associated with the HTT transition temperature. This "return point memory" effect has been seen before in magnetic systems, but never for CDWs.