BROOKHAVEN NATIONAL LABORATORY

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LDRD 2021 Annual Report: Laboratory Directed Research and Development Program Activities

J. Anderson, L. M Flynn

April 2022

Director's Office

Brookhaven National Laboratory

U.S. Department of Energy

Laboratory-Directed Research and Development (LDRD)

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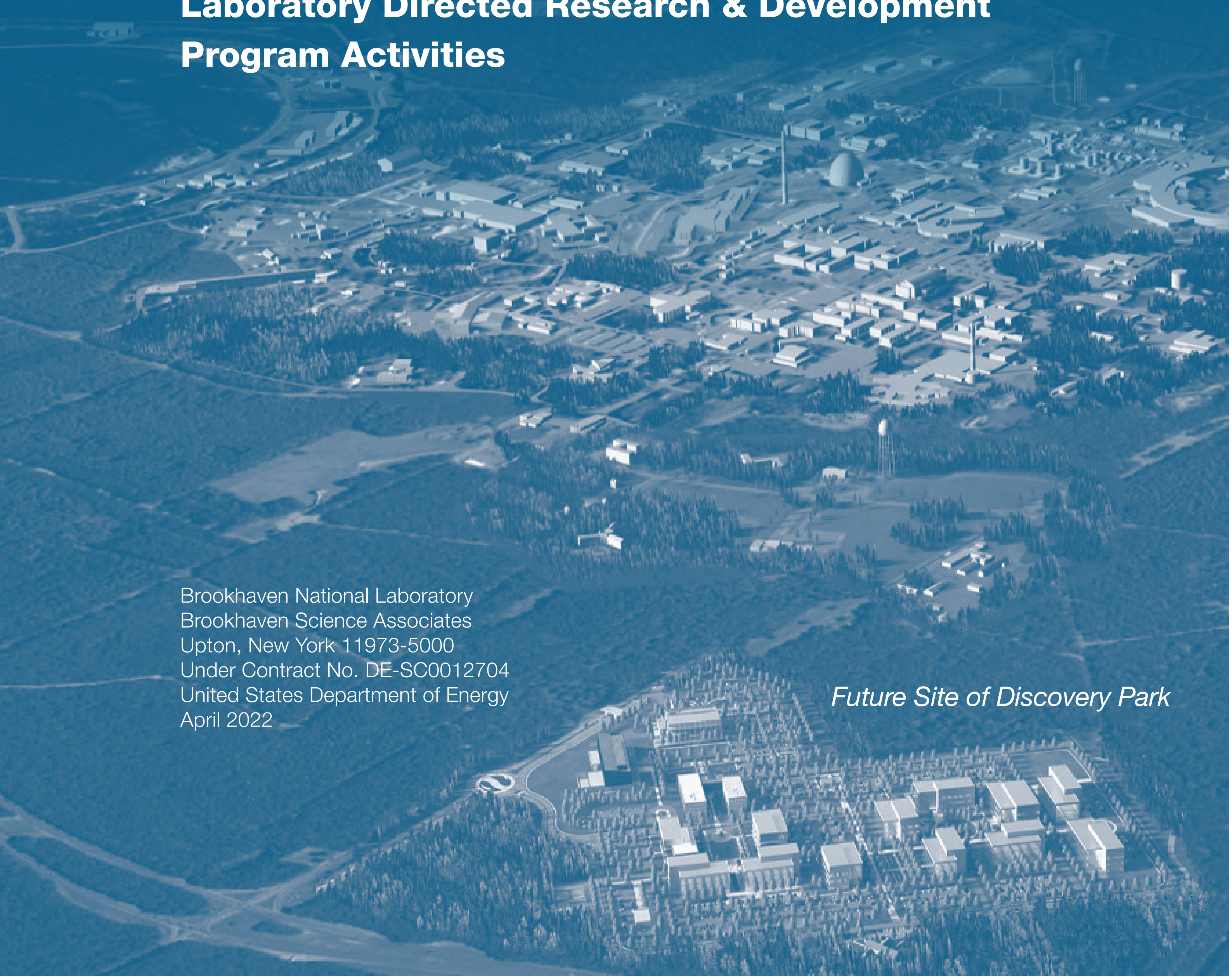
Brookhaven National Laboratory

BNL-52351-2021

2021 Annual Report

Laboratory Directed Research & Development Program Activities



Brookhaven National Laboratory
Brookhaven Science Associates
Upton, New York 11973-5000
Under Contract No. DE-SC0012704
United States Department of Energy
April 2022

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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 2021 report were coordinated by Liz Flynn and edited by Kathi Barkigia. Jack wishes to thank Kathi Barkigia, Antoinette Russo, Charles Baldassare, Janine Mehlinger 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 project annual reports.

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Introduction

Each year, Brookhaven National Laboratory (BNL) is required to provide a report of its completed Laboratory Directed Research and Development Program (LDRD) projects to the Department of Energy (DOE) Office of Scientific and Technical Information in accordance with DOE Order 413.2C Chg1 (MinChg) dated August 2, 2018. This report provides a detailed look at the scientific and technical activities for each of the LDRD projects funded by BNL in FY 2021, in fulfillment of that requirement. In FY 2021, the BNL LDRD Program funded 68 projects, 24 of which were new starts, at a total cost of $17.1M.

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. This requires simultaneous excellence in all aspects of BNL's work – from science and operations, to external partnerships with the local, state, and national communities, and beyond. This is enabled by safe, efficient, and secure operations; by an unwavering commitment to a diverse, equitable, and inclusive environment, including workforce development, both with staff and reaching out to the community; and by a strong focus on renewed infrastructure.

The six scientific initiatives are:

- Nuclear Physics: The Electron Ion Collider
- Clean Energy and Climate
- Quantum Information Science and Technology
- Discovery Science Driven by the Human-AI-Facility Integration
- High Energy Physics: Building for Discovery
- Accelerating Isotope Production: Ensuring the Nation's Supply is Secure.

The funded projects support BNL's six scientific initiatives and priority programs as well as new areas of research and competencies at the Laboratory that are consistent with the Laboratory's vision and mission. In total, these LDRD investments supported 80 postdoctoral researchers and graduate students in whole or in part and resulted in 136 publications and 3 awards.

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

2021 PROJECT SUMMARIES

Analysis on the Wire (AoW)

LDRD # 18-002

D. Katramatos

PURPOSE:

In the era of Big Data, more data can potentially be found at any given moment in transit than in storage. The Analysis on the Wire (AoW) project examines the feasibility of performing general-purpose computations on streaming data on the wire, i.e., while in transit in the network and develops a framework to enable such computations. AoW further evaluates the feasibility and effectiveness of the approach in a number of use cases (e.g., Smart Grid, cybersecurity). On-the-wire processing can provide real or near-real time information to speed up decision making processes, e.g., in steering computation; in optimizing and prioritizing information routing and correlation; and offering additional processing cycles to free data center resources. AoW could also be beneficial for High Energy Physics and National Synchrotron Light Source II (NSLS-II) experiments by facilitating decision making through the early processing of acquired data.

APPROACH:

One or more (federated) network devices needs to be programmed (and/or coordinated as a distributed computing platform) to recognize specific data flows and transparently apply certain types of computations, using suitably designed algorithms, on the data of network flows before forwarding them to their destination. The approach followed is three-fold: to investigate and evaluate existing and upcoming network hardware and technologies and to establish a suitable testbed for experimenting and evaluating an AoW system; to develop an efficient AoW software framework for proof of concept; and to study a number of real-world use cases through the development of suitable algorithms that can be run on the wire.

TECHNICAL PROGRESS AND RESULTS:

In prior work, we evaluated the capabilities of networking hardware to determine its suitability for use in the project, explored a worst-case performance scenario with available Software Defined Networking (known as SDN) software components, and developed an efficient future grid load prediction algorithm based on Smart Meter data. In FY18 and FY19, we developed a basic framework, established a testbed for experimentation and evaluation of analysis algorithms, and considered a number of use cases (streaming analysis of NSLS-II beamline images, Smart Grid load forecasting, Smart Grid state estimation) with promising results.

In FY20, we:

- Continued to expand the testbed with newer hardware
- Developed a basic Field Programmable Gate Array-based mechanism for accelerating the processing of layer 2 network packets and a mirroring mechanism aimed at fully stand-alone analysis nodes
- Developed and evaluated a new Smart Grid load forecasting algorithm based on Collective Echo State Networks
- Started two follow-on projects based on this project (funded by the National Nuclear Security Administration, Office of Nonproliferation Research and Development [NA-22] and the Office of Cybersecurity, Energy Security, and Emergency Response [CESER], shown in Figures 1 and 2 respectively).

In FY21 (the project was concluded on 2/28/2021), we:

- Continued the testing, debugging, and evaluation of AoW software

- Made modifications to the framework to facilitate its use in the new projects
- Repurposed the testbed nodes to be used in the new projects.

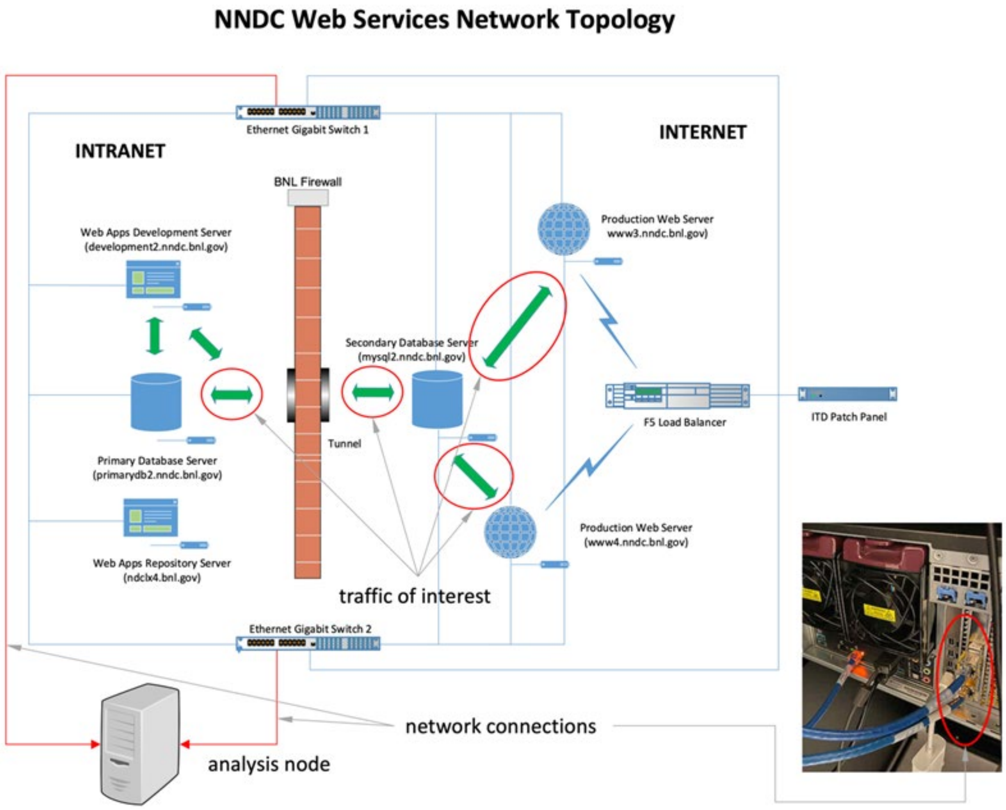


Figure 1. AoW testbed in Hidden Network Surveillance (NA-22 project). Traffic from the network is captured by the analysis node for preprocessing, filtering, and lightweight correlation.

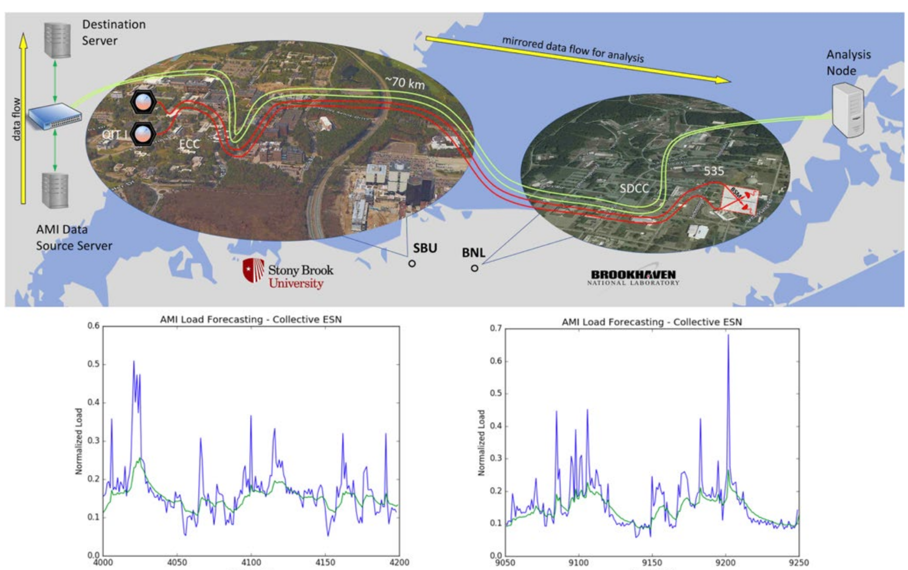


Figure 2. AoW testbed using the hybrid Quantum/Classical network prototype for Smart Grid data analysis (CESER project). Traffic is sent to analysis node to generate predictions in real time as shown in the two forecast plots**.**

MILESTONES:

None. This project concluded in FY21.

Electrolyte Flow Battery for Smart Grid Application

LDRD # 18-017

E. Takeuchi, A. Marschilok, L. Wang

PURPOSE:

The objective of this project is to investigate electrolyte systems that may be suitable for batteries destined for grid scale applications. Aqueous-based electrolytes with different anion compositions and different concentrations are being explored. This approach may allow exploitation of materials classes as negative and positive electrodes and suitable electrolyte formulations for aqueous battery systems for grid applications.

APPROACH:

This project is targeted to develop technology platforms for grid energy storage applications at Brookhaven National Laboratory. Renewable energy generation, such as solar energy and wind energy harvesting, requires the development of reliable, sustainable, and low-cost energy storage systems. Lithium (Li)-ion batteries are the state-of-the-art technology for energy storage today. However, the cost of Li-ion batteries is too high due to the use of low earth abundant transition metals as part of their electrode system. Moving to aqueous-based systems to reduce cost and environmental impact is needed. Aqueous rechargeable zinc (Zn) ion batteries (ARZIBs) have recently received significant attention due to their low cost, innate safety, and the desirable ability to scale-up. The use of manganese (Mn)-based systems represents a promising cathode for ARZIBs due to low cost and low environmental impact. Here, we demonstrated a strategy to explore these systems.

TECHNICAL PROGRESS AND RESULTS:

Research progress made in earlier fiscal years: Dissolution of solid manganese oxide was demonstrated as a critical attribute of Zn/alpha-Mn oxide batteries.

Research progress made in the present fiscal year: results acquired on Zn/Mn cells used a MnO_2@C (manganese oxide and carbon) and C-only (carbon only) electrodes with Zn metal as the negative electrode. The MnO_2@C electrode delivered 73% greater areal capacity relative to the carbon-only analog (Figure 1a). The cell demonstrates significant energy density improvements relative to previously reported related designs. The MnO_2@C electrode also demonstrated superior long-term cycling over 15 cycles relative to a C-only analog where Coloumbic efficiency (CE) of the C-only electrodes decreases by 13% from cycle 1 to cycle 15; CE remains at 100%

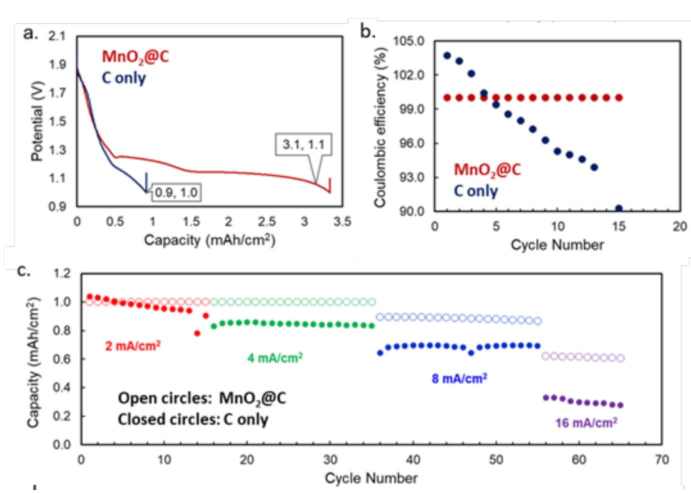


Figure 1. a) First cycle discharge of experimental Zn/Mn cells with MnO_2 and Carbon electrode (red) and C-only electrode (blue). b) Coulombic efficiencies. c) Rate capability data.

for the MnO_x@C electrode (Figure 1b). The three-dimensional (3D) MnO_2@C electrode also demonstrates superior rate capability (Figure 1c). In contrast, the C-only delivered lower capacity during the test (Figure 1c). In summary, the 3D MnO_2@C electrode delivers higher energy density, superior cycle life, and rate capability relative to C-only electrodes.

Research during the past year demonstrated that the carbon substrate plays a critical role in the Mn dissolution-deposition mechanism during the charge step. Additional Zn/Mn cells with either graphene paper, carbon cloth, or carbon nanotube (CNT) substrates were tested. Notably, the deposition for the CNT substrates showed greater amounts of deposition (Figure 2a). On first discharge, the CNT substrate delivered two to five times greater areal capacity than the other carbon types (Figure 2b). From cycle 1 to cycle 11, a decrease in oxidation potential is observed for the carbon cloth and carbon, indicating a change in the deposition mechanism on charge (Figure 2c). Notably, the tenth discharge demonstrates a similar trend to the first discharge, where delivered capacity is CNT > carbon cloth > graphene paper (Figure 2d). Electrochemical impedance spectroscopy measurements before cycling and tracking of substrate mass and before and after cycling were recorded and aided in the rationalization of the electrochemical trend. The graphene paper Zn/Mn cell demonstrated the largest impedance and deposits the least amount of MnO_x on the substrate. In contrast, ~ten times and ~thirty times more MnO_x is measured on the carbon cloth and CNT after the eleventh charge, respectively. Both carbon cloth and CNT conditions have reduced impedance relative to the graphene paper. Interestingly, the CNT impedance is larger relative to the carbon cloth impedance, yet more MnO_x is deposited on the CNT substrate. These results suggest that factors in addition to cell impedance are contributing to the Mn deposition process.

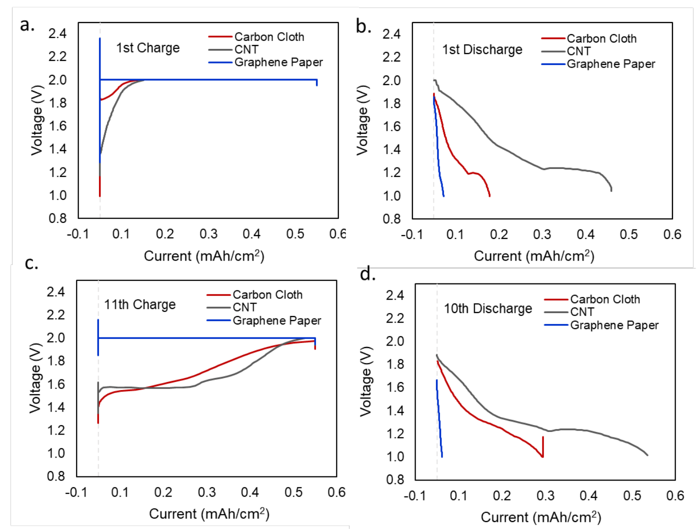


Figure 2. a) First charge. b) First discharge. c) Eleventh charge. d) Tenth discharge of Zn/Mn flow cell with graphene paper (blue), carbon cloth (red), and CNT (black) cathode substrate.

A paper is in preparation summarizing the results.

MILESTONES:

None. This project completed in FY21.

Electron Beam Formation via Ionization Injection for Next Generation Accelerator Research & Development

LDRD # 18-026

M. Palmer, R. Kupfer, M. Fedurin, M. Babzien

PURPOSE:

Laser wakefield acceleration (LWFA) is a method of harnessing the extremely strong electric fields of high intensity ultrafast lasers for efficient acceleration of electrons by driving a high amplitude nonlinear plasma wake. Despite the progress of LWFA in recent years, the bunch quality and total charge of LWFA are still sub-par compared to conventional radio frequency (RF) accelerators, as the large phase space volume of trapped electrons results in a large emittance and limited applicability. In the two-color ionization-injection scheme, the tasks of "driving of plasma wake" and the "injection of electrons" into this wake are separated and assigned to two different laser pulses with vastly different wavelengths. Specifically, a high-a_0 (normalized vector potential), low intensity, long wave infrared (LWIR) laser is used for driving the wake, and a low-a_0, high-intensity near infrared (NIR) laser is used to ionize electrons from a specifically chosen high-Z gas target and to inject the resulting electrons into a beam of high quality and low emittance. The overall objective of this project is to develop the experimental infrastructure for the first realization of two-color ionization injected LWFA, which could then be the basis for an all-optical electron source at Brookhaven National Laboratory.

APPROACH:

The experimental apparatus utilized the Accelerator Test Facility's (ATF) state-of-the-art ultrafast hybrid CO_2/solid state LWIR laser as the driver and the newly commissioned ultrafast titanium-sapphire (Ti:Sa) NIR laser as the injector in a transverse injection scheme (Figure 1). Using krypton gas as the target, we measured the ionization states produced at foci volume of the NIR and LWIR lasers using time-of-flight spectroscopy. We developed alignment and timing procedures for sub-2 ps/10 μm spatiotemporal overlap of the laser pulses and set up electron bunch diagnostics to probe the spatial profile and energy spectrum of the accelerated electrons.

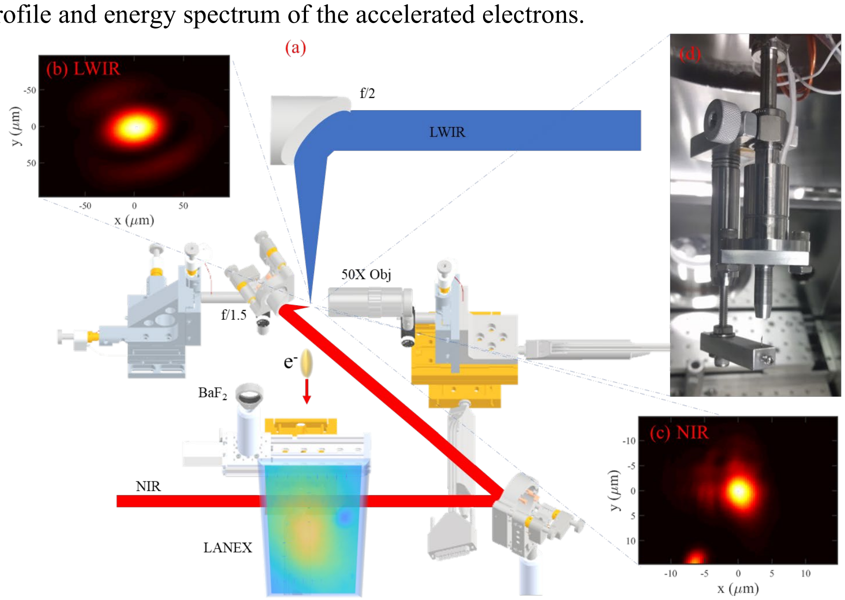


Figure 1. (a) Experimental layout near the interaction point: LWIR pulses were focused using a 90^{O} f/2 off-axis parabolic mirror, while the NIR pulses where focused to the same transverse position using a 60^{o} f/1.5off-axis parabolic mirror. A 50 times imaging objective was used to optimize the NIR focus and timing overlap with LWIR. (b) and (c) representative low energy LWIR and NIR focus spots, respectively. Generated electrons were recorded by a LANEX screen and imaged with an external CCD camera. (d) a photo of the gas nozzle and valve assembly.

TECHNICAL PROGRESS AND RESULTS:

Temporal synchronization and timing of LWIR and NIR was achieved by locking both oscillators to a common master oscillator clock. Temporal overlap was measured by observing (the heavily attenuated) NIR focal spot as it just strikes the surface of a metal pin placed at the interaction point, while firing the LWIR laser at low energy (~50 mJ). Early NIR pulses that arrived at the interaction point ahead of the LWIR pulses passed through, while late ones were blocked by the over-dense surface plasma generated by the LWIR pulses. Fine timing was done by electronically adjusting the Ti:Sa oscillator phase in a "binary search" until the temporal window for passing/blocking was reduced to ~2 ps.

To verify the NIR pulse ionized higher charge states of krypton than the LWIR pulse, separate time-of-flight measurements of the ionization at the tight focus of both NIR and LWIR were performed. For the LWIR pulse with an energy of 3.2 J, only the first ionization state of krypton, Kr^{+1} was detected. The NIR pulse at low energy (~2 mJ) produced ionization states up to Kr^{+6} with a higher amplitude of Kr^{+3} than observed for Kr^{+2}. At higher NIR pulse energies, splitting of each ionization state peak was observed due to space-charge effects at the tight laser focus. This splitting grew with increasing NIR energy up to 5 mJ where saturation was reached. This suggests that the maximum possible ionization was reached at the focus and that the laser intensity was not enough to pass the energy gap for the next ionization states.

While observing the transverse profiles of the accelerated electrons with a LANEX screen imaged to a charged-coupled device (CCD) camera, we scanned for self-injection via the LWFA process with only the LWIR driver and identified the onset of a well-defined electron bunch. Then, operating just above the onset of self-injection, we applied the NIR injector. We performed a timing scan of the NIR injection and found a noticeable improvement in the y-axis divergence for negative timing delays (i.e., the NIR pulse arrives ahead of the LWIR pulse). A collection of electron profiles without and with NIR injection is presented in Figure 2. While we did not observe any effect at zero or positive NIR delays, which is expected since the plasma structures for our LWIR laser parameters were quite small and completely overlapped with the LWIR pulse, we have demonstrated the first all optical seeding of a LWIR- driven LWFA by NIR ionization of a density perturbation ahead of the wake.

Collaborative follow-on funding with Stony Brook University, University of Texas–Austin, and University of California, Los Angeles was included in the renewal submission "Plasma Wakefield Research at ATF and FACET II" (DE-FOA-0002546, DOE-HEP).

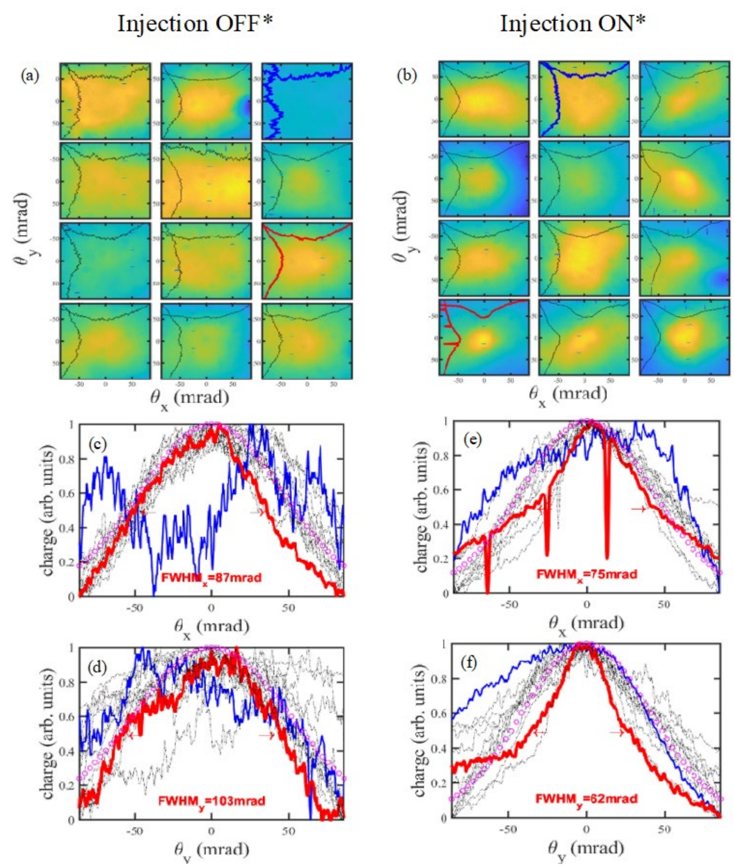


Figure 2. Comparison of the divergence of unseeded and seeded LWFA: (a) and (b) show representative transverse electron profiles for (a) unseeded self-injection LWIR-driven LWFA and (b) seeded LWIR-driven with $3.75 < E_{LWIR} < 4.3J$, $E_{NIR} = 4$ mJ, $\Delta t = -4$ ps. (c-f) X and Y lineouts of transverse electron profiles for self-injected LWFA and ionization-seeded LWFA. Best and worst profiles are highlighted in red and blue, respectively. The mean divergence is shown with purple circles. The mean divergences for seeded shots were $\langle \Theta_x \rangle = 98$ mrad and $\langle \Theta_y \rangle = 97$ mrad, compared to $\langle \Theta_x \rangle = 108$ mrad and $\langle \Theta_y \rangle = 158$ mrad for unseeded shots.

MILESTONES:

None. This project completed in FY21.

Micro-Pattern Gas Detectors for Electron-Ion Collider (EIC)

LDRD # 18-033

A. Kiselev, C. Woody

PURPOSE:

The primary goal of this project is the application of novel one-dimensional (1D) and 2D readout plane designs and modern high-precision printed circuit board (PCB) manufacturing technologies to a wide variety of Electron-Ion Collider (EIC) Micro-Pattern Gaseous Detector (MPGD) types with the goal of building a "family" of high-performance detectors featuring optimized spatial resolution, low channel count, and minimal non-linearity of response for coordinate measurement, for a wide range of charge cloud footprints.

APPROACH:

The conventional approach to achieve high spatial resolution in the planar gaseous tracking detectors like gas electron multipliers (GEM) and micromegas is to increase the readout plane granularity, namely, to use a strip pitch (1D) and/or pad size (2D) a few times smaller than a typical charge cloud footprint. This approach causes an increase in the electronics channels, which is usually the main cost driver for such detectors. However, if in a typical configuration with straight strips (1D) and/or square pads (2D) the readout plane granularity becomes too coarse, not only does this cause severe spatial resolution degradation, but also causes a substantial differential non-linearity in the detector response. By designing highly interleaved readout electrodes, one can optimize the charge sharing properties among neighboring strips and/or pads and therefore drastically improve the detector positional accuracy without increasing the electronic channel count.

TECHNICAL PROGRESS AND RESULTS:

We demonstrated experimentally that this approach applies to virtually any type of avalanche schemes anticipated for EIC planar gaseous tracking detectors, including more modern micro-resistive well (μRWELL) ones. We also showed that the developed readout board design principles are applicable to the capacitively-coupled Micro-Channel Plate photomultiplier tubes, in particular, to so-called Large Area Picosecond Photon Detectors (LAPPDs) manufactured by Incom (USA). These devices can become a prime choice for an affordable high performance photosensor for the EIC.

During the first year of this project (FY18), we successfully designed, manufactured, and tested with beam particles, 1D zigzag strip readout boards in GEM and micromegas configurations. For the first time, laser ablation was used as a primary technique to build MPGD readout planes.

In FY19, we performed systematic comparisons of 1D strip configurations for chemical and laser etching for various types of avalanche schemes (GEM, micromegas, μRWELL) in a wide range of the zigzag parameter space of interest for typical EIC tracking. We designed and manufactured the first samples of laser-etched 2D readout boards. As part of the project, we also facilitated the design of a high intensity X-ray test stand at Brookhaven National Laboratory.

In FY20, we collaborated with the CERN gaseous detector workshop in building low mass and low channel count kapton-based MPGD readout board prototypes with 2D charge sharing strips and pads (see Figure 1, left). We used a fine (1.5 mil) chemical etching process for these boards, and have the detectors implemented in both quadruple GEM and μRWELL configurations.

In FY21, we tested these 2D prototypes using an X-ray scanner in the lab, and performed measurements with the particle beam at Fermilab. It was confirmed that one can achieve spatial resolution better than ~55 microns in both U- and V-projections at once (see Figure 1, right), with a relatively small remaining differential non-linearity. At Fermilab, we also tested a pair of medium size (40x40 cm^2 active area) flat micromegas 1D zigzag detector prototypes.

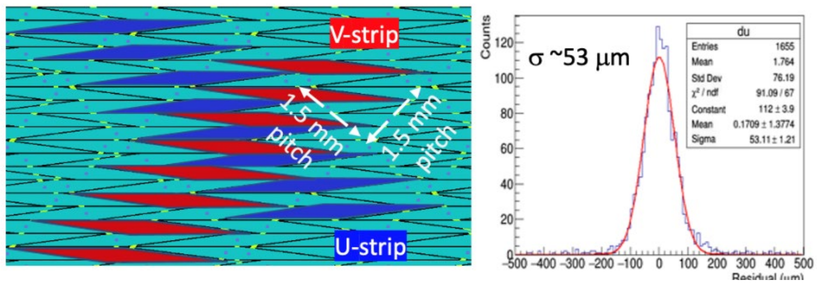


Figure 1. (Left) 2D strip layout of the MPGD readout plane with 1.5 mm pitch, realized on a two-layer kapton substrate. A pair of U- and V-strips in the top layer is highlighted in blue and red, respectively. Strip segments are interconnected by traces in the bottom layer (shown in yellow) through chemically etched blind metallized vias (small blue dots in the picture). (Right) Beam test data with the μRWELL prototype (difference between a measured U-plane coordinate and a reference proton track projection).

We built a LAPPD test stand, equipped with a semiconductor laser with focusing optics, motorized translation stages, and 256 5 GS/s digitizer channels (see Figure 2, left). We designed and built several LAPPD readout boards with a variety of square and charge-sharing pixels, and successfully performed the world's first beam test measurements with a finely pixelated capacitively-coupled LAPPD photosensor in a Cherenkov ring imaging configuration (see Figure 2, center and right). Lab measurements with segmented charge-sharing pixels similar to the ones shown in Figure 1 (left) clearly indicate that one can maintain sub-mm single photon spatial resolution with these sensors, but drastically reduce effective electronics channel count.

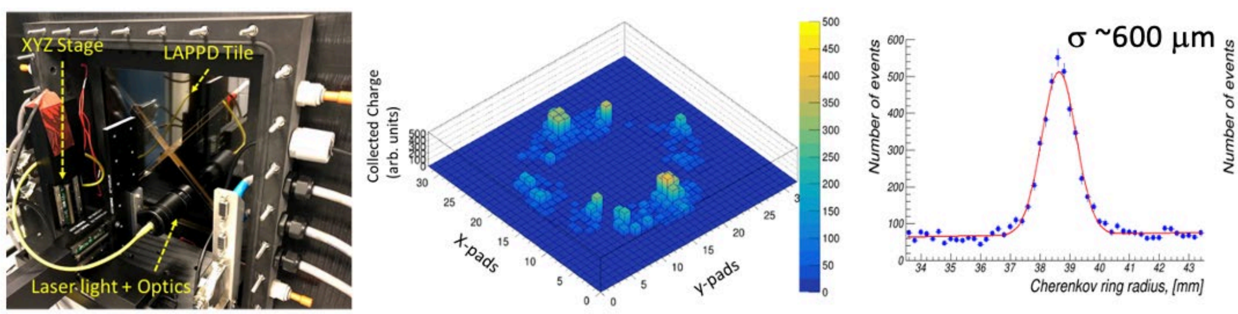


Figure 2. (Left) LAPPD test stand layout in the lab configuration (laser as a single photon source). (Center) Beam test data (a single event display, with a ~76 mm diameter ring composed from multiple clusters produced by Cherenkov photons generated by 120 GeV protons passing through a focusing quartz aspheric lens). (Right) Measured single photon ring radius distribution. ~600 micron resolution is obtained with a 4 mm square pad matrix shown in the central panel.

MILESTONES:
None. The project completed on January 31, 2021.

Finding a Lifshitz Point with the Beam Energy Scan II

LDRD # 18-036

R. Pisarski, A. Tsvelik

PURPOSE:

This project will analyze the phase diagram of Quantum Chromodynamics (QCD) as measured through the Beam Energy Scan II at the Relativistic Heavy Ion Collider. We are particularly interested in the possibility of a regime with spatially inhomogeneous phases, and an associated Lifshitz point (or regime).

APPROACH:

We began with a large N expansion in a scalar model and discovered a "Quantum Pion Liquid." In the past year, we greatly extended these results in several directions.

TECHNICAL PROGRESS AND RESULTS:

First, Rob Pisarksi studied the effects of a Quantum Pion Liquid in the equation of state of nuclear matter. With data from experiments, such as the Laser Interferometer Gravitational-Wave Observatory (known as LIGO) and the Neutron star Interior Composition ExploreR (known as NICER), it is clear that there must be a rather large, and unexpected, increase in the speed of sound, at densities which are probed by neutron stars. An ω_0 condensate, as first suggested long ago by Zeldovich, is the easiest and most natural way of getting an increase in the speed of sound. However, this must match onto the quark equation of state, so the problem is getting it to decrease. This happens if a Quantum Pion Liquid is formed, as that leads to a sharp increase in the mass of the ω_0.

Second, Rob Pisarski and Fabian Rennecke studied the phenomenological signals which might be produced by a "moat" regime. A moat regime is the precursor to a Quantum Pion Liquid. The great interest in a moat regime is that it can occur in a much larger region of the phase diagram than the standard signal, which is being searched for presently, which is a Critical End Point. We proposed measuring the ratio of two particles to the square of single particle correlations, as a function of momentum. In a simple model, we showed that the enhancement can be greater than four orders of magnitude in the signal.

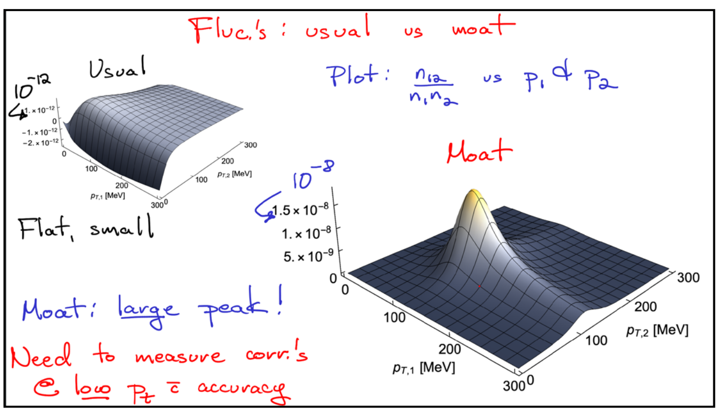


Figure 1. Fluctuations without (left) and with (right) moat spectrum.

This is illustrated above in Figure 1. Plotted is the ratio of the fluctuations in the two point function, $n_{12}(p_1, p_2)$, as a function of the momenta p_1 and p_2, divided by the product of single particle functions, $n_1(p_1)$ and $n_2(p_2)$. One sees to the left, "usual", the standard result, which is 10^{-12}. To the right, "moat", is the result with a moat spectrum. One sees the result is enhanced by 10,000 from the "usual" to the "moat."

Lastly, with Marton Lajer and Robert Konik, Rob Pisarski and Alexei Tsvelik studied the equation of state of nuclear matter in 1+1 dimensions. While the spectrum is rather involved, by looking at just the lightest excitations near the Fermi surface, we could show several remarkable facts. For a single flavor, the theory reduces to a "Luttinger" liquid. This is a free, massless boson, where the speed of propagation is less than the speed of light, with an overall coefficient which is nontrivial. Using the methods of Conformal Field Theory, the solution for the Luttinger liquid could be computed explicitly, for all values of the chemical potential, in several special cases. This included a single flavor and any number of colors, and two flavors and three colors.

We were also able to relate this to the behavior of the quarkyonic phase of nuclear matter in 3+1 dimensions. We demonstrated that for a large number of colors, there are two quarkyonic phases: one dominated by pion/kaon condensates, and one dominated by quark condensates. Our analysis in 1+1 dimensions applies to the latter and shows that it exhibits non-Fermi liquid behavior. This is extremely important for the transport properties of a neutron star, which we illustrated by computing neutrino emission.

We were very successful in our approach. We have discovered several fundamental and novel phenomena, found experimental signals in heavy ion collisions, and solved nuclear matter in 1+1 dimensions, with direct applications to 3+1 dimensions in the quarkyonic regime.

MILESTONES:
None. This project concluded in FY21.

Forward and Backward Tracking at the Electron-Ion Collider using Small Strip Thin Gap Chamber Detector

LDRD # 18-037

L. Ruan

PURPOSE:

The purpose of this project is to have measurements of the position resolution of the small strip Thin Gap Chamber (sTGC) for charged hadrons from cosmic ray and beam tests, and at the Solenoidal Tracker at RHIC (STAR) experiment. The goal is to demonstrate the tracking performance, including position resolution, momentum resolution, and tracking efficiency by performing simulation in the Electron-Ion Collider (EIC) framework, and to provide the conceptual design of sTGC disks, including the disk location, detector size, and readout for an EIC experiment.

APPROACH:

The 2015 Long Range Plan for Nuclear Science in the U.S. recommended a high-energy high-luminosity polarized EIC as the highest priority for new facility construction after the completion of the Facility for Rare Isotope Beams. For the first time in our field, the EIC will enable us to measure gluonic substructures in nucleons and nuclei. The facility will be located at Brookhaven National Laboratory (BNL) and is expected to be online around year 2030. The Principal Investigator (PI) plans to take a leading role in conducting scientific research at the EIC. In this project, the PI proposes to develop key instrumentation for the tracking system at forward and backward rapidity $1<|\eta|<3.5$ at the EIC.

The PI focuses on detecting all charged hadrons and studying the tracking performance, such as the tracking efficiency and momentum resolution. Specifically, the PI is interested in detection of electrons from the collisions. The planned research will be the first step in developing an sTGC-based detector for an EIC experiment. It will be combined with other tracking detectors to provide a complete tracking system for a future EIC experiment.

The PI is collaborating with Shandong University (SDU) in China. The high-energy group at SDU has been constructing the sTGC detectors for the tracking upgrades at ATLAS (A Toroidal LHC Apparatus), which is beneficial for muon identification. The nuclear physics group at SDU has been constructing the sTGC detectors for the STAR forward upgrades. The goal of this project is to provide a conceptual design for multiple disks of an sTGC detector as part of the forward and backward tracking for an EIC experiment.

The PI uses the sTGC design at ATLAS as a starting point. For the EIC forward tracking experiment, two layers of an sTGC module with strips perpendicular to each other will be combined into one sTGC disk, which will provide two-dimensional position reconstruction from strip charge read out. In addition, the PI proposes to do a minimal re-design of the sTGC for an EIC experiment, for example, a square shape of 60 cm x 60 cm, with a 3.2 mm strip pitch perpendicular to the wire direction. An sTGC with a square shape of 60 cm x 60 cm will be significantly easier to construct mechanically. At ATLAS, an application-specific integrated circuit is used for strip and wire readouts of the sTGC detectors. This readout option could be considered for the sTGC for an EIC experiment in the future. For a prototype, we are using the current STAR Time Projection Chamber electronics.

TECHNICAL PROGRESS AND RESULTS:
In 2020, SDU produced three sTGC prototypes with a square shape of 60 cm x 60 cm. The test at SDU showed that the efficiency was above 98% with the optimal gas, which is a mixture of 45% n-pentane and 55% CO_2. The position resolutions along the x- and y- directions were found to be 142 and 117 μm, respectively without any alignment corrections. The alignment correction will be implemented for a final technical paper. One of the three prototypes was shipped to BNL in May 2020. However, because of COVID-19, BNL was in mini-safe mode and in the phased opening, the installation and test were delayed. The prototype was eventually installed at STAR in April 2021. In addition, tremendous efforts went into the design of a system for using a flammable gas mixture safely at STAR. We passed the safety committee review and had the gas system installed for running the sTGC in Run 2021. After the run and during the summer shut down maintenance period, we found that the material used for the gas tubes in the control panel was not compatible with n-pentane. We consulted with an sTGC expert in the ATLAS collaboration and replaced the gas tubes. The gas system has been running successfully in 508 GeV p+p collisions in Run 2022 for the STAR sTGC system. We will obtain the position resolution and efficiency from Run 2022.

In the past year, we performed the sTGC tracking simulation in the EIC framework. We basically used the detector concept laid out in the EIC Yellow Report. For the tracking, in the all-silicon detector concept scenario, a silicon tracker will be used in the central barrel region, while silicon disks will be used in the forward and backward regions. An investigation in the Yellow Report studied the impact of an auxiliary tracking station in front of or behind the ring-imaging Cherenkov (aka RICH) detector. Two options were considered for the auxiliary detector: silicon with 20 μm position resolution or gas electron multiplier (aka GEM) with 70 μm position resolution. It was found that an auxiliary tracking station provided substantial benefit over none (the baseline). In our study, we replaced the GEM station with an sTGC tracking station. We found that an sTGC with 100 μm resolution provides an equivalent tracking (momentum resolution, transverse distance of closest approach) compared to the GEM with 70 μm resolution. We also found that the performance does not depend strongly on the sTGC resolution to better than 200 μm. These conclusions hold for the Brookhaven eA Solenoidal Tracker (known as BEAST) (3T magnetic field), shown in Figure 1 and BaBar (1.4T magnetic field) detector concepts.

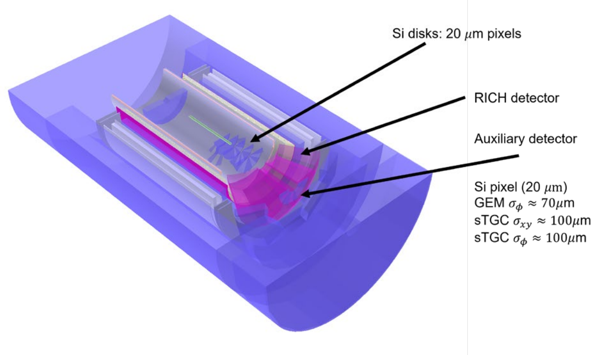


Figure 1. The BEAST detector configuration.

MILESTONES:
None. This project completed in FY21.

Ultra-fast High-granularity Silicon Sensor Technology for Photon Science

LDRD # 18-038

A. Tricoli

PURPOSE:

In various physics research areas and technical applications, there is growing demand for sensors that combine ultra-fast timing, i.e., a few tens of picoseconds, and excellent spatial resolution, i.e., at the micron level. These areas comprise timing and tracking detectors for high energy physics (HEP) experiments, photon science, and imaging. This program aims at developing a novel silicon-based sensor with excellent and unique capability in both space and time domains: Low Gain Avalanche Detectors (LGADs) that can achieve a time resolution of a few 10s of picoseconds, and a spatial resolution of a few 10s of μm, with excellent signal-to-noise ratio. This new technology could drive major advances in HEP detectors as well as in photon science, for optical and soft X-ray detection and other imaging applications, such as quantum information. We have optimized the in-house production of LGADs, and with this project, Brookhaven National Laboratory (BNL) has become a leading contributor to the development of these innovative devices for several scientific applications. In addition, we have led internationally the design and fabrication of a novel sensor that inherits the fast time property of LGADs and can also provide fine spatial resolution as a pixel detector, i.e., the Alternating Current-coupled LGAD (AC-LGAD). With this project, we have planted the seed for a long-term and strategic expansion of the R&D effort in the Physics Department and Instrumentation Division in silicon research for ultra-fast timing detectors.

APPROACH:

Together with BNL co-investigator Gabriele Giacomini, post doc Gabriele d'Amen, technician (and former student) Enrico Rossi, and several summer students, we have started a new field of research at BNL in fast timing silicon detectors and have quickly become world leaders in this field. This research is also carried out in collaboration with Andrei Nomerotski (BNL) for his expertise on photon detection; the University of California at Santa Cruz (Abe Seiden, Hartmut Sadrozinski) on LGAD design and testing; Stony Brook University (Cinzia da Via) and Prague (Stanislav Pospisil and collaborators) for their expertise on pixel readout using the commercial TimePix3 chip; Fermilab (Ron Lipton, Artur Apresyan) for test-beams and studies of new sensor designs; and the Stony Brook Chemistry Department (Thomas Allison) for applications on photoelectron spectroscopy. More specifically our approach is to:

- Develop, fabricate, and characterize fast-time LGAD sensors
- Adapt and optimize the LGAD design from current HEP applications to photons and low-energy electron detection
- Design, fabricate, and test AC-LGADs to allow simultaneously fine spatial and time resolution
- Prototype an LGAD-based fast optical camera for imaging applications.

BNL, with its specific competencies, is in a unique position to control the full process, from simulation and sensor fabrication to experimental characterization of the performance for several applications, including HEP, photon science, photonics, photoelectron spectroscopy, and imaging.

TECHNICAL PROGRESS AND RESULTS:

Previously, a new laboratory for fast-timing silicon sensor characterization was set up at BNL that has positioned BNL as a major research center for LGADs in the world. Several LGAD structures were simulated, designed, fabricated, and tested in-house, and showed excellent performance. We also led the design and fabrication of a novel concept for pixelated LGADs, i.e., AC-LGADs, that allows fine spatial resolution in addition to timing for four-dimensional (4D) detectors.

Several wafers of AC-LGADs were fabricated at BNL with varying pixel and strip sizes. In order to develop small-scale demonstrators, for the first time an AC-LGAD pixel sensor (55 x 55 μm^2 pixel size) was assembled with a fast-time and commercial readout chip (TimePix3), and an AC-LGAD strip sensor of 100 μm pitch was assembled to a dedicated application specific integrated circuit (ASIC), named ALTIROC0, that was developed for the ATLAS experiment at the Large Hadron Collider. While the assembly with TimePix3 needs further investigation, the assembly with ALTIROC0 was extremely successful, as it showed for the first time that an AC-LGAD can be read out by an ASIC and the performance is compatible with expectations. The tests were conducted at BNL with an infrared (IR) laser and beta particle beams, and an article is being written to document those results in a peer-reviewed international journal.

Further characterization of AC-LGADs with beta-particle and IR laser beams showed that a time resolution compatible with LGADs (~30 ps) can be achieved with appropriate doping of the intrinsic gain layer, as we reported at international conferences and workshops. The spatial resolution of AC-LGADs was tested with 120 GeV protons in a new test-beam campaign at Fermilab, with an improved set-up that allowed a more precise space resolution. It was found that we can reach 30 ps time resolution and few micron space resolution (as low as 6 microns) with a 100 μm-pitch strip AC-LGAD sensor (see Figure 1). The article that documents these results will be submitted to the Journal of Instrumentation.

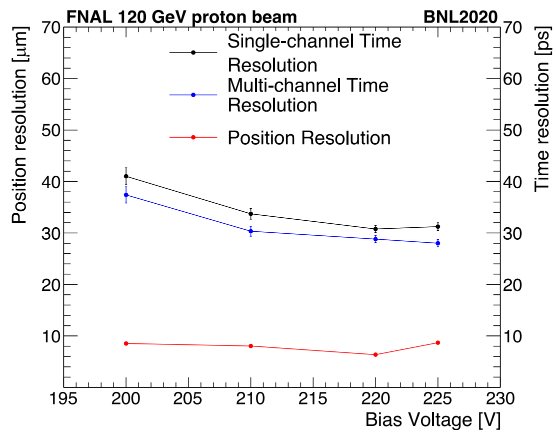


Figure 1. Measurements made at the Fermilab test-beam of time and space resolution for a BNL-made AC-LGAD sensor as functions of the bias voltage.

New AC-LGADs with optimized segmentation were fabricated to exploit the signal sharing between pixels/strips and further improve the space resolution, e.g., optimizing silicon layer doping, inter-strip gap sizes, and strip pitches. Improved designs of AC-LGADs were also studied in simulations and the first prototypes fabricated to make the built-in gain layer deeper and thinner, and in turn more radiation-hard. First results at the Fermilab test-beam facility are positive and prove that such a device is functional; however further tests are needed to characterize its performance.

The experimental tests of AC-LGADs conducted in this project proved that such sensors can be used as 4D detectors and new AC-LGAD concepts were developed aiming to make them more resilient to radiation effects and more efficient at detecting photons and low-energy particles.

MILESTONES:
None. This LDRD concluded in FY21.

Studying Confinement and Nuclear Structure through Correlations and Quantum Entanglement at an Electron-Ion Collider (EIC)

LDRD # 18-039

A. Deshpande, D. Kharzeev, T. Ullrich, R. Venugopalan

PURPOSE:

The Electron-Ion Collider (EIC) at Brookhaven National Laboratory (BNL) is a key component in the future program of the nuclear physics community in the U.S. This project addresses two distinct physic topics that will broaden the EIC's program beyond what was detailed in the EIC White Paper: (i) the study of entanglement entropy to gain insight into the mystery of confinement, one of the most fundamental questions in modern physics, and (ii) the investigation of short-range nuclear forces that originate in the underlying quark-gluon dynamics of Quantum Chromodynamics (QCD). Both studies are based on a common experimental technique, the measurement of correlations in the target fragmentation (forward) region. From these studies, the feasibility of performing such important measurements at the EIC in terms of detector and machine requirements is carefully examined and will impact the ongoing efforts in the conceptual design of EIC detectors.

APPROACH:

In diffractive Deep Inelastic Scattering (DIS), a clean separation develops between the fragmentation region of the electron and that of the nuclear target. Correlations among nucleons in the target fragmentation region have the potential to provide new insights into the underlying quark-gluon correlations that generate short-range nuclear forces. One promising idea is to look at the breakup mechanism of light nuclei (e.g., deuterons) in diffractive J/ψ meson production. Correlations between the breakup nucleons can provide us with essential information on the short-range nucleon-nucleon dynamics. The underlying cross-sections strongly depend on the final-state proton-neutron system, which can be used to probe different spatial and momentum configurations of the bound deuteron in the initial state. To study this mechanism, a Monte Carlo generator of electron+proton (e+p) and electron+nucleus (e+A) collisions, named "BeAGLE", was developed with Mark Baker (MDB Physics and Detector Simulations LLC). Here, it is vital to correctly model the deuteron in terms of its intrinsic spectral functions.

In the absence of e+A data from an EIC, data from deuteron+gold (d+Au) ultra-peripheral collisions (UPC) recorded with the STAR experiment, can be used as a proxy to test various techniques and hypotheses. Instead of virtual photons in a wide range of virtuality as produced in e+A collisions, we use almost real photons emitted from the Au nucleus to probe the deuteron structure. The observed cross-section of diffractive J/ψ production can provide valuable experimental constraints on those we expect to see at the EIC. This study will significantly improve our understanding of the deuteron wave-function in exclusive processes, which is expected to be sensitive to the short-range dynamics arising from the underlying quark-gluon correlations. Furthermore, the separate study of coherent (deuteron stays intact) and incoherent (deuteron breaks up) diffractive J/ψ production in UPCs, will further provide experimental insights into model calculations and physics extractions. For example, the coherent differential cross section $d\sigma/dt$, where t is the momentum transfer, will be directly sensitive to the gluon density distribution of the deuteron, which has never been measured before.

Understanding the origin of confinement of quarks and gluons inside nucleons is one of the fundamental questions in physics. A recent proposal is to investigate the underlying mechanism of color confinement via entanglement entropy inside a proton. However, the parton model that successfully describes the quark-gluon dynamics does not consider effects of entanglement among partons. Therefore, it is urgent to test for effects of quantum entanglement inside a nucleon. The idea is to compare the entanglement entropy derived from the region probed by the electron in DIS with the von Neumann entropy of the rest of the proton, or alternatively in p+p collisions at high energy. If there is strong entanglement, the entropy from different regions is expected to be the same. Various DIS event generators are investigated, and a global analysis of data from the Large Hadron Collider is studied in terms of entanglement entropy. The e+p DIS data from the H1 Collaboration at HERA is also under careful investigation in collaboration with Stefan Schmitt (DESY), where important information about the entropy in the current and target region can be extracted.

TECHNICAL PROGRESS AND RESULTS:
The experimental data analysis of the STAR UPCs in d+Au was finished and submitted to Physical Review Letters, and we have received very positive referee reports. Thus, this important experimental measurement of the gluonic structure of the deuteron will be published soon. This is the first time we have measured the diffractive J/ψ production off the deuteron at high energy, which constrains the leading models in describing these types of processes in such collisions. This will serve as the best experimental measurement in this kinematic region until the EIC.

Related to the exclusive J/ψ measurement, we have recently published an analysis on investigating the incoherent backgrounds of coherent diffractive J/ψ productions in e+Pb collisions at the EIC. It is an important study to show that with EIC forward detectors, we may be able to veto the incoherent events experimentally to measure the coherent diffractive events – a golden channel at the EIC for gluon imaging and saturation dynamics. This study was accepted in Physical Review D for publication.

In addition to exclusive probes, we have investigated a tagged DIS process in deuterons at the EIC, which can be used to extract the free neutron structure function. The modification of the partonic structure and the free nucleon structure are the two sides of the same coin; this follow-up study of the short-range nuclear correlations analysis from last year focuses on the other end of the spectrum in spectator kinematics: low nucleon momentum. This was recently accepted by Physical Review C for publication. Another follow-up study related to the tagged DIS in deuterons has just begun, which is beyond the scope of this project but initiated by it.

Two other analyses on the topic of entanglement have reached important milestones. First, the H1 data analysis on entanglement entropy was finally published in the European Physical Journal C, where the high-precision results have drawn attention from experts in the field and many model/theory calculations were done to compare with our data. One such paper claimed they have seen evidence for maximally entangled low-x partons in DIS based on this data. The second analysis is to investigate a new idea on spin entanglement using Bell-type inequalities of Lambda particles in high energy collisions. We have utilized the state-of-the-art quantum computer (IBM Q) to perform a quantum computation. This work was submitted to Physical Review Letters and is being reviewed. The impact of this work is that it has opened a new paradigm in studying QCD strings with parton correlations, quantum-to-classical transitions, and nucleon spin structure. A new Laboratory Directed Research and Development (LDRD) project will investigate this novel aspect using the STAR data by measuring Lambda polarization.

This project has delivered all promised physics analyses and adapted the original idea with what we have learned to new physics problems. The novel measurements and phenomenology studies also stimulated interdisciplinary collaborations, laying the experimental foundation for understanding the color confinement at the EIC. This project ended with eight high impact journal publications with already over 57 citations since 2020. There are three more analyses in preparation and planned for publication in FY22. Finally, this project has helped Z. Tu (hired as a Goldhaber Fellow) obtain a tenure-track scientist position at BNL, where he successfully secured LDRD funding for a continuation study on the EIC physics.

MILESTONES:
None. This project completed in FY21.

Physics-Informed Autonomous Synthesis of Self-Assembling Materials

LDRD # 19-001

K. Yager

PURPOSE:

Self-assembled nanomaterials hold the promise of enabling next-generation technologies by allowing researchers to easily generate precise nanomaterials. However, exploring this materials synthesis paradigm is challenging, owing to the vast space of possible material combinations and processing conditions. This project is developing physics-informed autonomous synthesis for self-assembling materials. We are advancing beyond existing work in autonomous experimentation (by ourselves and other groups) by coupling the known physics of the material under study deeply into the autonomous experimental loop, in particular using simulations to model the pathway dependence of assembly. This input is fed into a machine-learning algorithm that can make experimental decisions, which in turn controls an X-ray scattering beamline, featuring a photo-thermal platform for real-time material processing. By coupling physical models into real-time experimental synthesis and control, we will demonstrate previously impossible exploration of self-assembling materials.

APPROACH:

Self-assembly is a powerful paradigm for materials synthesis, wherein nano-components organize themselves into well-defined patterns. The Center for Functional Nanomaterials (CFN) is a leader in synthesis-by-assembly, wherein new nanomaterials are generated not by chemical reactions (conventional chemical synthesis), but by combining nanoscale components to yield new materials with distinct emergent properties. The CFN is developing paradigms for non-equilibrium self-assembly, where processing history plays a crucial role in controlling the final structure that forms. In recursive self-assembly, a self-assembled structure is used as a template to control the ordering of another self-assembling material. Applied iteratively, this allows the fabrication of complex architectures with targeted structure and material makeup. However, the associated materials parameter space is enormously large, owing both to the combinatorial complexity (the vast number of possible material combinations) and the processing complexity (the many possible processing histories that can be applied to control the material). The multi-dimensional parameter spaces underlying recursive self-assembly cannot be explored using traditional experimental paradigms. This project is developing autonomous synthesis of self-assembling materials, where real-time experimental control is used to explore materials libraries, and ultimately to control material processing in real time, in order to fabricate a target structure. The methods developed will also be applicable to a broad range of materials science problems — especially those characterized by large and complex parameter spaces.

The technical approach consists of making improvements in: 1) the experimental setup; 2) the modeling of these materials; and 3) physics-based decision-making. Experimentally, we will combine fabrication of gradient material libraries (achieved using flow-coating and electrospray deposition of polymer materials) with a novel photo-thermal annealer (laser-based temperature control) that can operate directly on an X-ray scattering beamline (the Complex Materials Scattering beamline at National Synchrotron Light Source II). Gradient libraries allow us to explore the composition spaces rapidly, while the photo-thermal platform allows us to control material processing in real time during structural measurement using X-rays. With respect to modeling, we will deploy coarse-grained molecular dynamics in a high-performance computing (HPC) context in order to generate an extensive database of material physics predictions. These will be aggregated as a prior for subsequent real-time autonomous guidance algorithms. With respect to machine-driven experimental decision-making, we will investigate complementary strategies. On the one hand, modeling priors can be fed into Bayesian models that leverage known information to intelligently suggest subsequent experiments. On the other hand, the experiments can be treated as a real-time control problem, where we tune a controller, based on known physics. Combined, the above advances will enable us to demonstrate a platform for real-time synthesis and study of novel nanomaterials. We anticipate advances in fundamental knowledge of self-assembling materials, as well as progress in generic machine-learning methods applicable to a broad range of autonomous experiments.

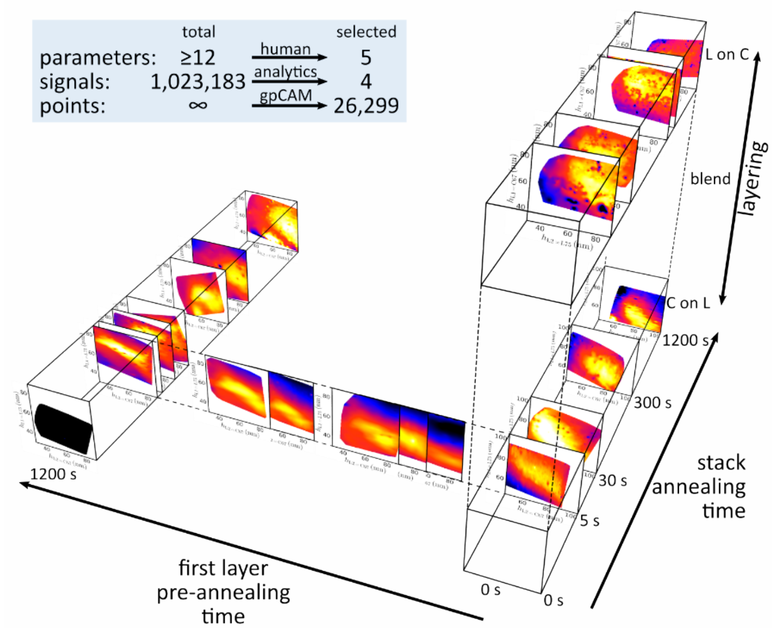


Figure 1. Outcome of an autonomous X-ray scattering experiment exploring a 5D parameter space. The space represents the full range of possible processing conditions for the selected self-assembling nanomaterial. A machine-learning decision algorithm (based on GP methods) was used to select the sequence of measurement points in order to reconstruct material ordering in this full space.

TECHNICAL PROGRESS AND RESULTS:

We previously demonstrated a coarse-grained molecular dynamics approach to simulating non-equilibrium evolution of order in block copolymers (BCP), developed a new methodology for non-equilibrium self-assembly through layering of BCP films, and performed proof-of-principle experiments where the ordering of such systems was explored by deploying an autonomous algorithm to select measurements from combinatorial samples. Recently, we have elaborated on these results considerably. We have deployed the autonomous experimentation decision-making methodology to the selection of simulations to be performed in an HPC environment. We advanced our studies of layered assembly of BCP materials, demonstrating how material selection and pre-ordering of layers can be used to generate novel non-equilibrium nanostructures. We performed a large-scale exploratory autonomous experiment at a synchrotron X-ray scattering beamline, wherein our Gaussian Process (GP)-based decision-making algorithm was tasked with exploring a five-dimensional (5D) parameter space for material ordering (Figure 1). Moreover, we deployed semi-supervised machine-learning methods to cluster the resultant dataset, allowing the underlying structural evolution in this 5D space to be probed. Combined, these results represent a landmark study in the ability of autonomous experimentation to tackle complex materials science problems.

MILESTONES:

None. The project concluded in FY21.

Physics-Guided Optimization of Quantum Gate Operations

LDRD # 19-002

M. Lin

PURPOSE:

Noisy intermediate-scale quantum (NISQ) computers suffer from noise and decoherence, which constrain the depth of quantum circuits and thus limit the implementation of sophisticated quantum algorithms. This is true for both kinds of mainstream quantum computers, such as those based on superconducting qubits (IBM, Google, and Rigetti) and those on trapped ions (IonQ). Similar to classical computing, it is very important to optimize the way quantum circuits are implemented at the low level on the given quantum architecture, known as quantum compiling. We will investigate and implement an optimizer for quantum compilers by taking into account the physics properties of the target quantum computer architecture. The eventual goal of this project is to improve the performance of the NISQ-era quantum computers.

APPROACH:

Quantum software applications are typically written either in high-level programming languages, such as Python, or in a graphic quantum circuit composer. They then are compiled to an intermediate representation, such as quantum assembly language, followed by transformation to hardware-specific instructions for running on the target quantum devices (quantum computers). On many quantum computing architectures, the hardware-specific instructions are generated as microwave pulses and electrical signals. Quantum compilers are tasked with optimizing the gate synthesis of a given input quantum algorithm, and mapping the synthesized gates to microwave pulses that correspond to the actual hardware implementation of the gates.

Our project primarily focuses on optimizing the control pulses of the quantum devices, aided by possible quantum error mitigation strategies on top to further reduce the impact of imprecise control. The time evolution of the quantum states of qubits can be described by a phenomenological Lindblad master equation. In order to solve the Lindblad master equation, we need to define the Hamiltonian that describes the quantum computing hardware. In our case, we want to optimize the pulses for the IBM Q systems. We obtain the Hamiltonian and all the necessary parameters, including the coherence error information from the IBM Q backends and encode the approximate Hamiltonian into QuTiP. We then employ the QuTiP Lindblad master equation solver to obtain the optimized control pulse amplitudes. The postdoctoral researcher hired through this project, Dr. Elisha Siddiqui, focuses on pulse optimization, while a doctoral graduate student, Yanzhu Chen, from Stony Brook University investigates the error mitigation strategies. We then test and benchmark our optimizations on the IBM Q systems provided through Q Hubs at Oak Ridge National Laboratory and Brookhaven National Laboratory.

TECHNICAL PROGRESS AND RESULTS:

In FY19 and FY20, we developed the code needed to solve the Lindblad master equation, using QuTiP to obtain optimal control pulses for our target IBM Q devices. In order to find the best optimization strategies, we explored different optimization algorithms, including Limited-memory-Broyden–Fletcher–Goldfarb–Shanno (BFGS) (LBFGS), and Simultaneous Perturbation Stochastic Approximation (SPSA). We discovered that LBFGS converges faster and gives much smaller fidelity error than SPSA. Therefore, we chose LBFGS optimization for our subsequent work. We also developed code using IBM's Qiskit OpenPulse Application Programming Interface to employ the optimized pulses on the IBM Q hardware. This requires the construction of custom single-qubit and two-qubit basis gates using the optimized pulses, which was done in Qiskit. We verified that our optimized pulses implement the target gates through quantum process tomography. We also devised an optimization method based on probability error cancellation (aka PEC) to estimate and possibly mitigate (or even cancel) certain kinds of quantum noise in a systematic fashion.

In FY21, we focused on quantifying the improvements the optimized control pulses provided to the quantum gate fidelity. We did this through two types of measurements. The first was interleaved randomized benchmarking (IRB), which measures average error per Clifford gate on random quantum circuits and also allows us to interleave our custom gates in the randomized gate sequence used in the standard randomized benchmarking. The second measurement was the measured probability of the output state of the custom gate. Since we know the theoretical probabilities of these output states for the custom gates, we can also use this as a guide to determine how well the custom gates perform. We implemented the optimized pulses for three single-qubit gates, the NOT (X) gate, the Square Root NOT ($\sqrt{X}$) gate, and the Hadamard (H) gate, and the two-qubit control NOT (CX) gate. We compared the randomized benchmarking results for the custom gates with the default gates provided by IBM and found that the optimized pulses were able to reduce the error rate as measured by IRB for the X and $\sqrt{X}$ gates, and marginally for the CX gate. The custom H gate, on the other hand, resulted in a larger error rate compared to the default H gate (see Table 1). We hypothesize this may be due to the fact that the H gate is not a basis gate on the IBM Q system. Instead, it gets transpiled into the $\sqrt{X}$ gate and two $\pi/2$ virtual Z rotations. The CX gate involves two qubits, and one of the major issues is the uncertainty in the Hamiltonian, which makes it hard to generate optimal pulses using the gradient-based methods that we implemented.

Gate	IRB error rate, custom pluses	IRB error rate, default pulses	Improvement (custom over default pulses)
X	2.0(5) x 10e-4	2.8(5) x 10e-4	29%
$\sqrt{X}$	2.4(8) x 10e-4	6.5(1.4) x 10e-4	63%
H	26(4) x 10e-4	5.0(8) x 10e-4	N/A
CX	5.6(9) x 10e-3	6.2(1.3) x 10e-3	10%

Table 1. Comparison of error rate per gate with and without optimized custom pulses as measured by IRB on IBM Q devices. Results for the X, CX, and $\sqrt{X}$ gates were obtained on the IBM Montreal system. Results for the H gate were obtained on the IBM Toronto system.

We are writing a paper documenting the results of the project.

MILESTONES:
None. The project concluded in FY21.

Building an Integrative Forecast System to Address Challenges Facing Renewable Energy Forecast

LDRD # 19-003

Y. Liu

PURPOSE:

To address the challenges of forecasting renewable energy over a range of horizons, this project aims to build an integrative weather forecast system that organically integrates physics-based modeling, data-driven modeling, and measurements. The project has four primary objectives: 1) develop a physics-based forecast system; 2) develop a data-driven forecasting system; 3) integrate the physics-based and data-driven models as an integrative forecasting system; and 4) prepare state-of-the-art high-resolution four-dimensional (4D) data and perform model evaluation to quantify the model accuracy and uncertainty.

APPROACH:

1) Develop a physics-based forecast system — Weather Research and Forecasting (WRF)-Renewable: It is well known that the fidelity and accuracy of physics-based models depend primarily on the parameterization of unresolved subgrid processes and data assimilation to enhance initial and boundary conditions. Previous studies have treated the forms of solar energy, wind, and hydropower largely in isolation; however, physical processes determining related quantities such as solar irradiance, wind speed, and precipitation are all closely interconnected. Thus, the first objective is to upgrade the widely used community WRF model for solar energy forecast (WRF-Solar) to WRF-Renewable to improve forecasting of the key variables, including solar radiation and winds. We plan to build first on the available WRF-Solar model to further improve the parameterizations of physical processes essential to improving wind forecasting, including planetary boundary layer (PBL), turbulence, shallow convection, and land surface processes. WRF-Solar is the WRF with improved parameterization of radiative transfer and warm cloud microphysics specifically for forecasting solar energy. The current parameterization schemes in WRF-Solar are mostly derived for simple flat terrains, with a low order of turbulence closure, and are thus not adequate for complex terrains involved with offshore or mountainous wind farms. We propose to explore ways to unify the treatment of PBL and shallow convection, to examine high order turbulence closure, and to improve the estimate of the turbulent dissipation rate.

2) Develop data-driven forecasting system: Complementary to improving the physical model, this approach is to learn from measurements or higher resolution/more accurate physical models that reflect the physics we wish to encode by taking advantages of the advancements in the field of machine learning (ML) and big data science.

3) Develop an integrative forecast system: The physics-based and data-driven models should properly be viewed as complements, not competing alternatives, in the domain of forecasting and prediction. For example, we expect that over short lead times (e.g., less than an hour), observational-based ML models should outperform physics-based models, while the reverse should be true for longer lead times. Thus, the optimal strategy for forecasting should be to seamlessly unite these two approaches into a single closed system for forecasting over a wide range of space-time scales, allowing the data-driven model to dominate where it is best suited, and vice versa for the physics-based model.

4) Model evaluation and high-resolution 4D data: Adequate model evaluation/verification to quantify forecast accuracy, data assimilation, and training/testing ML models all call for high-resolution, high-quality measurements. An important subtask related to this objective is to select the test sites and associated measurements. Based on the location of representative wind farms (e.g., offshore vs. land-based) and availability of quality measurements, we have focused on different types of sites over New York State, including offshore Long Island and a land-based site over complex terrain, and extended this to two sites with measurements of vertical wind profiles.

TECHNICAL PROGRESS AND RESULTS:

One set of foci was on evaluating the updated WRF-Solar in simulating surface winds under different land types, including Long Island and offshore sites, and vertical wind profiles at a coast site and a mountain site, to discern the plausible physical causes underlying wind forecast biases. It is found that the model biases exhibit diurnal variations, with details depending on land types. WRF-Solar generally overestimates winds at continent sites, whereas at the island sites where sea-land breeze circulation is active, the wind speed is underestimated during the daytime but overestimated during nighttime. Furthermore, the offshore sites exhibit generally higher wind bias and lower temperature bias than sites over land, without clear correlations between wind biases and other biases, suggesting that the large wind bias may be due to lacking wind measurements assimilated into the initial conditions used to drive the model over the ocean. A comparative evaluation with different metrics of numerical simulations of vertical wind profiles against measurements near a coast site and a mountain site further reveals different deficiencies in representing diffusive turbulent PBL processes and counter-gradient convective transport at the different types of sites. High resolution simulations were also conducted for physical understanding of small-scale turbulence effects and exploring the utility of a 4D data cube.

The other set of foci was on developing data-driven forecasting models. Key to data-driven models is their ability to recover/use the multiscale temporal and spatial variabilities hidden in observational data. We examined and compared a number of statistical and deep learning models trained with the measurements of wind speed, temperature, draw point, pressure, and precipitation of five historical days and then forecasted the wind speed of 1-12 future hours. The models examined include linear regression, polynomial regression, eXtreme Gradient Boosting (XGBoost), Long Short Term Memory (LSTM), Convolutional LSTM (ConvLSTM), Spatial-Temporal Dynamic Network (STDN), and the novel Dynamic Switch-Attention Network (DSAN). The results show different levels of forecast accuracy among these models, and the novel DSAN outperforms the other models due to its outstanding ability to capture spatio-temporal information. We also explored the potential of using data-driven models to enhance data assimilation, and of optimally blending the data-driven models and WRF-Solar for forecasting both wind and solar energy.

MILESTONES:

None. This project completed in FY21.

Integrating Multimodal Experiments using Advanced Data Analytics Developed with Microscopic Theories for Quantum Materials

LDRD # 19-008

E. Stavitski, S. Campbell, T. Caswell, E. Dooryhee, M. Hybertsen, M. Liu, D. Lu, X. Qu, J. Sadowski, D. Stacchiola, I. Waluyo, S. Yoo

PURPOSE:

This project aims to develop platforms for combinatorial material discovery, focusing on the growth of thin films of transition metal (TM) silicides. These materials have high potential significance for Quantum Information Systems (QIS). We will optimize alloy compositions and conditions of strain for superconductivity in metal-silicon (Si) thin films and search for new compositions in the MxM'1-xSi family that exhibit topological fermion states, predicted but not yet observed. Understanding structure-function relationships and driving optimization require multiple data sources, including structure at the atomic, nano, and meso scales. These silicide materials are important for the Laboratory goals in QIS, require an integrated combinatorial discovery platform, and represent a real application and baseline opportunity for the multimodal data analysis techniques to be developed.

APPROACH:

To achieve the goals of the project, several research pathways will be pursued: (1) Materials growth to synthetize combinatorial samples; (2) Multimodal characterization experiments; (3) Theory and simulation to provide large structure and computed property data sets for training the data analytics approaches and to rationalize the structure-property relationships; and (4) Data analytics development to discover the most effective tools and materials descriptors, and to deliver deep-learning-based tools for material optimization.

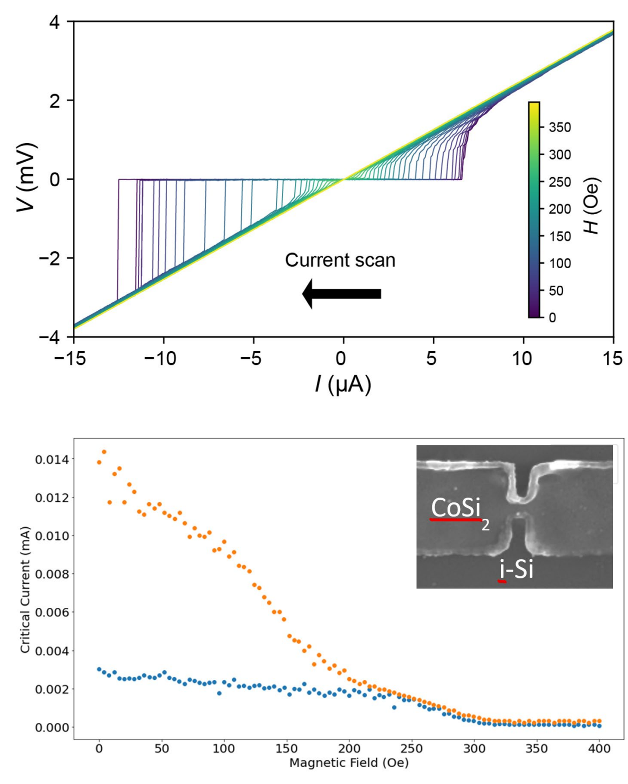


Figure 1. (Top) Voltage-Current (V-I) curves collected for a constricted $CoSi_2$ junction device, showing I_c varying over the sweeping magnetic (H) field. (Bottom) Extracted Ic – H relation shows a small oscillation. (Inset) Scanning Electron Microscope image of a constricted $CoSi_2$ junction. The constriction is about 100 nm in width.

TECHNICAL PROGRESS AND RESULTS:

New facility developments: this fiscal year, a Quantum Design Dynacool 12T Physical Property Measurement System (PPMS) with an optional He-3 insert was installed at the Center for Functional Nanomaterials. The device enables low temperature (from 350 K down to 350 mK) electrical transport measurements of various quantum materials, including TM silicides, under a magnetic field up to 12 T.

Combinatorial synthesis of TM silicides, multimodal characterization, and quantum device fabrication: through the combinatorial thin film synthesis and multimodal characterization at the National Synchrotron Light Source II with high-throughput X-ray Absorption Spectroscopy (at the Inner-Shell Spectroscopy beamline) and Wide-angle X-ray Scattering (at the Complex Materials Scattering beamline), we have determined the optimized condition for fabricating $CoSi_2$, a promising quantum material that has a superconducting transition temperature at 1.4 K and is lattice matched to silicon. Using the epitaxial $CoSi_2$ thin film on silicon as the starting material, we were able to fabricate Josephson junctions that feature either a coplanar $CoSi_2$/Si/$CoSi_2$ superconductor/normal/superconductor structure or a $CoSi_2$ constriction structure. The latter features two larger $CoSi_2$ islands connected by a narrow constriction that has dimensions comparable to the superconducting coherence length of $CoSi_2$ (~100 nm), so that the Josephson effect can be achieved (Figure 1, inset). Using the newly installed PPMS, we conducted electrical transport measurement on these devices at low temperatures under various magnetic fields. The preliminary results suggest that the fabricated $CoSi_2$ thin films are indeed superconductors. We also demonstrated that supercurrent flows through the junctions, with the critical current varying over the sweeping magnetic field (Figure 1). The I_c – H relation shows a small oscillation that is consistent with the Fraunhofer diffraction expected for Josephson junctions. Further studies are underway to confirm that the Josephson effect is observed.

Theory: a Ph.D. student, Xuance Jiang, funded by the project, performed first-principles studies to analyze the multimodal experimental data of the zinc titanate system. Research was mainly directed to unravelling the atomic structure of zinc titanate at low titanium (Ti) concentrations using density functional theory and corroborating the structure models with experimental X-ray diffraction and X-ray absorption measurements. We generated representative atomic models by creating single, double, and layered Ti dopants in wurtzite ZnO; each defect is formed by a zinc (Zn) substitution plus an interstitial oxygen. We found that there is a thermodynamic driving force for the Ti dopant to aggregate, which supports the picture that a mixture of Ti point defects and layered defects co-exist in zinc titanate at low Ti concentrations.

Simulated diffraction patterns from the structure models show that the main diffraction peak has a lower angle than $2\theta = 34.1^{\circ}$ of ZnO (002) and shifts to even lower value, as the Ti concentration increases. The qualitative trend is consistent with the experimental diffraction data. We simulated the Ti K-edge X-ray Absorption Near Edge Structure (XANES) spectra using the Ti defect structure models and compared the simulated spectra of single, double, and layered defects with the spectral component of low Ti zinc titanate extracted from the Multivariate Curve Resolution (MCR) analysis, labeled as MCR C4. We found that the simulated spectra reproduce very well the main spectral features at the pre-edge (near 49700 eV), white line (near 4987 eV), and post-edge (between 4990 and 5000 eV). Therefore, our structure analysis is validated by multimodal (X-ray diffraction and X-ray absorption) experimental data.

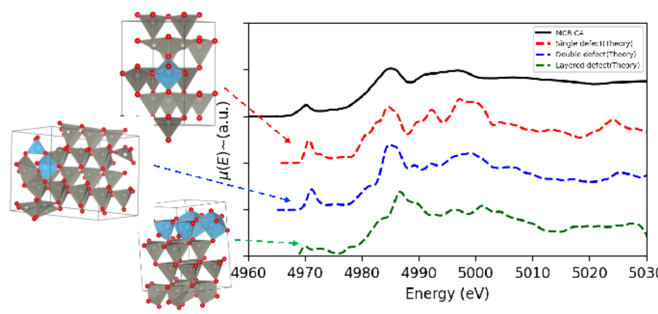


Figure 2. Comparison of the simulated Ti K-edge XANES on representative zinc titanate at low Ti concentrations with the experimental spectral component (MCR C4) extracted from MCR analysis.

MILESTONES:
None. The project completed in FY21.

X-ray Vision of Topologically Protected Bosons and Their Decay Mechanisms

LDRD # 19-010

M. Dean

PURPOSE:

Although all materials are at their basic atomic level governed by quantum mechanics, this is often hidden from view. This is seen in the widespread success of classical theories that, to varying degrees, ignore quantum mechanics, but are nonetheless often successful in describing the macroscopic properties of matter. Quantum information science (QIS) is based on storing, manipulating, and transporting information while protecting and preserving quantum coherence. Provided we can realize this, QIS is set to revolutionize many technologies. One major strategy in QIS is to exploit topology to preserve quantum coherence. Kitaev, Levin, Wen, and others devised correlated quantum models that feature non-local topological quasiparticles with emergent protected anyonic statistics. Topological band structures can be used in different ways to protect quantum information, while it is transported around a lattice. Fermionic topological band structures in condensed matter have been considered in this regard. Bosonic topological band structures in condensed matter have, on the other hand, only just been discovered. Analogous bosonic physics has already been realized in engineered metamaterial-based topological phononic band structures, which has even been used to route quantum entangled photons through a microscopic device. Fully understanding bosonic topological band structures in condensed matter might one day be used to perform analogous transport of phonons or magnons scaled down to the nanoscale. Moving quantum information encoded in bosons (as well as fermions, anyons, etc.) is also a key component for quantum computation. This project will establish a new research effort to probe bulk and surface topological bosons in condensed matter using X-ray scattering and establish Brookhaven National Laboratory (BNL) as a leader in this area.

APPROACH:

Our approach combines BNL's expertise in X-ray spectroscopy with symmetry-based indicator theories and compatibility relations in order to predict and experimentally verify topological bosons. A key element of our strategy is to exploit the state-of-the-art X-ray facilities at BNL.

TECHNICAL PROGRESS AND RESULTS:

In condensed matter physics, the age of topology has led to the discovery of Weyl and Dirac fermions in electronic band structures with great potential for new types of electronic transport and topological quantum spin liquids with novel quantum entanglement properties. In the final stage of this project, we have studied the novel quantum spin liquid $Ba_4Ir_3O_{10}$ with resonant inelastic X-ray scattering (RIXS). Our results, plotted in Figure 1, show that emergent one-dimensional (1D) spinon excitations can arise from the 2D magnetic layers in $Ba_4Ir_3O_{10}$, due to the frustrated inter-chain (intra-trimer) interactions. The highly suppressed magnetic order can be easily recovered by disturbing the subtle balance of the frustration, confining the spinons into magnons. These results indicate that, instead of forming an isotropic quantum spin liquid state, magnetic frustration can effectively reduce the system dimension, suppressing the magnetic order and realizing deconfined spinons in a unique way.

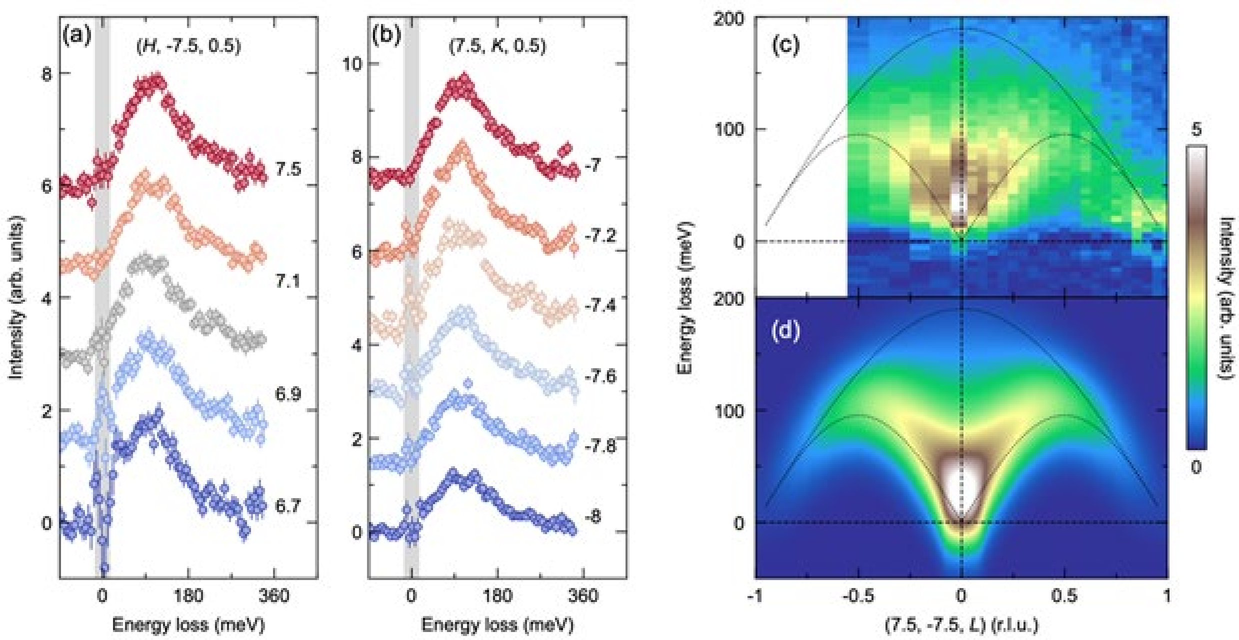


Figure 1. (a) Spinon magnetic excitations in undoped $Ba_4Ir_3O_{10}$ at 8 K. (b) Magnetic excitations at different Q along the H and K directions, respectively, showing essentially dispersionless behavior. All the RIXS spectra are presented with constant background, quasi-elastic line, and high-energy dd excitations subtracted to highlight the magnetic contributions. The vertical gray bars indicate the quasi-elastic regime, the widths of which represent the energy resolution. The values of H/K are indicated for each curve and the spectra are shifted along the y axis for clarity. (c) Color plot of the magnetic excitations along the L direction. (d) Calculated spinon excitations of an antiferromagnetic spin-1/2 chain with J_{chain} = 55 meV and Δ = 1.3, convoluted with a Gaussian profile with full-width at half-maximum of 60 meV along the energy loss axis to account for the line shape that is not fully captured by the current model. The dotted lines are the calculated excitation boundaries without broadening and the dashed lines are guides to the eye.

The "age of topology" has led to the discovery of Weyl and Dirac fermions in electronic band structures with great potential for new types of electronic transport. We have measured bulk topological phonon states and produced two published papers. We have more recently studied magnetic excitations with interesting topological properties in an iridate with a paper submitted.

Phononic Helical Nodal Lines with PT Protection in MoB_2, T. T. Zhang, H. Miao, Q. Wang, J. Q. Lin, Y. Cao, G. Fabbris, A. H. Said, X. Liu, H. C. Lei, Z. Fang, H. M. Weng, and M. P. M. Dean Phys. Rev. Lett. 123, 245302 (2019).

Observation of a chiral wave function in the twofold-degenerate quadruple Weyl system BaPtGe, Haoxiang Li, Tiantian Zhang, A. Said, Y. Fu, G. Fabbris, D. G. Mazzone, J. Zhang, J. Lapano, H. N. Lee, H. C. Lei, M. P. M. Dean, S. Murakami, and H. Miao, Phys. Rev. B 103, 184301 (2021).

Emergence of spinons in layered trimer iridate $Ba_4Ir_3O_{10}$, Y. Shen, J. Sears, G. Fabbris, A. Weichselbaum, W. Yin, H. Zhao, D. G. Mazzone, H. Miao, M. H. Upton, D. Casa, R. Acevedo-Esteves, C. Nelson, A. M. Barbour, C. Mazzoli, G. Cao, and M. P. M. Dean, submitted to Phys. Rev. Lett. (2022).

MILESTONES:
None. This project completed in FY21.

In operando Imaging and Dynamics of Two-Dimensional (2D) High Temperature Superconductor Based, Dense Q-bit Arrays

LDRD # 19-013

C. Mazzoli

PURPOSE:

This project will determine the feasibility of new Q-bits based on high temperature superconductors. Current solid-state Q-bits are essentially based on conventional superconductors (SC). This project aims at understanding the opportunities of alternative solutions represented by advanced materials, characterized by a higher superconducting transition temperature. Some limitations (coherence length) are expected, and the advanced electronic investigation by soft X-ray-based microscopic techniques proposed here will help in understanding their impact and hopefully work out mitigating strategies, in order to relax some of the constraints currently impacting applications and devices. Considering the attention on quantum supremacy and its implications, the potential impact is enormous.

APPROACH:

Quantum Information Science (QIS) is expected to revolutionize computing by enhancing information processing and security, far beyond what is allowed by current silicon-based technologies. Quantum computing (QC) is already a reality with a number of competitors from several continents investing to provide web-based end-user services. In their solid-state realization, current QC platforms are based on conventional SC, which suffer coherence time limitations (a few microseconds). This affects their scalability as only small quantum calculations can be effectively performed before error propagation takes over. After the recent advances in quantum material nanofabrication, van der Waals (vdW)-encapsulated high temperature SC (HTSC) stacks are considered of great potential to overcome these limitations. The use of complex materials and advanced fabrication techniques will require a detailed microscopic understanding of their structure and electronic dynamics before being used in large-scale applications.

X-rays represent the ideal probe for these crucial investigations. National Synchrotron Light Source II (NSLS-II) provides a unique, world-leading, set of experimental tools to image the spatial arrangement and temporal evolution of collective electronic orderings in complex quantum systems and on relevant space and time scales. Successful preliminary test experiments have been performed on both the Hard X-ray Nanoprobe (Y. Chu, X. Huang, H. Yan) and the Coherent Soft X-ray Scattering (S.Wilkins, W. Hu) beamlines, showing extremely promising results. On the sample side, we leverage the experience of our partners (P. Kim, N. Poccia at Harvard) to build hybrid heterostructures combining HTSC, insulators, and metallic building blocks in order to realize dense arrays of quantum calculation units, called Q-bits. Further electronic investigation of these unique samples with infrared (L. Carr, NSLS-II) and their advanced modeling (S. Carretta, G. Campi, Italy) will also be an integral part of this project. We aim at studying their structure, coherence, and dynamics under pump-probe and in operando conditions using NSLS-II and Transmission Electron Microscopy in the Condensed Matter Physics and Materials Science Department (Y. Zhu) to complement their investigation.

The goal of this project is to build an on-campus expertise on quantum coherence investigation in solid state advanced devices for Q-bits, and to drive a coordinated evolution of the on-site sample environments and their control in order to fully address the problem. We plan to nano-image the active electronic states in HTSC Q-bit arrays under time-resolved, in operando conditions for the first time. This will answer the question of whether dense, vdW-stacked, HTSC based Q-bits can become future QIS devices. If positive, these studies will drive their engineering development. This project will strengthen the collaboration between the institutions involved and will help establish Brookhaven National Laboratory and its partners as leaders in the field of quantum material-based devices for QIS in the years to come.

TECHNICAL PROGRESS AND RESULTS:
Unfortunately, this project was seriously impacted by COVID-19. Due to the various restrictions (work area access and travel), sample provision was a serious limiting factor. We tried to mitigate the degeneration of the available samples by storing them in our low temperature (-80 deg C) refrigerators available on site and by procuring a new glovebox for handling and local storage of the samples under moderately protective low temperature (-20 deg C refrigerator inside it). An air-sealed suitcase compatible with the beamline loadlock will also be purchased. We have already developed and tested a simple procedure, limiting the sample exposure to air down to few seconds. This is an important step for our beamline, not only for this project, as it opens investigation to a vast series of mildly air-sensitive samples, which couldn't be effectively prepared before. Most of FY21 was spent wrapping up available results as planned. This culminated in participation in an international conference, (Resonant Inelastic and Elastic X-ray Scattering 2021 (aka RIXSREXS) and a submitted paper. Another paper is in preparation that will summarize the results obtained during the project. The most relevant results are:

- First ever low temperature mapping of structural and electronic orderings in a HTSC flake and their correlation with the superconducting temperature transition.

The chase for the charge density wave signal continues, as it is at the very limit of our current capabilities. In order to extend it, the beamline procured a unique Silicon Drift Detector tailored to our needs. The detector is expected to be delivered, installed, and commissioned during the first half of FY22 and the data produced will be integral part of the final project publication.

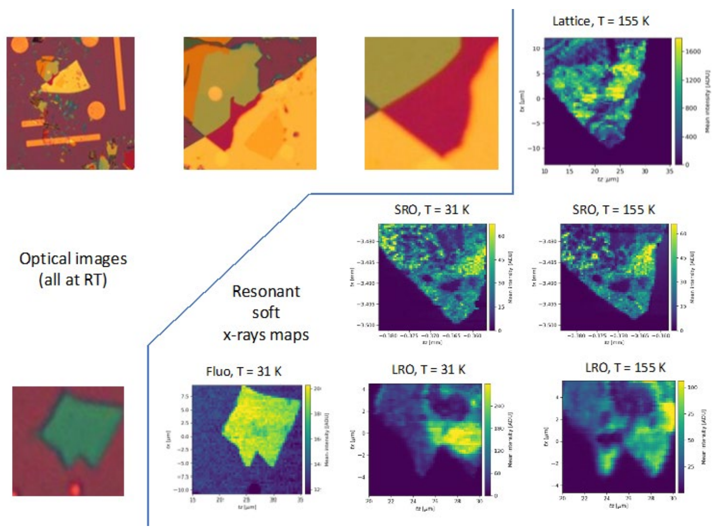


Figure 1. Optical (left) and resonant soft X-ray maps (right) taken at the Cu L3 edge of two Bismuth strontium calcium copper oxide (BSCCO) flakes. Top, thin flake (8 uc) with (bright yellow zone) and without hexagonal boron nitride coverage (light color in the center of the optical image), at growing magnification (left to right). Note the various Au (orange) markers (dots and lines) to guide the search for specific flakes on the silicon chip. Bottom, a naked small and extra-thin (3 uc) flake (note the optical and fluorescence maps correspondence). The soft x-ray signals collected (structural, Short Range Order (SRO) and Long Range Order (LRO), from top to bottom, respectively) are acquired at different temperatures below (left) and above (right) the superconducting transition temperature (Tc ~ 60 K). The evolution of the electronic correlations above a static structural landscape is evidence for the LRO signal, while the weaker and more fragile SRO signal seems insensitive to the superconducting order development.

MILESTONES:
- Glovebox use; new detector procurement, installation, commissioning, and use. Finalize data acquisition, analysis, implementation, and publication.

Linking Experiments to Algorithms for Solving Single-Particle Cryo-Electron Microscopy (Cryo-EM) Challenges

LDRD # 19-014

S. McSweeney, Q. Liu, Y. Lin, L. Wang

PURPOSE:

Our aim is to develop an automated particle picking mechanism using experimental data to determine unbiased templates for model building from cryo-electron microscopy (cryo-EM) images. We use small angle X-ray scattering (SAXS) from the National Synchrotron Light Source II beamline 16-ID (Life Sciences X-ray Scattering [LiX]) to establish a molecular envelope (a low resolution representation of the molecular structure), leveraging the expertise within the Biology Department and the Computational Science Initiative to apply deep learning to autonomously recognize all particles in the micrographs and to refine these selections so that highly accurate two-dimensional (2D) views of the particles can be created. We expect these "class-averages" will form the basis for more accurate 3D structures to be built.

APPROACH:

Single particle cryo-EM is used to determine the structure of proteins and macromolecular complexes without the need to grow crystals. Recent advances in detector technology and software algorithms have allowed structures to be determined at near-atomic resolution matching or exceeding those achieved through X-ray diffraction methods. With the assistance of New York State, and support for operations from the Department of Energy Office of Biological and Environmental Research, the Laboratory for BioMolecular Structure (LBMS) began operations for peer-reviewed experiments in January 2021.

Leaving aside the difficulties associated with obtaining pure preparations of the molecule(s) of interest and then, high contrast electron micrographs, a cryo-EM structure determination project relies on the labor-intensive process of particle picking. The quality of these particles is a major factor in the subsequent analysis. To include too many poor particles may preclude successful structure determination, while inclusion of a template based on a preconceived idea of the structure, (aka researchers' bias), may result in erroneous models being constructed.

Our initial work set out to challenge this conception by: 1) developing rapid and accurate particle picker software; these tools (we have prototypes for several methods) must be able to recognize particles in projection with little or no extra input; 2) creating a workflow building on the hypothesis that an unsupervised 2D class average may be used iteratively for gradually improving a machine learning (ML) model for highly efficient particle picking; and 3) augmenting the methods developed by creating the basis for picking from experimental data, in this case the molecular models, from SAXS measurements of proteins in solution.

TECHNICAL PROGRESS AND RESULTS:

In previous work, we demonstrated that we had developed a workflow to select particles and used them for training a three-layer convolutional neural network, implemented by using the Keras (http://keras.io) ML framework with TensorFlow (https://www.tensorflow.org) as the backend. The effectiveness of this approach was demonstrated using a number of publicly available datasets (obtained from the Electron Microscopy Public Image Archive (EMPIAR https://www.ebi.ac.uk/pdbe/emdb/empiar/) along with data collected from our instrument at LBMS.

In FY21, we sought to address some challenges in ML-based particle picking by using auxiliary measurements from SAXS experiments. The shape of protein molecules in solution determined by SAXS is used to produce a deep learning-based particle picker. The resulting particle picker can then be applied to micrographs containing the same protein, to facilitate single particle analysis and eventually lead to a 3D structure. The benefit of our approach is that a useful collection of particles can be identified with less bias and much less manual effort compared to previous approaches.

Our methodology begins with the creation of a low-resolution bead model from a protein of interest, obtained from SAXS. Interpreting this bead model as a 3D density map, we generate a dataset of synthetic 2D micrographs using many random projections of the bead model. This results in a set of images with known particle locations and orientations as the training set for a deep-learning-based particle detector built on RetinaNet. During the training procedure, we track the mean average-precision of the detector on a validation set and stop training when validation performance plateaus. After this training phase, the model is applied to real micrographs to identify particles of the protein of interest. These then serve as the input for 2D particle classification, ab initio 3D map generation. We have evaluated our approach using as a test system Rabbit Aldolase, forming an Aldolase bead model from SAXS measurements taken at the LiX beamline. Applying the trained model to a set of Aldolase micrographs yielded a large set of particles that are of similar quality to particles obtained by more manual approaches to particle detection.

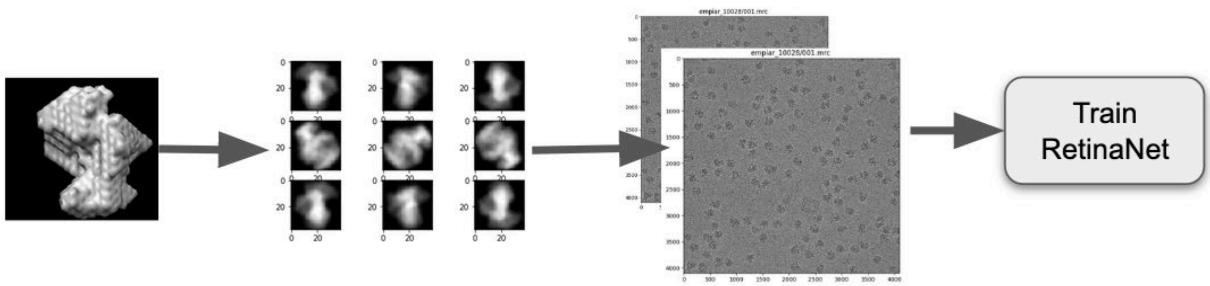


Figure 1. Schematic workflow of our approach. A low resolution model based on the SAXS measurements is used to create projections of the molecule, which are subsequently used to create synthetic micrographs that are then used to train the particle picking network. Subsequently the trained network picks particles from micrographs obtained from the electron microscopes.

MILESTONES:
None. This project completed in FY21.

Triplet-Driven Solar Energy Capture

LDRD # 19-015

J. Miller

PURPOSE:

This project will investigate ways to improve the production of triplet excited states (triplets) of π-conjugated organic molecules. Their usage is currently limited in energy-related technologies, such as organic photovoltaics (OPV). This research advances and tests the hypothesis that triplets of π-conjugated organic molecules can better harvest solar energy than the currently used singlet excited states (singlets). It seeks expanded knowledge of triplets that might aid many technologies and could lay the groundwork toward triplet-based OPV with the potential to be more efficient than the current generation of singlet-based OPV. Photo-generated spin-correlated radical pairs (SCRP) in electron donor-bridge-acceptor (D-B-A) molecules are key intermediate species in the conversion of photons to electricity. Photoexcitation initially produces singlet SCRPs (^{1}RP). A ^{1}RP can undergo spin evolution to form a triplet (^{3}RP), primarily catalyzed by hyperfine couplings, which then can recombine into local triplets. This pathway is a major energy loss mechanism in the current singlet-based OPVs. Delocalization of spin and charge can produce intriguing effects in SCRPs that are not yet understood. We hypothesize that charge recombination to triplets proceeds more efficiently when a charge can fluctuate in space. We will develop and investigate a new series of model D-B-A molecules to examine how fluctuating spatial delocalization of spin-charge affects the coherence and the production of triplets; the former is the focus of our recent Quantum Information Science (QIS) proposal. We will exploit the unique capabilities of the Brookhaven National Laboratory (BNL) Chemistry Division's Laser Electron Accelerator Facility (LEAF) at the Accelerator Center for Energy Research (ACER) to study the spin dynamics of SCRPs and the triplets, together with laser-based pump probe spectroscopy. We will implement a new experiment setup in the LEAF to couple a small electromagnet with ultraviolet-visible-near infrared (UV-vis-NIR) detection, enabling us to study the external magnetic field dependence of the spin dynamics that could disentangle the spin dynamics and charge recombination within SCRPs.

Overall, the project strengthens BNL's research in energy and quantum information sciences. This collaborative project established a basis for a joint appointment for Tomoyasu Mani, an Assistant Professor at the University of Connecticut (UConn), at BNL to collaborate in QIS and solar energy with the Principal Investigator, John Miller, and the Electron- and Photo-Induced Processes group. Long term, we are optimistic that a joint appointment based on this project and QIS efforts will receive favorable consideration for Office of Basic Energy Sciences program funding.

APPROACH:

In this project, inspired by the studies of π-conjugated polymers in OPVs, we test the hypothesis by examining how spin/charge delocalization affects the coherent spin evolution, and subsequent charge recombination (or coherence time) within SCRPs to produce triplet excited states.

Synthesis: we are developing a new series of artificial "unconventional" D-B-A type molecules where delocalization in conjugated bridges enables charges/spins to fluctuate in space. They are based on rigid ladder-type oligophenylenes. Such new molecules are currently being synthesized and characterized in the Mani laboratory at UConn.

Magnetically affected reaction yield (MARY) spectroscopy coupled with Pulse Radiolysis: we are investigating how the fluctuation of spin/charge delocalization affects the spin dynamics by conducting MARY spectroscopy coupled with laser spectroscopy (UConn) and pulse radiolysis (BNL).

TECHNICAL PROGRESS AND RESULTS:

Nitrile-functionalized ladder-type oligophenylenes: we have successfully synthesized and characterized a new series of nitrile-functionalized ladder-type oligophenylenes (L3P, L4P, and $(L3P)_2CN$). Time-resolved IR spectroscopy coupled with pulse radiolysis showed that the linewidth of the nitrile vibration bands in the radical anions of these more rigid oligophenylenes becomes sharper than those of oligofluorenes that possess flexible dihedral angles. This is only true when an electron has room to fluctuate in space. We observed a significant ~5 cm^{-1} difference in the case of L4PCN and F_2CN. Analyses of the IR bands of the neutral and radical anions species showed that the IR intensities of the nitrile bands are affected by the size of the molecules (Figure 1). In particular, the intensities of the anion species correlate with the degree of electron delocalization: the intensities become larger in molecules having more delocalized electrons; we observed up to 10,000 $M^{-1}cm^{-1}$ for the radical anion of $(L3P)_2CN$, which is the largest known for the ν(C≡N) band. We attributed these IR intensity amplifications to two processes: 1) coupling between the redistribution of charges and vibrational motion, and 2) mixing between a low-lying electronic transition and a vibrational transition (vibronic coupling). Combined, we demonstrate that the IR intensity and linewidth can also provide unique and complementary information on the nature of charges. Quantifications of IR intensity and linewidth in a series of nitrile-functionalized oligophenylenes reveal that the C≡N vibration is coupled to the nuclear and electronic structural changes, which become more prominent when an excess charge is present. The paper summarizing these findings was published (Yan et al., *Chemical Science*, 2021 12, 12107-12117).

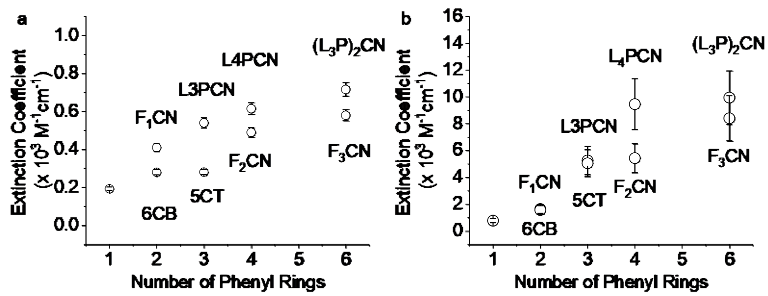


Figure 1. Extinction coefficients of (a) neutral species and (b) radical anions in tetrahydrofuran.

D-B-A Molecules: We have synthesized new D-B-A molecules based on nitrile-functionalized ladder-type oligophenylenes, and are now characterizing their optical properties.

In summary, we have continued our efforts to study SCRPs in a series of new “unconventional” D-B-A type molecules, clarifying the effect of charge delocalization on triplet formation.

MILESTONES:

None. This project completed in FY21.

Demonstration of Feasibility of Sub- nanometer (nm), Picosecond Electron Microscope for the Life Sciences

LDRD # 19-016

T. Shaftan, L. Doom, S. McSweeney, Y. Zhu, Q. Liu

PURPOSE:

Determination of sub-nanometer (nm) resolution structures of whole cells would be transformative for the life sciences. To do so with picosecond time resolution to outrun radiation damage, the ability to study cellular structures and dynamics would be truly game changing. This project aims to build on the lessons learned from earlier work with high-energy electrons for ultrafast electron diffraction (UED) to demonstrate the feasibility of achieving such a breakthrough using relativistic electrons. The project is split into phases. This project will develop a low-resolution microscope (<200 nm) to demonstrate the feasibility of this approach, taking advantage of the existing set up in the Accelerator Test Facility (ATF). This phase was complete by the end of FY20. Later phases would develop a stand-alone instrument, targeting sub-nm spatial resolution and a few picosecond temporal resolution. The goal of this project is to demonstrate imaging of thick and large objects utilizing pulsed relativistic electrons with energy of 3 MeV, but with relatively low resolution (<200 nm).

APPROACH:

This project builds on the development and completion of the Laboratory Directed Research and Development (LDRD) project "100 femtosecond single-shot electron beam slicing technology towards ultra-fast imaging." We have gained considerable experience in UED/Ultrafast Electron Microscopy (UEM) physics including in the performance of optimized quadrupole condensers for focusing – the key element in the UEM design. Such a condenser is now operational in the UED set up.

Design opportunities being addressed include the development of adjustable field permanent magnet quadrupoles, small vacuum chamber beam tubes on the order of 4 mm diameter with associated disconnect able interfaces, low-cost remote positioning of the permanent magnet quadrupoles, high magnification flag optics, and a sample chamber with goniometer and sample holder.

Other contributors to this project include: Xi Yang, Victor Smalyuk, Danny Padrazo, Bernard Kosciuk, King Wilson, Charlie Spataro, Dave Bergman, Chris Cullen, Mikhail Fedurin, Mark Palmer, James Rose, Michael Fulkerson, Frank Lincoln, and Marcus Babzien.

TECHNICAL PROGRESS AND RESULTS:

- The UEM magnet system manufacturing tolerances and field measurements were specified via start-to-end simulations. All fifteen permanent-magnet quadrupoles were adjusted and measured to meet required specifications.
- We completed the start-to-end simulation of the entire beamline, including the interaction of the electron beam with a sample.
- In FY21, we completed the UEM design, manufacture, and assembly. The UEM instrument (Figure 1, left) was installed in Building 912 (ATF) and was ready for integrated testing and commissioning in August 2021. Unfortunately, multiple failures at the UED facility prevented an adequate electron beam from being provided for UEM commissioning.

- At the beginning of the project, we determined that the UEM resolution is dominated by the beam energy spread. The UEM resolution was estimated as a function of the energy spread by numerical simulations. From this analysis, we found that jitter above 1 x·10^{-3} will significantly complicate the commissioning of the UEM. For reference, similar radio frequency (RF) gun facilities operate with an energy jitter of 2·x 10^{-4}.
- Starting in 2019, we carried out several tests to characterize the electron beam quality at the ATF UED facility. Beam-based measurements of energy jitter (Figure 1, right) produced values of 2·x 10^{-3} in July 2019 and 4·x 10^{-3} in August 2020 and May 2021.

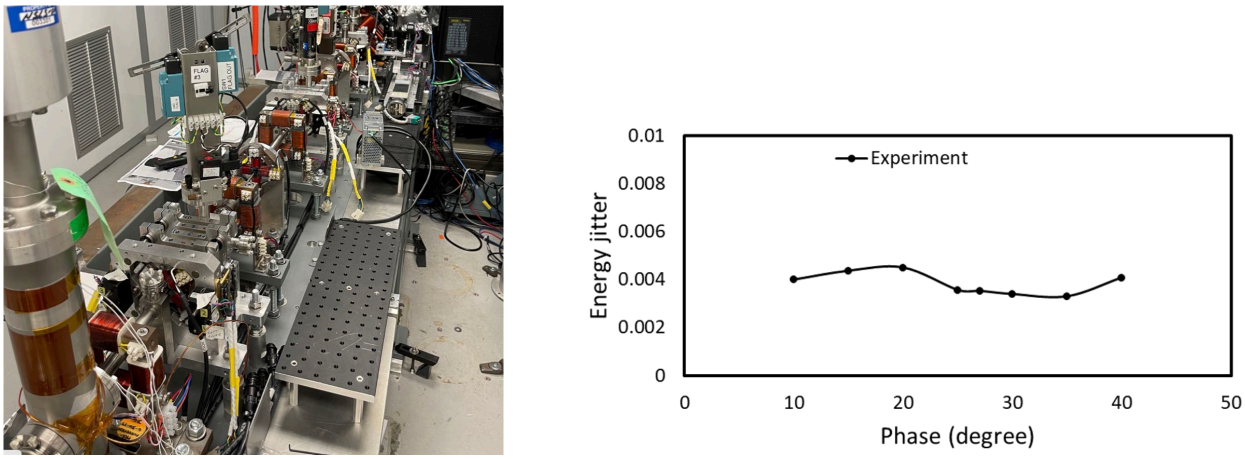


Figure 1. (Left) UEM installaton at building 912. (Right) Shot-to-shot energy jitter at 5 Hz.

- The RF jitter measurement in February 2020 showed phase jitter of >1° and amplitude jitter of 1.3·x 10^{-3}. The klystron phase jitter of 1° dominates the overall phase jitter. The klystron and kilowatt amplifier contribute similarly to the amplitude jitter.
- We have developed a detailed plan of the UEM Phase-I commissioning to determine if the ATF electron beam quality currently meets the requirements of this project. Jitter and slow drift of the beam energy at the level of 1·x 10^{-3} and beam pointing jitter smaller than 15 urad are needed.
- Although the project is closed, additional costs and schedule delays were incurred due to COVID-19.

In summary, we completed manufacturing of the UEM in January 2021, the magnet measurements and alignment in June 2021, and installation of the UEM assembly in Building 912 in August 2021. The UEM is ready for integrated testing and beam commissioning. We have a detailed plan for the UEM Phase-I commissioning to check if the ATF beam quality meets the UEM requirements. The main results of this research were published in journal articles and presented at the 12th International Particle Accelerator Conference (IPAC'21).

MILESTONES:
None. This project completed in FY21.

Cryo-Complementary Metal–Oxide–Semiconductor (CMOS): Enabling Technology for Scalable Quantum Processors

LDRD # 19-020

G. Carini, V. Manthena, M. Dabrowski, E. Raguzin, S. Rescia, J. Echevers

PURPOSE:

The objective of this project is to develop a new Application-Specific Integrated Circuit (ASIC) in complementary metal–oxide–semiconductor (CMOS) technology operating at cryogenic temperatures for quantum control and readout. The biggest challenges are: 1) the absence of simulation device models, which reliably describe the behavior of devices in extreme cryogenic operation (at 4 K); 2) avoiding interfering with the qubits; and 3) achieving the challenging low noise performance required without exceeding the tight power budget compatible with the cooling capacity of the cryogenic environment (which scales exponentially from ~1 W at 4 K to ~400 µW at 100 mK).

APPROACH:

In the Instrumentation Division (IO), we have been carrying out research on developing qubit readout ASICs. In the first step of building qubit readouts, we have characterized transistor test structures at temperatures as low as 77 K. Based on the measured results, models are extrapolated to 4 K and used to develop the design of a microwave signal generator using an LC (a parallel connection of inductor [L] and capacitor [C])-based voltage-controlled oscillator (VCO) and phase locked loop (PLL) working at 6 GHz. Based on this approach, we will be able to build small circuits operating at 6 GHz and to characterize them at 4 K. Using the knowledge on test structures and models, a full quantum control and read out circuit can be developed.

TECHNICAL PROGRESS AND RESULTS:

The test chip, an array of 133 different test structures of MOS devices with different flavors, was characterized at a temperature of 77 K. Based on the models at 77 K, an ASIC incorporating fundamental blocks of microwave signal generation, such as charge pump, frequency dividers, ring and LC based oscillators was developed. The main functional blocks are:

- A test current mode logic (CML) divider, which will divide incoming clocks up to 10 GHz by 2
- The VCO with a 6 GHz output, the microwave signal needed to control qubits
- A digital logic controls/configuration block to adjust the VCO parameters for optimization.

An ASIC was designed, simulated, laid out, and fabricated to test these functions at a temperature of 4 K. A printed circuit board (PCB) was designed to power, configure, and provide inputs and outputs to the ASIC. The Cryo-CMOS ASIC was wire bonded to the PCB in-house at the High Density Interconnect Laboratory. Figure 1a shows a picture of the fabricated ASIC. Figure 1b shows the ASIC bonded to the PCB.

Testing was then conducted for the new chip at room temperature, and all power consumption was normal. A Field Programmable Gate Array (aka FPGA) system was developed to read and write to the digital logic using two different interfaces compatible with other systems, and their operation was confirmed. A signal generator (Rhode and Schwarz SMW200A) was used to generate the GHz range signals. An artifact of the PCB substrate material having insufficient transmission properties for high-speed signals above a few GHz, was observed and impeded testing of the VCO functions above 5.5 GHz. Figure 2a shows the Cryo-CMOS ASIC testing setup.

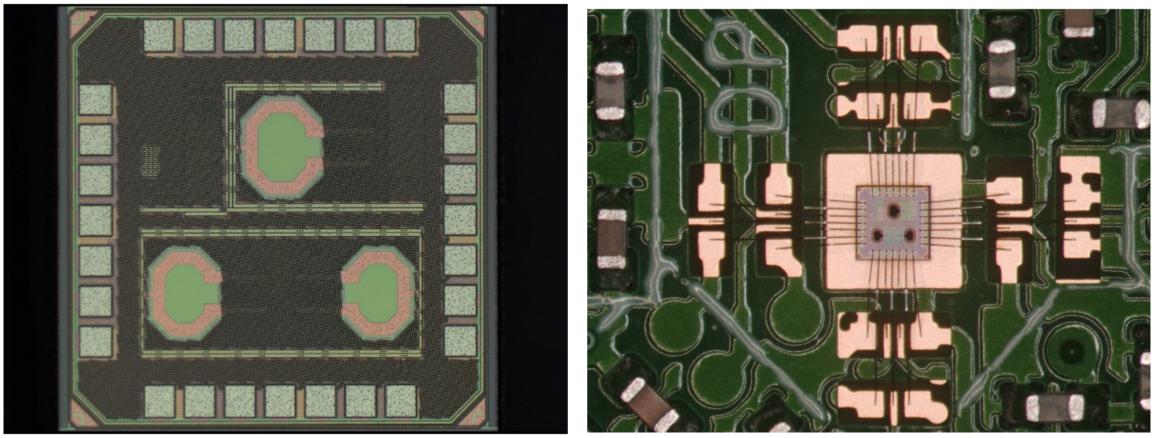


Figure 1. a) Cryo CMOS ASIC close-up picture. b) Cryo-CMOS ASIC bonded to test PCB.

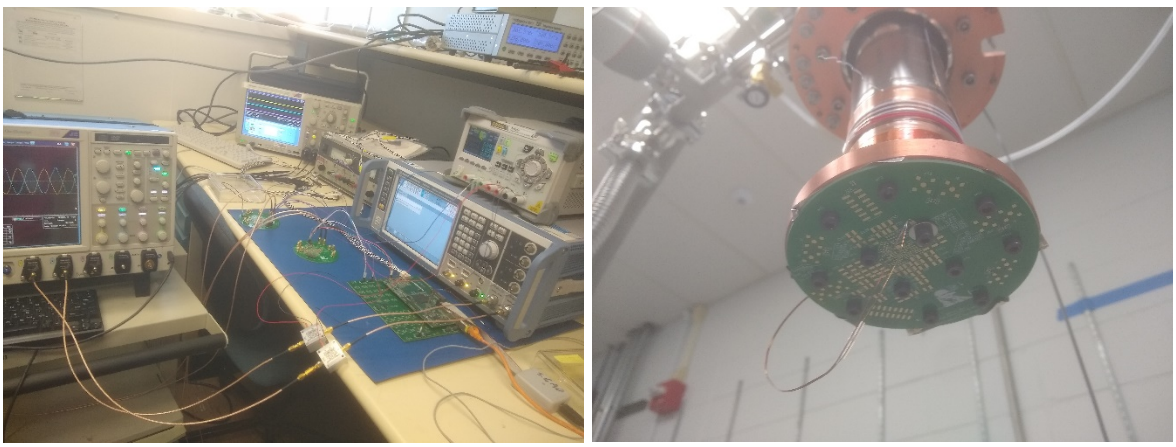


Figure 2. a) Cryo-CMOS ASIC testing setup. b) Cooldown of PCB on cryo-system.

Additionally, an Advanced Research Systems cryostat capable of cooling down to 4 K was procured and commissioned, with feedthroughs for signals up to 10 GHz. A test cooldown was done with the Cryo-CMOS ASIC PCB, seen in Figure 2b, and the ASIC area of the PCB was found to cool down only to ~14 K. With this feedback, a plan to improve the ASIC testing was developed (e.g., a new PCB with the proper high speed and thermal properties).

We have successfully carried out characterization of CMOS structures at cryo-temperatures and developed sub-circuits of microwave signal generation in the CMOS 65-nm process, which work at cryo-temperatures. We have successfully submitted a test chip with LC-VCOs, CML frequency divider, and digital logic circuits. A paper on the 6 GHz dual charge pump PLL was accepted at the IEEE Ubiquitous Computing, Electronics & Mobile Communication Conference (known as UEMCON) and a talk on the design of PLL for quantum control and readout was given. We also received best presentation award for the talk titled “A 1.2-V 6-GHz Dual-Path Charge-Pump PLL Frequency Synthesizer for Quantum Control and Readout in CMOS 65-nm Process.” The ASIC was fabricated and wire bonded, and parts of its functionality were confirmed at room temperature. No issues were found with the ASIC so far. IO now has capability for high-speed electronic design methodology and testing down to 4 K.

MILESTONES:
None. This project completed in FY21.

Silicon Drift Detector with Internal Gain and Ultra-low Noise Charge Preamplifier for Single Photon Detection

LDRD # 19-021

G. Giacomini

PURPOSE:

This project plans to develop and fabricate a detector that comprises a Silicon Drift Detector (SDD) with a small internal amplification and its dedicated readout, i.e., an ultra-low noise Charge Sensitive preAmplifier (CSA). The amplification in the sensor will boost the signal by a small factor and, since the noise will increase less than the amplified signal, single photon detection will be possible. Currently, single photon detection is achieved using Silicon PhotoMultipliers (SiPM). However, SiPMs have several drawbacks: crosstalk, high capacitance, and low fill-factor. Silicon Drift Detectors don't present these disadvantages and can achieve noise levels of a few electrons when read-out by a low-noise CSA. However, if no gain is present, single photon detection is still beyond their reach. We plan also to investigate new techniques that can increase the quantum efficiency for the detection of photons in a broad spectral range, from soft X-rays to visible light.

Single photon detectors find applications in the most disparate fields and can enable new scientific paradigms. For example, some experiments, such as the next Enriched Xenon Observatory (aka nEXO) and DarkSide, rely on the detection of ultraviolet (UV) photons. Quantum information, detection of some candidates for dark matter (e.g., axions), and ultra-high resolution spectroscopy rely on such capability as well.

APPROACH:

Currently, single photon detection is achieved using SiPMs, which are arrays of Single-Photon Avalanche Diodes (known as SPAD) that work in Geiger mode and deliver a signal with a gain of about 10^5-10^6. However, SiPMs have a number of drawbacks: crosstalk (due to the light generated during the avalanche that triggers nearby cells), high capacitance (which greatly complicates read-out and bias filtering), and low fill-factor (due to geometrical reasons).

An SDD with internal gain does not suffer from any of the SiPM drawbacks and could be a candidate for replacing the SiPMs. To do so, SDDs need a small amplification of the signal. After tests conducted during the first year of this project, we concluded that the gain layer should be placed under the entrance window.

A low-noise single channel CSA needs to be developed in parallel, to read out the small signal generated within the SDD. As commercially available technology nodes become smaller (and to some extent cheaper), we wanted to test if a low-noise CSA in Taiwan Semiconductor Manufacturing Company Ltd (known as TSMC) 65 nm technology is achievable, as smaller nodes usually come at a price of degraded analog performance.

To detect photons with small penetration depth (such as UV) with enhanced efficiency, an engineered entrance window made by nanotextured silicon can be explored. We have used a technology developed at the Center for Functional Nanomaterials, making use of nanocones. Such a window showed broadband total absorption in optical devices (black silicon).

TECHNICAL PROGRESS AND RESULTS:
Silicon fabrication of past years pointed to the fact that an SDD with a gain layer on the thin uniform entrance window can give an amplification on the order of a few tens for low-penetrating particles. The layout of a silicon wafer populated with SDDs has been designed and the process flow to be followed in the clean room defined (Figure 1, left). Fabrication has started.

The CSA after careful design (in TSMC 65 nm technology) aimed at the optimization of the input transistor to minimize the noise, was fabricated (Figure 1, right). The read-out board for the initial tests has been designed and fabricated as well. Preliminary results showed correct functionality and expected performance of both the CSA and the board. Further tests will measure the noise of a detection system made by an SDD (already fabricated by Brookhaven National Laboratory) connected to the CSA chip.

Regarding the fabrication of the broadband antireflective entrance window, nanocones necessitate a passivation layer to decrease the surface leakage current induced by the high density of defects due to the silicon etching. A layer of alumina oxide has been tested as a possible passivation method. It has been deposited on test wafers featuring diodes with nanocones in their active area. A funded project will continue such activity.

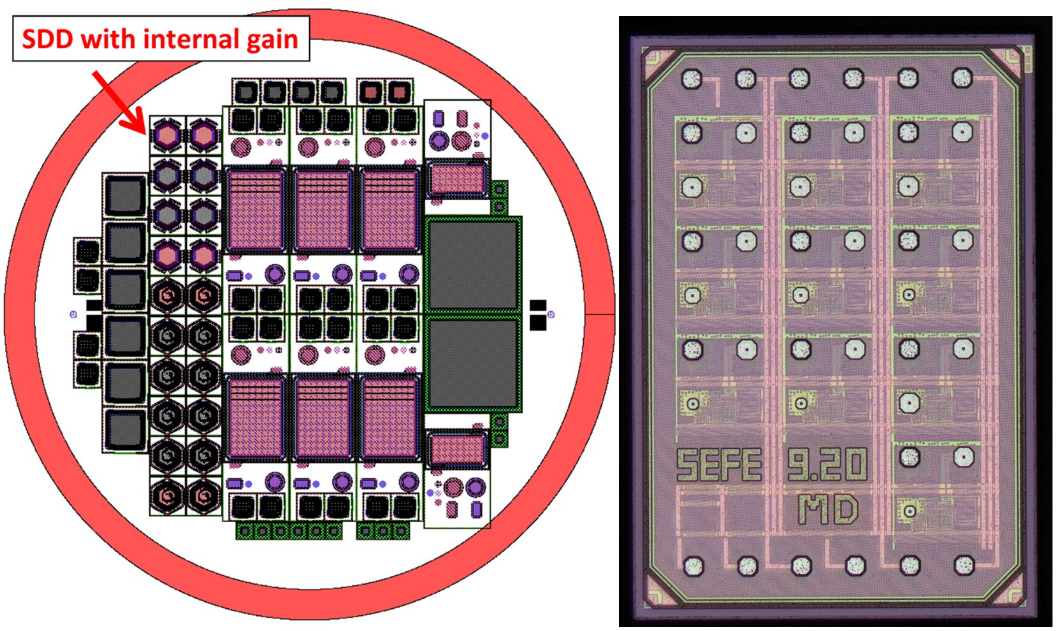


Figure 1. (Left) Layout of the silicon wafer populated with, among other structures, SDDs with internal gain. (Right) Laser microscope picture of the low-noise CSA fabricated in TSMC 65 nm technology.

MILESTONES:
None. The project completed in FY21.

Sensors and Electronics for Next-Generation Dark Energy Surveys

LDRD # 19-022

P. O'Connor, A. Slosar

PURPOSE:

The objective of this project is to develop new instrumentation and methods for exploring the physics of Dark Energy, inflation and neutrinos through observations of the cm-, mm-, and nm-wavelength emission from extragalactic sources. Following ongoing agency prioritization panels (2020 Decadal Survey of Astronomy and Astrophysics (ASTRO2020), Snowmass 2022), we expect to receive guidance towards funding to be made available to pursue investigations leading to feasibility of a large-scale cosmic survey project.

APPROACH:

Future Dark Energy discoveries will come from large-scale cosmic surveys. Current and near-future surveys use optical imaging (Legacy Survey of Space and Time (LSST)/Rubin Observatory), optical spectroscopy (Dark Energy Survey), or intensity mapping in the millimeter-wavelength radio band (Stage III Cosmic Microwave Background experiments). In the late 2020s, these surveys will be complete. This project seeks to develop hardware and software techniques that will enable next-generation surveys, targeting, in particular, the needs of a new cosmic survey using the 21-cm intensity mapping technique – an approach that is gaining recognition as having significant cost and speed advantages over competing technologies.

TECHNICAL PROGRESS AND RESULTS:

During FY19-20, the Principal Investigator and collaborators reconfigured the Baryon Mapping Experiment (BMX) radio telescope from a single-dish spectrometer to a four-dish interferometer with upgraded receiver electronics. We developed a new data analysis pipeline and implemented autonomous acquisition, processing, and archiving of observations. The facility has now operated continuously for over 14,000 hours except for periods of maintenance or weather-related downtime.

We began studying a new, highly integrated radio frequency RF signal processing platform (Xilinx ZCU111 RF System-on-Chip [RFSoC]) as a potential upgrade to the BMX receivers and as a low-cost path towards larger arrays. We submitted a whitepaper and response to a request for information from the ASTRO2020 prioritization panel, in which we outlined the science case for the Packed Ultra-wideband Mapping Array (PUMA), a large interferometric array, and cited the BMX experiment as an early pathfinder.

In FY21, data from an unmanned aerial vehicle (UAV)-based calibration campaign taken in March 2020 was analyzed. We obtained maps of the BMX angular beams across the frequency band 1100-1500 GHz in both linear polarizations. Data was acquired using a commercial UAV equipped with a broadband transmitter developed by Yale University collaborators. Beam calibrations were also performed by analyzing the passages of Global Navigation Satellite System (GNSS) satellites (GPS, GLONASS, GALILEO, and BEIDOU constellations) over BMX. The positions of the satellites, which transmit navigation data in the band observed by our telescope, are found using the orbital elements and projected on a map of the overhead sky. By comparing the predicted times of transits with the observed times the satellite signals are seen, we can get a measurement of the beam offsets from the zenith. The angular rates and predicted tracks of the satellites over BMX are shown in Figure 1.

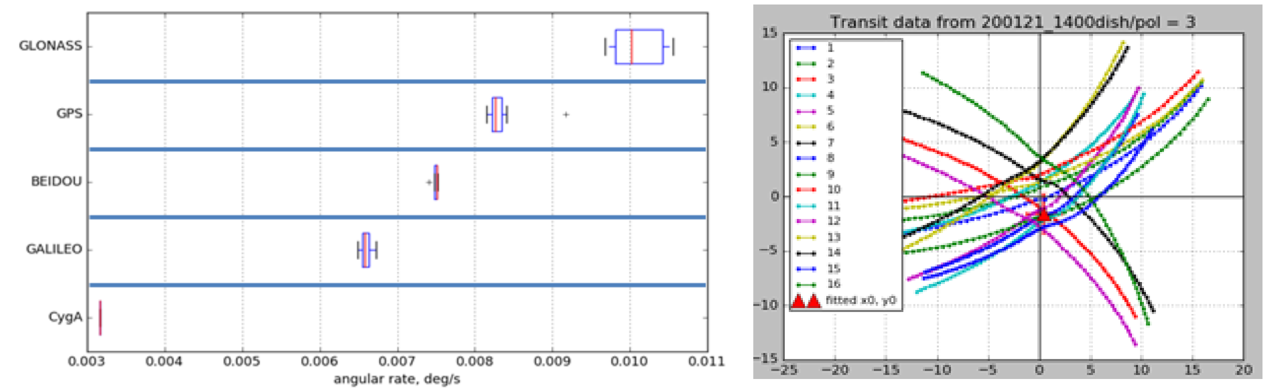


Figure 1. Results from fitting GNSS satellite observations. (Left) Angular rates by GNSS constellation (orbital altitude determines rate). (Right) Predicted tracks over BMX. Red triangle indicates beam center derived from track fitting.

The prior two beam calibration methods used single-dish signals in spectrometer mode. By interferometrically measuring the passages of the active galaxy Cygnus A as it transits above BMX, we were able to derive beam pointings and widths, as well as array geometries. For a sequence of 60 days observing, we find the fitted beam parameters repeatable to better than 0.2 degrees (RA offset and rms width) and baseline lengths to 1.3%.

To counteract the effect of computer-generated RF interference, during Feb. – May 2021 we designed a new shielded, weatherproof enclosure to house the BMX data acquisition electronics. With the new enclosure, we found a significant improvement in signal integrity and our uptime increased from 49% to 70%.

Results of our BMX calibration measurements were reported at the SPIE Astronomical Telescopes and Instrumentation conference and in the Conference Proceedings [O'Connor, Paul, et al. "The Baryon Mapping eXperiment: a 21cm intensity mapping pathfinder." *Ground-based and Airborne Telescopes VIII*. Vol. 11445. International Society for Optics and Photonics, 2020.] A more complete presentation was given at the Coordinating Panel for Advanced Detectors Instrumentation workshop in March 2021.

In FY21, we completed the construction of the BMX 21-cm intensity mapping pathfinder. We demonstrated robust operation with 70% uptime and found signal integrity and repeatability through on-sky and UAV-based calibrations (both critical for the success of future 21-cm cosmology experiments). The data logged over two years of operation amounts to around 1.6 TB, most of which has not been completely analyzed and is available to interested cosmologists and students. BMX will continue to operate in automated mode delivering 28 GB data per day through the foreseeable future and can be made available as a facility with unique characteristics for field trials of new experimental techniques.

MILESTONES:
None. The project completed in FY21.

Development of Low-Background Interconnects

LDRD # 19-023

E. Raguzin

PURPOSE:

Upcoming state-of-the-art physics detectors in neutrinoless double beta decay and dark matter detection, most notably the next Enriched Xenon Observatory (nEXO) project, as well as DarkSide, require radiopure components and electronics in the cryogenic detection area to achieve the signal resolution required. Previous experiments have required either radiopure detectors and cables that transmit the signal to warm electronics outside the detector for processing or have allowed cold electronics with commercial materials in the detector area without concern for radiopurity. The combination of both requirements means that electronic techniques that have been effective and taken for granted (Printed Circuit Boards (PCB) made from glass-reinforced epoxy laminate material (known as FR-4) chips that are encased in plastic housings, ceramic capacitors, solder, plastic connectors, and cables) are all too radioactive for these new experiments' high sensitivity requirements. The objective of this research is to develop new techniques for connecting detectors, electronics, and cabling that satisfy radiopurity requirements and electrical requirements (high voltage for detectors, current draw for electronic chips, high speed transmission for digital communication). The goal will be to demonstrate solutions to all technical bottlenecks with methods/materials that are feasible in terms of procurement, cost-effectiveness, and assembly time/yield. Brookhaven National Laboratory (BNL) is a pioneer in the field of cold electronics and given the push for low-radioactive background physics experiments on the horizon, continuing our status as an innovator and leader in the discipline will enable us to take part in future experiments and aggressively pursue funding opportunities.

APPROACH:

The nEXO experiment is a major contender for the Department of Energy (DOE) Office of Science U.S.-led ton-scale neutrinoless double beta decay experiment for the 2020s. As the effort is pre-funding, BNL's Instrumentation Division has joined nEXO as a collaborator and contributed to the extent that project and internal funding allow. The collaboration has radioassaying capabilities, and we have results for the radioactivity of solder, wire bonds, substrates, capacitors, and application specific integrated circuits (ASICs). By comparing the results of available materials to the thresholds that the detector will be held to, the low-background interconnects have been deemed a high risk component of the nEXO design during DOE reviews. While we could plan to manufacture traces on radiopure fused silica in the fashion of a PCB and wire bond the output contact of a chip to a radiopure polyimide cable that allows 1 GHz transmission speeds, none of these assumptions has ever been physically demonstrated, much less at cryogenic temperatures.

The scope of this project is to conceptualize a comprehensive list of possible interconnection methods that use only acceptably radiopure materials, prototype them, characterize them, and improve them to the point where they can be integrated into a low-background detector design, such as in nEXO. The only rigid material that meets radiopurity requirements and is cost-effective is fused silica. Similarly, the only flexible material that is feasible is polyimide (Kapton). We are most concerned with the fused silica-to-flex connection and a flex-to-flex connection to allow cable breaks, which greatly assist in detector assembly. Interconnections between electronic components, such as ASICs and capacitors to the substrate, are also within the scope.

To achieve this goal, we will develop prototypes to test one or multiple concepts of connection (wire bonding, bump bonding, compressive methods), using the actual radiopure material. We will connect the components using the Instrumentation Division's High-Density Interconnect Lab, and characterize them for robustness, test them to make sure they can survive cryogenic cycling, and analyze them for electronic properties. Since connections used for high speed digital transmission are expected to be higher risk, those properties will be tested first. With multiple iterations, we will converge on designs that fulfill the requirements, while also making the connections as dense and cost-effective as possible. We will develop the institutional knowledge and experience to understand the trace width and spacing that are needed on fused silica to achieve a consistent differential impedance at the values required over our connection method, and measure them through Bit Error Rate Tests (BERT). This bandwidth limitation will drive the electronic design of ASICs and the digital architecture for the nEXO experiment.

TECHNICAL PROGRESS AND RESULTS:
For the fused silica to flex bonding effort, we procured metallized fused silica from vendors and scientists within BNL with our desired test patterns. We tested (see Figure 1) our interconnection concepts (wire bonding, bump bonding, soldering, mechanical compression) at low temperatures and redesigned the most promising types for high speed transmission. The project ended before final assembly and measurement could be done, and we are looking for nEXO bridge funding to complete this work. Numerous lessons were learned in tolerancing and specifications when procuring these materials. This project allowed us to purchase a formic acid oven, which has shown good potential for radiopure bonding of fused silica.

In parallel, polyimide cables to test the flex-to-flex connection concepts were fabricated. During the cryogenic cycling tests, some connection methods were shown to successfully maintain a connection (the "fuzz button" and "crimp-flex" and direct compression methods). We then developed a second, refined cable design with controlled impedance differential traces. We purchased a Time Domain Reflectometer, designed a custom field programmable gate array (aka FPGA)-based BERT, and confirmed that these connections successfully passed 1 GHz signals at a cryogenic temperature.

Also, radiopure silicon resistors and capacitors were procured and characterized for performance with wire bonds/solder in cryogenics. A number of materials (components/solders/bonding, etc.) were purchased for potential radioassay. Weekly communication was maintained with the nEXO collaboration regarding projected radioactivity; estimates are routinely revised and circulated, and potential final substrate/layout designs are conceptualized based on our results.

We reached all of our milestones, aside from final assembly and measurement of the flex to quartz connection. This largely eliminated the interconnection risks of the nEXO low-radioactivity electronics and helped with a successful down select and additional BNL funding and responsibilities in the project plan. Much of our work in characterizing the transmission lines along the experiment helped enable other work to begin, for example, with ASIC driver studies.

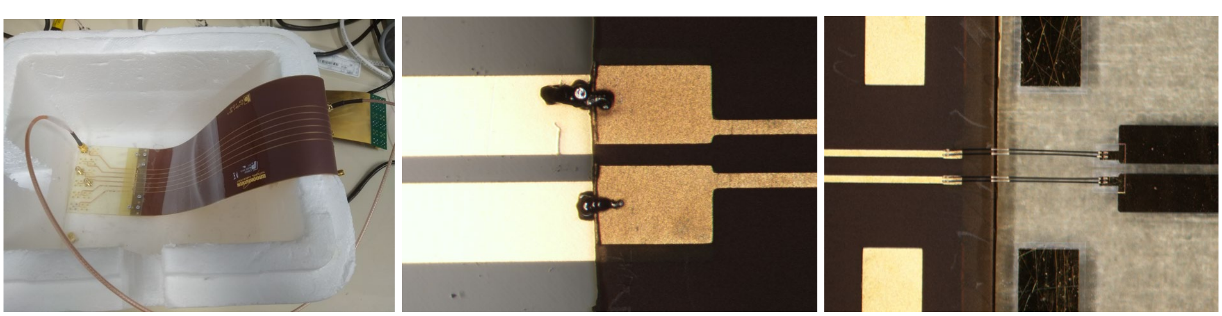


Figure 1. Images demonstrating cryogenic testing of the fused silica to Kapton connection.

MILESTONES:
None. This project completed in FY21.

Quantum Ultraviolet (UV) Sensors Based on Superconducting Nanowire Single Photon Detector

LDRD # 19-024
S. Rescia

PURPOSE:
This project aims at realizing a vacuum ultraviolet (VUV) sensor based on Superconducting Nanowire Single Photon Detectors (SNSPDs) closely coupled with its readout electronics. Currently, there are no photovoltaic type detectors capable of detecting VUV radiation with wavelength $\lesssim$ 150 nm. Such wavelengths are important for astronomy and for detecting scintillation light in liquified noble gases (liquid argon, xenon etc.) used in time projection chambers (TPC) for neutrino detection and in Dark Matter searches. A VUV large area detector/readout system could benefit Brookhaven National Lab's efforts in developing noble liquid TPCs (e.g., for the next Enriched Xenon Observatory [nEXO]), DarkSide (liquid-argon based dark matter search experiment), Deep Underground Neutrino Experiment [known as DUNE], liquid-argon based) and in developing new VUV detectors for space astronomy.

APPROACH:
SNSPDs have demonstrated very low dark count rates, fast reset times (a few nanoseconds), and very good timing resolution (single photon timing resolution $\cong$ 10 ps). We plan to develop quantum sensors for the ultraviolet (UV) using high temperature superconductors (e.g., yttrium barium copper oxide [YBCO] and bismuth strontium calcium copper oxide [BSSCO]) in collaboration with the Berggren group at MIT.

TECHNICAL PROGRESS AND RESULTS:
A YBCO sensor was characterized. It shows a superconducting transition even at a temperature of 65 K and a strong response to 1550 nm light, with a fast 13 ns response with a good signal to noise ratio.

The YBCO sensor superconducting transition temperature after patterning is still too low for operation to detect noble liquid scintillation light, and the BSSCO material has been explored as a suitable candidate to achieve a higher temperature of operation (the goal is $T_c > 90$ K for a patterned device).

BSSCO is a high temperature superconductor very sensitive to the environment. A new attempt was carried out in FY20 to build a BSSCO SNSPD. A BSSCO flake was encapsulated with UV transparent graphene and Boron Nitride (BN) as depicted in Figure 1. Once protected, the material is patterned with the helium (He) ion beam by slightly increasing its energy that goes through the protection layers (Figure 2).

The new BSSCO SNSPD shows a superconducting transition at $T_c = 83$ K, although it is hampered by an excessive contact resistance between the gold (Au) pads and the BSSCO flake (of the order of a kilo-ohm). Although the BSCCO pilot device is sensitive to optical irradiation, the high resistance of the Au pads (2.7 kΩ) limits the amplitude of the output voltage pulse and thus the sensitivity of the sensor.

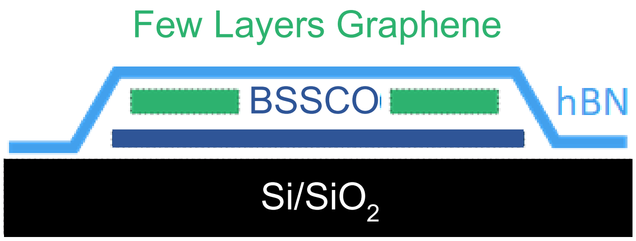


Figure 1. BSSCO SNSPD detector stack-up. Patterning is performed through the protection layer by increasing the He ion energy.

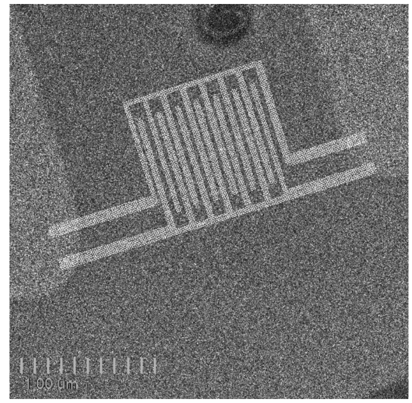


Figure 2. Patterned BSSCO superconducting nanowire device. The clearer gray areas are oxygen depleted non-superconducting BSCCO.

Among the areas studied:

- Minimize contact resistance of Au pads used to connect the sensor to the external readout electronics
- Optimize the parameters for using a He focused ion beam to pattern the "meander" structure to define the SNSPD sensing area
- Select and test electronic devices capable of cryogenic operation for sensor readout.

MILESTONES:
None. This project is expected to complete in early FY22.

Cathode Development and Deployment for the Electron-Ion Collider (EIC)

LDRD # 19-026

M. Gaowei, J. Smedley

PURPOSE:

The Electron-Ion Collider (EIC) project will involve a high current unpolarized electron source for strong hadron cooling. This project will push the state of the art for high-average current photoinjector sources, with average currents on par with and potentially in excess of the demonstrated performance of these sources (75 mA for 8 hours of operation). The goal of this project is to develop a robust photocathode that can meet the high current operation requirement for the strong hadron cooling of the EIC project. The technical goal is to be achieved both by engineering cathode material with larger grains and better longevity and by adapting a commercial delivery option to the needs of the EIC to realize large volume, inexpensive commercial production.

APPROACH:

The Relativistic Heavy Ion Collider (RHIC) at Brookhaven National Laboratory (BNL) is the nation's premier quantum chromodynamics research facility. Beginning in FY19, RHIC implemented hadron cooling using electron beams to improve the luminosity; further, such a cooling scheme is anticipated for all EIC projects at BNL. While the details of the electron coolers vary, one common aspect is the requirement for a high average current photoinjector, with most designs requiring >100 mA. Alkali antimonide photocathodes have demonstrated high average current at Cornell University; however there is very limited operational experience in how long such a cathode can operate before needing a replacement. The current estimate is that this cathode may need replacement every day, which leads to a practical engineering challenge to produce robust, reliable, and reproducible photocathodes in sufficient quantities.

There are three technical approaches planned for this project: the first is to utilize the unique capability of the state-of-the-art synchrotron X-ray techniques at BNL to develop large grain semiconductor photocathodes and characterize the longevity of the material. Second, we will leverage our Nuclear Physics Small Business Innovation Research-funded efforts (by members of this team) to develop commercially produced accelerator photocathodes in self-contained vacuum systems. Third, we will deliver the encapsulated photocathodes into the electron cooling systems, such as Low Energy RHIC electron Cooling (LEReC) and Coherent electron Cooling (CeC) at RHIC and test the gun with these cathodes for high current performance.

TECHNICAL PROGRESS AND RESULTS:

In FY19, we compared the sequential growth method with the co-evaporation method for a cesium telluride photocathode and proved the latter produces photocathodes with better stoichiometry, larger grains, and higher quantum efficiency. The LEReC alkali antimonide photocathode recipe was characterized along with its heat decomposition process. In FY20, we discovered the nucleation of cesium telluride on graphene substrates. The comparison of co-evaporated cesium telluride layers on a silicon substrate and on graphene reveals that cesium telluride nucleates at an earlier stage on graphene under the same growth conditions. Progress in the design and construction of the growth and sealing chamber of the sealed capsule was significant. The hardware design of the emittance measurement system progressed.

In FY21, we had one beamtime session at National Synchrotron Light Source II and discovered evidence for the nucleation of cesium antimonide (Cs_3Sb) photocathode material on hexagonal silicon carbide (SiC). In situ and real time X-ray characterization study was performed to reveal the compositional, structural, and surface evolution of the growth process. The film was grown to near optically dense thickness, and X-ray diffraction revealed the alignment of the cubic Cs_3Sb to the substrate peaks of the 4H-SiC single crystal and the formation of large grains. X-ray reflectivity revealed that the resulting film is remarkably smooth. High quantum efficiency (QE) was measured for this cathode, with a peak QE at 18.2% and a green QE of 7% at 532 nm. Figure 1 shows the diffraction pattern captured by an EIGER detector for a 20 nm thick, highly textured Cs_3Sb film on the 4H-SiC substrate (left) and the integrated X-ray Diffraction (XRD) plots comparing the Cs_3Sb film on SiC and on Si (right), indicating a much better crystallinity of the former. The grain size of the Cs_3Sb cathode is estimated to be 10 nm for a 20 nm total film thickness.

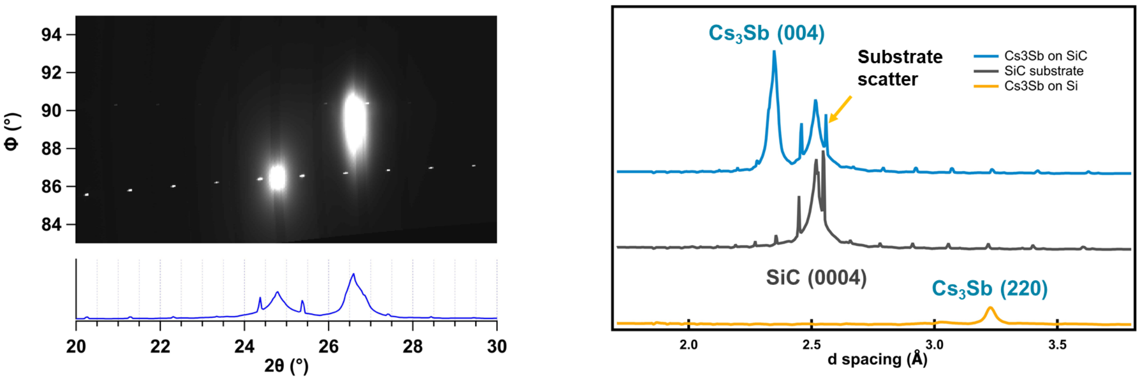


Figure 1. (Left) The diffraction pattern of a 20 nm thick, highly textured Cs_3Sb film on the 4H-SiC substrate. (Right) The integrated XRD plots comparing the Cs_3Sb film on SiC, the bare SiC substrate, and a similar film grown on Si.

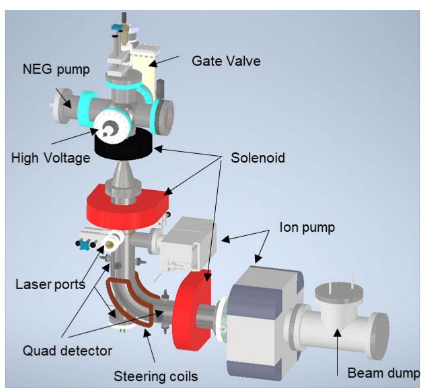


Figure 2. Hardware design of the CeC compact DC gun.

In FY21, the assembly of the growth and sealing chamber of the sealed capsule was completed. The transfer procedure for the sealed capsule from the growth chamber to the sealing chamber was established. The emittance measuring capability was developed as part of the CeC compact direct current (DC) gun for testing photocathodes in high current conditions (Figure 2). During the emittance measurement, only the second solenoid located 45 cm away from the cathode is used to perform a waist scan and find the emittance. The emittance measurement is not completed due to the delayed construction of the CeC compact DC gun.

MILESTONES:

None. The project completed in FY21.

Real-time Particle Tracking with Deep Learning on Field Programmable Gate Arrays (FPGAs)

LDRD # 19-027

V. Cavaliere, S. Yoo, M. Begel, S. Tang, J. Huang, W. Kalderon, H. Abidi

PURPOSE:

Hardware-based trigger systems, which lie at the heart of hadron collider detectors, must make physics decisions within a few microseconds to ensure good data is written to tape. Field Programmable Gate Arrays (FPGA) are the preferred technology for these high-throughput low-latency situations and offer flexibility as they can be updated to meet new experimental needs and can incorporate the latest technological advances.

Charged-particle tracking is one of the most important aspects for event reconstruction at hadron colliders enabling an early rejection of background and more signal-like events written to tape. Silicon-based, high-granularity tracking sensors detect ionization charge deposited by particles as they propagate through the magnetic field of the detector. Tracking algorithms use this information to measure the curvature of particle trajectories and thus deduce the particles' charge and momentum. The capability to reconstruct detached vertices is an essential tool, for example, for identifying b-jets in all cases when the new particles have a preferential decay to heavy quarks, like the Higgs boson.

In this project, we are exploring the applicability of advanced machine learning algorithms to reconstruct tracks in FPGAs at high data rates and with low latency. In the process, we will acquire crucial knowledge of ultra-fast different machine learning algorithms to be deployed on hardware architecture, such as FPGAs, and establish Brookhaven National Laboratory (BNL) as a worldwide leading center for this research. The competencies developed will be critical to the Lab for the trigger design at facilities of the next generation.

APPROACH:

We plan to solve the high-throughput low-latency particle track reconstruction problem for ATLAS upgrades and other detectors, by using deep learning, which is accelerated by FPGAs. We are targeting two steps of tracking reconstruction.

- A Neural Network, that based on hit coordinates (vector of x/y/z position coordinates) in the detector, provides rejection against fake hit combinations
- A second network that uses the hit coordinates and a set of initial seeds in the innermost layers to determine the coordinates of the hits in the subsequent layers.

Several network architectures are being studied. Neural networks are typically over-parameterized with extremely huge number of weights and operations. Since reducing the weights can significantly reduce the memory cost and the arithmetical operations, pruning will be studied. More in detail the expected results will include:

- A high-speed FPGA program of a deep neural network (DNN) suitable for real time particle tracking of large-scale streaming data
- Successful execution of this project that will lead to BNL being the main production site for development of the High Luminosity Large Hadron Collider HL- LHC) track trigger boards and that will be useful for sPHENIX track-trigger implementation.

TECHNICAL PROGRESS AND RESULTS:

We designed a first low precision neural network, which classifies tracks as true or fake using a DNN framework. The inputs are hits, which belong to a track seed extracted with a standard Hough Transform algorithm. The NN score (shown in Figure 1, left) is extracted and an overlap removal algorithm is implemented. The output tracks are compared, if they shared more than 5 hits, the one with the lowest NN score is selected.

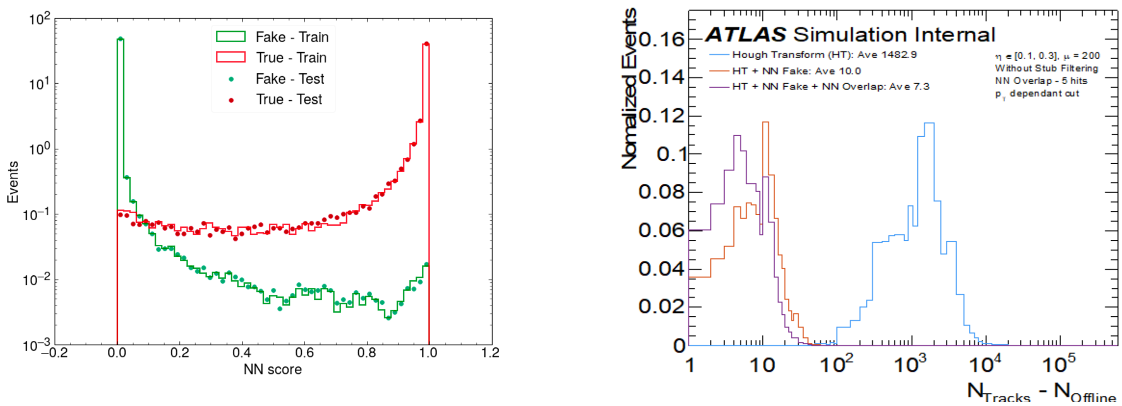


Figure 1. (Left) NN score for the network that classifies tracks as being either true or fake. (Right) Number of tracks in output of the first step of tracking without (blue) and with the application of the NN (red and purple). Lower number of tracks means less resources needed for the subsequent steps of tracking.

These steps significantly reduced the number of combinations that need to be fitted in the subsequent steps of the tracking algorithm as shown in Figure 1, right.

To run NN inference on an FPGA, we need to quantize and synthesize it. The hls4ml framework is used for this purpose. QKeras is used to train with quantized-aware nodes: weights and bias are quantized while training to allow for a translation on an FPGA without any loss. The first estimate of the resource usage of the NN inference for an Alveo U250 card shows that the NN does not take significant space on the FPGA, even without optimization.

Based on the work done with this project, ATLAS decided to change the scope of the tracking at the trigger level for the HL-LHC program. This NN has become part of the new baseline tracking algorithm for the Event Filter Tracking and BNL is among the leaders of the project.

We are working now on a second network that would replace the standard Hough Transform Algorithm to select the track seeds to be fed to our overlap removal network and subsequent fitting. We studied different architectures for the NN and decided to use a Recurrent Neural Network. First studies show that we can predict the coordinates with small errors.

MILESTONES:

- Deploy the compressed neural networks on FPGA nodes and test on the large-scale dataset.

High-Throughput Advanced Data Acquisition for Electron-Ion Collider (EIC), Particle Physics and Cosmology Experiments

LDRD # 19-028

J. Huang

PURPOSE:

With the advances in high-speed programmable electronics, the paradigm of data acquisition (DAQ) for large-scale physics experiments is transforming from the traditional event-oriented triggered DAQ to time-stamped streaming DAQ with real-time digital data processing. The goal of this project is to prototype the next generation continuous and dead-timeless DAQ, which features a data recording rate of about 100 Gbps, roughly one order of magnitude higher than the current experiments at the Relativistic Heavy Ion Collider at Brookhaven National Laboratory (BNL). This development will be the critical step to establish the foundation for the DAQ infrastructure for Electron-Ion Collider (EIC) experiments, and it will also be applicable to a wide spectrum of projects at BNL, including the Deep Underground Neutrino Experiment (DUNE) detector, the next Enriched Xenon Observatory (nEXO), and a 21-cm radio interferometer (the Packed Ultra-wideband Mapping Area Array [aka PUMA]). The Front-End Link eXchange (FELIX)-based DAQ in these applications will minimize the custom hardware design and simplify the system architecture to utilize commercial components as early as possible. It will also provide a flexible and scalable interface to different far detector technologies with the option to include hardware compression and buffering without incurring excessive cost.

APPROACH:

We will develop a firmware, timing, and data distribution concept with prototype DAQs based on the BNL-led hardware development of the FELIX card series for the above applications. The FELIX card bridges customized detector front-end electronics and commodity computing. Such architecture is planned to be used in many major Large Hadron Collider detector upgrades from the early to mid-2020s. Using the existing sPHENIX tracking and calorimeter detector test stand as a platform, we will develop the timing and FELIX firmware, demonstrating a full stream readout system that will allow recording all detector hits without deadtime or trigger bias at the EIC. Meanwhile, a FELIX based-DAQ test bench is being established for a DUNE DAQ system demonstrator. It focuses on data flow of detector readout, hit-finding in FELIX firmware, interface to the software-based trigger primitive selection, and data buffering.

TECHNICAL PROGRESS AND RESULTS:

Since the project started in June 2019, we have developed the FELIX-based DAQ concept for the EIC. For the DUNE application, we demonstrated a full chain from front-end electronics through the FELIX server to the high-speed switch. New lines of investigation also started in the generic streaming digitizer based on the recently available Radio-Frequency System-on-a-Chip (known as RFSoC) chips and Artificial Intelligence-based real-time data reduction for the FELIX streaming DAQ. We continued these tasks in FY21, with high level progress summarized below:

- EIC DAQ: we achieved the major goal of establishing the foundation for the DAQ infrastructure for the EIC experiments. The FELIX-style DAQ system designed by this project was referenced in the EIC Conceptual Design Report and Yellow Report, as well as being used as the reference DAQ design in all three EIC detector proposals: ATHENA, CORE, and ECCE detectors.

- Artificial Intelligence/Machine Learning in DAQ: in the FELIX-style streaming DAQ system at the EIC, sufficient bandwidth can be provided to readout detector signal, noise, and background hits. We quickly realized that the main challenge is to reduce the data sufficiently and safely, so they fit into the bandwidth for permanent storage. In collaboration with the Computational Science Initiative and the University of California, Los Angeles, major progress was made this fiscal year in designing and demonstrating a Machine Learning-based data compression based on a new algorithm named Bicephalous Convolutional Autoencoder (BCAE) as shown in Figure 1. We published our first paper on this topic [arXiv: 2111.05423] and gave four invited talks.

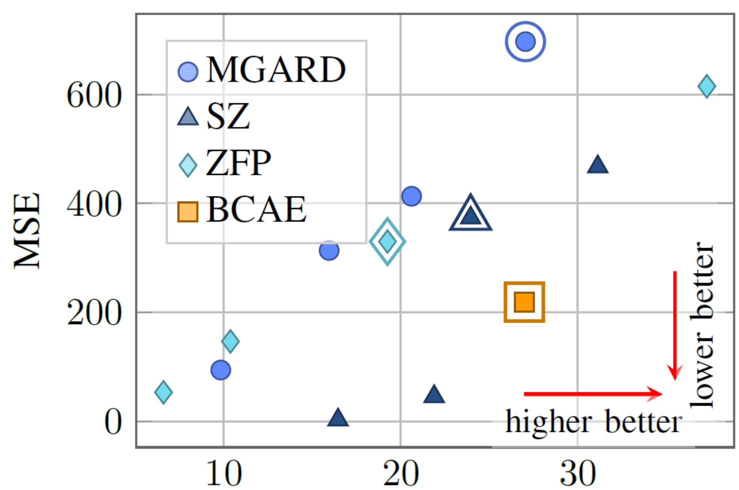


Figure 1. The BCAE algorithm (yellow square) demonstrated superior performance over traditional lossy data compression, as given by mean square error (MSE) vs compression ratio on the emulated sPHENIX FELIX streaming data from the Time Projection Chamber detector [arXiv: 2111.05423].

- DUNE-related FELIX firmware development: in collaboration with the DUNE project, we continued to support DUNE DAQ requests for FELIX Phase-II firmware features and improvements to ease system integration. Progress was achieved on multiple fronts, including a programmable number of chunks were combined, the mainstream baseline was stripped down, the trigger primitive was generated, and the FELIX RegMap was optimized.

- Next generation FELIX Phase-II hardware: in collaboration with the ATLAS FELIX project, we supported Nikhef (the Dutch National Institute for Subatomic Physics) performing studies of Xilinx Versal Prime resources, to understand how the current FELIX firmware can be migrated to the FLX181 board for Phase-II. We also supported the 25 Gb/s links development, with bug reports, fixes, tests, and benchmarks of Nikhef's Core1990 soft-core implementation of the Interlaken high-speed serial protocol for the ATLAS Phase-II Trigger and Data Acquisition system (aka TDAQ) Upgrade.

MILESTONES:

FY22: ending June 2022

- Demonstration of real-time high throughput data compression in Artificial Intelligence.

Single Atom Barium (Ba)-Identification (ID) for the next Enriched Xenon Observatory (nEXO) Using Electron Microscopy

LDRD # 19-029

J. Warren, M. Chiu

PURPOSE:

This objective of this project is to develop the technique for identifying single atoms of barium (Ba) using Scanning Transmission Electron Microscopes (STEM) for the next Enriched Xenon Observatory (nEXO) experiment. In nEXO, we are searching for neutrino-less double beta ($0\nu\beta\beta$) decays of ^{136}Xe, which leave a remnant Ba++ ion. Being able to capture that remnant atom after a decay, and identifying it as barium, would enable nEXO to unambiguously identify that the decay event came from ^{136}Xe, rather than a background decay such as uranium (U) or thorium (Th). Being able to do this "Ba-Tagging" of decays makes the experiment essentially background free, and vastly improves the sensitivity of the experiment. The discovery of $0\nu\beta\beta$, would for the first time, prove that the neutrino is a Majorana particle, and that lepton number is violated. All of these discoveries would rewrite the fundamental laws of physics regarding the neutrino.

APPROACH:

Ba-Tagging was proposed over 28 years ago as a way to make a nearly background-free measurement of the double-beta decay of ^{136}Xe, which would vastly improve the sensitivity of a $0\nu\beta\beta$ search. Since then, there has not been a successful demonstration of Ba-Tagging, despite concerted effort by several groups. Part of the difficulty lies in being able to produce a unique signature of Ba at the single atom level with enough signal to unambiguously identify the Ba, and that the signal is consistent with the expectation from a single atom of Ba. Another difficulty lies in being able to devise a system that is inherently free of contamination, particularly since Ba can often be embedded inside bulk materials.

To solve both the contamination and single atom identification challenges, we proposed using existing, high-performance STEM to image and identify a Ba atom captured onto a graphene sheet. Because graphene is a single layer of carbon, it avoids many of the contamination issues found in bulk substrates. The thin material also makes it ideal as a substrate for transmission electron microscopes, which have in recent years been developed by commercial companies, such as Hitachi and Nion, to provide almost turnkey capability in imaging and to identify single atoms very robustly.

TECHNICAL PROGRESS AND RESULTS:

We have recently been able to achieve very robust imaging and identification of single Ba atoms adsorbed on graphene using a Hitachi 2700C STEM. There are two techniques for the identification – Energy Dispersive X-ray Spectra (EDSX) and Electron Energy Loss Spectroscopy (EELS). Our analysis of the efficiency of the two techniques shows that EELS is a more powerful identification technique for barium. The analysis also gives us quantitative estimates for the amount of time required to perform the identification, input that is essential to designing the Ba-Tagging system for nEXO. From here, the next stage is to work on the ability to capture the Ba ion onto a test probe while in liquid xenon (LXe).

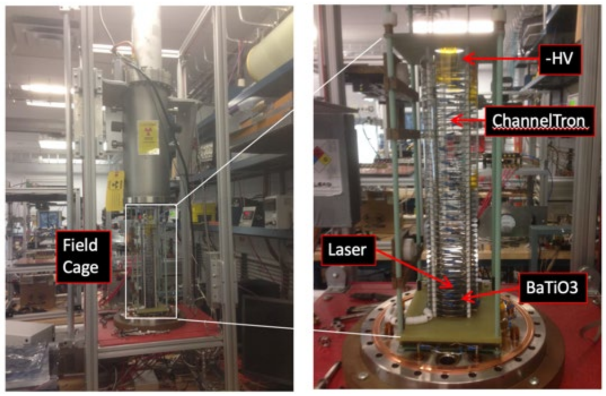


Figure 1. Vacuum Ba beam system developed at Brookhaven National Laboratory for testing the efficiency of Ba attraction onto various capture probe designs. Once successful, the next step is to do the same tests in LXe.

A vacuum Ba beam system was developed to test our capture probe designs, as shown in Figure 1. The Ba beams are created through laser desorption on a $BaTiO_3$ target, and guided by the electric field cage, where they are detected by the channeltron or captured on our test probe. The beam system was simulated inside the COMSOL package. The probes were also designed within COMSOL, and many iterations of designs were made until we had good optimization. Various prototype probes have been built and tested, work which is currently on-going. When the capture probe is built and successfully tested, we will move to doing the same tests in LXe.

Prior year milestones are as follows. In FY19, we measured the EDSX and EELS from isolated Ba atoms to establish the strength of the Ba identification capability using these signals. In FY20, we designed an attractive probe to electrostatically attract the Ba ion in LXe. The probe consists of a device made from suspended graphene, with leads to apply an electric bias to it. We also built a test setup to create a beam of Ba ions in a vacuum system for testing the attractive probe. In FY21, we worked on publishing a paper on the capability of STEM to do single Ba atom identification, using measurements made in FY19. We also developed simulations of the probe in the vacuum system and measured the efficiency of the attraction and identification process for single Ba atom imaging using STEM via simulation.

MILESTONES:
None. This project completed in FY21.

Towards Brookhaven National Laboratory (BNL)-Stony Brook University (SBU) Scalable Quantum Processing and Network Enabled by Fast Imaging of Single Photons

LDRD # 19-030
A. Nomerotski

PURPOSE:

Over the last decade, intense research has been devoted to finding physical systems that can become the backbone of future quantum technologies. The combination of the properties of collective excitations of atoms and quantized fields of light has become one of the strongest candidates to achieve quantum communication due to their ability to be optically controlled and their ease to interface with one another. One of the most pursued implementations in this area of science is the creation of photonic long-range quantum correlations and its mapping to long-lived atomic arrays. This project will focus on quantum networking and quantum sensing.

Quantum networking: We will aim to prototype a long distance quantum network using a modular approach based upon the development of three technological building blocks: a) quantum memories with high efficiency and low noise; b) entangled photon sources with high brightness and wavelength compatible with atom-based memories; and c) characterization devices for imaging and time-stamping of entangled photons, with nanosecond time resolution, capable of evaluating in real time the quantum network performance.

Quantum sensing: We will investigate how the quantum systems entangled at long distances can be applied to sensing of particles and fields by observing changes in the wave function for the entangled system.

APPROACH:

Both directions employ High Energy Physics (HEP) technologies to advance Quantum Information Science through real-time imaging of entangled photons and have the promise to produce new experimental methods for HEP, using a quantum network of entangled quantum sensors or telescopes separated by long-distances. The fast camera proposed for the experiments is a hybrid pixel detector with an optical sensor developed by Nomerotski at Brookhaven National Laboratory.

TECHNICAL PROGRESS AND RESULTS:

Scalability is an important consideration for quantum memories based on ^{87}Rb cells developed by the Stony Brook University group. A single cell can accommodate multiple dual rail laser beams, and using a single camera to detect multiple beams, makes the system truly scalable. Even a 256 x 256 pixel sensor, as in the fast Tpx3Cam camera, could allow simultaneous detection of multiple photon beams, since they will be received in parallel by independent pixel areas of the sensor, corresponding to multiple qubits. This was demonstrated in the first measurements for an 8 x 8 grid using the Tpx3Cam. The left part of Figure 1 shows the experimental setup with two orthogonal acousto-optical modulators (AOMs) to deflect a laser beam into multiple beams. The right part demonstrates 8 x 8=64 beams from AOMs and the bottom insert shows AOM beams composed as letters to demonstrate flexibility of the beam rastering. We also built an AOM controller in a rack-mounted unit for four independent devices.

The microchannel plate (MCP) inside the camera intensifier amplifies the single photon signals. The MCP output signal is very fast and can be used to improve the timing resolution, in principle, to the 30 ps level. We tested this approach by employing an intensifier with MCP output connected to a fast 380 MHz ORTEC amplifier after a 1 MHz high-pass filter. The timing resolution was determined using a fast laser beam split into two paths, with one of them delayed by 200 ns. The time difference between the two pulses, about 202 ns, measured experimentally, is shown in Figure 2, which has resolution for three cases a) MCP readout of the intensified Tpx3Cam; b) Single-photon avalanche diode (SPAD)-based single photon counters; and c) superconducting nanowire single photon counter. These results are encouraging and demonstrate the potential of the approach.

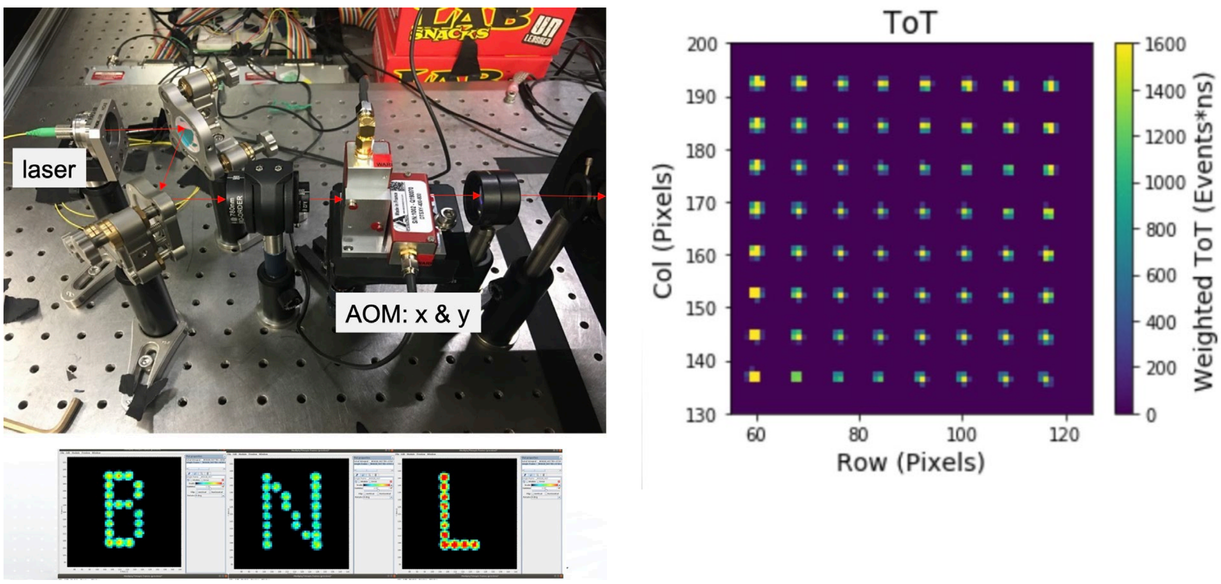


Figure 1. (Left) Experimental setup with two orthogonal AOMs to deflect a laser beam into multiple beams. (Right) 8 x 8 beams from AOMs. (Bottom) AOM beams composed as letters to demonstrate flexibility of the beam rastering.

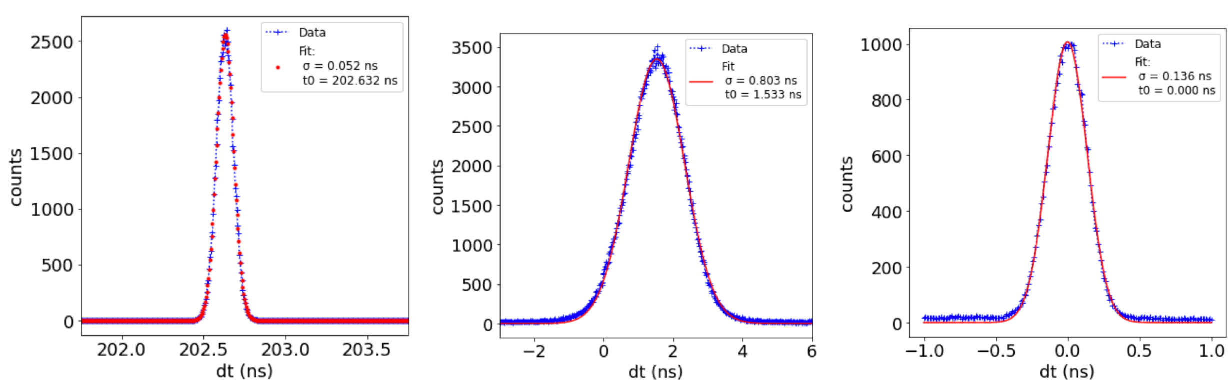


Figure 2. Time resolution for three types of single photon detectors used for the benchmarking measurements. a) MCP readout of the intensified Tpx3Cam. b) SPAD-based single photon counters. c) superconducting nanowire single photon counter. The time difference distributions are fit with a Gaussian function, with its sigma presented in the plots.

These and other project results were published in six peer-reviewed papers in 2021.

MILESTONES:

None. This project completed in FY21.

Novel Approaches for Self-Assembly of Bio-Nanomaterials and Enabling Their New Functions

LDRD # 19-035

O. Gang

PURPOSE:

The goal of this work is to develop novel approaches for self-assembly of bio-nanomaterials and enabling their new functions. We aim to explore new methods for organizing enzymes and proteins in three-dimensional (3D) arrays for manipulating chemical cascades, directing and enhancing reaction pathways, packing enzymes and proteins in a highly dense form, and enhancing their performance. The work aims to establish a platform for engineering the organization of bio-macromolecules (enzymes) in confined 3D nanostructures. To support the analysis of bio-molecular structures, cryo-Electron Microscopy, X-ray scattering, spectroscopic, optical, and microfluidics methods will be used and developed.

APPROACH:

Our method addresses unresolved questions on the effect of micro-environments on the activity of enzymes located on engineered nanoscale scaffolds. These questions are of high importance for the field of nanoscale chemistry, bio-catalysis, sensing, biomaterials, and self-assembled nanomaterials. Our work introduces a new transistor-based methodology for real-time label-free tracking of enzymatic activity with significantly increased precision over the conventional methods. We placed an enzyme at different locations of a 3D octahedral DNA scaffold (with a size of ~50 nm). This design allowed us to modulate the local environment of the enzyme. The combination of the newly designed chemically active nano-architecture and the novel probing methodology revealed the effect of the local environment on enzyme activity and enables us to characterize its magnitude.

TECHNICAL PROGRESS AND RESULTS:

DNA nanotechnology has increasingly been used as a platform to scaffold enzymes based on its unmatched ability to structure enzymes in a desired format. The capability to organize enzymes has taken many forms, from more traditional 2D pairings on individual scaffolds to recent work introducing enzyme organizations in 3D lattices. As the ability to define nanoscale structure has grown, it is critical to fully deconstruct the impact of enzyme organization at the single-scaffold level. Here, we present an open, 3D DNA wireframe octahedron, which is used to create a library of spatially arranged organizations of glucose oxidase (GOx) and horseradish peroxidase (HRP). We explore the contribution of enzyme spacing, arrangement, and location on the 3D scaffold to cascade activity. The experiments provide insight into enzyme scaffold design, including the insignificance of scaffold sequence makeup on activity, an increase in activity at small enzyme spacings of <10 nm, and activity changes that arise from discontinuities in scaffold architectures. Most notably, the experiments allow us to determine that enzyme colocalization itself on the DNA scaffold dominates over any specific enzyme arrangement.

A 3D wireframe DNA origami geometry (Figure 1) enables a versatile addressable space that allows exploration of how a two-enzyme cascade depends on the enzyme arrangement, both in relation to each other and to scaffold structure, including spacings and binding locations. We explored these factors and their relative importance by using a library of DNA scaffolds based on a 3D DNA frame topology that allows for 1) the ability to vary enzyme binding locations over different wireframe edges and at different relative positions to each other; 2) a large testable edge length, enabling spacing tests outside the size regime of the enzymes (>5 nm); and 3) a discontinuous scaffold structure in order to study its effects on substrate channeling. We utilize general and widely used enzyme functionalization and attachment methods to explore these effects by varying enzyme binding locations on a 3D scaffold, while allowing for all possible orientations of enzyme relative to the attached strands. This approach provides a systematic study over averaged enzyme rotation conformations for well-defined positions on 3D scaffolds. We investigated and compared reactions for 42 spatial arrangements of GOx and HRP cascades, as discussed below.

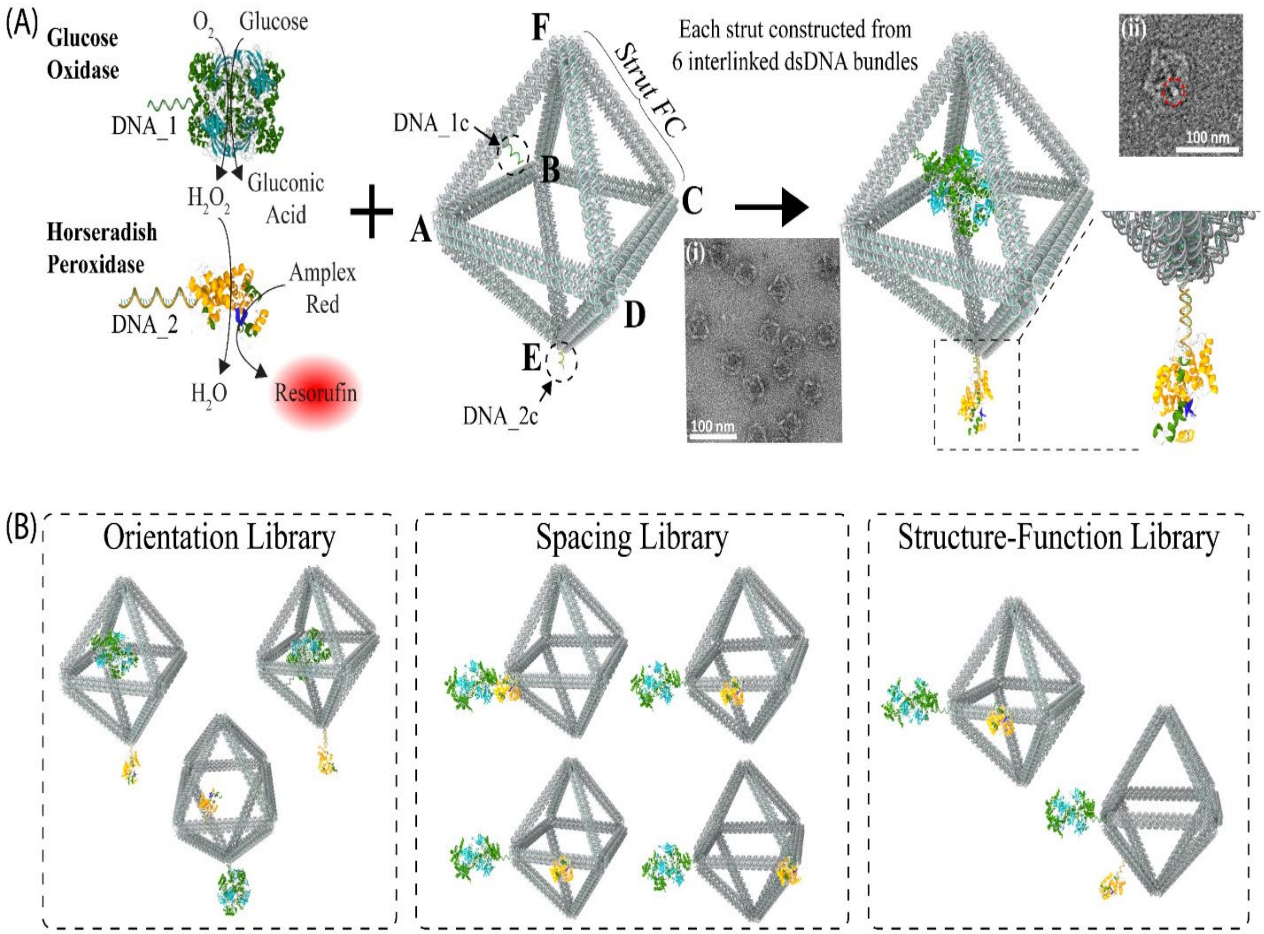


Figure 1. Overview of experimental system and scaffolded enzyme libraries. (A) Scheme of enzyme reaction and mechanism of enzyme attachment to a DNA origami octahedron. Labeling of vertices is performed using letters A-F, with struts being designated by their vertex termini. Transmission Electron Microscope (TEM) images (i) and (ii) depict origami before and after enzyme functionalization, respectively. Enzyme attachment was confirmed through TEM, where (ii) GOx is circled in red, demonstrating that the wireframe structure was maintained through enzyme incubation. (B) Overview of enzyme-origami library classes used in the study to determine relative influences of enzyme arrangement at a given distance, enzyme spacing with fixed binding orientation of the enzyme attachment sites relative to the octahedron scaffold, and the origami structure between enzymes on the enzymatic cascade function.

MILESTONES:

None. This project completed in FY21.

Advancing Atmospheric Prediction Capabilities in Urban Areas for Energy Resiliency and National Security

LDRD # 20-002

A. Vogelmann

PURPOSE:

While 55% of the world's population live in cities with a significant fraction (38%) living within 50 km of the coastline, considerable gaps exist in the prediction capabilities of urban coastal regions that affect our abilities to simulate extreme events, either caused by climate/weather phenomenon (e.g., heatwaves) or by accidental or malicious gas releases. Our goal is to produce quantifiable improvements in high-resolution simulations (i.e., street- and building-resolved) of the New York City (NYC) urban environment, driven-by unique insights from urban-scale datasets acquired by the Brookhaven National Laboratory (BNL) Center for Multiscale Applied Sensing (CMAS). A distinct challenge with this undertaking is adequately treating the extreme heterogeneity of a coastal urban landscape that significantly complicates wind patterns in and around the city, particularly at high-resolution. This project builds on investments in CMAS through the unique alignment of expertise in atmospheric science observations and high-resolution modeling necessary to impact our cross-disciplinary objectives. The project supports the Lab Plan Initiative "Making Sense of Data at the Exabyte-Scale and Beyond" and builds capabilities that complement the expertise within the BNL Tracer Technology Group, who use perfluorocarbon tracers as tools for studying long range atmospheric transport and dispersion, and the BNL Nonproliferation and National Security Department. The project will put BNL in a position to pursue funding from the Department of Energy, and to develop Strategic Partnership Projects with New York State and national security and intelligence agencies.

APPROACH:

The highly heterogenous urban environment is one of the most complex and least understood systems. This project aims to fill knowledge gaps driven mainly by the lack of comprehensive urban-scale measurements of these diverse environments, and the lack of integrated efforts needed to accelerate data-driven improvements in urban prediction capabilities. The project will use BNL-CMAS stationary and mobile atmospheric sensors to evaluate a high-resolution urban atmospheric model. Doppler lidar measurements are of particular use, as they can obtain vertical profiles of winds but have been underutilized in urban studies. These observations will be used to evaluate a state-of-the-art climate model called the System for Atmospheric Modeling (SAM) that is being modified to represent the impact of buildings and other urban features on gas dispersion in a version called SAM-Urban. It is a versatile atmospheric model that can simulate regional-scale weather systems at horizontal resolutions down to a few meters, needed to resolve atmospheric circulations within urban street canyons. The objectives are to use unique CMAS observations to evaluate SAM-Urban, and then apply the model to examine localized (street-level) stresses for heat waves and support evaluation of emergency response models of gaseous dispersion. The project has the requisite expertise in the areas of remote sensing (Dr. Katia Lamer), instrument and software support (Andrew McMahon), artificial intelligence (Ed Luke), the author of the SAM-Urban model (Dr. Marat Khairoutdinov, Stony Brook University [SBU]), and the Director of CMAS (Dr. Pavlos Kollias, BNL/SBU).

TECHNICAL PROGRESS AND RESULTS:

The project started last year and activities focused primarily on the initial development of SAM-Urban, acquisition of model inputs (e.g., building heights), and performance testing. Model output was used to develop an optimal scan strategy for a Doppler lidar to determine the winds within the restrictive urban canyon geometry. This year, the numerical approach used to represent buildings in the model was refined and verified using wind tunnel data. A new, one-of-a-kind, mobile observatory was designed, developed, and deployed for measuring environmental properties within the urban landscape.

The Quasi-Solid Box Method was developed for simulating wind around obstacles in the SAM-Urban atmospheric model. The method is computationally efficient and was verified using wind tunnel data around a single building. A manuscript is in preparation.

A new, one-of-a-kind, mobile observatory (Figure 1) was used for the measurements. The CMAS instrumentation includes a Doppler lidar, to measure wind profiles around buildings, and a weather station, for surface meteorological observations. A diesel generator enables off-grid operations so the observatory can be moved rapidly to different sites, necessary for sampling the heterogeneous urban landscape. The instrumentation can be controlled by an onboard user from a computer, or by a remote user over a Wi-Fi connection. A smart sampling paradigm was developed to optimize data collection within the obstacle-laden urban environment.

The CMAS mobile observatory was deployed to NYC. It was deployed twice, on June 1 and June 28-29, during which wind profile measurements were obtained for study at the four faces of the One Vanderbilt skyscraper, and within Manhattan street canyons. Surface-air temperatures from a transect between Upton and Manhattan indicated an urban heat island effect.

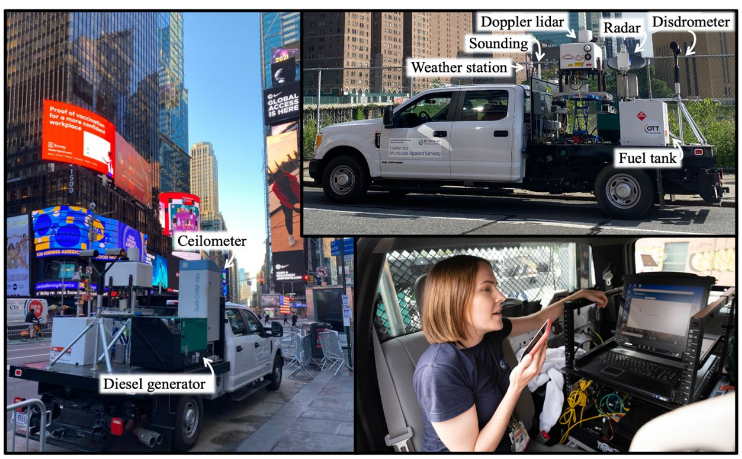


Figure 1. The CMAS Mobile Observatory. Images are during deployments to New York City with labels indicating instrument types and locations on the truck bed. The observatory design, development, and mobile deployments were led by Dr. Katia Lamer, pictured in the lower right image at the computer console within the cab.

MILESTONES:

- Deploy the CMAS Doppler lidar as part of the Department of Homeland Security (DHS) Urban Threat Dispersion Experiment in NYC in October 2021
- Use data from the June and DHS deployments to evaluate SAM-Urban
- Submit manuscripts describing the Quasi-Sold Box Method, the CMAS mobile observatory, observational analyses of airflow around One Vanderbilt, and use of the observations to evaluate SAM-Urban.

Power Efficient Plasma Device for Removal of PFAS, 1,4-Dioxane & Other Contaminants of Emerging Concerns from Water Supply & Wastewater at Record Water Disinfection Rates

LDRD # 20-008

A. Hershcovitch

PURPOSE:

The objective is to develop power efficient, cost effective, environmentally friendly water treatment that removes all polyfluoroalkyl substances (PFAS) and 1,4-Dioxane from the water supply, deactivates all pathogens including COVID-19, and degrades all pharmaceuticals and chemical contaminants, by in-water plasma generation, at an unprecedented rate, which no existing technique can do. The plasma is an in-water vortex stabilized water plasma, unlike other plasmas and electron beams, which are not stabilized. The project is designed to address existing water contamination in the U.S. (Brookhaven National Laboratory (BNL) and Long Island). It has far-reaching potential for removal of all contaminants of emerging concern, pathogens, and pharmaceuticals from the water supply and wastewater at low cost and large water disinfection rates, thus increasing the potable water supply, since good quality drinking water is a dwindling resource for significant segments of the world population, including parts of the U.S.

APPROACH:

As a BBC http://www.ft.com/cms/s/2/8e42bdc8-0838-11e4-9afc-00144feab7de.html quoted article claims, water shortage is a bigger problem than climate change. Certainly, it's a more immediate problem. Aggravating the problem are contaminants of emerging concern - PFAS & 1,4-Dioxane (synthetic industrial chemicals in multiple products). PFAS have extreme resistance to degradation, exhibit bioaccumulation and have high mobility. Consequently, PFAS has been detected in drinking water and wastewater. Human exposure to PFAS has been linked to cancer, immune suppression, etc. Although there is a large effort for PFAS detection, little or no activity exists in developing novel methods for their removal; current technologies have limitations: adsorption using activated carbon and membrane technologies does not remove all PFAS and is expensive due to aggressive fouling, requiring replacement and regeneration. Although there is no evidence of problematic COVID-19 in water yet, it's prudent to anticipate. One option for increasing the water supply is to treat polluted water and recycle wastewater by inexpensive, environmentally friendly methods. Present water disinfection is done chemically and/or using Ultraviolet (UV) light. Chemical use is limited due to residual toxicity; UV consumes much electricity and has other problems. Plasmas in water are attractive for water disinfection due to UV and an advanced oxidation process (AOP) that generates chemicals (hydroxyl, ozone, etc. that are known to kill pathogens, and degrade antibiotics and organic dyes). Present and contemplated techniques utilize unstabilized discharges and electron beams, requiring large amounts of costly power. This project utilizes vortex-stabilized steady state multi-Ampere, low voltage (generating large diameter arc) plasmas. In stabilized plasmas, ions and electrons are well-confined. Consequently, for the same power, orders of magnitude larger quantities of UV and AOP chemicals are generated and greater water disinfection rates can be achieved.

No cost collaborators: Suffolk County Water Authority (SCWA) agreed to test water samples; a company in Oxnard, CA (PVI) that was contracted to fabricate the water vortex device agreed to optimize the water vortex device and its in-water arc at no cost. PVI engineers have been guiding

John Halinski (the BNL designer) in designing the test stand, all electrodes, and diagnostic ports. Arjun Venkatesan from Stony Brook University agreed to prepare spiked water samples.

TECHNICAL PROGRESS AND RESULTS:

Although a vortex-stabilized water plasma device was planned to be operational by now, there have been COVID-19 delays in fabrication of various project components, due to closures and supply chain issues. PVI ignited an arc, at their expense, in a ¼ scale vortex tube model, which generated significantly important data for cathode protection and large diameter in-water arcs; this required design changes, causing further delays (large diameter in-water arcs were never generated).

Figure 1. Photo of the disinfection device.

Fabrication of all components including the test stand and all diagnostics ports was completed. Figure 1 is a photo of the water vortex disinfection device including test stand, which has a feature allowing arc length variation with anode motion (on the bottom). The top flange controls the cathode's radial movement for adjusting the arc diameter. The total system height is 121 inches. Presently, initial plasma discharges are conducted at PVI. Optimization of the internal vortex shell indicates that the poly(methyl) methacrylate internal shell must be replaced by quartz with different slits. Figure 2 shows the ¼ scale vortex device. Extrapolating to a 1 m long 45 A (prototype reactor), 11.5 kW is needed for operation (half of what was predicted; orders of magnitude below other plasma/electron beam experiments). These results strongly suggest low power operations (i.e., low cost) feasibility.

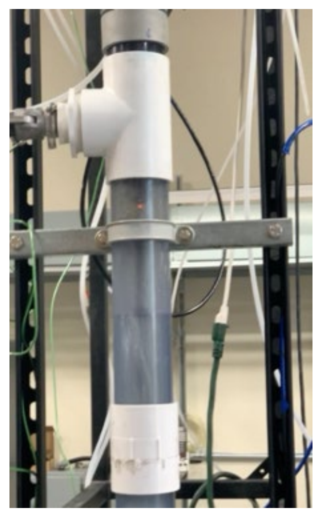


Figure 2. The ¼ scale vortex device.

MILESTONES:

- Work on vortex stabilized plasma device optimization at PVI; to be completed by March 31st, 2022
- Finish all PVI optimization: generate water vortex stabilized in-water steady state plasma with a current of at least 45 A and a diameter of at least 3 cm; to be completed by April 30th, 2022
- Move and set up water vortex stabilized discharge device at BNL. Commence device operation; to be completed by May 31st, 2021
- Optimize device operation and commence UV plasma characterization. Complete safety review; to be completed by July 31st, 2022
- Initiate spiked PFAS injection and measure decrease in its concentration as it passes through the plasma center; to be completed by August 31st, 2022
- Initiate spiked 1,4-Dioxane injection and measure and optimize decrease in its concentration as it passes through the plasma, while monitoring UV generation; to be completed by August 31st, 2022
- Treat and optimize treatment rate of combined PFAS and 1,4-Dioxane contaminated BNL and SCWA wells; optimize germicidal UV emission; to be completed by September 30th, 2022.

Developing Multi-Terawatt Carbon Dioxide (CO_2) Laser Capabilities

LDRD # 20-010

I. Pogorelsky

PURPOSE:

This project investigates scientific opportunities to compress 2 ps, 5 TW pulses delivered by the Accelerator Test Facility (ATF) carbon dioxide (CO_2) laser system down to sub-picoseconds ($<10^{-12}$ sec). This is the first ever attempt to attain a several-optical-cycles, multi-terawatt regime with long-wave infrared (LWIR) (λ ~8-15 μm) lasers. Achieving this regime has been identified as a prime strategic upgrade of the ATF capabilities to support users on their frontier research in advanced particle accelerators. The first prototype of a sub-picosecond LWIR pulse compressor is a primary deliverable for this project.

APPROACH:

Our approach is based on leveraging experimental and simulation methods towards delivering a demonstrator for the first implementation of a nonlinear post-compression (NLPC) technique for high-power LWIR lasers on their breakthrough to a sub-picosecond pulse regime. Theoretical studies supported by simulations show the opportunity for post-compression of 2 ps, 9.2 μm laser pulses down to 100 fs, using self-phase modulation (SPM) and dispersive compression (DC) in bulk materials. At the same time, a preliminary survey has revealed significant gaps in our understanding of nonlinear material properties under the high intensity irradiation required for such demonstration. In close coordination with a parallel effort conducted by Brookhaven National Laboratory (BNL) and the University of Central Florida on measuring properties of candidate materials, which is supported by the Department of Energy Accelerator Stewardship program (Principal Investigator M. Polyanskiy, BNL), the project team is working on identifying materials suitable for NLPC and proceeding with a sub-picosecond compressor demonstrator.

TECHNICAL PROGRESS AND RESULTS:

The project's first milestone on the proof-of-principle NLPC demonstration was completed in FY20, when a sub-picosecond pulse structure was observed after a single, 10 cm thick NaCl crystal was used for simultaneous SPM and DC. In the second year of the project, we proceeded with optimizing the NLPC components, using results of material studies supported by the Accelerator Stewardship Program. This resulted in completing another experimental milestone where KCl and BaF_2 slabs were used to improve the pulse compression efficiency by partly separating SPM and DC functions.

The principal optical diagram of the experiment and its results are illustrated in Figure 1. To reduce the influence of irregularities in the laser beam profile, we diaphragmed the beam and, after several meters of propagation, produced the Gaussian radial distribution. The pulse compression after the NLPC has been analyzed with a single-shot autocorrelator within the central portion of this distribution. The observed compression to 300 fs was reproducible and agrees with in-house computer simulations. Our results have been published and presented at conferences. In FY22, we will attempt post-compression of the entire 10 joule laser energy. We will conduct our experiments in vacuum to allow for the laser beam's spatial filtering and, ultimately, its long-distance propagation to user experiments. Vacuum chambers are being installed and experiments resume in Spring 2022. Completing this milestone (#5 in the table below) will mark a new achievement in the LWIR laser field and put our team in the position for completion of this project with a practical NLPC engineering design.

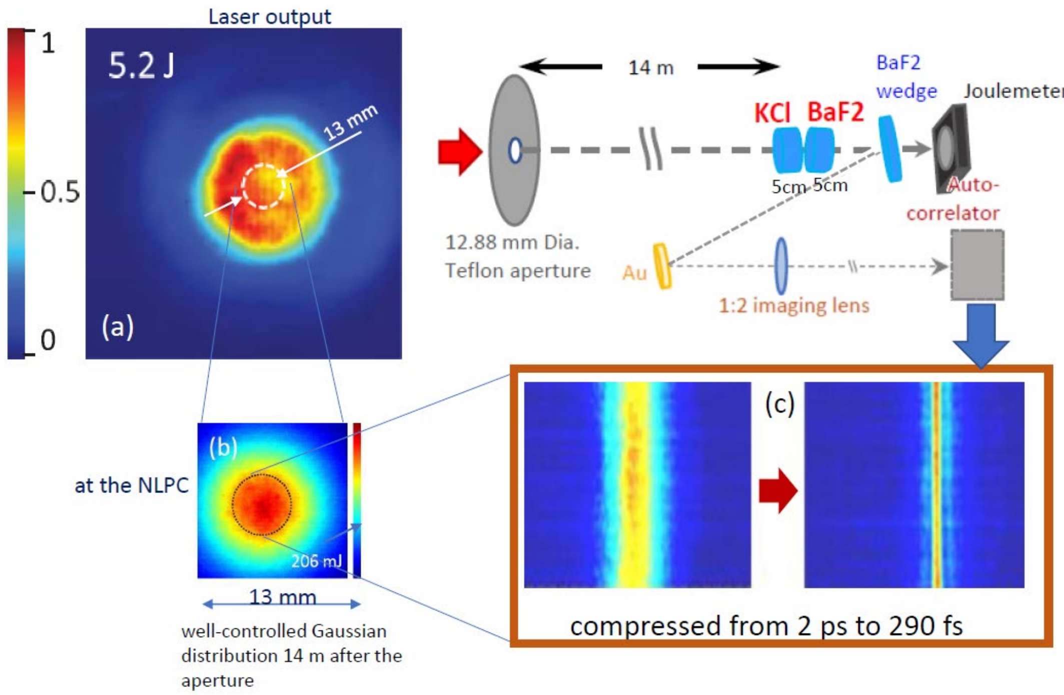


Figure 1. (a) Principal diagram of the 2021 experiment with embedded sample beam profiles at the laser output. (b) At a long distance after the 13-mm diaphragm. (c) Autocorrelator images.

MILESTONES:

Task#	Description	Milestones and Deliverables	FY 2022	FY 2023
2	Installation and commissioning of the NLPC vacuum chambers.	Chambers installed.		
3	Predictive simulations of the nonlinear optical materials action in order to optimize the NLPC test configuration.	Materials and regimes are selected for experimental testing.		
5	Conduct in-vacuum NLPC tests optimizing the energy conversion efficiency and the beam quality.	Sub-picosecond beam characterized and optimized.		
6	Coordinate test results with simulations to confirm relevant models.	Theoretical models are fine-tuned and ready for use in engineering a production system.		
7	Develop a conceptual engineering design for the ATF's high-power CO_2 laser pulse compressor from 2 ps down to sub-picoseconds.	Formulate directions for future R&D; Prepare and submit publications; Write a final report.		

* Milestone numbering according to FY20 Report (milestones #1 and #4 completed)

Building a Quantum Repeater Prototype Connecting Brookhaven National Laboratory to New York City

LDRD # 20-018

E. Figueroa

PURPOSE:

Quantum communication using entanglement — especially with its promise of revolutionizing current communication systems — is capturing interest across science, industry, and the global internet community. Despite this promise, quantum networking is still in a nascent phase, and many challenges remain before the full potential of large quantum communication systems can be realized. One of the greatest challenges is to demonstrate the preservation of entanglement after its propagation, using the current fiber infrastructure. The objective of our project is to demonstrate the transmission of entanglement at telecom wavelengths over long-distances (over more than hundred kilometers) in a real testbed, using currently available fiber infrastructure. These developments are part of our wider Brookhaven National Laboratory (BNL) research program that targets the construction of the first quantum-repeater-assisted, entanglement distribution network in the U.S.

APPROACH:

We have used a multi-pronged approach to address this challenge. First, we have created a quantum technology development program aimed at producing the first instances of telecom entangled sources and single photon detection systems that are compatible with fiber infrastructure (in collaboration with Paul Stankus and Julian Martinez, BNL Instrumentation Division scientists). Second, we have developed quantum enabling technologies that will allow us to manage and control (using the classical internet) the entanglement distribution process over a long-distance fiber network (in collaboration with Dimitrios Katramatos, BNL Computational Science Initiative scientist). Last, we have procured and established a commercial fiber testbed that connects the BNL Instrumentation Quantum Information Science and Technology laboratory to the Stony Brook University (SBU) Quantum Internet Laboratory and a new node using the Commack (CMK) mindShift collocation facility. Our target is to combine all these developments to form the first U.S.-long-distance entanglement distribution experiment.

TECHNICAL PROGRESS AND RESULTS:

In the last two years, we have developed (starting from an empty laboratory), the technical requirements needed to attempt the envisioned large entanglement distribution experiments.

Telecom laser infrastructure: we developed two tunable tapered-amplified telecom laser systems operational at the original band of telecom transmission. The lasers were tuned to a wavelength of 1324 nm. Additionally, one of the telecom systems was integrated with a second harmonic generation system able to produce visible radiation at a wavelength of 662 nm. These systems are now used in the production of entangled photon pairs and the stabilization of cavities to enhance the rate of photon pair production. We also developed specialized two-photon absorption spectroscopy setups that allowed the stabilization of the tunable lasers to atomic transitions in the telecom.

Telecom-band entangled photons sources: we developed a polarization entanglement source operational at telecom wavelengths. In this source, an input pump laser tuned to a wavelength of 622 nm is used to generate photon pairs centered around the 1324 nm 5P1/2 -> 6S1/2 transition of rubidium atoms. Photon pair production was achieved with two Periodically Poled Lithium Niobate (aka PPLN) non-linear Type-I crystals. We are further developing the system to produce a Bell state of the form: $|HH\rangle + |VV\rangle$.

Single Photon Detection: we developed two operational Superconducting Nanowire Single Photon Detectors (SNSPD) systems. One of them comprises four channels sensitive at telecom wavelengths (~1300-1500 nm) and four channels sensitive at near-Infrared (NIR) (~800 nm), and the second one (portable) has two telecom channels and two NIR channels.

Enabling technologies for long distance quantum communication: we developed the classical control mechanisms needed for initialization, management, and online control of the quantum network. We developed classical communication systems, including servers for data analysis and storage, optically-connected switches forming the backbone of the control networks, wave-multiplexing systems to manage the traffic of classical and quantum information in the same fibers, and White Rabbit time synchronization systems for optical clock distribution among the network nodes.

Fiber infrastructure: Our fiber network infrastructure has been expanded to cover more nodes across Long Island, including multi-purpose quantum nodes at BNL, SBU, and CMK. This three-node quantum network (Figure 1) is the first of its kind in the U.S. and is the testbed to distribute entanglement over 140 km of commercial fiber.

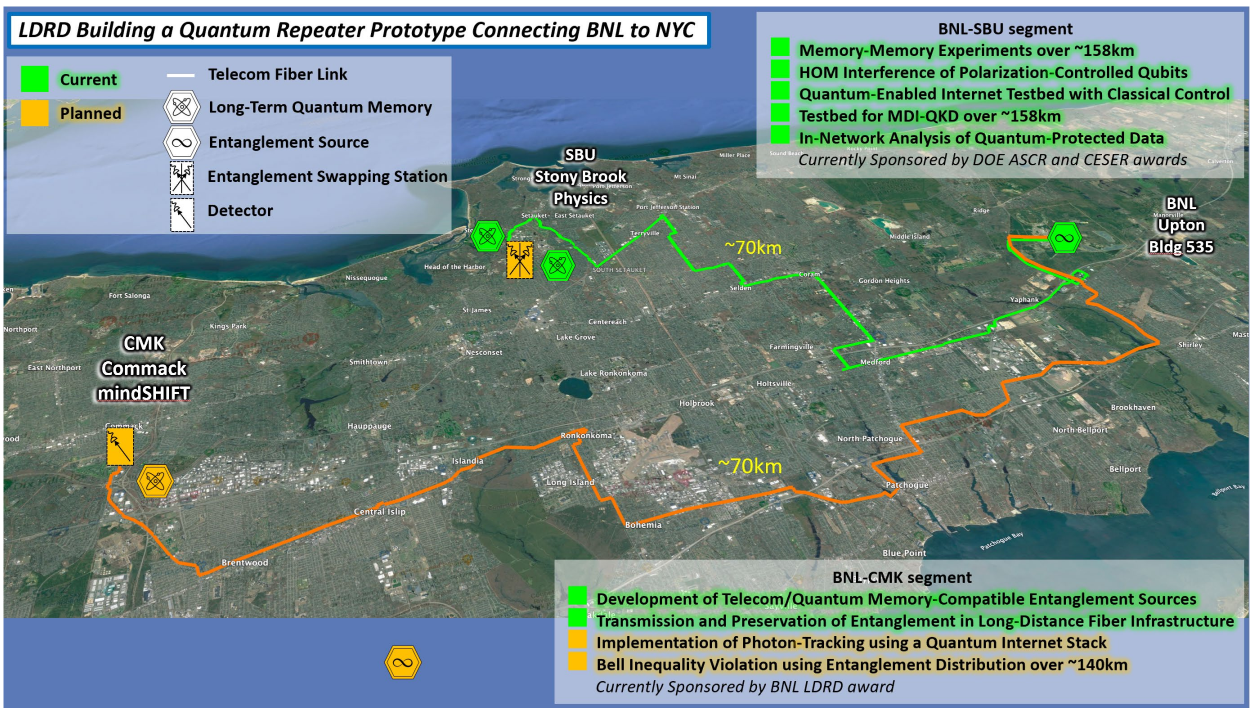


Figure 1. The current existing entanglement distribution testbed connecting the BNL campus to the SBU campus and the CMK mindShift collocation facility over a fiber distance of 140 km. In green, we show the current experiments being performed in both quantum communication branches and in orange, we show the targeted developments for 2022 that will converge in a long-distance demonstration of entanglement distribution.

MILESTONES:

In the last year of the project, all the mentioned parts will come together in a large experiment. We will produce telecom entangled photons at BNL and we will distribute them (in a network control fashion) to distantly-located nodes in SBU and CMK. Here, remotely controlled SNSPD systems will analyze the distributed quantum state and perform quantum tomography to determine the fidelity of the distributed entangled states. Figure 1 shows the current status of the testbed, including the current developments and the envisioned milestones towards achieving our proposed objectives.

Topological Quantum Error Correcting Codes in the Noisy Intermediate-Scale Quantum (NISQ) Era

LDRD # 20-022

L. Hormozi

PURPOSE:

The work is an interdisciplinary effort that aims at taking advantage of the progress made in noisy intermediate-scale quantum (NISQ) devices to build a two-way bridge between quantum information science (QIS) and the physics of complex quantum systems. On one hand, we aim at utilizing the characteristics of exotic (topological) phases of matter to design quantum error-correcting (QEC) codes that are scalable and more efficient than the existing codes. On the other hand, we design quantum circuits that can realize topological phases in existing testbeds and reveal their remarkable properties (such as non-Abelian statistics) – a goal that despite much effort has not yet been realized in any platform. This work ties into the Department of Energy's mission of advancing science and technology and is aligned with the Office of Science on-going programs on advancing interdisciplinary research between QIS and the Offices of Basic Energy Science, High Energy Physics, and Advanced Scientific Computing Research.

APPROACH:

Background: Quantum computers are famously prone to errors, as quantum states are susceptible to losing their coherence, due to interactions with the environment. Thus, the development of QEC methods is a fundamental component of QIS. One of the most promising approaches to QEC is inspired by the so-called "topological phases of matter" – a class of low-energy quantum states that, owing to their internal structure, are inherently protected from the environment. While these systems are rare in nature (the only observed examples are fractional quantum Hall states) and are very hard to engineer, their intrinsic immunity to errors can be utilized in the idea of topological QEC codes. In these codes, the protected ground state of the topological phase is effectively simulated on a system of qubits by carrying out standard quantum gates. This simulated "code space" then inherits the intrinsic protection of the parent topological phase and can be used to store and manipulate quantum information in a fault-tolerant way.

The simplest of topological codes, the so-called surface code, is currently the front-runner among existing QEC codes. This code provides the highest known error threshold (the maximum error rate the system can tolerate without spoiling the quantum computation) of ~1% and the relative simplicity of the underlying (Abelian) topological phase provides a straightforward recipe for the implementation and analysis of this code. However, this internal simplicity is also a peril since the underlying topological phase is not capable of universal quantum computation. This leads to the need to carry out the resource-extensive process of "magic state distillation," which results in the code's extreme inefficiency. This problem can, in principle, be circumvented by implementing the more complex "non-Abelian" codes, which are based on lattice implementations of non-Abelian topological phases and are capable of universal quantum computation.

Scope and Methods: Here we aim at addressing the basic properties of non-Abelian codes and their implementation schemes. The work consists of two parallel projects: (1) to design a blueprint for the circuits required to simulate the ground state and excitations of non-Abelian codes and reveal their non-Abelian statistics (in collaboration with Richard Allen, a student at Harvard and Steven Simon at Oxford); and (2) to carry out a comparison between Abelian and non-Abelian codes by estimating error thresholds and the resources required for non-Abelian codes using a combination of analytical and numerical tools (in collaboration with Evan Philip, a postdoc at the Computational Science Initiative [CSI]). This work has also led to new collaborations with Ning Bao (CSI), Andreas

Weichselbaum (Condensed Matter Physics and Materials Science), Tzu-Chieh Wei (Stony Brook University) and the IBM theory group on related projects. In both projects, the main risk stems from the fact that even minimal blueprints might require resources beyond the reach of current or near-future NISQ devices.

TECHNICAL PROGRESS AND RESULTS:
For the first project in FY20, we devised a method to systematically simulate the ground state of a simple topological model known as a “Fibonacci” string-net model and wrote a code that designed the corresponding circuits for implementation on generic trivalent lattices. In FY21, we devised a specific minimal model, consisting of six qubits sitting on the edges of a tetrahedron (Figure 1(a)), and devised circuits for creating and moving its (plaquette) excitations. We also devised an operator to distinguish the two fusion channels of the Fibonacci excitations and thus reveal their fundamental non-Abelian characteristic (Figure 1(b)). As of FY22 we are ironing out the details of the final measurement circuit and simultaneously working on a manuscript, summarizing our results for publication.

The second project took off in FY21, with the arrival of a new postdoc. We wrote codes to reproduce the existing threshold calculation for the Abelian surface code and tested very small systems of a simplified Fibonacci code. The threshold for the Fibonacci code was calculated by another group in January 2021, so we directed our focus instead to resource estimation, more specifically, estimating circuit-depth lower bounds for the Fibonacci code, hoping to compare the relative efficiency and feasibility of Abelian and non-Abelian codes. The idea was to treat the code as a topological state and to simulate the computation with native logical braid gates, thus cutting down the compiling cost and enhancing the circuit efficiency. Unfortunately, the project was halted by the abrupt departure of the postdoc in early August 2021, and we have not yet been able to find a suitable replacement.

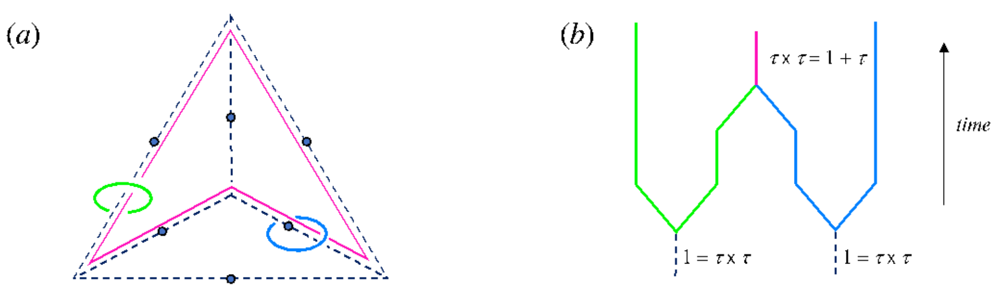


Figure 1. (a) Qubits, excitation generating, and measurement operators. The blue dots represent the arrangement of six qubits on the edges of a tetrahedron. The dashed lines represent the ground state of the Fibonacci code. The blue and green rings are operators that create pairs of excitations on the four plaquettes. The magenta ring illustrates the measurement operator to distinguish the two fusion channels of Fibonacci excitations. (b) An illustration of the excitation generation and measurements. Two pairs of Fibonacci plaquette excitations are generated, then the total charge of the middle two is measured. The outcome is either total annihilation (1) or another excitation (t). Demonstration of these two fusion channels reveals the non-Abelian nature of the Fibonacci code.

MILESTONES:
First project: For the remainder of FY22, we expect to finish and publish our results on the design of a minimal quantum circuit to reveal the non-Abelian properties of the Fibonacci code and hopefully optimize the code based on the characteristics of specific NISQ devices. We also hope to investigate native applications of these codes, such as implementation of the Jones polynomial.

Second Project: We hope to revive this project as soon as we are done with the first project by early 2022 and carry out a comparison of the efficiency and feasibility of the Abelian and non-Abelian codes, considering the properties of specific NISQ devices.

High-density Superconducting Interconnect for Quantum Control

LDRD # 20-023

S. Rescia

PURPOSE:

Scaled-up superconducting quantum computers require thousands of microwave-frequency signal lines to connect the cold (~10 mK) processor to control and readout electronics at higher temperature stages (4 K to 300 K). Superconducting quantum hardware imposes unique requirements on the wiring solution: 1) Low insertion loss up to 10 GHz; 2) Low thermal conductance at cryogenic temperatures; 3) Good isolation between neighboring signal lines, and between the signal lines and the environment; 4) Small physical footprint; and 5) Low cost and ease of manufacture. The prevailing solution employed by industrial and academic research groups alike is individual, connectorized coaxial cables. These become impractical beyond the present scale of quantum hardware, i.e., about 50 qubits and some hundreds of control lines. As a result, there is a near-immediate need for an advanced cryogenic microwave interconnect.

APPROACH:

To address the previously listed requirements, high-density flex interconnects with normal or traditional low-temperature superconducting conductors have been proposed and demonstrated in recent years. This project aims at developing flex interconnects using the yttrium barium copper oxide (YBCO) high-temperature superconductor (HTS). YBCO is a ceramic HTS that offers the ideal conductor characteristics even beyond liquid nitrogen temperatures (77 K): zero resistive losses, and exponentially suppressed electronic thermal conductance. In the context of quantum computer hardware, the key benefit of HTS wiring is that it allows placing the hot end of the interconnect, and therefore the semiconductor control electronics, at a higher-temperature stage of the cryostat. In practice, this would be the first pulse tube cryocooler stage (~50 K) instead of the second (~3 K) stage. This, in turn, will allow the semiconductor control electronics designers to target a higher operation temperature with typically thirty times larger cooling capacity. The critical current density of HTS cables is also very large, allowing smaller conductor linewidths, in principle.

The proposed solution is to use HTS YBCO films that are exfoliated from the growth substrate and then transferred to a low-loss Kapton substrate. After the transfer, the film is patterned into individual conductors that form the microwave waveguides using a laser. Importantly, both the exfoliation and the laser striation are standard processes used in the manufacture of superconducting wiring for accelerator magnets. Micrometer-scale resolution has been demonstrated with laser striation, allowing very dense wiring patterns. This process also allows reel-to-reel processing to be employed, which enables long cables to be made economically. We make use of the existing expertise and facilities available at Brookhaven National Laboratory (BNL) across various departments, together with a close industry partner, Brookhaven Technology Group (BTG).

TECHNICAL PROGRESS AND RESULTS:
In the first year of the project (FY20), a short (140 mm) prototype, using a microstrip configuration (signal conductors over a ground plane), was constructed and tested. The results of these initial tests were described in a paper submitted to the Applied Superconductivity Virtual Conference (ASC 2020).

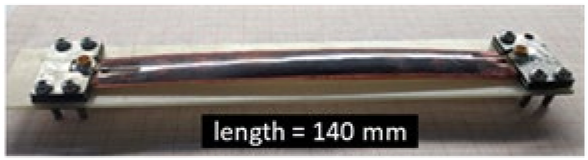


Figure 1. A short prototype test cable.

The technical design aspects of the cable consist of the geometry of the waveguide and the design of the connectors at the ends of the cable. In the first year, the test cables were made in a stripline geometry. The cable was terminated with custom end blocks that exposed coaxial ports. The cables were manufactured by BTG under an R&D subcontract. Microwave testing at 77 K (immersion in liquid nitrogen) was performed at the BNL Instrumentation Division. The main technical achievement is the demonstration of < 1 dB/m attenuation constant up to 6 GHz in a flexible cable with no metallic conductors.

In the second year (FY21), an updated version of the cable was contracted to BTG. It consists of a short (140 mm) cable with three conductors in a stripline configuration (signal conductors sandwiched between two ground planes), with ground top and bottom ground planes shorted together by multiple vias to reduce signal crosstalk and electromagnetic interference.

This project has demonstrated the feasibility of the YBCO-Kapton cable for microwave transmission in a cryogenic environment and the goal of a reduced thermal load. The main positive results are low microwave losses and sufficient mechanical robustness to allow normal handling and temperature cycles between cryogenic and room-temperature environments. However, additional advances are needed before the technology is mature enough for use in a quantum computer. Our results also indicated that the transition from the microwave connector (or copper printed circuit board traces) to the flexible segment needs to be improved in terms of both mechanical robustness and microwave impedance variation to reduce reflections of the microwave signals used to control qubits in quantum computers. The design of the more complex stripline waveguide geometries needed to improve the crosstalk between neighboring signal lines, and to better isolate the signal lines from the external environment experienced some challenges.

MILESTONES:
None. This project completed in October 2021.

Quantum Machine Learning for Dissipative Dynamics of Noisy Intermediate-Scale Quantum (NISQ) Devices

LDRD # 20-024

Y-C Chen, M. Lin

PURPOSE:

Efficient simulation of noisy intermediate-scale quantum (NISQ) computers is critical for scientists, engineers, and application developers to understand, analyze, and improve the device performance. However, even though today's NISQ devices have only ~50 to 100 qubits, simulating their dynamics already presents a significant computational challenge. Machine learning (ML) aims to provide an effective model that has the representative power for an important, finite-number of features in a huge parameter space. Inspired by ML, quantum neural network (QNN) states, such as restricted Boltzmann machines (RBM), have been successfully applied to describe closed quantum systems, but not much has been done for open quantum systems (those with noise, loss, and dissipation, such as the NISQ devices). Following this line of thought, we aim to combine wisdom from both the fields of ML and quantum physics and investigate such ML-inspired approaches to simulate complex systems more efficiently (e.g., using less computational resources, exploiting potential quantum speedups, etc.), with the primary target application being the simulation of NISQ devices. Specifically, this project will explore 1) classical ML architectures for fast and reliable computation of quantum dynamics, especially for the generation of quantum circuit architectures, and 2) the capabilities and limitations of quantum ML architectures for various tasks.

APPROACH:

We will develop a flexible theory tool (with a software implementation) for studying the expressive power of QNN representations and other kinds of quantum ML architectures, with the application to open quantum systems in mind. Then we will construct a hybrid tensor network and quantum circuit ML models to leverage the advantages from both the classical and quantum world. In addition, we will apply the classical reinforcement learning (RL) method to tackle the quantum circuit architecture search problem. We will implement an OpenAI Gym for quantum circuit architecture search in this part of the work. The packages built can be further used in many future research topics related to quantum circuit design or optimization. We also explore other ML-inspired approaches that will benefit quantum information science and strengthen Brookhaven National Laboratory's capability and visibility in this field. In particular, we seek quantum ML architectures that permit a hybrid classical-quantum workflow - training on quantum computers and optimizing on classical computers, for example. Finally, we have strong synergy with the ongoing Department of Energy High Energy Physics Quantum Machine Learning (QML) project (led by Dr. Shinjae Yoo) and the Computational Science Initiative's (classical) ML efforts.

TECHNICAL PROGRESS AND RESULTS:

In FY20, we collaborated with a senior Ph.D. student, Xin Zhang, from Duke University to study QNN representations. We expressed the quantum mechanical wave function of a closed quantum system in terms of an RBM, then trained the neural network parameters using variational Monte Carlo. To generalize this approach to open quantum systems, we expressed the density matrix of the system using two independent copies of RBMs (for the bra and ket states), and the variational approach follows, with twice the number of parameters to train. We have shown its applicability by finding good agreement with exact calculations (within an accuracy of about 10^{-3}) for the steady-state properties of a simple system with nonlinear bosons driven coherently. We also developed the quantum Long Short-Term Memory (Q-LSTM) architecture, which replaces the classical neutral networks by a quantum version to allow natural implementation on quantum computers. Q-LSTM offers faster convergence, better error behavior and prediction than that from the classical LSTM with roughly the same number of neurons, particularly when learning complex temporal behaviors.

In FY21, we continued the efforts on a QNN and the development of a scalable quantum machine learning package metaQuantum. The results are demonstrated in quantum supervised learning tasks, such as a quantum convolutional neural network (QCNN) and a quantum graph convolutional neural network (QGCNN). The numerical simulations imply that the quantum models have advantages over classical models in terms of: 1) reducing the number of parameters; 2) reaching the optimal performance in fewer training epochs; and 3) achieving higher testing accuracies. We also showed that the QML models can be trained in a federated manner, meaning that the training data is not shared across different nodes, while the trained models are the only things exchanged. We demonstrated via numerical simulations that the federated QML training can preserve not only the data privacy but also the classification accuracies. Along this direction of privacy-preserving QML, we also presented QML with differential privacy. We showed that the differentially private optimization algorithm can be used to optimize the quantum models and the quantum model demonstrates advantages over classical models with higher accuracies. To fully leverage the capabilities of NISQ devices, we studied the data embedding schemes on quantum circuits and showed that the learnable data encoding scheme is robust on noisy real quantum computers. We also showed that certain quantum-inspired classical algorithms, such as tensor networks, can be used to compress the high dimensional classical data into smaller data vectors suitable for NISQ devices. We demonstrated via simulations that such a hybrid architecture can beat the traditional dimensional reduction method, such as PCA (Principal Component Analysis). Finally, we presented an automatic quantum circuit architecture search method based on deep reinforcement learning (DRL). We showed that the artificial intelligence (AI) agent can determine the Bell and Greenberger—Horne—Zeilinger (aka GHZ) states without any encoded physics knowledge under the influence of device noise.

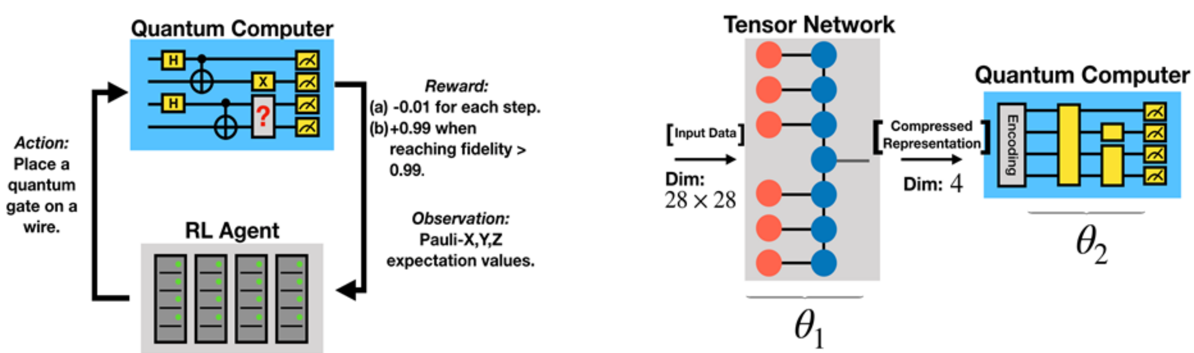


Figure 1. (Left) Quantum circuit architecture search via deep RL. (Right) Hybrid Tensor Network-Quantum Computer end-to-end ML model.

MILESTONES:

- Continue the research on the quantum circuit architecture search problem. We will focus on the development of AI agents which can determine the optimal quantum circuit with changing or drifting noise patterns. The agent is expected to update its knowledge of building circuits automatically when the device's noise patterns change.
- Explore AI/ML based methods for encoding and decoding in a quantum error correction (QEC) workflow. We will first investigate the RL-based decoders under changing or drifting noise. Then we would extend this to include both the encoder and decoder circuits. Part of the work is to leverage the superior searching capabilities of RL to design the QEC circuits.

Unraveling the Elusive Active Site Structures of Membrane Bound Non-Heme Diiron Enzymes

LDRD # 20-029

M. Ertem, Q. Liu, J. Shanklin

PURPOSE:

The primary research goals for this study are: (i) to develop a computational protocol for the prediction of enzyme diiron active site structures and mechanisms; and (ii) to employ the developed methodology together with quantum chemical calculations and biochemical studies to investigate the activation, substrate binding, and oxidative reactivity of a specific enzyme, namely AlkB. The project aims to achieve a breakthrough in the understanding of structure and reactivity of membrane bound non-heme diiron enzymes and establish Brookhaven National Laboratory (BNL) leadership by collaboration of chemical and physical biosciences expertise and capabilities. This cross-disciplinary study in theoretical chemistry, biochemistry, and structural biology aligns well with fundamental research goals of the Office of Basic Energy Sciences-Chemical Sciences, Geosciences, & Biosciences (BES-CSGB) Division, which funds two of the current principal investigators through different programs in Chemistry and Biology and which is encouraging inter-program synergistic research. The joint research will create a compelling seed project that could potentially attract funding from the BES-CSGB Photochemistry and Biochemistry program. In addition, the fundamental mechanistic understanding derived from this work will open the door to potential funding by the National Institutes of Health. This study also aligns well with the BNL institutional strategy via establishing a strong foundation for synergistic effort between the Chemistry Division and the Biology Department utilizing National Synchrotron Light Source II (X-ray crystallography) and Cryo-electron microscopy (Cryo-EM) resources.

APPROACH:

The functionalization of carbon-hydrogen (C-H) bonds in hydrocarbons in an environmentally benign manner is one of the greatest challenges of chemistry in the twenty-first century. Membrane bound non-heme diiron enzymes catalyze C-H functionalization reactions, which are crucial for lipid metabolism in bacteria, plants, animals, and humans, and provide a model for bio-inspired activation of C-H bonds under mild conditions. However, there is a lack of mechanistic understanding of the diiron active sites, linked to the difficulties of structural determination of such membrane-bound enzymes that prevent their exploitation for novel challenging catalysis and pharmaceutical applications.

The present study aims: (i) to develop a general computational methodology to uncover the elusive active site structures and mechanisms of membrane bound non-heme diiron enzymes based on available spectroscopic data; and (ii) to apply the developed approach specifically to study the AlkB enzyme, a membrane bound non-heme diiron-containing monooxygenase which catalyzes a critical step in the conversion of hydrocarbons to high-value bioproducts.

The first stage of the study focuses on developing a computational protocol, based on density functional theory (DFT) calculations, which relates the molecular coordination environment and redox/spin states of the iron (Fe) centers of diiron systems to the available spectroscopic data (e.g., Mössbauer, electron paramagnetic resonance, Extended X-ray Absorption Fine Structure spectra). The second stage of the study involves a synergistic effort of application of the developed predictive computational protocol and biochemical and biophysical analysis to uncover the active site structure of AlkB, a specific non-heme diiron enzyme. In this stage, the developed computational protocol will be employed to assess the validity of our proposed active site model structures with respect to published spectroscopic data and will provide a theoretical basis for further experimental validation by site-directed mutagenesis. In the final stage of the study, we will simulate oxygen activation and following the substrate oxidation, steps to determine the mechanism of the enzyme activity and pursue pathways for enhancing the catalytic activity with further mutational, biochemical, and biophysical characterization (X-ray crystallography, Cryo-EM).

TECHNICAL PROGRESS AND RESULTS:

DFT computations to predict Mössbauer parameters for a given structure proved to be quite useful to interpret the complex nature of spectral values, such as isomer shift δ and quadrupole splitting $|E_Q|$, and to understand the electronic structure and coordination environment of the Fe centers. In FY20 and FY21, a computational protocol based on a specific DFT method, B97-D3, to predict Mössbauer parameters δ and $|E_Q|$ of non-heme diiron complexes was developed. A linear relationship was constructed from the electron density obtained from DFT calculations at the Fe nuclei and experimental δ values, which displayed an excellent predictive ability with a R^2 of 0.98 (Figure 1a). We further tested the developed computational protocol to study the Mössbauer parameters δ and $|E_Q|$ of model structures generated from X-ray structures of non-heme diiron enzymes and observed similar robust predictive power (Figure 1b). This latter result indicates that the developed approach could be more generally applied to non-heme diiron systems beyond the original training set of 20 complexes. We have constructed active site models for the AlkB enzyme and started computing their Mössbauer parameters, which together with other spectroscopic data, both computational and experimental, have been used to develop models for understanding the elusive active site structure and mechanism of AlkB.

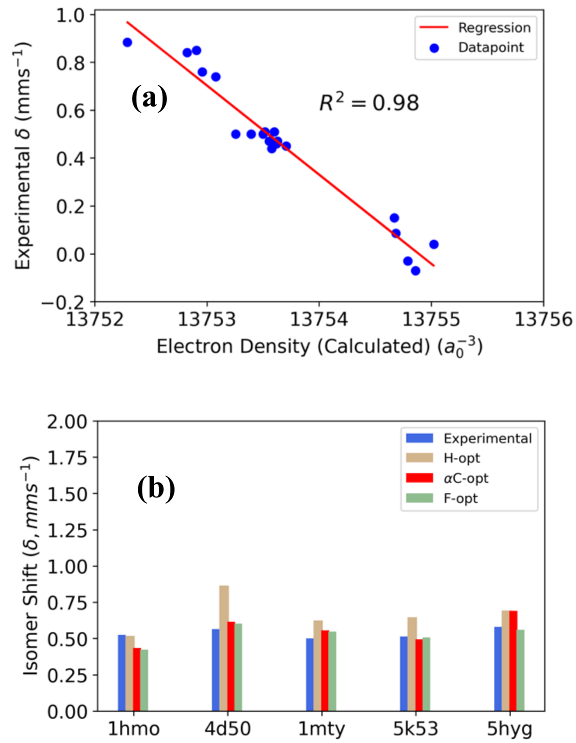


Figure 1. (a) Linear regression plot for calculated electron density on Fe centers and experimental δ. (b) Prediction of δ for nonheme diiron enzyme active site models.

In summary, we developed computational methods for structural prediction of diiron complexes from spectroscopic data and applied those methods to determine AlkB model complexes. We have also submitted the first manuscript on this work.

MILESTONES:

Implement model complexes to understand AlkB enzyme active site and conduct mechanistic and site-directed mutagenesis studies.

Robust Physics-informed Machine Learning Application in Spectro-imaging and Microscopy

LDRD # 20-030

M. Ge, T. Flynn

PURPOSE:

This project is targeted at addressing challenges faced by the scientific X-ray imaging community, with a focus on improving the accuracy in quantitative imaging analysis. We propose to develop new algorithms to incorporate physical constrains into machine learning (ML) models to solve various problems, including missing angle tomography and quantitative two-dimensional (2D)/3-dimensional (3D) X-ray Absorption Near Edge Structure (XANES) imaging analysis. With successful execution of the project, we expect to deliver capabilities to: 1) restore the missing information in the missing-wedge; 2) incorporate a self-absorption correction in X-ray fluorescent imaging; and 3) denoise low signal X-ray images into high resolution with high fidelity as a prerequisite for quantitative XANES image analysis. We expect that our methods would largely benefit all the imaging beamlines at National Synchrotron Light Source II (NSLS-II), and keep NSLS-II competitive and help maintain its leading role in the X-ray community for both scientists and industrial researchers.

APPROACH:

The first task is to solve the problem of missing-angle tomography. We apply an ML-based method to restore the missing information that arises from incomplete data collection due to experimental constraints. As convolutional neural networks (CNN) are capable of finding hidden connections and restoring the missing information, we are building a two-step CNN model to fill in the missing information in both sinogram and tomogram domains to achieve self-consistence.

The second task is to correct the self-absorption problem in X-ray fluorescent imaging. Self-absorption is universal in X-ray fluorescent imaging. In the experiment, the excited X-ray fluorescence will be partially absorbed by the material itself before reaching the detector, inducing significant artifacts for quantitative analysis. A few existing approaches attempt to correct the self-absorption with an oversimplified 2D model. Here, we are developing a realistic 3D model to fully describe the problem mathematically. We have achieved preliminary results demonstrating the feasibility. In this part, we are collaborating with the beamline scientist at the Hard X-ray Nanoprobe beamline.

The third task is XANES image denoising. A XANES image is a collection of X-ray absorption images taken at different X-ray energies. We try to embed the physical constrains, which link the image contrasts to a specific material property into the ML denoising model.

TECHNICAL PROGRESS AND RESULTS:

The project is on the right track as planned with expected progress:

- For the missing wedge tomography project, the ML model and updated results are in Figure 1.
- For the self-absorption correction project, built upon the achievement we made last year, we have conducted statistical analysis to evaluate the method's performance (Figure 2).
- For the XANES image denoising project, we have built the ML model. Denoising results are shown in Figure 3.

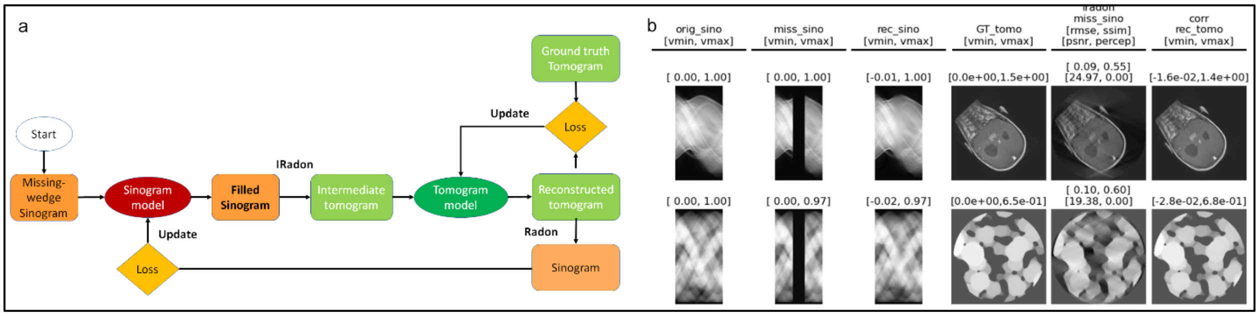


Figure 1. (a) Proposed ML model, including two CNN models (sinogram model and tomogram model). (b) Recovering the missing-wedge sinograms using ML methods. From left to right are: ground-truth, missing-wedge sinogram, recovered sinogram using ML, ground truth image, inverse Radon transform of missing-wedge sinogram, inverse radon transform of recovered sinogram.

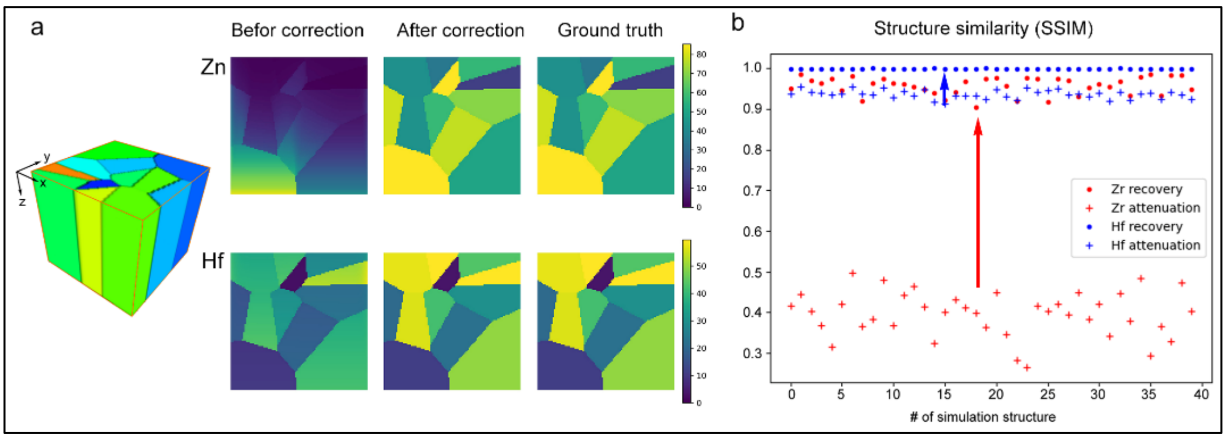


Figure 2. (a) Fluorescence image of a simulated zinc-hafnium (Zn-Hf) alloy before and after absorption correction. (b) Statistical analysis of structure similarity (SSIM) for over 40 diffenrent Zn-Hf compositional models. The red and blue arrows indicate the improvement of SSIM after correction.

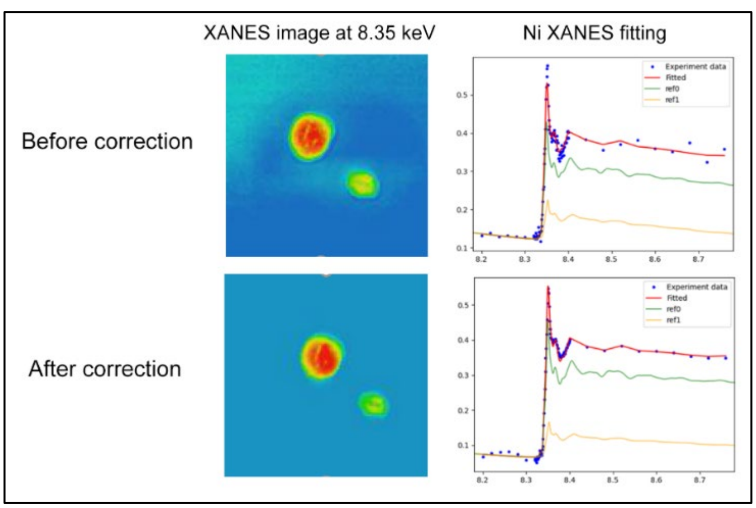


Figure 3. Nickel (Ni) XANES images and fitting comparison. The first row shows the raw image data and the region of interest spectrum fitting results. We clearly see the non-uniform background in the image (left), which results in the noise spectrum shown on the right (blue dots). The second row shows the results after ML denoising. We see the uniform background and smoothed spectrum.

MILESTONES:

2020: comprehensively test existing methods and identify feasible methods; propose our own models if necessary (completed); 2021: prototyping code and production implementation; publish our work in journals; 2022: deploy the package to users and open-source platforms; interact with users to improve the code; seek potential external partnership opportunities.

Intelligent Quantum Dot Growbot for High Throughput Targeted Quantum Materials

LDRD # 20-031

S. Ghose

PURPOSE:

Colloidal lead halide perovskite quantum dots (PQDs) have recently emerged as a highly efficient single-photon emitter with record optical coherence. This project is focused on developing a machine learning (ML) guided approach to the predictive colloidal synthesis of semiconductor lead halide perovskite nanocrystals. Establishing a predictive science of PQD nanocrystal synthesis is a major challenge in nanoscale manufacturing due to the chemical complexity of colloidal crystallization reactions. Therefore, we seek to use ML to predict reaction conditions (e.g., temperature, reagent concentrations) that can yield final nanocrystal ensembles with a desired peak absorbance/emission wavelength. From there, reaction conditions that can optimize key photophysical properties, such as quantum yield and emission linewidth for a given peak emission wavelength, will be targeted. The success of this project will help further establish user facilities for multimodal in situ X-ray scattering experiments at National Synchrotron Light Source II (NSLS-II) and advance this project's focus area of applying artificial intelligence (AI) to quantum materials discovery at Brookhaven National Laboratory (BNL).

APPROACH:

In situ characterization of nanocrystal synthesis in flow reactors has been explored in recent years as an effective tool for high throughput data collection for screening synthesis conditions and monitoring the synthesis. While the advantages of such an approach as compared to batch flask synthesis for efficiently screening different reaction conditions is substantial, the full parameter space of all reasonable reaction conditions for a given synthesis goal is still quite large. Consequently, the application of ML techniques to help guide the search for optimized reaction conditions in nanocrystal synthesis, which historically has largely been developed empirically, is a rapidly expanding field.

Our approach focuses on developing a flow reactor for lead halide perovskite nanocrystal synthesis, where different residence times or reaction timepoints can be measured at a given distance along the reactor path for a set flow rate. Such reactors have been demonstrated by our collaborator Prof. Milad Abolhasani at North Carolina State University using ultraviolet-visible (UV-Vis) absorbance/emission spectroscopies. However, the use of synchrotron X-ray diffraction techniques which provide complementary structural data on atomic and colloidal length scales has not been explored. We will combine UV-Vis absorbance/emission spectroscopies and the use of synchrotron X-ray scattering techniques to benchmark high throughput multimodal screening of reaction conditions. This will provide a training set of data for ML to yield a predictive relationship between reaction conditions and nanocrystal product photophysical properties alongside complementary structural details at different length scales from X-ray scattering measurements. The decision-making algorithm is being developed by BNL Computational Science Initiative scientist Dr. Ai Kagawa. Once the whole workflow is successfully tested, this equipment could be used for autonomous experiments for predictive nanocrystal synthesis at BNL.

TECHNICAL PROGRESS AND RESULTS:

The project depends on two major parts: a Growbot (design, build, test, and commission the flow reactor and diagnostic setup at NSLS-II beamlines) and an ML Algorithm (develop, test and implement decision-making algorithms).

Flow Reactor Growbot: FY21 work was delayed partially by COVID-19 pandemic restrictions and partially by workforce loss. In FY21, we focused on benchmarking the flow reactor growbot designed in FY20 with in situ synchrotron X-ray and lab photophysical characterization of $CsPbX_3$ perovskite flow synthesis. A custom designed X-ray flow cell for in situ X-ray scattering measurements of $CsPbX_3$ formation was tested at multiple NSLS-II beamlines, including using multimodal characterization Small-angle X-ray Scattering (SAXS)/X-ray Total Scattering Pair Distribution Function (PDF), UV-Vis absorbance, and photoluminescence (Figure 1). A manuscript on the proof-of-principle results from the in situ scattering experiments at NSLS-II beamlines is being prepared for publication.

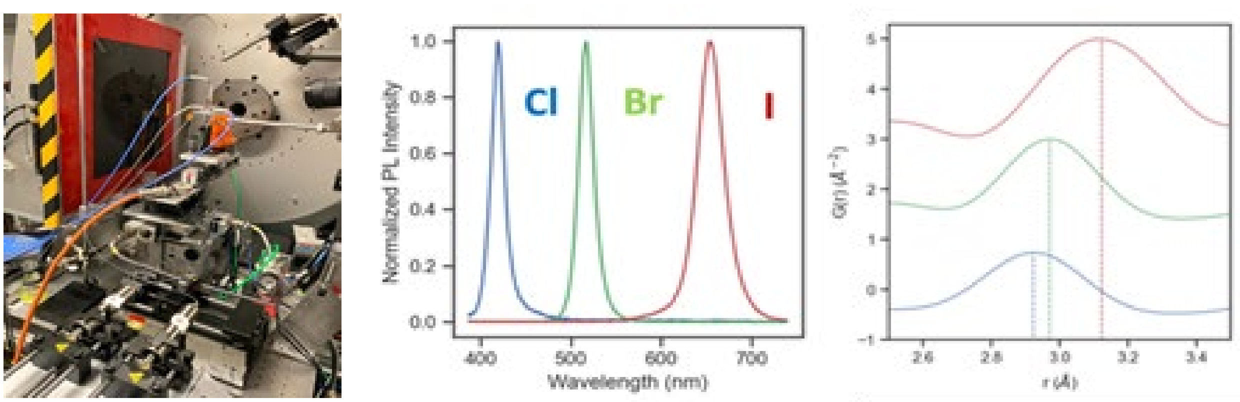


Figure 1. (Left) X-ray flow cell at X-ray Powder Diffraction beamline at NSLS-II. (Right) In situ photoluminescence band gap tuning of $CsPbX_3$ PQDs.

ML Algorithm: in FY21, we investigated a multi-objective Bayesian optimization approach with a hypervolume improvement algorithm to improve the optimization results from the weighted single objective algorithms worked on in FY20. We compared three multi-objective algorithms: a random algorithm (Sobol), an evolutionary algorithm (qParEGO), and an expected hypervolume improvement algorithm (qEHVI). Preliminary results are promising and a manuscript is in preparation.

MILESTONES:

FY22: Synthesize nanocrystals with different input parameters and collect in situ data for UV-Vis Absorbance/Emission and synchrotron X-ray SAXS/PDF during synthesis. Use the experimental data to train the machine and apply the multi objective hypervolume improvement algorithm to guide the experiment for targeted synthesis of PQDs. Build all the interfaces to integrate the AI/ML algorithm into the NSLS-II Data Science and Systems Integration-developed data acquisition and analysis protocols.

FY23: Optimize the complete workflow for an autonomous experiment in synthesizing targeted PQDs. Compile the results and prepare manuscript for publications.

Accelerating Materials Discovery with Total Scattering via Machine Learning

LDRD # 20-032

D. Olds, S. Campbell, T. Caswell

PURPOSE:

The goal of this project is to develop tools and protocols, integrated with the Bluesky control software, to perform prompt analysis of streaming data via Artificial Intelligence/Machine Learning (AI/ML) methods. This is an advancement from the current state of the field where analysis is largely separate and after-the-fact from measurement on beamlines. We will prototype these methods by developing AI/ML methods for characterizing nanoparticle size/shape information from X-ray atomic Pair-Distribution Function (PDF) data. These new AI/ML methods have the potential to produce results in mere seconds that previously took months of computational effort with conventional approaches. Success in this work can lead to expanded analysis capabilities at all beamlines running the Bluesky software, greatly improving user operations and accelerating the pace of discovery at National Synchrotron Light Source II (NSLS-II) and all Department of Energy (DOE) Office of Basic Energy Sciences lightsource user facilities.

APPROACH:

At NSLS-II, materials discovery from in situ measurement is often bottlenecked not by measurement speed, but subsequent analysis time. For example, conventional analysis of total scattering data via PDF analysis involves a fitting of models to measured data - a classical example of an inverse problem. A single instance of the calculation required to derive a PDF from a model scales as the order of the square of the number of atoms, making it computationally challenging for large-scale structures. However, this procedure is made even more costly when employed in the refinement of data, where many, many repetitions of this calculation can be required.

Recent work has described a method to extract precise details of nanomaterial size, shape, faceting, and polydispersity through careful analysis of the characteristic damping envelope that encompasses the measured PDF, which is referred to as the shape-function. This approach has been utilized in the studies of several hierarchical systems including layered MXene materials, platinum-palladium nanoalloy catalysts, gold nanoparticles, layered δ-MnO_2 (birnessites), alumina nanoplate products such as Boehmite and gibbsite, and a particularly noteworthy study that was able to fully resolve the faceting of TiO_2 nanocrystals.

Collaborating with Josh Lynch of the Data Acquisition, Management and Analysis group (NSLS-II), we are developing AI/ML methods to characterize the shape-function of materials from PDF data, aligned with the Bluesky suite of beamline control software.

TECHNICAL PROGRESS AND RESULTS:

During FY21, while we continued development of the previous agents, we also began to explore and develop other AI/ML methods to meet the emerging needs of the beamline. Particularly, these drivers were the result of newly identified critical capabilities required to optimize data collection during remote operations – now emphasized by the ongoing pandemic. As a result, we developed a suite of AI/ML-driven data collection and analysis agents including a Reinforcement Learning (RL)-based agent designed to optimize data collection across multiple samples, and a new unsupervised learning method called Constrained Matrix Factorization (CMF) that helps users quickly and efficiently understand their data during in situ experiments on the beamline. We used the infrastructure developed during the previous year of the project to deploy these methods on the PDF beamline. Figure 1 demonstrates the result of CMF during a gas flow measurement, which was only possible due to the agents developed as part of this project. This work has resulted in multiple publications, and future directions to further enhance high-throughput data collection on the beamlines at NSLS-II, which we will explore in the final year of this project.

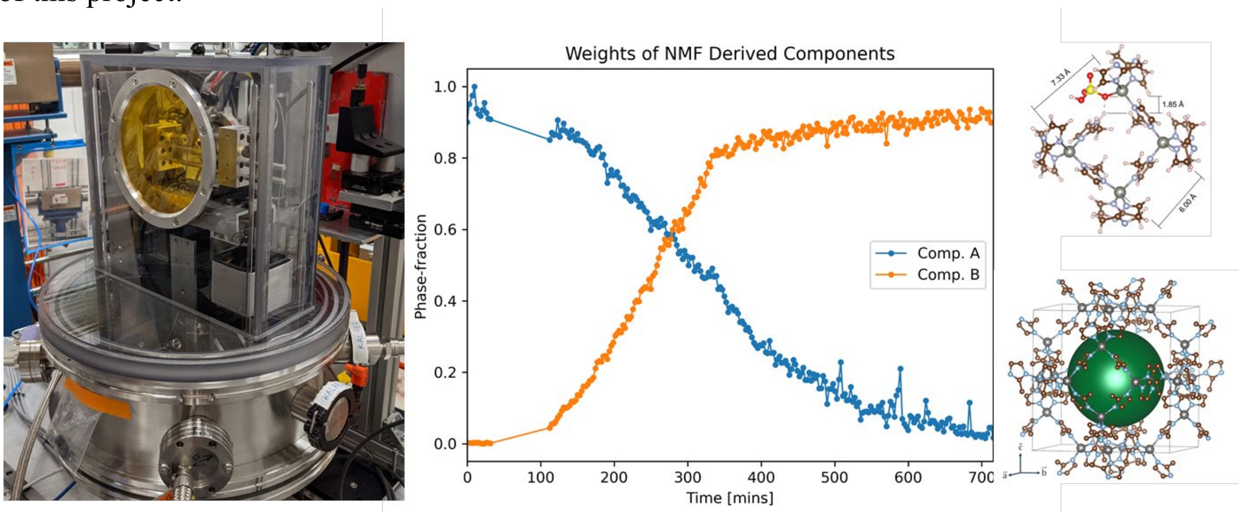


Figure 1. (Left) A hazardous gas flow sample environment on the PDF beamline, under which the degradation of the atomic structure of Metal Organic Frameworks was studied during flows of nitrogen oxide gases (NOx) using x-ray total scattering. The precise structure of the degraded product and reaction kinetics was unknown prior to the measurement, making conventional analysis difficult. Using CMF, we were able to monitor the progression of the experiment (Center) without a priori knowledge of the two components we were monitoring (Right).

To summarize, in 2021, we developed CMF; tested employing CMF on the PDF beamline during user experiments; expanded use of AI/ML "hooks" for Bluesky onto other beamlines (X-ray Powder Diffraction); further developed high-throughput measurement capabilities with BadSeed. The results of this project formed the basis of a DOE Early Career Proposal.

MILESTONES:

- Further develop and deploy the CMF method on additional beamlines
- Publish "best practices" guidance on deploying AI/ML methods on beamlines
- Re-scope "ShapeML" agent goals to fit the needs of the community
- Re-submit DOE Early Career Proposal using results from this project.

Storage Rings for Quantum Computing

LDRD # 20-035

K. Brown

PURPOSE:

The technical objective of this project is to perform a design study for a specialized type of storage ring, called a circular radio frequency quadrupole (CRFQ), that will be used to create ion Coulomb crystals. These crystals could be the basis for a prototype large scale quantum computer, capable of containing thousands of quantum bits (qubits). Such an approach is novel and has never been attempted. The appeal of this approach is it builds on a large body of research and knowledge developed in the ion trapping and particle accelerator communities. A CRFQ is essentially an unbounded ion trap, in which the ions circulate with some fixed velocity. Since ions are stationary (on average) in an ion trap, laser cooling systems can operate in all directions to reduce the motional modes of the ions in the trap. The result is a stable ion Coulomb crystal. In a CRFQ, the ions are not stationary, so new methods need to be developed to cool the ions in all directions as well as to utilize the ions as qubits. Using ions for qubits means the systems for performing quantum operations are true quantum systems. Such a system would take Quantum Information Science (QIS) beyond noisy intermediate-scale quantum (NISQ) systems and would be capable of solving large scale and "true" quantum computing problems. Many QIS approaches aim for commercial NISQ systems. These tend to be smaller scale systems, with a handful to a few dozen qubits at the most. Some systems can scale to thousands of qubits. These perform quantum computations based on quantum annealing or using specialized quantum materials. Such QIS systems are sufficient for many problems. However, a large-scale QIS system (on the order of tens of thousands to hundreds of thousands of qubits) that goes beyond NISQ systems that can perform any kind of quantum computation is an ambitious endeavor. The focus of this project is to explore the potential of storage ring technologies to reach such a goal.

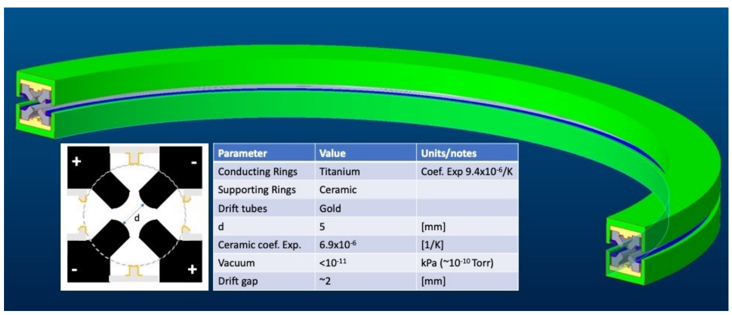
APPROACH:

In the field of quantum computing, many technologies are being studied as platforms for QIS, each of which has its own challenges and limitations. It is generally expected that a practical quantum computer be scalable, have means for initializing qubits, allow operations within decoherence times, have methods for a universal set of operations, and allow qubits to be easily read. Quantum computers that might be compared to a storage ring system would be those that attempt to operate at the atomic level. A trapped ion quantum computer will confine ions or other charged particles in some free space using electromagnetic fields. The qubits are the quantum states associated with each ion. Ion trap systems are scalable, in principle, and hold much promise as quantum computing systems. A storage ring system is very similar to an ion trap. A significant difference is that the particles will be confined into a circular electromagnetic guide. Storage ring ions will have finite average velocities, although velocity is a free parameter that can be optimized. Ions in a storage ring can be entangled, as they can in an ion trap. A storage ring quantum computer (SRQC) can contain large numbers of ions' (and so is scalable) and potentially can be a platform for large numbers of qubits. With the ability to entangle the ions internal quantum states, the system will have methods for a universal set of operations.

Quantum computing is a growing field, still very early in its development. The idea of using storage rings for quantum computing is novel and new, although traditional storage rings cannot establish the proper conditions, since creating and preserving ion Coulomb crystals in storage rings require conditions that are not disruptive to the crystalline structure, such as strong changes in fields for focusing and the dispersive optics from bends. CRFQs produce ideal conditions for creating and maintaining such crystalline structures. Since the main difference between the CRFQ and a linear Paul trap is that the ions in the CRFQ have a nonzero velocity, the focus of this work is to explore the design of a CRFQ that can be optimal for creating very low temperature crystalline beams. Prof. Boris Blinov from the University of Washington, who is an expert in ion traps, joined as a collaborator and is interested in the design and methods for cooling the ions to create ion Coulomb crystals.

TECHNICAL PROGRESS AND RESULTS:

This is a new project that is gaining interest worldwide. For this project, much of the work focused on exploring the unique problems associated with using a storage ring as an unbounded ion trap. We have explored the design of a CRFQ (Figure 1) with optimal conditions for ion Coulomb crystals with various ion species (magnesium, lithium, beryllium, ytterbium (Yb+), barium (Ba+)). The ion trap community has found many ions that have very long coherence lifetimes. Yb+ and Ba+ are prime candidates since they are known to have internal eigenstates with lifetimes exceeding one hour. We have explored methods for addressing ions with nonzero velocity and how to change and measure the internal eigenstates. Work on this problem continues and is promising. We have studied ion Coulomb crystal theory and dynamics and how to locate and identify ions traveling in a circular trap. We have studied ion beam cooling methods and have developed a potentially promising new three-dimensional cooling method using an electrostatic wiggler. This work is ongoing and progressed in the second year of the project. Finally, we have developed a preliminary theoretical description on mapping quantum circuits to ions in an SRQC. The work from this year's efforts produced a very important study that explored the challenges and directions for research and development (published in Phys. Rev. Accel. & Beams), as well as two new inventions, soon to be submitted for patent potential. Finally, efforts have spun off from this work to address problems that are common between ion traps and storage rings and are of high value to a storage ring system. These include studies on detecting qubit states and ways to study quantum excitations in test bed accelerators, such as at IOTA at Fermilab and at DESIREE at Stockholm University in Sweden.



Parameter	Value	Units/notes
Conducting Rings	Titanium	Coef. Exp 9.4×10^{-6}/K
Supporting Rings	Ceramic	
Drift tubes	Gold	
d	5	[mm]
Ceramic coef. Exp.	6.9×10^{-6}	[1/K]
Vacuum	$<10^{-11}$	kPa ($\sim 10^{-10}$ Torr)
Drift gap	~2	[mm]

Figure 1. Circular Radio Frequency Quadrupole Computer-aided Design Model (for ½ of the ring) and cross-section showing the electrodes and ceramic frame (supporting rings) with gold plated drift tube electrodes.

We have engaged a larger community into the discussion of this approach, with many scientists showing great interest from the ion trap community, among quantum information scientists, accelerator physicists, and many others. Of note were notable scientists who signed a letter of intent at the most recent Snowmass reviews in the particle physics community presented in September 2020. At the International Particle Accelerator Conference (IPAC 2021), held virtually from Campinas, Brazil, there was significant interest in the paper we presented on storage rings for QIS. There continues to be great interest, as seen in the upcoming Innovation Fostering in Accelerator Science and Technology (iFAST) Work Package 5 Workshop on Extreme Storage Rings "ESRW22", which includes a complete session on crystalline beams for quantum computing.

QIS is a very competitive area of research, although no one else is working on using a CRFQ for QIS. We have submitted proposals to DOE calls. The comments from reviewers are very positive and extremely helpful. We are addressing those comments in new proposals. As this project ended, we continue to pursue new funding opportunities. However, this project has tremendously helped to jump start this specialized field of research. Groups around the world are looking at the use of storage rings for QIS and our preliminary work is helping them to direct their efforts. Our own collaboration remains mostly in place and several research projects will lead to publications in the coming year.

MILESTONES:

None. This project completed in FY21.

The Rice of the Future: How Growing Practices Can Decrease Human Exposure to Toxins

LDRD # 20-037

R. Tappero, A. Seyfferth (University of Delaware)

PURPOSE:

Rice is grown on every habitable continent and is an important source of calories for billions of people worldwide. Most rice is grown under flooded soil conditions, which mobilize the toxic and cancer-causing element arsenic (As) that gets absorbed by the roots of rice plants. Our aim is to discover sustainable and cost-effective best management practices (BMPs) to reduce the loading of toxic metal(oid)s (e.g., As and Cadmium [Cd]) into edible grains. Our objective is to gain a predictive-level understanding of how specific agronomic practices (silicon [Si] amendments, water management) influence rhizosphere chemistry and ultimately the abundance and speciation of toxic metal(oids) in edible grain. To date, plant science initiatives at Brookhaven National Laboratory (e.g., the Environment, Biology, Nuclear Science & Nonproliferation's Quantitative Plant Science Initiative) have targeted the plants themselves; however, the rhizosphere (soil immediately adjacent to plant roots and directly influenced by roots and associated microorganisms) is the next frontier for plant science and the key to global food security and human health.

APPROACH:

Extensive research has been conducted on the fate and transport of toxic metals in soil and the biogeochemical processes controlling metal(oid) "bioavailability." From this level of understanding, it is possible to devise agronomic practices to minimize the loading of toxic metals into edible grains of wetland rice. Our approach is to develop BMPs that create favorable biogeochemical conditions in soil to minimize metal(oid) bioavailability at key points during rice cultivation (e.g., ripening). A critical part of this development process involves spatio-temporal monitoring of toxic metal(oid)s and plant nutrients (e.g., sulfur [S]) in rice grain and rhizosphere. To this end, a technical development in solid-phase speciation was needed to combine "tender" and hard X-ray measurement of both "light" and "heavy" elements in plants or soils.

TECHNICAL PROGRESS AND RESULTS:

We completed the conceptual design, fabrication, and testing of the imaging system for combined "tender" and hard X-ray measurements at the X-ray Fluorescence Microprobe beamline at National Synchrotron Light Source II. In FY21, we utilized this new capability to image Si, S, and As in rice grain yielded from the various BMPs, developed a method to preserve wetland roots and rhizosphere for elemental imaging (Seyfferth et al., 2021; doi: 10.3791/62227) and utilized this new method to characterize the rhizosphere soil for different BMP soil amendments (Linam et al., 2021 in press). Collectively, these results (see Figure 1, for example) will provide mechanistic understanding of the role of BMPs in reducing metal(oid) bioavailability in soil and subsequent loading into edible grains.

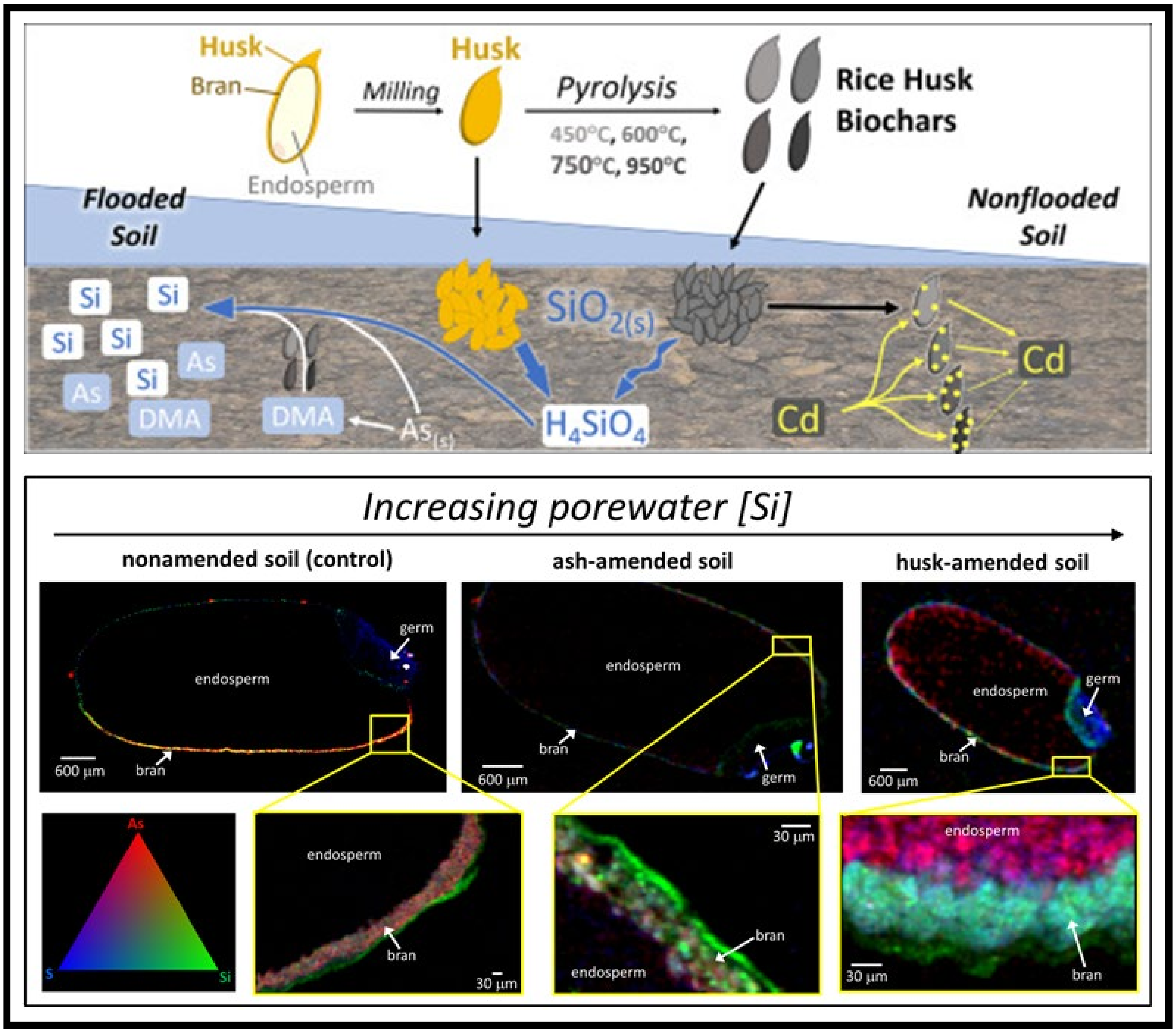


Figure 1. (Top) Depiction of the biogeochemical cycling of As and Cd in the rice rhizosphere as influenced by different Si-rich soil amendments (e.g., husk, biochar). (Bottom) Differences in As loading into rice grains obtained for the different soil amendments, showing that husk amendment altered metal(oid) uptake and localization in rice grain. As = red, Si = green and S = blue.

MILESTONES:

None. This project completed in October 2021.

Machine Learning for Real-Time Data Fidelity, Healing, and Analysis for Coherent X-ray Synchrotron Data

LDRD # 20-038

A. Barbour, A. DeGennaro

PURPOSE:

The intent of this project is to develop and apply computational tools that automate the processing and analysis of X-ray Photon Correlation Spectroscopy (XPCS) data. The motivation for this is the “big data” challenge. User facilities (e.g., National Synchrotron Light Source II [NSLS-II]) are capable of generating a significant amount of raw experimental data, but the utility of this data is currently limited by the speed at which human engineers and scientists can post-process and evaluate it. XPCS is a technique that produces complex results with data similar in size to the input raw data. Existing community attempts to process and evaluate the results are limited to heuristic curve fits and ad hoc solutions that are not sufficiently robust across all datasets and often require expert intervention, parameter tuning, and supervision. A solution that automates this analysis and post-processing is needed in order to accelerate the pace of light-source-based science and to make this science more accessible to non-expert users. Artificial Intelligence/Machine Learning (AI/ML) techniques will be investigated and developed to solve the problem of human-in-the-loop decision making that requires expert knowledge. To our knowledge, AI-guided XPCS methods have not been published, let alone reported to exist within any X-ray beamline’s data analysis pipeline. With an AI-guided XPCS data analysis pipeline at NSLS-II, we can better understand how to address similar problems outside of XPCS.

APPROACH:

AI-guided XPCS analysis will have a large impact on international X-ray Synchrotron Science from the point of view of users and scientific staff. As the first step, we are investigating technical methods from the field of ML. We are using convolutional autoencoder neural networks as the backbone of our approach to the data processing. The goal of an autoencoder is to learn a compressed, low-dimensional representation of the training data that is presented to it. Because this representation is compressed, it is approximate, and therefore the output of the autoencoder can be interpreted as a “smoothed” version of the original input datum. This black box smoothing operation is one of the most important steps of the XPCS data post-processing, as it can be used to remove corrupting artifacts from unusual/unexpected experimental conditions and other forms of undesirable noise.

It is also the case that there are distinct categories of data that could be fed into our data processing algorithms. For example, data collected from systems in equilibrium look very different from data collected from non-equilibrium systems. Using a single autoencoder to post-process both types of data is perhaps asking too much; one could possibly do better by first labeling a given datum (e.g., as either “equilibrium” or “non-equilibrium”) and then apply to the result one of several autoencoders that has been specially trained on only that category of data. This represents a two-step flow of work, and both steps (the labeling and smoothing) can be handled by convolutional neural networks.

TECHNICAL PROGRESS AND RESULTS:

The project’s progress in the last four months of FY20 until the end of FY21 was the hire of Tatiana Konstantinova (Research Associate). Data and traditionally computed results for equilibrium, non-equilibrium, and static dynamics from the Coherent Soft X-ray Scattering (Andi Barbour) and Coherent Hard X-ray Scattering (Lutz Wiegart) beamlines were aggregated and labeled by Konstantinova. In general, we have a few models for a variety of dynamics (50% published) that remove noise (artifacts). The models are accompanied by recently developed modular software (Konstantinova and Anthony DeGennaro) that enabled testing on user data and future deployment at NSLS-II beamlines for automated data analysis (FY21). The latest models for non-equilibrium dynamics (mid-FY21) were essential to reliable quantification of dynamics, which are only limited by the measurement’s time resolution. Konstantinova developed code for parallel XPCS fitting and results extraction. This not only enabled real time, high throughput automated analysis, but also opened possibilities for autonomous XPCS experiments or databases of reliably labeled XPCS results for future use by complex AI models (analysis or materials discovery). The application of these new models and scientific software began in earnest (late FY21).

In collaboration with other ML Laboratory Directed Research and Development projects since FY21, Konstantinova developed an anomaly toolkit that utilizes a one-dimensional signal that labels data as "good" or "bad." The draft manuscript is finalized and pending a collaborator's institution approval.

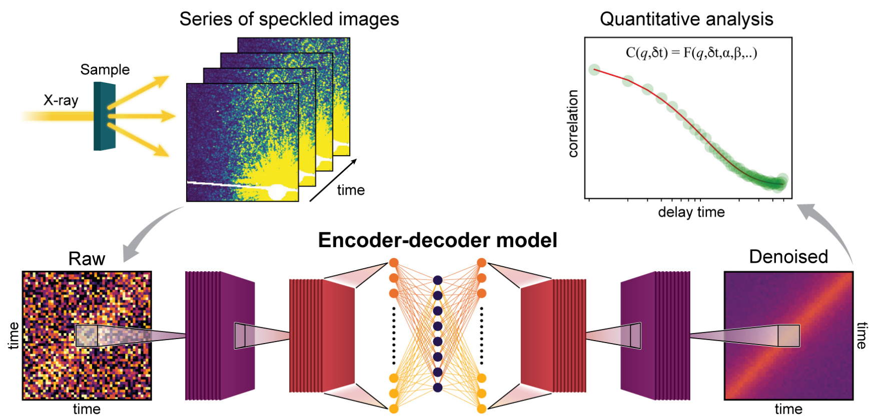


Figure 1. Workflow for XPCS experiment with a denoising autoencoder model incorporated into the analysis. During an XPCS experiment, a series of scattered speckled images is recorded and the (raw) intensity-intensity correlation function is calculated. The function is passed through the denoising model and the (denoised) output is used for further analysis. Domain-specific measures are employed for quantification of the model uncertainty and making decisions about the use of its output. The denoised version of the correlation function is used for fitting to a certain functional form. The trustworthiness of the results is determined by their fit error and their relative values.

Resulting Publications:
Campbell, S. et al. Outlook for artificial intelligence and machine learning at NSLS-II. Mach. Learn. Sci. Technol. (2020); Konstantinova, T. et al. Noise reduction in X-ray photon correlation spectroscopy with convolutional neural networks encoder–decoder models. *Sci Rep* **11,** 14756 (2021).

MILESTONES: (as perceived by end users)

- FY21: beamlines with ML embedded in Data Acquisition, Management and Analysis infrastructure to start testing artifact identification with NSLS-II users and understand how to improve training sets (on going)
- FY21: begin to understand artifact removal (done)
- FY22: optimized binning of time-resolved, out-of-equilibrium dynamics using ML (done)
- FY22: begin effort to remove artifacts from simulated data
- FY23: routinely use artifact identification at beamlines and implement it to screen data prior to automated analysis (on going)
- FY23: automated data analysis for extracting physical quantities from large sets of correlation functions (done)
- FY23: start testing artifact removal with NSLS-II users (on going)
- FY23: perform an experiment on an unknown scientific sample with induced dynamics due to consequential experimental conditions.

Electrochemical Systems for Large Scale Energy Storage

LDRD # 20-039

E. Takeuchi, A. Marschilok, D. Bock, L. Wang

PURPOSE:

The project will engage in the design and assembly of new electrochemical cell designs suitable for in situ and in operando synchrotron-based research in consideration of various cell architectures. Traditional cell materials and architectures can be studied using synchrotron techniques. However, this is not the case for novel and scalable architectures. The research will consider the science of scalable energy storage systems and the needed methodology for their characterization. In situ and operando cell designs will be designed, constructed, and tested electrochemically. When electrochemical function is demonstrated, then the cell designs will be used for the in situ and operando collection of data at National Synchrotron Light Source II (NSLS-II). This will enable time and spatially resolved collection of electrochemical behavior.

APPROACH:

Initial work is focused on the development and demonstration of suitable test cells for sample evaluation under electrochemical use conditions. The test cells are validated electrochemically to ensure reasonable agreement with performance data collected in previously demonstrated cell configurations. The test cells are then used at NSLS-II as appropriate to demonstrate the viability of material analysis at the anode and cathode of the cell. Cells are designed and fabricated consistent with the space requirements of the end stations at NSLS-II. The sample electrodes are sized consistent with the target set of experiments. The potentiostat controlling the current and/or voltage of the cell will be implemented into the beamline data acquisition system.

TECHNICAL PROGRESS AND RESULTS:

Extended Cycling Capability: In the past year, we successfully designed, fabricated, and demonstrated a planar cell design suitable for in situ and operando diffraction and spectroscopy measurements, with a lithium (Li) metal negative electrode and metal oxide cathode that were effective over extended cycling. Figure 1 shows Li/ $LiNi_{0.8}Mn_{0.1}Co_{0.1}O_2$ (Li/NMC811) batteries that were cycled to different voltage limits (4.3 and 4.7 V) for 100 cycles in both a conventional coin cell design (Figure 1a) and in the developed in situ pouch cell design (Figure 1b). The delivered capacities of the two cell types are very similar, verifying that the electrochemical behavior using in situ design is not compromised over high cycle numbers.

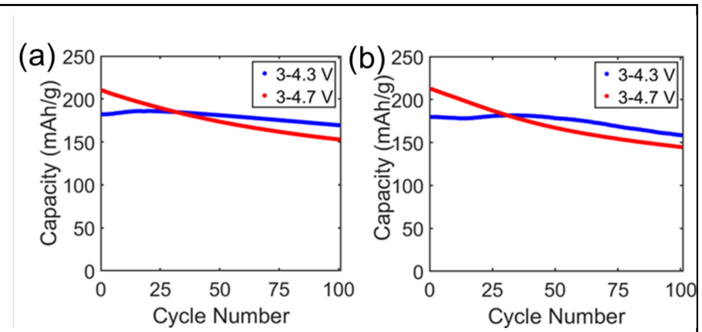


Figure 1. (a) Representative galvanostatic cycling of Li/NMC811 coin cells. (b) Representative galvanostatic cycling of Li/NMC811/Li in situ pouch cells.

Full Cell Design: An in situ cell design was developed that is suitable for a full cell configuration, with graphite anode instead of Li metal anode and NMC cathode material. This cell design is the type of configuration used currently in commercial Li-ion batteries. Excellent electrochemical performance of these cells was observed at both 1C (60 minute charge time) and 4C (15 minute charge time) charge rates with a 1C discharge (Figure 2) with excellent capacity retention observed over 150 cycles. This new design enables operando probing of battery electrodes at extremely fast charging rates. This achievement was critical for successful progress of several funded programs where fast charge rates, such as 15 minutes, are a key variable.

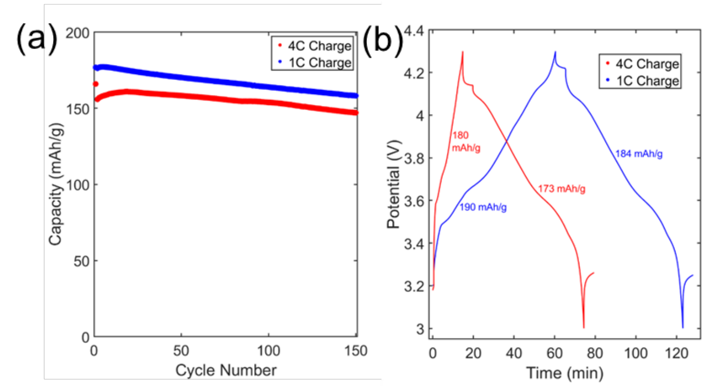


Figure 2. (a) Capacity versus cycle number. (b) Representative voltage profiles for in situ graphite/NMC811 full cells cycled with 60-minute charge time (blue) and 15 minute charge time (red).

Demonstration at NSLS-II: Both the lithium anode cell arrangement and the full cell design were demonstrated successfully at NSLS-II using the operando methodology. The new cell design was used to collect simultaneous operando synchrotron X-ray diffraction (XRD) and X-ray absorption spectroscopy (XAS) of Li/NMC622 cells, which had previously been cycled 150 times prior to beamline measurement (Quilty et al., 2021). Li/NMC811 cells were measured operando using the multi-cell holder using XAS, with analysis of both X-ray Absorption Near Edge Structure (XANES) and Extended X-ray Absorption Fine Structure (EXAFS) to understand changes in oxidation state and local atomic structure as a function of cycle number and cell charging voltage (Quilty et al., Multi-modal X-ray Probing…). The full cell design was used to collect operando XRD and XAS data under extremely fast charging rates to understand cathode aging mechanisms at high rate (Quilty et al., Elucidating Cathode Degradation…). Thus, the cell designs developed under this project have enabled improved understanding of the structural degradation modes in state-of-the-art Li-ion batteries.

Resulting Publications

- Quilty, C. D.; Wheeler, G. P.; Wang, L.; McCarthy, A. H.; Yan, S.; Tallman, K. R.; Dunkin, M. R.; Tong, X.; Ehrlich, S.; Ma, L.; Takeuchi, K. J.; Takeuchi, E. S.; Bock, D. C.; Marschilok, A. C., Impact of Charge Voltage on Factors Influencing Capacity Fade in Layered NMC622: Multimodal X-ray and Electrochemical Characterization. *ACS Applied Materials & Interfaces* **2021,** *13* (43), 50920-50935.
- Quilty, C. D.; West, P. J.; Dunkin, M. R.; Ehrlich, G. P. W.; Ma, L.; Jaye, C.; Takeuchi, E. S.; Takeuchi, K. J.; Bock, D. C.; Marschilok, A. C., Multimodal X-ray Probing of The Capacity Fading Mechanisms of Nickel Rich NMC – Progress and Outlook. *(under review)*.
- Quilty, C. D.; West, P. J.; Wheeler, G. P.; Housel, L. M.; Kern, C. J.; Tallman, K. R.; Ma, L.; Ehrlich, S.; Jaye, C.; Fischer, D. A.; Takeuchi, K. J.; Bock, D. C.; Marschilok, A. C.; Takeuchi, E. S., Elucidating Cathode Degradation Mechanisms in NMC811/Graphite Cells Under Fast Charge Rates Using Operando Synchrotron Characterization. *(under review)*.

MILESTONES:

Develop cell holder that can accommodate multiple cells simultaneously.

Conceptual Design Options for Future Upgrade of National Synchrotron Light Source II (NSLS-II) Facility

LDRD # 20-041

V. Smalyuk

PURPOSE:

The purpose of this project is to deliver a pre-conceptual design for the National Synchrotron Light Source II (NSLS-II) upgrade, which when implemented, will result in a large gain in the facility performance compared to today's operations. Under all upgrade scenarios, the upgraded NSLS-II would have the highest coherent fraction among the U.S. synchrotron light sources and dominate in brightness in the photon energy range between 1 keV and 10 keV. These pre-conceptual designs will enable us to be prepared for the start of the Department of Energy (DOE) Critical Decision process.

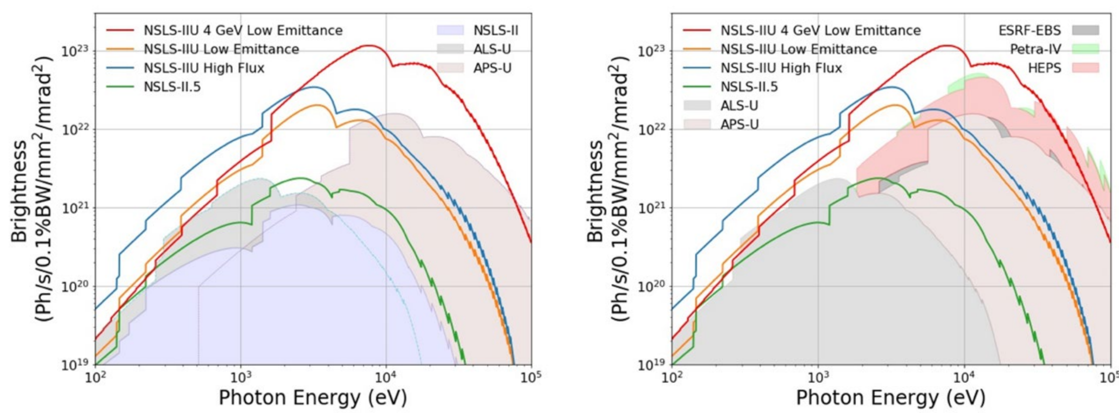


Figure 1. (Left) Brightness of the NSLS-II upgrade (NSLS-IIU) options compared to the present NSLS-II, Advanced Photon Source Upgrade (APS-U), and Advanced Light Source Upgrade (ALS-U). (Right) Compared to the light source projects worldwide.

APPROACH:

Three approaches are considered for the upgrade of the NSLS-II facility: 1) a medium-scale project replacing just a part of the existing NSLS-II hardware; the expected gain in the facility brightness is about 4-5 times; 2) design of a new low-emittance lattice with the emittance close to the diffraction limit at 10 keV; this will drive Brookhaven National Laboratory to the forefront of synchrotron radiation science; and 3) design of a high-current synchrotron delivering unprecedented intensity with state-of-the-art brightness. We will develop a set of lattice designs together with the assessment of the required changes in the NSLS-II accelerators and subsystems. Figure 1 shows the brightness of the upgrade options compared to NSLS-II and other U.S. synchrotron light sources and other light source projects worldwide.

TECHNICAL PROGRESS AND RESULTS:

We completed an assessment of the medium-scale upgrade option in FY20. The final lattice provides the beam emittance of 300 pm with moderate modification of the NSLS-II hardware.

In FY21, we worked on the lattice design, alignment and stability specifications, and collective effects studies for the low-emittance upgrade of the NSLS-II storage ring:

- Completed the assessment of a high-flux upgrade option with maximum beam current of 2 A and emittance of 100 pm·rad. We analyzed the beam-induced heating of vacuum chamber components, single-bunch and multi-bunch instabilities, beam lifetime, and injector requirements. Finally, we concluded this upgrade option is not optimal due to performance risks and high cost.

- Developed a hybrid multi-bend achromat lattice, which adopts a layout similar to the European Synchrotron Radiation Facility-Extremely Brilliant Source (ESRF-EBS) lattice. The emittance was reduced to 28 pm, and the nonlinear beam dynamics were optimized to achieve a dynamic aperture sufficient for the off-axis injection scheme.
- Proposed a Hybrid Complex Bend Achromat lattice providing an emittance of 24 pm, dynamic aperture sufficient for the off-axis injection, and a momentum aperture large enough to result in a beam lifetime of several hours.
- Analyzed the longitudinal beam dynamics in the Double Complex Bend Achromat 25 pm lattice. For filling patterns with five and ten bunch trains, a bunch lengthening by a factor of three is achievable, together with a moderate shift of bunch centroids and small non-uniformity of bunch lengths along the train.
- Reviewed alignment and stability specifications for ALS-U, APS-U, Diamond-II, ESRF-EBS, NSLS-II, SIRIUS, MAX IV, SOLEIL-II to determine the future requirements for alignment and stability of the upgraded NSLS-II.
- To expand the capabilities of NSLS-IIU, we started to consider a free-electron laser (FEL) project using a low-emittance electron beam of NSLS-IIU that is synergistic with storage ring operations. A new Laboratory Directed Research and Development project (LDRD 22-028) "Assessment of FEL options for NSLS-II upgrade" has been approved and funded.

In FY21, two post-doctoral researchers worked on this project. A summary of the accelerator physics R&D needed for the NSLS-II upgrade has been written and included in the White Paper proposal to DOE. The main results of this research were published in journal articles and presented at the 12th International Particle Accelerator Conference (IPAC'21).

MILESTONES:
FY22-23

- Converge the lattice options being considered for the high-brightness upgrade into a single lattice with the best performance
- Work on the best combination of the beam emittance, intensity, and energy to realize the highest brightness of undulator radiation
- Analyze collective effects, beam-induced heating, magnetic field errors, and misalignments to specify the engineering requirements
- Work on the cost and schedule estimate.

Development of Wavelength Conversion Techniques for Generation of Coherent Radiation at the Extreme Ultraviolet (XUV) to Long-wave Infrared (LWIR)

LDRD # 21-001

R. Kupfer, T. Rao, J. Wishart, M. Babzien, M. Polyanskiy, and I. Pogorelsky

PURPOSE:

This project aims to develop wavelength conversion techniques based on Raman conversion in solid crystals or ionic liquids followed by sum/difference-frequency generation (S/DFG) in wide-bandgap semiconductor nonlinear crystals pumped with chirped near infrared (NIR) pulses. These techniques can be used for generation of either a broadband long-wave infrared (LWIR) seed or high energy nanosecond mid-infrared (mid-IR) pulses with a scalable repetition rate. By avoiding gain-narrowing from regenerative CO_2 amplification, this approach will provide an immediate and cost-effective path for generation of sub-picosecond, multi-Joule, LWIR laser pulses that can drive blowout-regime laser wakefield acceleration and high harmonic generation (HHG).

APPROACH:

We are addressing three specific wavelength needs: 1). Development of a solid-state broadband LWIR seed source; 2). Development of a wavelength conversion method from NIR to mid-IR based on Raman conversion in ionic liquids; and 3). Development of an XUV/soft X-ray source based on HHG that is driven with either an NIR or LWIR source. Part 1 is necessary to achieve femtosecond-scale, Joule-level LWIR pulses. Part 2 can be used for optical pumping of CO_2 and is critical for repetition rate increase and beam quality improvements. Part 3 will use the upgraded short pulse LWIR to generate XUV/soft X-rays at higher energy and photon flux.

TECHNICAL PROGRESS AND RESULTS:

Raman and UV-Visible-NIR spectra of several ionic liquid (IL) families containing common IL cation and anion motifs were measured to evaluate their suitability for Raman conversion (Figure 1). Criteria included being liquid at room temperature, having a strong Raman band corresponding to a desired target wavelength, and low absorbance at the pump (532 or 1064 nm) and Stokes wavelengths.

The results of the survey indicated that the desired Raman characteristics could be designed into the IL by using anions containing nitrile groups (C-N triple bonds) that have intense Raman bands in the region of 2200-2230 cm^{-1}. Examples include the dicyanamide (DCA, $[N(CN)_2]^-$), tricyanomethanide (TCM, $[C(CN)_3]^-$), and tetracyanoborate (TCB, $[B(CN)_4]^-$) anions. While the TCB samples available in our lab from earlier projects were very promising, TCB ionic liquids are no longer commercially available in the required quantities. We therefore focused on DCA and TCM ILs.

Our survey also showed that IL absorption in the 1064 nm region is primarily due to overtones of C-H stretching bands in the aliphatic regions of the cations. To minimize that absorption, the best IL cations are those with the smallest numbers of aliphatic C-H bonds. Examples are aromatic (imidazolium or pyridinium) cations with the shortest aliphatic side chains that still form room-temperature ionic liquids (e.g., 1-ethyl-3-methylimidazolium (emim)) or have aromatic side chains (e.g., 1-benzylpyridinium (BzPy)) since aromatic C-H bond overtones absorb in another, less important region. Based on these concepts, we have acquired large enough quantities of three candidate ILs for Raman conversion tests (emim DCA, emim TCB, and BzPy DCA).

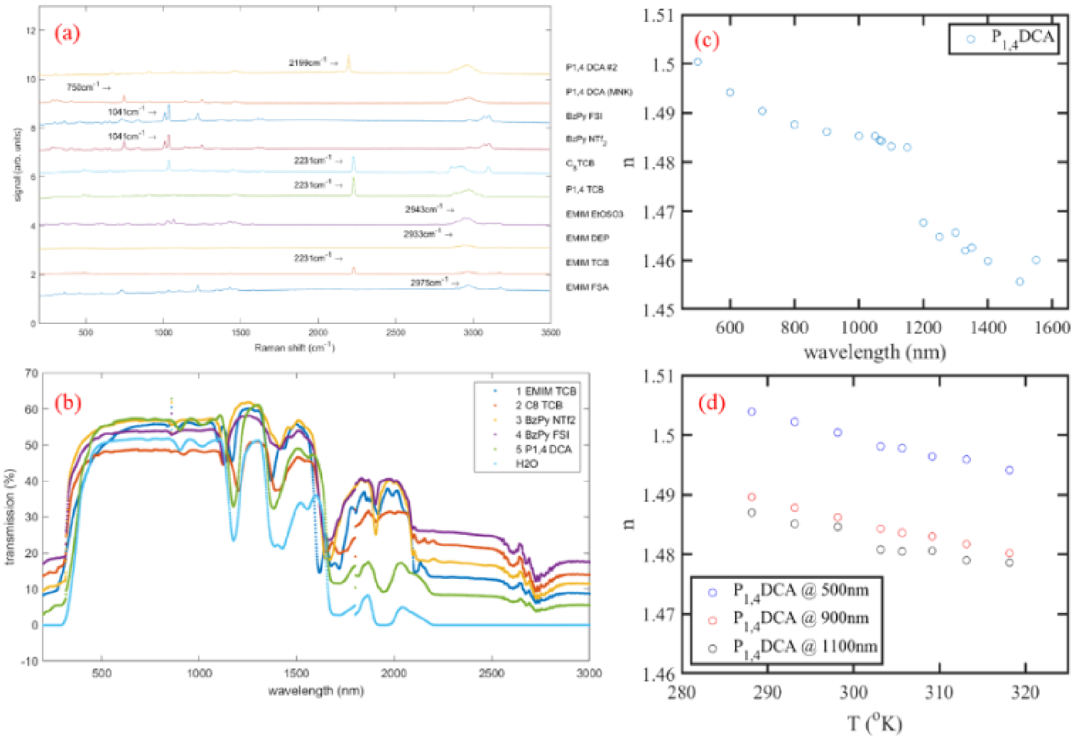


Figure 1. (a) Raman and (b) optical transmission spectra of various ILs. (c) Refractive index variation around optical absorption peak and (d) temperature variation of the refractive index in $P_{1,4}$DCA.

We also characterized the Raman conversion process in Calcite using chirped 800 nm pulses (Figure 2) and found that conversion efficiency to the first Stokes can be as high as 40%, depending on the crystal orientation with good energy scalability. Special attention should be given to the optical power to avoid saturation, red shifting, and spectral narrowing. We also verified the timing for the following DFG process by cross-correlation in barium borate. With the arrival of $BaGa_4S_7$ (BGS) crystals (early 2022), we should be ready to demonstrate conversion to 9.2 µm.

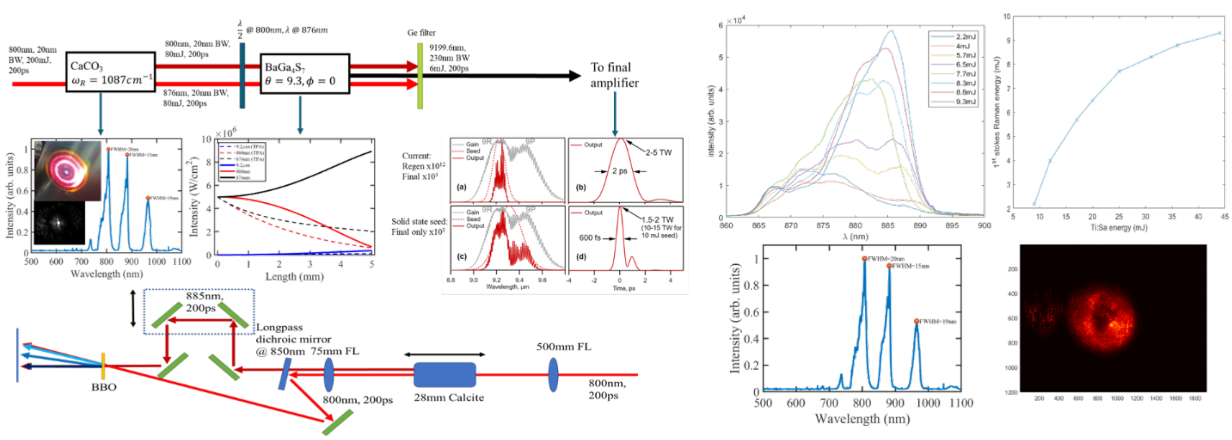


Figure 2. (Top Left) Schematic of the two-step conversion method. A chirped 800 nm pulse is partially Raman converted to 876 nm in Calcite and the two colors are frequency differentiated in a wide bandgap BGS crystal to generate a broadband chirped 9.2 µm pulse. (Bottom) Schematic of a test setup for the Raman conversion stage. (Right) Energy scalability of the Raman process, spectral red-shift due to saturation at higher energy and the resulting 876 nm beam profile.

MILESTONES:

- Perform full scale experiments of Raman conversion in ionic liquids
- Develop a prototype LWIR seed generator at ~100 µJ level
- Prepare the experimental apparatus for HHG with LWIR driver.

Development of an Alexandrite Based Gain Module for Direct Amplification of 780 nm Light for Polarized Electron Beam Applications

LDRD # 21-005

P. Inacker-Mix

PURPOSE:

This project focused on the simulation of the lasing properties of Alexandrite in a bar-based gain module, the required thermal attributes of a heatsink, and optimal pump light utilization. Furthermore, a compact heatsink design capable of supporting the pumped Alexandrite crystal for use as a laser gain medium in future lasers was designed.

The purpose of this exploratory project was to determine the feasibility and cost for such a gain module, which can accept different forms of pump light due to the lack of commercially available, high power pump diodes at the required wavelength at this point. The success of this project opens a path to further develop this technology critical for polarized electron beam production and enables the construction of a full prototype laser system.

APPROACH:

My field of research has been solid state laser systems and I have worked on Nd:YAG laser systems for the past seven years. I was approached three years ago to develop a laser system for a polarized electron gun on short notice that could support prototyping work for the Electron-Ion Collider. The most feasible way to achieve the needed system parameters in the given amount of time was to use a Nd:YAG laser system and convert the infrared radiation to 780 nm light using two different conversion techniques. While this approach was successful, the method is cumbersome, sensitive to environmental factors, and relatively inefficient.

Ideally one would create and amplify pulses of 780 nm light directly, which could reduce cost and size of the current system dramatically, while improving on stability and system capabilities. Developing a laser amplifier from scratch would have taken too much time for the project back then. Ready-to-deploy, off-the-shelf Gain Modules exist for Nd:YAG systems, which fast tracked the laser development significantly.

The idea for this project was to kickstart the development of a gain module that, analogous to its cousins for Nd:YAG, can support 780 nm light and fast track future laser deployments needing this wavelength. A two-punch approach was chosen to develop this device. In a first project, the feasibility for such a design was to be evaluated and a heat sink design created capable of handling the heat load, while tightly controlling the temperature of the gain crystal. The success of this project would then kickstart a second project to build and deploy a prototype laser system to test and expand on laser capabilities. The finished product would be a gain module optimized for manufacturability and flexibility that can be widely marketed and deployed across different laser applications.

TECHNICAL PROGRESS AND RESULTS:

The Gain medium has been simulated using Matlab to obtain information about the needed pump light profiles and powers, informing the design for the heatsink. A three-dimensional (3D) model of the heatsink was designed in Autodesk Fusion360. Together with a machinist, the design was optimized for manufacturability and a prototype was manufactured (Figure 1).

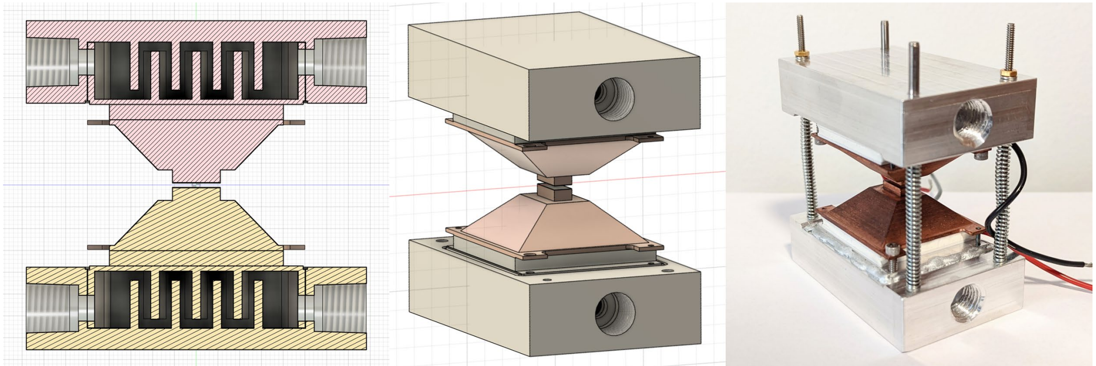


Figure 1. 3D Design and manufactured prototype.

Simulations of the thermal performance of the heatsink were performed using Ansys 2021 (Figure 2). Using this insight, the design was further tweaked to increase the homogeneity of the temperature profile across the laser crystal. These optimizations enabled relaxation of material requirements for the heatsink. All parts of the heatsink can now be manufactured from the same aluminum alloy further simplifying the manufacturing process.

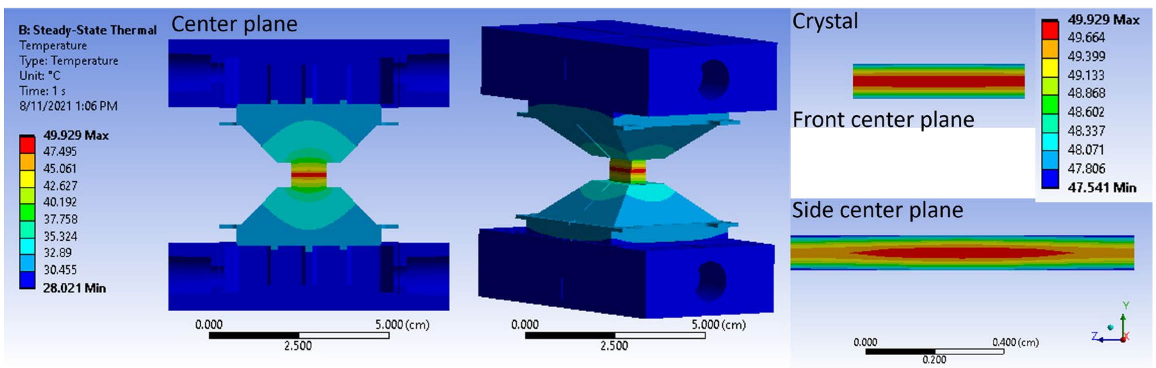


Figure 2. Thermal simulation results.

The result is an easily manufacturable heatsink design capable of handling higher than advertised thermal loads. Technical advances over standard heat sink designs include:

- The use of thermo electric coolers as heat valves to regulate laser crystal temperature
- A spring-loaded design to control the clamping force on the laser crystal in any orientation
- Minimally obstructed view in the crystal plane for direct access to all crystal faces.

In summary, the design and simulation of the gain medium Alexandrite and the heatsink was successful and sufficient information about the feasibility and cost of a prototype laser system was obtained. The next step for the development of a working laser will be the construction of the full prototype for laser performance testing, including pump optics and laser resonator, which exceed the scope of this project.

MILESTONES:
None. This project completed in FY21.

In Vitro Analysis for Isotopes

LDRD # 21-006

V. Sanders

PURPOSE:

The purpose of this project is to establish protocols for the post-production analyses of potential imaging and therapeutic radioisotopes produced at Brookhaven National Laboratory (BNL). Analyses such as high-performance liquid chromatography (HPLC) and instant thin layer chromatography (ITLC) are the most common radioanalytical techniques used for assessing the amount of radionuclide bound to a molecule, known as the labeling yield. In addition to developing radioanalytical techniques, its purpose is to assist in the forward progression of the use of the medical isotopes produced at BNL. This is done through the development and publication of methodologies such as dose calibrator evaluations, optimization of generator systems, preliminary radiolabeling to standard ligands, and stability studies. While the production of novel isotopes is essential to the mission of the medical isotope research and production (MIRP) group and to the medical model of personalized medicine, the establishment of validation protocols is vital to the translation of these isotopes to preclinical and clinical studies. The data and techniques developed will aid researchers within the community in the utilization of the isotopes as well as increase the interest in these radionuclides. In short, a handbook of methods will be developed as a published guide to those interested in using these isotopes to advance the safe application of these novel isotopes.

APPROACH:

The Brookhaven Linac Isotope Producer (BLIP) has been instrumental in the production of medical isotopes for several decades. The BLIP facility and the MIRP group focus on the production, processing, and shipment of clinically used isotopes to supplement the needs of the nation. Although this is an essential mission, the MIRP group also seeks to develop novel isotopes to further expand the field. Following the production of these isotopes, they are typically conjugated to biomolecules, which act as targeting vectors directing their localization. When complexing the radionuclide to biomolecules, it is necessary to determine the amount of the radionuclide bound to the molecule, as unbound or free radionuclides have different pharmacokinetics than those bound to targeting vectors. The ability to evaluate these novel radionuclides at BNL allows for immediate validation.

Production strategies, such as the generator production of isotopes, can be optimized to provide the most ideal separations between the parent isotope and the isotope of interest. Thus, a portion of this project focuses on the optimization of the $^{44}Ti/^{44}Sc$ generator system.

Novel isotopes are typically conjugated to organic moieties for use in biological applications. The resulting complexes must have a high radiolabeling yield and must be stable after radiolabeling under biological conditions. To determine proper radiolabeling protocols, common ligands will be used to perform preliminary radiolabeling studies. The resulting complexes with be evaluated via in vitro or in vivo studies to determine the stability of the complex. A portion of these studies was completed in collaboration with Prof. Lynn Francesconi (Hunter College) and Jason Lewis (Memorial Sloan Kettering Cancer Center). This work involved complexing ^{227}Th to the HOPO chelator, evaluating the stability in vivo, and establishing a dosimetric baseline versus unbound ^{227}Th.

Following radiolabeling studies, radioanalytical techniques are used to assess the labeling yield. However, prior to determining the yield of a radionuclide complex, protocols for separating the unbound radionuclide from the bound radiocomplex must be developed. As a growing isotope of interest, ^{225}Ac complexed to the universally used DOTA chelator was used to evaluate various ITLC systems.

TECHNICAL PROGRESS AND RESULTS:

During FY21, a protocol for ^{227}Th purification and radiolabeling with the 3,4,3-(LI-1,2-HOPO) chelator was developed. The purification allowed for a radionuclidic purity of 97.5 % and subsequent radiolabeling studies resulted in a yield of 59.3 %. The dosimetric studies show that unbound ^{227}Th localizes in the bone but this is mitigated through coordination to the HOPO chelator.

With the development of ITLC protocols for ^{225}Ac, various coordinating and non-coordinating solvents were evaluated as well as three different TLC plates. In all cases, the radiolabelled complex stayed at the baseline when TLC SG-60 plates were used, moved with the solvent front when iTLC-SG plates were used, and partially moved with the solvent front (R_F = 0.3) when iTLC-SA plates were used. TLC SG-60 plates take the longest to develop (15 min) where iTLC-SG plates develop quickest (2 min), and iTLC-SA plates are in the middle taking 9 minutes. This suggests that the mechanism by which the ^{225}Ac-DOTA complex moves is driven by kinetics of the solvent moving up the plate rather than coordinating strength or interaction of the complex with the solvent used. It was determined that the optimal developing solution is 0.9% NaCl/10 mM NaOH and the optimal TLC plate is the iTLC-SG (Figure 1). This ITLC trace shows excellent separation of unbound ^{225}Ac (at origin) and ^{225}Ac-DOTA (at solvent front). This work is ongoing and will investigate HPLC separations.

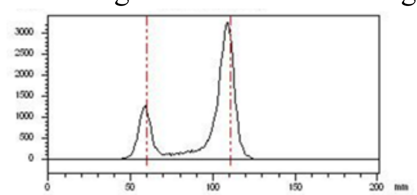


Figure 1. iTLC-SG, of ^{225}Ac-DOTA developed in 0.9% NaCl/10 mM NaOH.

A novel resin has been synthesized (AM) specifically for use in a $^{44}Sc/^{44}Ti$ generator system, which employs a methyl substituted hydroxamate group as the chelating moiety. The longest lasting generator (20 µCi, 300 µL BV) has been successfully eluted 48 times without any sign of ^{44}Ti breakthrough (Figure 2). This generator has been duplicated to ensure consistent results, and the second generator has been eluted 34 times without ^{44}Ti breakthrough and similar activity of ^{44}Sc eluted. The activity of ^{44}Sc eluted can be increased by eluting with 2 M HCl; this generator (20 µCi, 300 µL BV) has been successfully eluted 14 times eluting 70% of the available ^{44}Sc. A higher activity generator (1000 µCi, 1000 µL BV) has been loaded and eluted 13 times without any ^{44}Ti breakthrough, and remarkably elutes quantitative ^{44}Sc activity in 2 mL of 2 M HCl. The eluent of this generator has been used in subsequent radiolabeling experiments, giving 100% radiochemical yield demonstrating the purity of the eluted ^{44}Sc. This generator system has lasted five times longer than the commercial competitor (ZR resin) and elutes twice the activity of ^{44}Sc in the same volume. This work is ongoing to further optimize the generator size, elution volume, and elution solution.

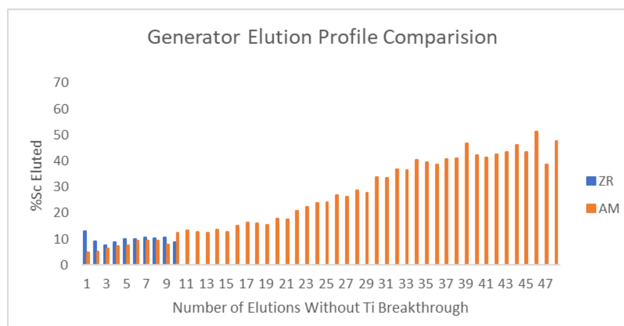


Figure 1. Elution profile comparison of synthesized resin (AM) vs the commercial resin (ZR).

MILESTONES:

FY21/FY22: Establish the biological dosimetry for ^{227}Th-HOPO as well as unbound thorium. Publish results in relevant journal. Submit proposal to build on project.

FY21/22: Optimize $^{44}Ti/^{44}Sc$ generator system. Submit proposal to build on project. Develop ^{225}Ac ITLC and HPLC methods. Publish results in relevant journal.

Near-threshold Production and the Mechanical Properties of the Proton

LDRD # 21-009

Y. Hatta

PURPOSE:

The purpose of this project is to study the gravitational form factors of the nucleon in general, and the so-called D-term, in particular. The D-term is a fundamental conserved charge of the nucleon, but its value has remained totally unknown to date. I will develop a theoretical framework, based on first-principle calculations in perturbative Quantum Chromodynamics (QCD), to compute the cross section of near-threshold quarkonium production in electron-proton scattering. I will clarify the impact of the nucleon D-term on the differential cross section of this process. I will also calculate the nucleon D-term in the Sakai-Sugimoto (SS) model. This work will better justify and motivate the future measurements of this process at the Electron-Ion Collider (EIC) at Brookhaven National Laboratory.

APPROACH:

It has been suggested by Kharzeev et al. in the late 90s that near-threshold quarkonium production, such as J/ψ and ϒ (Figure 1) can shed light on the origin of the nucleon mass. Then, in 2018, my collaborator and I showed that the same process is also sensitive to the nucleon D-term. These observables partly motivate the ongoing experiment at the Jefferson Lab, and possibly also at the future EIC. However, the theoretical frameworks used in previous approaches are not first-principle calculations with controllable approximations. I will study whether QCD factorization in terms of generalized parton distributions (GPD) is possible for this process, and if so, what the role of the D-term is in this factorization approach.

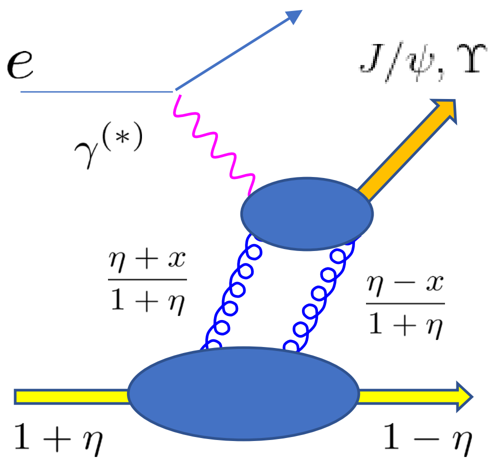


Figure 1. Quarkonium production near threshold in electron-proton scattering. x is the momentum fraction of the incoming proton and η is skewness.

In addition to the original proposal above, I have started a new collaboration with M. Fujita, S. Sugimoto and T. Ueda. We calculate the nucleon D-term in holographic QCD using the SS model. Previously, the D-term was calculated in a number of low-energy effective models of QCD, including bottom-up holographic models. The SS model is by far more sophisticated than any of these models. It features infinitely many pseudoscalar mesons, vector and axial mesons, and also glueballs with various spins. I will study how these degrees of freedom contribute to the D-term.

TECHNICAL PROGRESS AND RESULTS:

Together with Mark Strikman (co-Principal Investigator), I published a paper in Physics Letters B during the previous fiscal year. We showed that the scattering amplitude of near-threshold quarkonium production in the limit of large photon virtuality or large quark mass is dominated (about 80%) by the contribution from the gluon energy momentum tensor. The off-forward matrix element of the energy momentum tensor defines the gravitational form factors, including the D-term, so our observation proved the direct sensitivity of the gluon D-term to this process. This is remarkable because, in principle, infinitely many twist-two operators contribute to this process in a GPD-type factorization approach. It is also in contrast to the situation in Deeply

Virtual Compton Scattering (DVCS), which has been considered as a promising process to probe the quark D-term. A corollary to our result is that the quark D-term cannot dominate the Compton form factor in DVCS. Our work paved the way for a systematic expansion of the scattering amplitude of quarkonium production around the point $\eta=1$ (η is the skewness parameter) and therefore the possibility of extracting the gluon D-term at the future EIC.

In the present fiscal year, I am mainly focused on the calculation of the D-term in the SS model. Using holographic renormalization, we showed that the D-term allows different physical interpretations, depending on the value of the momentum transfer k. When k is small, the D-term can be interpreted as arising from exchanges of infinitely many meson pairs. When k is larger, the "glueball dominance" picture emerged, in complete analogy to the vector meson dominance of electromagnetic form factors. We also started the numerical calculation of the D-term in the small-k region and discussed the connection with the stability of the nucleon.

MILESTONES:

First year:

We laid the foundation for a first-principle calculation of near-threshold quarkonium production in QCD. We made a crucial observation that near the threshold, the skewness variable is close to unity, and the scattering amplitude is dominated by the gluon energy momentum tensor. It accounts for 80% of the Compton form factor and is therefore more important than all the (infinitely many) twist-two operators combined. Our work provides a strong theoretical justification for the ongoing and future experimental efforts.

Second year:

We are carrying out the first application of holographic renormalization to the nucleon in the SS model. We showed that the nucleon D-term is dominated by the exchange of infinitely many 2++ and 0++ glueballs. However, when the momentum transfer is small, the glueballs become less visible, and the meson pair exchange picture takes over. We are numerically calculating the D-term in this regime and discussed the stability of the nucleon from a new perspective. We plan to publish the result within this year.

Development of an Integrated Multi-scale Bioimaging Capability

LDRD # 21-013

Q. Liu, L. Wang, X. Xiao, Y. Chu, Y. Yang, T. Paape, Y. Lin, J. Liu (Yale)

PURPOSE:

One fundamental question underlying all living organisms is to understand their hierarchical organizations and physical changes with the needed spatial and temporal resolutions under physiological or pathological conditions. This is a multi-scale challenge requiring imaging from sub-nanometers to micrometers in a cellular context. While the individual imaging techniques are available, there is a critical need to integrate them into a workflow capability. Our objective is to develop an integrated multi-disciplinary and multi-scale bioimaging capability at Brookhaven National Laboratory (BNL). The capability will expand BNL's existing facility operation program in bioimaging and place BNL in a leadership position in bioimaging research. The capability will also address grand challenges of Department of Energy (DOE) science programs for national bioenergy sustainability and security.

APPROACH:

A grand bioscience challenge is the understanding of hierarchical living organizations and physical changes with the needed spatial and temporal resolutions under physiological or pathological conditions. Addressing this challenge requires multiple disciplinary efforts in technology development for visualization of biological processes ranging from atoms, molecules, organelles, cells to tissues with spatial resolutions from sub-nanometers to micrometers in a cellular context. However, due to the highly heterogeneous, dynamic, and adaptive nature in living organisms, understanding such complexity is challenging mainly due to lack of multi-scale imaging capabilities that can visualize transient biological processes under the cellular context and under physiological conditions. Multi-scale imaging is recognized by the DOE Office of Biological and Environmental Research as a major grand challenge.

Multiple imaging technologies, for example light microscopy, X-ray imaging, and electron microscopy have been developed for addressing the spatial-temporal resolution challenge. However, there is no single imaging technology that could effectively address the multi-scale challenge in a cellular context. By leveraging BNL's expertise in X-ray tomography (National Synchrotron Light Source II [NSLS-II]), quantitative plant sciences (Biology), machine learning (Computational Science Initiative [CSI]), the newly established cryo-electron microscopy (cryo-EM) facility (Laboratory for Biomolecular Structure [LBMS]), and Yale's expertise in cryo-electron tomography (ET), we are developing a bioimaging capability integrating three different, but highly complementary imaging techniques (light, X-ray, and electron imaging techniques) to quantitatively explore cellular structures and dynamics at multiple scales in a cellular and physiological context. We are also developing machine-learning based computational methods for imaging data segmentation and aggregation for visualization of biological significance.

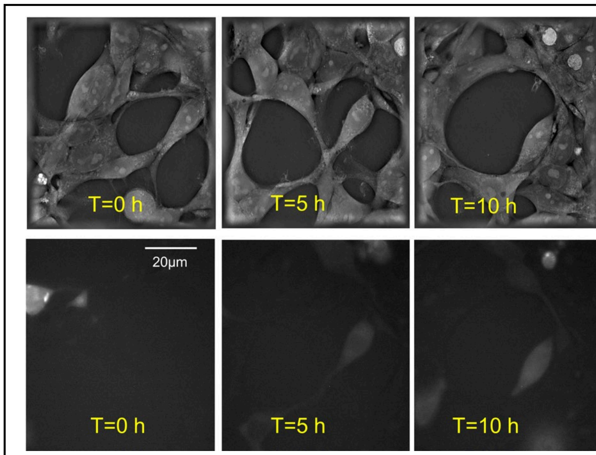


Figure 1. Live-cell imaging at indicated time. (Upper) Measurement of refractive index. (Bottom) Measurement of optical fluorescence.

TECHNICAL PROGRESS AND RESULTS:

The project started on April 1, 2021. We have made progress on different aspects of sample preparation, live-cell imaging, X-ray imaging, and electron tomography.

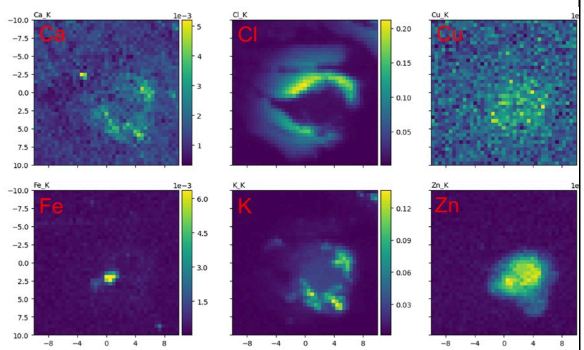


Figure 2. X-ray fluorescence measurement of indicated metals and ions for a single cell.

To support the live-cell imaging data collection, we have procured and tested a Nanolive 3-dimensional (3D) microscope. The microscope has a field view of 90x90x30 µm with a spatial resolution of 200 nm horizontal and 400 nm vertical. We have used the microscope for imaging engineered cells that are capable of up taking more zinc ions from environments. Figure 1 shows the morphology changes of growing cells and expression of green fluorescent protein (GFP)-tagged zinc uptake transporters.

On X-ray imaging, we have prepared cells on silicon nitride membranes and used the samples for both X-ray fluorescence imaging measurement at NSLS-II at the Submicron Resolution X-ray Spectroscopy (SRX) beamline (Figure 2) and X-ray tomography measurement at the Full Field X-ray Imaging (FXI) beamline (Figure 3). The X-ray fluorescence data reveal both the global and local distribution of not only zinc ions, but also other ions of potential biological interests. For the X-ray tomography, the absorption contrast is sufficient to reveal both overall cell shapes and cell-cell interactions and cellular and structural features, which may be correlated with X-ray fluorescence data for discovery of biological functions. Both SRX and FXI experiments were done at room temperature and worked quite well. We attempted to freeze cells for a cryogenic X-ray tomography experiment at FXI. However, the current setup showed vibrations which deteriorated imaging quality.

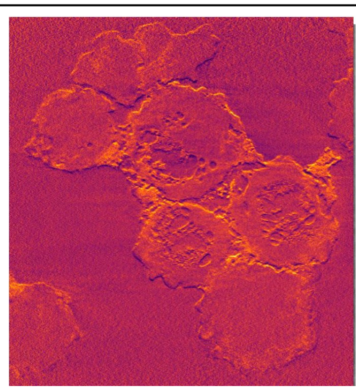


Figure 3. X-ray tomography measurement of multiple cells.

On electron tomography, we have installed and tested two tomography data acquisition programs EPU TOMO and SerialEM on the LBMS Titan Krios G3i instrument. Both programs are working and have been used to collect tilt series tomography data. We are preparing and optimizing real-life biological samples suitable for cryo- Focused Ion Beam (FIB) sample milling and cryo-ET measurement.

MILESTONES:

FY22-23: 1) Prepare cells and use them for X-ray absorption and fluorescence contrast imaging. 2) Produce thin lamellae samples for cryo-ET using cryo-FIB. 3) Develop high-throughput data acquisition for X-ray and electron tomography. 4) Demonstrate cryo-ET data collection and reconstructions of 3D tomograms at sub-nanometer resolutions. 5) Develop imaging analysis workflow for individual imaging techniques.

FY23-24: 1) Instrument cryo-cooling capability for X-ray tomography. 2) Demonstrate cryogenic X-ray tomography with 200-300 nm resolution. 3) Conduct live-cell imaging directed cryo-ET experiments for target identification and selection. 4) Conduct machine learning-based computational segmentation and assembly of multi-modal 3D images.

Transcriptional Co-regulation of Lignin Biosynthesis, Growth and Defense

LDRD # 21-014

M. Xie

PURPOSE:

This project aims to establish the transcriptional co-regulatory network of lignin biosynthesis, growth, and defense in the bioenergy crop poplar. This research will provide novel insights that will enable the biodesign of a better bioenergy crop with normal biomass production but low biomass recalcitrance, which is one of the missions of Brookhaven National Laboratory and the Department of Energy Office of Biological and Environmental Research.

APPROACH:

Lignocellulosic biomasses derived from plant secondary cell walls have been a promising source of renewable energy. To date, the major constraint in digesting this type of biomass into fermentable sugars is the prevalence of lignin polymers that protect biomasses from enzymatic or chemical deconstruction. Although the amount of lignin polymers in secondary cell walls can be reduced by genetically disrupting plant lignin biosynthesis, it is always accompanied by penalties on plant growth and defense, which dramatically impact biomass production. Using genome-wide approaches and next-generation sequencing techniques, we established a NAC-MYB transcription factors-centric transcriptional regulatory network. From the network, we identified genes that link the regulation of lignin biosynthesis and other physiological processes (e.g., growth and defense) in the bioenergy crop poplar. These genes provide potential bioengineering targets to reduce lignin content while maintaining other necessary cellular functions and biomass production.

TECHNICAL PROGRESS AND RESULTS:

Fiscal year 2021 is the first year of this project. In FY21, we established the poplar protoplast transient expression system to overexpress 16 NAC and MYB master regulators of lignin biosynthesis and performed chromatin immunoprecipitation-seq (ChIP-seq) to screen the genome-wide targets of these 16 master regulators in poplar cells. For each NAC and MYB master regulator, we selected the top 300 targets based on the q-value of their peaks in ChIP-seq data. Using the 16 NAC-MYB master regulators and their top 300 targets, we established a transcriptional regulatory network, which provides a better understanding of the architecture of the transcriptional network controlling lignin biosynthesis (Refer to the results for details). Additionally, we did gene ontology analysis using the discovered targets and observed gene enrichments in plant growth and defense mechanisms among the targets. This experimentally established transcriptional network enables the discovery of genes or transcription factors linking the regulation of lignin biosynthesis, growth, and/or defense, which are our candidates for bioengineering to compensate for, or even reverse, the growth and defense penalties in lignin biosynthesis disrupted plants.

Technical Progress and Results:

The establishment of the NAC-MYB transcriptional regulatory network: the network contains 2848 nodes and 5667 edges. Most of the genes for secondary cell wall biosynthesis (Red dots in Figure 1A), including lignin biosynthesis, phenylpropanoid pathway, and cellulose and xylan biosynthesis, are present. This network, for the first time, demonstrates a three-layer regulatory structure among the 16 master regulators (Green rectangles in Figure 1A): PtrWND2B forms the core, followed by its eight direct targets, and extends to the rest of the seven master regulators by indirect targeting. Among targets of these master regulators, we discovered the enrichment of genes involved in photosynthesis and stress responses in addition to lignin and secondary cell wall biosynthesis (Figure 1B).

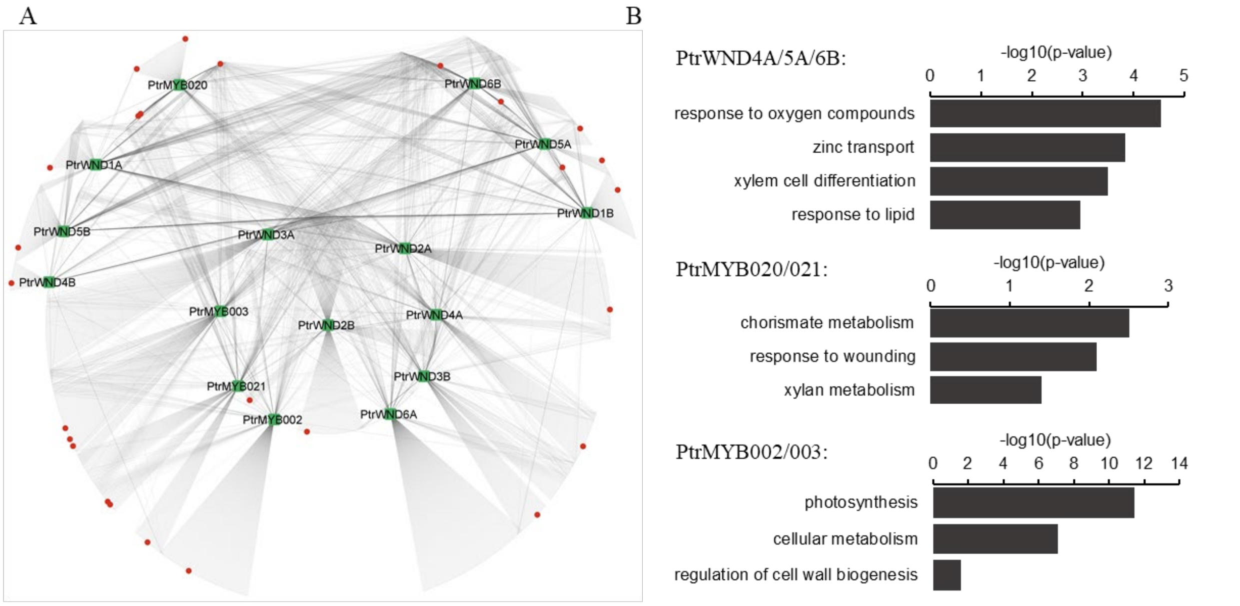


Figure 1. The NAC-MYB transcription factors-centric transcriptional regulatory network in poplar. (A) The overview of the transcriptional regulatory network. Green rectangles indicate NAC and MYB master regulators used in ChIP-seq analysis. Red dots indicate targets that are involved in secondary cell wall biosynthesis. (B) Representative gene ontology terms that are enriched in the top 300 targets of each master regulator.

MILESTONES:

- Experimentally validate top-ranked transcription factor-target associations in ChIP-seq
- Evaluate the recovery of growth and/or defense-deficient phenotypes via generating and phenotyping transgenic Arabidopsis.

Electron Microscopy Monolithic Active Pixel Sensors (EM-MAPS) for Structural Biology

LDRD # 21-020

G. Deptuch

PURPOSE:

The goal of this project is to explore the feasibility of building highly granular, least perturbing electron beam monolithic detectors, known as Monolithic Active Pixel Sensors (MAPS) capable of direct detection of electrons. The proposed detectors are to be developed using the specialized TowerSemi (formerly referred as Tower-Jazz) 65 nm process. This is the same process that is adopted for the upgrades of the ALICE Inner Tracking System 3 (ITS3) and ALICE 3 upgrades and for the tracking and vertexing layers in the Electron-Ion Collider (EIC). Highly granular, virtually massless Large Area Sensors are needed for the EIC detectors. This TowerSemi 65 nm process features unique enhancements added because of the collaboration with the CERN-led groups with the foundry. This enables design and fabrication of MAPS detectors without the typical restrictions, such as diffusion only charge collection and use of only one type of transistor in the pixel readout circuit of standard Complementary metal–oxide–semiconductor (known as CMOS).

Two main research targets were proposed in the EM-MAPS. The first one is Ultrafast Electron Microcopy (UEM), operating at energies up to 3 MeV, for which an integrating detector is needed to capture and store multiple electron impinging pixels in very short bursts. The second one is Cryo-Electron Microscopy (Cryo-EM), with a continuous electron beam of energies down to 100 keV, for which a virtually noiseless (detective quantum efficiency [DQE]=100%) detector, capable of cleanly counting incoming electrons per pixel and operating at effective frame rates of sub 100 kilo-frames per second is needed to yield high experimental throughput and to resolve the dynamics of specimens. Extremely small O(10 μm) pixels, readout with programmed regions of interest, pixel binning, and adjustment of the basic features, such as the gain of each pixel individually, are common needs. The program includes research on the ability of topological reconfigurability of the detector function so that the data is collected from where it is relevant to the experiment and in the way that it is needed.

The purpose of the project-covered by the EM-MAPS program is to ultimately develop demonstrators of the proposed functionalities embodied in small/medium-size prototypes. Beyond the scope of this project are steps that will require follow up funding for designing and fabricating large scale EM-MAPS detectors. The long-term goals of this program (beyond this project) are: 2K pixels per chip and more than 4K pixels seamlessly tiled. The Instrumentation Division team has significant experience in MAPS and other relevant pixel technologies.

APPROACH:

After signing the required Non-Disclosure Agreement (NDA) toward the end of last year, Brookhaven National Laboratory (BNL) acquired rights for downloading technology information contained in the Process Design Kit (PDK) and related Digital Design Kit (DDK) for the TowerSemi 65 nm process. This enabled the team to start, for the first time, studying, evaluating, and exploring analog and digital design flows in the TowerSemi 65 nm process. The first goal is to evaluate the specific features and new options offered by this process that will enable design optimization according to its strengths. The TowerSemi 65nm process is unique, as it features the smallest minimum gate length available on the market forimage sensors. It also includes the use of an increased resistivity substrate needed to operate the sensitive volume fully depleted. It provides an optimally profiled electric field and implantation of shielding wells for embedding electronic blocks. This removes restrictions for circuital solution and enables the design of optimal detectors for EM. To be able to best use the features of this process, more detailed knowledge of the technology, i.e., doping profiles built in the active volume of the sensor, is much desired. Direct contact with TowerSemi would provide the best channel to acquire the knowledge required for fruition of their process technology. In-house Technology Computer-aided design (TCAD) simulations of the conditions of charge collection resulting from the interaction of electrons with the medium are a key effort of this phase. In the next phase, work is planned to analyze and build circuit blocks and organize them into small and, successively, medium-scale prototypes that can be sent for fabrication. Measurements results will provide feedback for further iterations.

TECHNICAL PROGRESS AND RESULTS:
Signing the NDA was a major effort and caused significant slowdown in starting this work. Additionally, an effort to collaborate with Yale University is still pending finalization of a Cooperative Research and Development Agreement (aka CRADA). The first step, after signing the NDA, was to obtain the PDK and DDK databases from TowerSemi. Continuous interaction and participation in discussions with the community using this technology (e.g., around groups at CERN) is needed for information about database updates that occur regularly as a result of verification errors and deficiencies in meeting requirements for standardized design methods. In parallel, as a basis for further steps, direct contacts with the TowerSemi R&D (application support and sales in the U.S.) were initiated, aiming at establishing liaisons and requesting additional process information desired for optimization of the MAPS detector designs.

As the goal of this project is to implement original circuit design, test the devices produced, and provide the feedback necessary for the follow-up design steps, our efforts have been dedicated to studying in-pixel concepts and readout methodology for sending data off the pixel. The latter efforts benefit from synergies with other pixel designs that are under development in the Application Specific Integrated Circuit Group at BNL. Those projects aim at the development of hybrid pixels for X-ray detectors. As a result of the efforts on the readout procedures, a new ultra-fast and area efficient protocol with the associated circuit implementation has been developed. A Patent Cooperation Treaty (known as PCT) application for a World Patent is in preparation. Conceptual studies of the in-pixel circuitry are in an early stage, seeking appropriate test structures to be designed and later fabricated as small arrays of pixels.

MILESTONES:
Phase I: establish technology; basic circuit solutions; small prototype. The team is gaining familiarity with the capabilities of the process for detection and electric parameters, including carrying our circuit design.

Phase II: selected circuit solutions; medium size prototype for UEM and test results.

Phase III: full circuit solutions; medium size protype for Cryo-EM; tests results and proposal for large-scale device design and fabrication.

Free Space Optical Link for Entangled Photon Distribution Over Long Distances

LDRD # 21-021

J. Haupt, E. Barragan Figueroa

PURPOSE:

The objective of this project is to build an optical station capable of transmitting entangled photons through open air (free space) over the 20.5 km line of sight between Brookhaven National Laboratory (BNL) and Stony Brook University (SBU). The station involves semi-permanent infrastructure on the roof of BNL building 701 and on SBU's Health Sciences Tower, to be locally fiber-linked to the respective quantum networking labs of each institution. An experiment demonstrating this ability over a long non-marine baseline has yet to be achieved. This effort is well positioned to enable the first interstate quantum network in the U.S. and is expected to enable additional scientific programs and to have possible practical applications.

APPROACH:

The Principal Investigator's (PI) background in instrument development for optical astronomy has provided for a nearly seamless transition to free space optics, a field which deals with optical telescopes, systems integration, matters of optical quality, and atmospheric wavefront correction. The co-PI is a recognized quantum communications expert, thereby covering the needed knowledge spectrum for this endeavor.

A four-fold approach is employed for this effort: 1) creation of a sub-scale free space link "pathfinder" system to develop the instruments and procedures over short baselines, including bi-directional, multi-conjugate adaptive optics; 2) characterization of atmospheric metrics, which serve as proxies for optical transmission quality; 3) emulation of atmospheric effects in the laboratory; and 4) parallel infrastructure development for the rooftop quantum free space link facility.

Paul Stankus is valued for his contributions to this project as an experimentalist. Synergies also exist with Andrei Nomerotski's Quantum Astrometry project, wherein collaboration has been ongoing.

TECHNICAL PROGRESS AND RESULTS:

FY21 was the first year of this project. During this time, we've produced a sub-scale version of the 20 km system, which is mobile and can be aligned over baselines of our choosing. We incorporated a single-conjugate, receive-side-only adaptive optics system as a prelude to a more advanced system. We demonstrated an effective system telescope coupling optics with its associated alignment method and apparatus. We identified effective custom optical coatings and beam combining and splitting methods, and we began development of an automatic alignment and beam acquisition system (Figure 1). Optical fibers were installed linking the Free Space Link lab on the main floor of the Instrumentation Division with the basement quantum networking lab.

Additionally, we began development of an atmosphere characterization system (a scintillometer), which can be operated independently of the free space link equipment. Initial testing yielded believable, but as of now unverified measurements.

Lastly the design of the BNL rooftop lab was finalized with the Facilities and Operations Directorate, with construction scheduled.

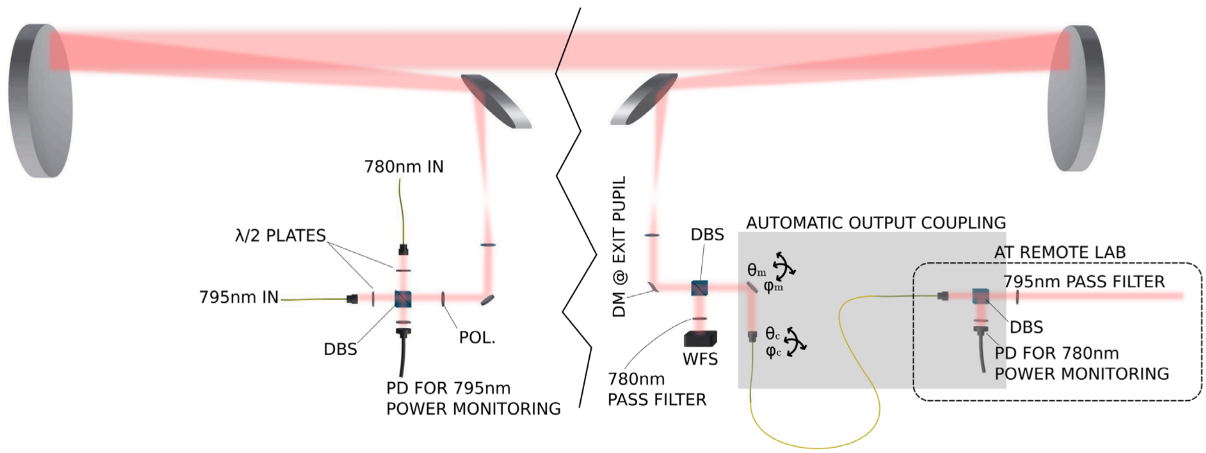


Figure 1. Automatic beam alignment and coupling system.

We've demonstrated free space coupling efficiencies in excess of 30% over 80 m in an indoor environment, while maintaining given polarization states.

MILESTONES:

Year 2:

- Rooftop infrastructure installed on building 701
- Bi-directional adaptive optics incorporated into the pathfinder system
- Automatic beam alignment/coupling system established for robust use
- Free space link demonstrations well in excess of 80 m demonstrated indoors using atmospheric turbulence emulation
- Outdoor free space link demonstrations achieved in excess of 500 m as validation of indoor testing
- Single photon experiments over indoor free space link demonstrated within the Instrumentation Division
- Custom optical components for full-scale system procured
- Scintillometer system validated and demonstrated over 20 km.

Year 3:

- Full-scale rooftop systems installed
- Free space coupling over 20 km BNL-SBU baseline demonstrated.

Precision Synchronization of Multi-Sensor Distributed Networks

LDRD # 21-022

V. Manthena, S. Zohar

PURPOSE:

Future detectors for radio cosmology and Antarctic neutrino research will involve sensor arrays needing ultra-precise timing synchronization over length scales of 1 - 10 km. Distribution of clock and frequency reference signals will need to compensate for phase fluctuations over time scales ranging from microseconds to hours, with precise knowledge of array element timestamps at the few hundred femtosecond level. Existing techniques to provide synchronization at this level are expensive and limited to shorter and less numerous links. We plan to investigate methods to propagate appropriate phase/frequency references over thousands - tens of thousands of optical links, augmented by global timing references from air- or space-borne natural and artificial sources. Successful demonstration of such techniques will enable new programs in dark energy/cosmic inflation research and can also be applied in studies of ultrafast phenomena and quantum communication.

The synchronization methods we plan to study are based on work over the past decade by national standards laboratories in transfer of highly stable frequency standards. It has been found that, with proper signaling, commercial telecommunications fiber links are capable of transporting frequency information with O(10-17) accuracy over hundreds of kilometers. Most work has been focused on use of "dark channels" in dense wavelength-division multiplexed (aka DWDM) fiber networks, so that new or unused ("dark") fiber runs are not necessary. Thus, the same fiber can simultaneously carry its normal data traffic along with the ultrastable frequency reference. For the scientific applications that we have in mind, this is also important as the fibers that are already planned for transport of sensor array data can also be used for synchronization.

APPROACH:

Developing powerful, cost-efficient synchronization methods is a key research and development (R&D) priority for a next-generation Dark Energy experiment based on 21 cm intensity mapping (IM). Successful demonstrations resulting from this project will remove uncertainties when IM proposals are being evaluated in the Snowmass and Particle Physics Project Prioritization Panel (known as P5) process in the Office of High Energy Physics. Additionally, these techniques have clear application to Time-of-Flight measurements in timing detector arrays at the Large Hadron Collider, the Electron-Ion Collider, and in atmospheric lidar.

The immediate goal of this project is to demonstrate cost-efficient methods for synchronizing arrays of 3-5 GHz waveform digitizers over km length scales with accuracy to better than 0.01 radian, and more broadly, to support the Instrumentation Division's efforts towards establishing expertise in the techniques of high-precision timing measurements in line with Brookhaven National Laboratory's science missions.

TECHNICAL PROGRESS AND RESULTS:

Our current approach is to estimate the time delay due to thermal expansion by using a neural network to extract the temperature information from thermal distortion present in the digitized transceiver signal. An order for a high-speed scope capable of observing small thermally induced changes in the detected signal is being purchased and four laptops with Graphics Processing

Units (aka GPU) capable of supporting machine learning frameworks have arrived. Additionally, two interns have been hired for this project.

MILESTONES:

Some of the future milestones for this project include:

- Down select between optical and non-optical clock synchronizations and distribution techniques which suit the current R&D work
- Develop baseline requirements using the proposed Packed Ultra-wideband Mapping Array (known as PUMA) 21 cm experiment as a potential target application
- Construct a characterization test bench using existing laboratory facilities in the Instrumentation Division
- Implement a White Rabbit network to characterize the present state of the art and develop characterization methods
- Explore quantum information technology and interface between electronics and optics.

Towards Edge Computing: A Software and Hardware Co-Design Methodology for Application Specific Integrated Circuits (ASIC)-based Scientific Neuromorphic Computing

LDRD # 21-023

S. Miryala

PURPOSE:

This project takes aim on a co-design approach for methodologies and their implementations of edge computing for optimal handling of data streams in scientific applications. Until now, most of the efforts in the scientific community, particularly those exposed to large but sparse datasets, as for example particle physics or nuclear physics, have been handled by building advanced and sophisticated Machine Learning (ML) code that runs on hardware platforms like Graphic Processing Units (GPUs), Artificial Intelligence (AI) Accelerators or supercomputers. Very little or no attention has been given to developing optimized hardware platforms, suitable for carrying edge computing in the form of neuromorphic processors on the front-end electronics.

The goal is to develop benchmarks for a novel software and hardware co-design approach that would result in energy-efficient, low-latency neural and neuromorphic networks that could be designed, fabricated, and deployed at the detector read out hardware either in Application Specific Integrated Circuits (ASIC) or Field Programmable Gate Arrays (FPGA).

Our software-hardware co-design provides a new approach for high volume spatio-temporal data processing applications. This new capability is extremely relevant for applications in event-based and data-streaming camera and detectors (e.g., sPHENIX, the next Enriched Xenon Observatory, the Electron-Ion Collider). Potential funding opportunities are within the Department of Energy, the Department of Defense, the Department of Homeland Security, and the Defense Advanced Research Projects Agency.

APPROACH:

In a multi-channel radiation detector readout system, waveform sampling, digitization, and raw data transmission to the data acquisition system constitute a conventional processing chain. The deposited energy on the sensor is estimated by extracting peak amplitudes, the area under pulse envelopes from the raw data, and starting times of signals or time of arrivals. However, such quantities can be estimated using ML algorithms on the front-end ASICs, often termed as "edge computing." Edge computation offers enormous benefits, especially when the analytical forms are not fully known, or the registered waveform suffers from noise and imperfections of practical implementations.

In this work, we aim to carry out waveform processing (such as predicting peak amplitude and time of arrival parameters) from a single waveform snippet. We would like to investigate various neural network algorithms, such as a Multi-Layer Perceptron (MLP) and a Convolutional Neural Network (CNN) by varying their model sizes, while maintaining good inferencing accuracy. To better fit front-end electronics, neural network model reduction techniques, such as network pruning methods and variable-bit quantization approaches, are necessary. Such parameter-efficient and predictive neural network models established feasibility and practicality of their deployment on front-end ASICs.

Conventional ML implementation frameworks, such as TensorFlow or PyTorch, are publicly available and rely on Python programming for training and designing various network topologies. The conventional semi-custom design for ASICs or FPGAs starts with hardware description language (HDL). There is a need for investigating commercial or open-source tools where the ML results can be seamlessly transferred to the HDL. For low power and low latency non von-Neumann architectures, such as in memory computing or beyond Complementary metal–oxide–semiconductor (CMOS) circuit elements, such as memristors, their fabrication and characterization is investigated as part of this project. The latter can perform synaptic functions of Spiking NN (SNN) nodes or handle instantaneous multiplication as needed in CNNs. The project includes evaluation of the commercial on-chip Artificial Intelligence implementation methodologies, including running selected processing of signals in detectors developed at Brookhaven National Laboratory (BNL).

Bringing BNL's experts in the Computational Science Initiative and Instrumentation Division, who have vast experience in developing ML algorithms to work together for the first time to co-design software and hardware as a neuromorphic computing methodology for scientific applications, will provide the collaborative platform needed for an interdisciplinary effort for this project.

TECHNICAL PROGRESS AND RESULTS:

- Developed a framework that models the behavior of an entire signal processing chain, composed of amplifier, shaper, and digitizer with an input response from the sensor. The sampled signal responses are used as labeled data sets (~10000 waveforms) for training of ML models.
- Studied the inferencing accuracy of various ML models designed for estimating the peak amplitude of the sample waveforms. We investigated two neural networks, CNN and MLP, for this study. The neural networks are optimized in terms of the number of layers, number of neurons on each layer, and bit precision of the weights. Overall results are quite promising.
- A design methodology is setup to translate the ML algorithms in Python to the HDL. The flow uses the open-source tool hls4ml available in the High Energy Physics community that translates python to C++. A commercial high level synthesis tool is then used to translate C++ into an HDL. The overall flow is automated and works well for both CNN and MLP.

MILESTONES:

Year 2021-2022

- Multi-modal regression, including waveform peaks and time-of-arrivals
- Synthesis and implementation of neural networks using semi-custom design flow
- Publication: S. Miryala et al., "Waveform processing using neural network algorithms on the front-end electronics", JINST, 2022.

Year 2022-2023

- Automated resource-aware network architecture search
- Investigation of novel devices, characterization, and other neural network architectures for low power and area applications.

Demonstration of Quantum Transduction from Superconducting Cavity to Atomic Vapor

LDRD # 21-025

P. Stankus, E. Figueroa

PURPOSE:

The goal of this project is to demonstrate the first essential step in a superconducting-to-atomic vapor quantum transduction capability. The final goal will be to have a superconducting (SC) quantum computing element, existing in a multi-component non-classical quantum superposition state, transfer its state coherently to photon degrees of freedom, which can then be transported without interaction across long distances at room temperatures. As a first step toward that goal, we will demonstrate that the field excitation of a SC resonator can be coupled to an atomic vapor ensemble, able to induce a transition across the vapor coherently, which can then coherently produce a single photon. The natural follow-on will be to demonstrate the same with the SC element cooled to the milli-Kelvin range, so thermal excitations will be sufficiently suppressed to demonstrate transduction from a well-defined initial quantum state. This will then, in principle, allow for quantum transduction into and out of a variety of quantum computing systems and a wide variety of potential applications and collaborations.

APPROACH:

The setup of this project is to establish the two components of i) a trapped, cooled, and optically driven rubidium (Rb) gas vapor; and ii) the magnetic field of a SC resonator with fundamental mode at a particular frequency (6.8 GHz) in the same spatial region. The challenges here are i) manipulating the atomic vapor within the cryogenic, high-vacuum environment, while ii) being able to supply optical drive fields from the outside into the cryo environment, without excessive heat loading. The local group spans the necessary expertise, including the Principal Investigators (PI) and our two new scientific staff hires, Dr. Julian Martinez and Dr. Joanna Zajac, along with two Stony Brook University Ph.D. students, Guodong Cui and Samet Demircan.

TECHNICAL PROGRESS AND RESULTS:

Our main achievement in the first year of the project was in designing and acquiring equipment, specifically the high-powered tunable lasers needed for atomic vapor cooling, together with associated optics; plus, we have specified and are supervising the purchase and installation of a new cryogenic system in the Instrumentation Division (IO), which this project can use, as well as other follow-ons. Pandemic-related supply delays have been considerable, and progress has been slowed for personnel reasons: our original co-PI Dr. Olli Saira left Brookhaven National Laboratory (BNL) employment in early 2021, and only after a considerable search, have we hired new staff.

At the present time, we are designing the SC resonator components, installing and aligning the vapor cooling lasers, and building the initial vapor cooling setup (Figure 1). Additionally, we are handling the infrastructure improvements for the new dilution refrigerator cryogenic setup (Figure 2).

Figure 1. Newly purchased tunable, high-power laser system manufactured by Toptica, being inspected on an optical table in the BNL IO Quantum Information Science (QIS) lab. The blue-cased unit houses two independent 780 nm beams, each of which is amplified to class 4 intensity. The red-handled units are controllers for the two systems. All components can be readily rack-mounted, and the assembly will ultimately be portable.

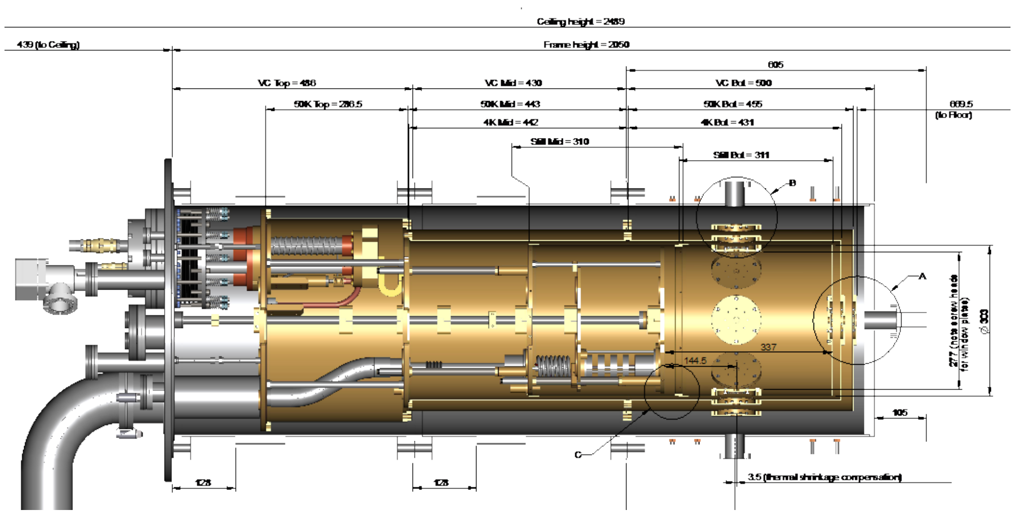


Figure 2. Highlight of the drawing showing the design of the milli-Kelvin dilution refrigerator cryostat, to be installed in the IO QIS lab (manufacturer: Bluefors). The cold plate and science section are to the right in the drawing, and the nine optical access ports and their windows can be seen. (When installed, the cryostat will be suspended with the cylinder long axis vertical, and with the science section at the bottom). We anticipate having the dilution refrigerator delivered in June 2022 and assembled soon thereafter.

MILESTONES:

FY 21:

- Acquire vapor cooling components and assemble (partially complete)
- Design SC resonator (partially complete).

FY 22:

- Complete vapor cooling setup and demonstrate in the lab
- Complete SC resonator, with test increments, and demonstrate in the cryo environment
- Design in-cryo optical and gas handling setup and complete installation of new cryo-system.

FY23:

- Build combined vapor and resonator setup in the cryo environment
- Demonstrate and measure resonator field coupling to Rb vapor hyperfine transition.

Bridging the Gap between Scientific Simulations and Experiments with Cycle-consistent Generative Models

LDRD # 21-029

Y. Ren, M. Lin

PURPOSE:

Many large-scale physics experiments, such as ATLAS at the Large Hadron Collider, the Deep Underground Neutrino Experiment (DUNE), and sPHENIX at the Relativistic Heavy Ion Collider, rely on accurate simulations to inform data analysis and derive results. Any systematic biases in the simulations can propagate through the analysis steps and result in systematic uncertainties. These biases may be detected and corrected using heuristics in the conventional analysis workflow, but they may become intractable when machine learning (ML) and artificial intelligence (AI) methodologies are applied, which tend to train on the simulations and infer on real data. Our goal is to develop a physics-informed ML framework that can bridge the gap between simulations and experiments. Potentially, this can be realized by applying generative adversarial networks (GANs) in an innovative way. We propose to construct a GANs-based cycle-consistent cross-domain (GC3D) simulation augmentation framework to improve scientific simulations so they will better represent reality. We will also investigate ways to encode relevant physics conservation laws and symmetries into the framework, while applying effective high-performance computing (HPC) techniques for scalability.

APPROACH:

This novel application of cross-domain GANs represents a paradigm shift from using GANs as surrogate models to reduce and remove systematic biases with real data knowledge. Unlike traditional uses of GANs that generate data from a random noise vector (Figure 1A), cycle-consistency GAN (cGAN) models use two GANs to form a closed cycle. Each GAN learns a mapping between two domains in the opposite direction and discriminates the generated examples from the other GAN (Figure 1B). There are two main advantages: data efficiency and no annotation. In Figure 1C, the cGAN is trained only on roughly 1,000 of Monet's paintings to be able to transfer an unseen photo. The scenes of a photo and a painting do not need to be matched, which is the crux for mapping simulations to experimental data as every experiment outcome is unique.

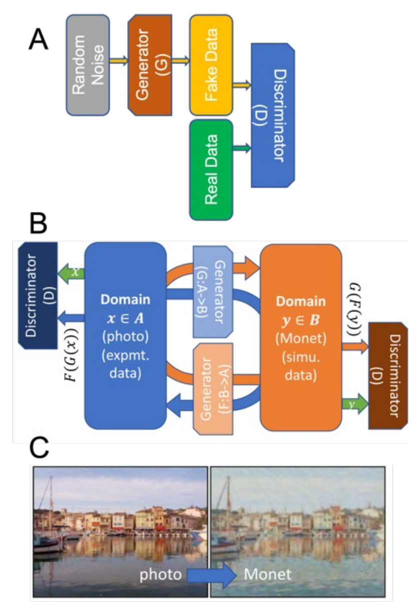


Figure 1. A) Standard GANs and B) cGANs). C) An example of applying the cGAN model on a scenic photo, where the model has been trained on unpaired real photos (Domain A) and Monet's paintings (Domain B).

Our multidisciplinary project team consists of experts in ML (Yihui Ren), HPC (Meifeng Lin), experimental nuclear physics (Jin Huang), and high-energy physics (Brett Viren and Haiwang Yu). We will focus on two major physics experiments, DUNE and sPHENIX. Brett Viren is the author of Wire-Cell, a liquid argon time projection chamber (LArTPC) simulation package, which will be used to generate several fidelity tiers of simulation data (https://lar.bnl.gov/wire-cell/). Jin Huang is a leading researcher, developing the sPHENIX test beam, data acquisition pipeline, and heavy-ion TPC simulator.

TECHNICAL PROGRESS AND RESULTS:

As the project began in mid-FY21, this annual status report covers the period between 06/01/2021–09/30/2021.

During this period, we have focused on training data generation and initial model implementation and validations. On the data generation front, we have created and verified two tiers of LArTPC analog-to-digital converter (ADC) data using one-dimensional (1D) signal and quasi-two-dimensional (q2D) signal response functions, respectively (Figure 2). These two domains will be our initial focus for studying AI-based domain transfer techniques.

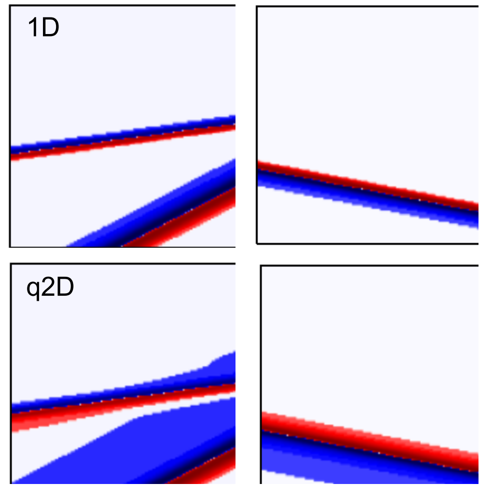


Figure 2. (First Row) Low-fidelity LArTPC simulation data samples using a 1D response function. (Second Row) Mid-fidelity LArTPC simulation data samples using a quasi-2D response function (colored in log-scale).

On the ML front, we have implemented two baseline model components, one for discriminating the two domains with very high accuracy. This shows there are enough differences between the two domains. Second, we have implemented an auto-encoding model to compress the ADC data into a low-dimension feature space. If successful, this will allow us to apply a more efficient transformation. After further inspection, the decoded image does not meet the scientific criteria. Thus, this idea currently is on hold.

MILESTONES:

FY22 (12 mo.): Develop visual-transformer model and improve model accuracy and shorten training time for the higher-tier complexity of DUNE data

FY23 (12 mo.): Integrate physics constraints and interpretation of the transforming process

FY24 (8 mo.): Generalize the approach on sPHENIX simulation and experimental data.

Designing Rechargeable Zn-air Batteries in Aqueous Electrolytes using Noble Metal-free Bifunctional Electro-catalysts for Grid-scale Energy Storage (GES)

LDRD # 21-031

L. Wang

PURPOSE:

This project focuses on developing porous $Mn_xMo_yO_z$-carbon nanotube (CNT) composites as noble-metal-free bifunctional electro-catalysts for oxygen reduction reactions (ORR) and oxygen evolution reactions (OER) that will allow for rechargeable Zinc-air battery (ZAB) operations in a cost-effective $ZnCl_2$-based water-in-salt electrolyte (WISE) system. This project aims to address two scientific targets: 1) optimize porosity of the catalysts to improve reaction kinetics and oxygen diffusion in viscous WISE and 2) determine optimal crystallinity and oxygen deficiency of $Mn_xMo_yO_x$ catalysts for improved catalytic activity. The proposed study will raise the stature of Brookhaven National Laboratory's new competency in grid scale energy storage systems and fits well with the new direction of the Department of Energy in working to develop new storage technologies by supporting research on energy storage.

APPROACH:

Commercial ZABs are replenishable primary batteries that require replenishing the zinc (Zn) anodes and electrolyte. Efforts towards rechargeable ZABs so far are mainly focused on the development of bifunctional catalysts and cathode materials for facilitating the ORR and OER, with alkaline electrolytes widely used to achieve fast electrode kinetics. However, Zn anodes are prone to form dendrites and suffer from passivation and corrosion in a basic environment. This situation is further complicated by the solid-solute-solid mechanism ($Zn–Zn(OH)_4^{2-}–ZnO$) and the degradation of alkaline electrolyte upon operation due to carbon dioxide (CO_2) poisoning and water consumption.

This work focuses on developing porous $Mn_xMo_yO_z$-CNTs bifunctional electro-catalysts that will allow for rechargeable ZAB operations in a cost-effective $ZnCl_2$-based WISE system that is free of CO_2 poisoning and can suppress Zn dendrite formation. Porous $MnMoO_4$ nanowires and MnO_x (MnO_2, Mn_2O_3, MnO) submicron spheres, as well as porous CNTs with controllable porosity are synthesized to provide insights into optimization of catalyst porosity, crystallinity, and oxygen deficiency for stable catalytic activity in ZABs. Two types of electrolyte systems are being investigated, including alkaline and near-neutral WISE. Functional aqueous cells are being designed to test the ZABs in various electrolytes and to allow for operando mechanistic studies at National Synchrotron Light Source II beamlines (Quick x-ray Absorption and Scattering (QAS), X-ray Powder Diffraction, X-ray Fluorescence Microprobe, and Submicron Resolution X-ray Spectroscopy) for further elucidating the failure modes of the cells and the impact of electrolytes on the ZAB systems. Two graduate students work on this project through collaboration with Dr. Esther Takeuchi, Dr. Kenneth Takeuchi, and Dr. Amy Marschilok at Stony Brook University.

The risks to the project are that the porosity of the $Mn_xMo_yO_z$-CNT catalyst needs to be increased to adapt to the highly viscous WISE system and the current collector needs to be resistant to the highly corrosive Cl-based electrolyte.

TECHNICAL PROGRESS AND RESULTS:
This project started in FY21 with synthesis of $MnMoO_4$, and its characterization using X-ray diffraction (XRD), scanning electron microscopy (SEM), and transmission electron microscopy (TEM). Material catalytic activity towards ORR and OER was determined by performing linear sweep voltammetry via a rotating disk electrode set-up. Additionally, various carbon additives were included to determine their impact on catalytic activity (Figure 1a-b). Determination of the material's performance in ZAB was initiated with progress towards electrode fabrication and battery set-up. Results included a catalyst-loaded gas diffusion electrode and its implementation into a ZAB set-up. Preliminary data suggested successful implementation of this catalyst in a functioning alkaline ZAB.

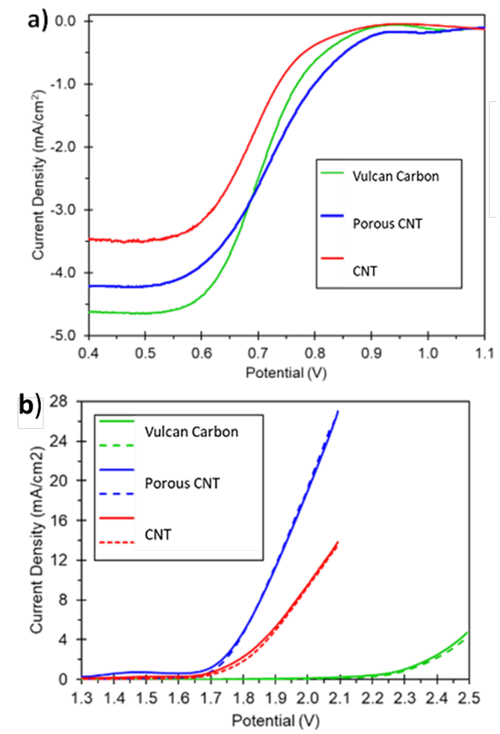


Figure 1. a) ORR and b) OER of $MnMoO_4$ with Vulcan carbon, porous CNT or pristine CNT.

In addition to the synthesis of $MnMoO_4$, porous manganese oxides, MnO_x were prepared by adjusting the annealing temperature of the precursor. Three manganese oxides were successfully synthesized (Mn_2O_3, MnO_2, and MnO) and characterized. ORR and OER studies as well as amperometry tests showed varying degrees of catalytic behavior and stability (Figure 2). Based on these studies, an alkaline ZAB was used to investigate the electrochemical performance using the best-performing Mn_2O_3 catalyst. For this set of studies, nickel foam and carbon thread current collectors were investigated as well as different carbon additives. The results of this study allowed the improvement of data collection by optimizing key components.

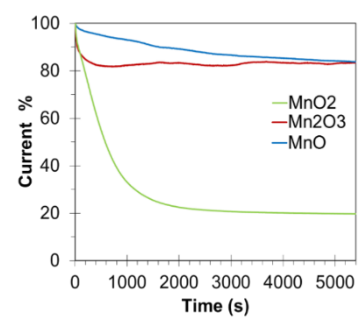


Figure 2. Current during ORR (stability) normalized where 100% for a sample is the initial current, measured at a potential of 0.5 V vs. a reversible hydrogen electrode as current versus time at 900 rpm.

MILESTONES:

- Proceed with testing in a $ZnCl_2$-based neutral electrolyte and further study the impact of electrolyte concentration on the performance of ZABs
- Tune oxygen defects and crystallinity of the catalyst materials and study the impact on OER, ORR, and ZAB performance in the various electrolyte conditions
- Conduct an in situ x-ray absorption spectroscopy experiment at the QAS beamline to track the changes in the local chemical environment of $Mn_xMo_yO_z$-CNTs catalysts during ORR and OER.

Development of a Planning, Operation, and Control Framework for Hybrid Energy Storage and Renewable Generation Systems

LDRD # 21-032

M. Yue

PURPOSE:

This project will develop a framework for planning, operation, and control of these “hybrid” energy systems containing high penetrations of renewables together with energy storage, non-wire alternatives, and conventional generating resources. We will leverage Brookhaven National Laboratory’s (BNL) expertise in energy storage technologies and developing probabilistic-based planning and control solutions for hybrid energy systems (battery, flywheel, etc.) together with machine-learning (ML) assisted, scalable stochastic optimization method. We will also partner with the New York Power Authority (NYPA) to utilize the grid simulation capabilities of their Advanced Grid Innovation Laboratory for Energy (AGILe) facility to refine and demonstrate the platform, and further strengthen BNL’s partnership with NYPA to develop the joint AGILe-CGI (Center for Grid Innovation) at BNL facility. This project will form the basis of a follow-on study to develop a hierarchical coordination and control structure for multiple standalone generation sources, such as energy storage systems (ESS) and/or hybrid renewables-ESS plants for frequency stability.

APPROACH:

This will be accomplished by focusing on: (1) a better understanding, modeling, and quantification of the different temporal and spatial scales of uncertainties, i.e., multi spatial temporal (MST) uncertainties; (2) synergistically sizing and siting a spectrum of technologies that includes, but are beyond different types of ESSs, e.g., relatively fast and more affordable generation units, such as gas turbines and demand response, in mitigating the multi-scale uncertainties, under the frequency dynamics constraints and MST uncertainties; and (3) a conceptual hierarchical coordination and control structure (HCCS) for the standalone or hybrid renewable-ESS plants for stabilizing grid frequency dynamics and provisioning synthetic inertia and adequate damping.

TECHNICAL PROGRESS AND RESULTS:

Uncertainty Modeling and Quantification: one of the key issues that needs to be addressed for the stochastic optimization of ESS capacity in a power grid is the development of scenarios that can capture the uncertainties related to renewable generation. While the Markovian approach can be used to model the wind generation evolution efficiently, the number of states will increase exponentially as the number of transitions increases. For long-term planning problems, the size of the Markov model becomes quickly intractable and an alternative way of uncertainty modeling for wind generation needs to be developed. A random field (RF) is a generalization of a stochastic process where the underlying parameters do not have to be real, or integer valued "time" but can instead take values that are multidimensional vectors or points on some manifolds. The major feature of the RF based approach is that it captures both the spatial and temporal correlations of wind speed information (and therefore, the wind generation) at different locations where the wind farms are built. The RF based uncertainty models are parameterized by using historical wind resource data. In this study, RF theory has been adopted and implemented to model the time-series wind generation for different penetration levels of renewables. The same approach can also be used to model the uncertainties related to solar irradiance and generation.

Stochastic Optimization for Sizing, Siting, and Operating Hybrid ESS and Synergistic Resources: to ensure optimal and reliable operation of power systems with energy storage in the presence of MST uncertainties, frequency dynamics constrained stochastic unit commitment problems are crucial. To achieve this, a probabilistic model considering MST uncertainties in wind generation is developed based on a multivariate discrete-time Markov Process. The statistical properties of wind speeds at different locations as well as their spatial and temporal correlations are defined by parameters of a time-varying Markov model established based on historical data. This wind speed model is then converted to a wind generation model by a nonlinear transformation based on wind turbine characteristics. The resulting stochastic model, however, introduces high complexity due to the large number of wind generation states required to accurately capture the MST uncertainties. To reduce complexity, area-perspective problems are formulated where in each problem, the stochastic model is simplified based on the area's perspective. To coordinate the area-wise solutions, a surrogate optimization-based framework is currently being developed.

Hierarchical Coordination and Control Structure for Hybrid Energy Systems: a droop-free distributed frequency control has been developed for the hybrid photovoltaic and battery energy storage (PV-BES)-based microgrid[1] (Figure 1). A distributed state of charge (SOC) balancing regulator achieves balanced SOC among the distributed generators (DG) with BES utilizing a distributed average SOC estimator and the power sharing regulator ensuring proportional power sharing among the PV-BES based DGs. These regulators generate two frequency correction terms, which are added to the microgrid-rated frequency to generate reference frequencies for the lower-level controllers. The performance of the proposed distributed control is validated through real-time simulations in OPAL-RT (parallel simulation and real-time software), which demonstrates the effectiveness of the proposed control in achieving frequency regulation, SOC balancing, and active power sharing in the hybrid PV-BES units under both islanded and grid-connected operation modes.

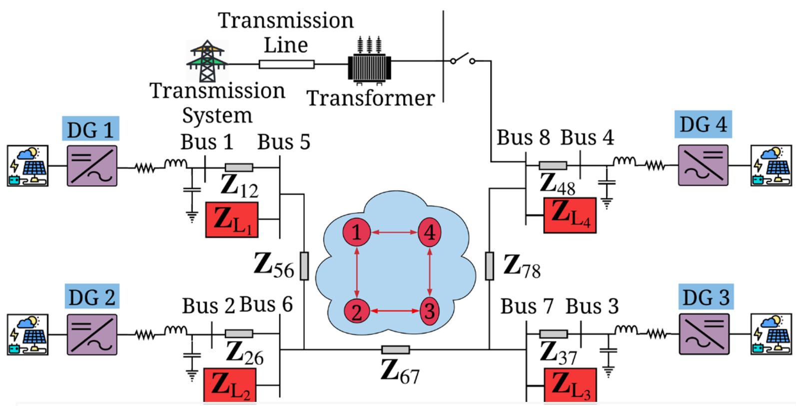


Figure 1. Schematic diagram of a 4-DG microgrid test system.

MILESTONES:

- Month 6: Data collection and enrichment complete
- Month 12: Methods, algorithms, and a suite of tools implemented for MST uncertainty modeling and quantification and online tuning scheme for optimizing HCCS/ESSs
- Month 20: Methods and tool development for optimal sizing, siting, and operation of ESS technologies and other resources
- Month 28: Enhanced ML-Surrogate Absolute-Value Lagrangian Relaxation (known as SAVLR) tool for optimal sizing, siting, and operation of ESS technologies and other resources
- Month 32: Prototype of the HCCS for hybrid energy systems.
- Month 36: Final report complete

[1] S. M. Mohiuddin, A. Yogarathnam, M. Yue, and J. Qi, "Droop-Free Distributed Frequency Control of Hybrid PV-BES Microgrid with SOC Balancing and Active Power Sharing," submitted to IEEE Power and Energy Society General Meeting 2022.

Interpretable Machine-Learning Aided Design of Dynamic Reaction Experiments

LDRD # 21-033

Q. Wu, J. Boscoboinik, X. Qu, H. van Dam, J. Rodriguez

PURPOSE:

There is an urgent need in catalysis science for new tools to understand catalyst design that can control chemical reactions more efficiently. We are pursuing a machine learning (ML) approach framework coupled with dynamic experimental facilities to address this need. Towards this goal, we: a) develop new experimental tools that can modulate reaction conditions and track kinetics responses in real time, i.e., transient kinetics spectroscopy; b) develop new ML tools to interact with experiments, directly deriving rate equations from the transient data and deciding if mechanistic understanding is reached or new experiments are needed; and c) use these tools to speed up the process of solving catalytic mechanisms. Our work will lead to the establishment of a new facility to design and exert external stimuli for dynamic experiments that will be used to reveal hidden mechanisms in complex chemical reactions. Moreover, dynamic catalyst control will be simulated and analyzed, leading to new experiments aiming at dramatically improving activity and selectivity in energy-related heterogeneous reactions.

APPROACH:

Rational catalyst design hinges on an accurate kinetic model of the catalytic reactions. Conventional kinetic modeling relies on approximating the complex reaction network with a system of ordinary differential equations (ODEs) whose solution is tuned to match experimental observations. This is a time-consuming process that is also prone to human biases. We use a novel ML technique, called Neural ODEs, to build a program that automatically extracts reaction kinetics from large time-series datasets. Concurrently, we develop new surface science instruments that can modulate reaction conditions and track kinetics responses in real time, making available significantly more (and more detailed) experimental data to be used with our ML method. Our project will be carried out in two phases. In the first phase, machine learning and experimental capabilities are developed in parallel in a coordinated matter with input from all team members. In the second phase, theory and experiment will be integrated into a single workflow, as shown in Figure 1.

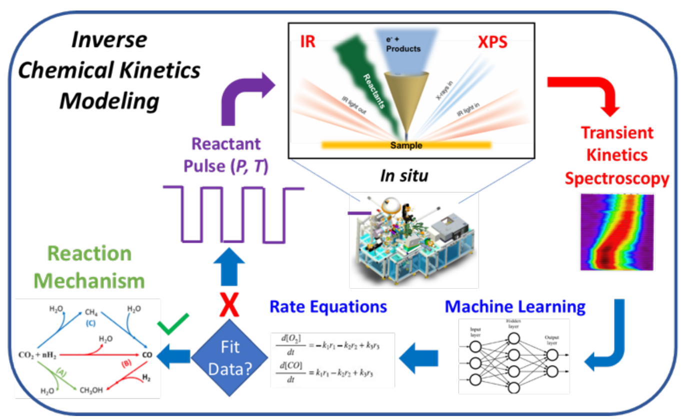


Figure 1. The workflow of our coupled experiment and machine learning approach to derive chemical reaction mechanisms.

TECHNICAL PROGRESS AND RESULTS:

In the first year of our project, our ML team developed a prototype software package that uses the Neural ODE method to derive chemical reaction rate equations from simulated transient concentration data. We built into the program our knowledge about chemical kinetics so that the output is easy to understand. We implemented the following important functionalities: a) it handles multiple datasets, including those with incomplete data points; b) it derives rate equations for hidden species whose data are completely missing; c) it does parallel computing; and d) it is hosted on a Github repository with continuous integration workflow. We successfully applied the code to simulated catalytic reactions that follow simplified Eley-Rideal and Langmuir-Hinshelwood models. To meet the challenge of increased reaction complexity, we started to train time-series classification algorithms as a screening tool to efficiently downsize the parameter space for our Neural ODE method. By training and validating 20 different methods using 20,000 data sets, we found that the Random Convolutional Kernel Transform (ROCKET) so far gives the best results.

Our experimental team started pulsing carbon monoxide (CO) gas molecules on a Pd(111) surface in multiple cycles and collecting transient infrared reflection absorption spectroscopy (IRRAS) data. We have achieved a 60-millisecond time resolution to follow the adsorption-desorption dynamics. This work involved the installation of a fast valve in the IRRAS instrument, writing a macro to execute the experiment and collect the data, and building electronics needed for triggering the valve. Following these developments, we have carried out experiments in which oxygen (O_2) is also present in the chamber, enabling transient kinetics studies of CO oxidation as a model reaction.

MILESTONES:

2022

- Implement mass spectrometry capabilities to complement the IRRAS
- Use the initial transient kinetics experimental studies of CO adsorption-desorption and CO oxidations to test and guide the ML tools being developed
- Carry out more complex reactions, with competing pathways, including nitric oxide (NO) disproportionation
- Test ML tools to understand the mechanistic aspects of these more complex processes, including obtaining rate constants for the different elementary steps.

2023

- Develop predictive tools that feed experimental conditions that will favor selectivity toward desired products. This will be tested with the aforementioned NO disproportionation reaction.
- Increase experimental capabilities to address more complex processes by adding more gas pulsing valves and writing LabView scripts to coordinate the different pieces of equipment involved in the experiments (infrared spectrometers, fast valves, mass spectrometer)
- Expand capabilities to an X-ray Photoelectron Spectroscopy System
- Release the software package to the public.

Laser Switching to "Hidden" Phases in Quantum Materials

LDRD # 21-037

J. Pelliciari

PURPOSE:

Photo-induced metastable quantum phases are of great scientific interest; however, their mechanism is not understood, due to the lack of experimental probes and data. Here, we propose to shed light on this topic by combining Resonant Inelastic X-Ray Scattering (RIXS) that is sensitive to spin, charge, orbital, and lattice degrees of freedom with ultrafast laser pulses triggering these "hidden" transitions. Currently, these types of experiments are not available at any synchrotron world-wide; thus, the goal of this project is to create a unique experimental setup and open a new research direction not only in the investigation of quantum materials and their ultrafast phase transitions but also in fields, such as quantum information science or optoelectronics, where ultrafast laser pulses are used to transmit information.

APPROACH:

Creating new metastable "hidden" phases that have no analog in thermodynamic equilibrium is becoming increasingly important in condensed matter physics and material science. The use of ultrafast laser pulses (30-150 fs) to trigger such quantum phases is extremely appealing as some of these can be long-lived, with a lifetime of hours or weeks and reversible, with temperature sweeping or extra laser pulses. The reversible switch of material properties (resistivity, electrical polarization, magnetism) is the key to innovative technological applications, such as memristive devices (change in resistivity), spintronics technologies (change in magnetism), and transient capacitors (appearance of ferroelectricity). Speculations on the mechanism of these transitions indicate the involvement of coherent excitation of phonons and charge transfer modifies the potential energy landscape. This effect leads to a new metastable ground state often not accessible in equilibrium conditions, thus a "hidden" state. However, despite a tremendous potential for novel applications and sustained efforts made by the ultrafast community, a mechanistic comprehension of the interactions underpinning these transitions is currently poorly understood.

We propose to combine ultrafast laser switching with RIXS at the Soft Inelastic X-ray Scattering (SIX) beamline of National Synchrotron Light Source II with the goal of uncovering the underlying mechanism of this new class of laser-induced phase transitions. More specifically, we will set up a laser-RIXS system aimed at the investigation of the laser-induced metal-to-insulator transition in $La_{2/3}Ca_{1/3}MnO_3$ (LCMO). RIXS has exquisite sensitivity to all the electronic excitations (spin, charge, and orbital) and electron-phonon coupling, which spans the range in explaining these phenomena. These capabilities make RIXS the ideal spectroscopic tool to investigate the interactions at play in these systems. Our goal is to perform proof-of-principle experiments of a laser-switching-RIXS-probe (see Figure 1c for a scheme) and perform a differential analysis of the excitations before and after the switching to reconstruct the switching mechanism. Knowledge of the detailed mechanism will be key for modeling these "hidden" phases, providing important clues on the future design and engineering of quantum devices.

TECHNICAL PROGRESS AND RESULTS:

The project started in FY21, and a postdoc was hired. During FY21, the postdoc was trained and performed an experiment on initial materials in the absence of laser pulses. We performed RIXS experiments on superconducting nickelates (Figure 1a, b) and LCMO (Figure 1c, d) uncovering

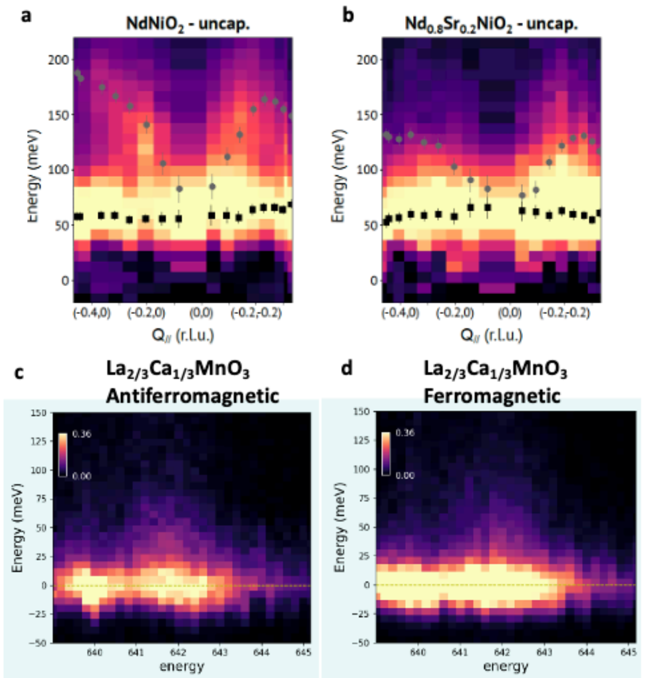


Figure 1. (a, b) Low energy RIXS spectra of parent and superconducting nickelates. (c, d) Low energy RIXS spectra of antiferromagnetic and ferromagnetic $La_{2/3}Ca_{1/3}MnO_3$.

the elementary excitations of these two compounds. We were able to identify magnetic excitations and differentiate between the different ground states. We performed data analysis and developed routines to analyze the data specific to this project. This is fundamental to efficiently perform the more challenging experiment using the ultrafast laser.

The delivery of the ultrafast laser was delayed to FY22; however even in the absence of the laser, we were able to design an experimental setup (depicted in Figure 2) to locate the laser. In this setup, we addressed some technical challenges, such as the entry point for the laser beam, the optical scheme of the laser, and the mounts for all these parts. Procurement is now underway. Simulations of the beam profile and its manipulation have been performed with commercial software and the optics has been specified and is being procured.

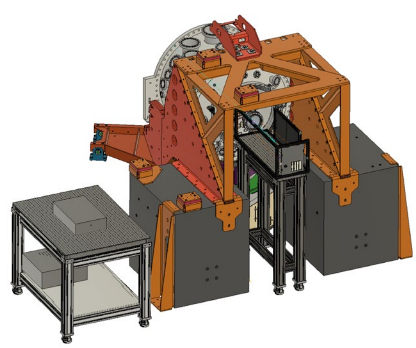


Figure 2. Design of the laser stand and focusing table.

MILESTONES:

We have scientific and technical milestones. The scientific ones will be the publication in FY22 of the conventional experiments performed on nickelates and LCMO. One manuscript (on the nickelates) is already in preparation, and we are about to submit it to a peer reviewed journal. In FY22, we expect to complete the laser setup and move on with its installation and first tests on the SIX beamline and SIX experimental chamber. We will schedule commissioning and perform experiments on actual materials (the LCMO case is particularly promising) in the second quarter of the year. At the end of FY22 and in FY23, we expect to publish the results of these experiments and extent this approach to more involved cases, such as titanates and cuprates.

Laying the Foundation for an Integrated Center for Sequence-to-Function Discovery

LDRD # 21-038

L. Yang, C. Blaby-Haas, H. van Dam, M. Fuchs, L. Wang

PURPOSE:

The National Synchrotron Light Source II (NSLS-II) structural biology beamlines, the Laboratory for Biomolecular Structure cryo-Electron Microscopy lab, and the computing resources at the Computational Science Initiative are Brookhaven National Laboratory's state-of-the-art assets for supporting cutting-edge life science research. Under this project, we aim to increase the efficacy and impact of our scientific support by integrating these technical capabilities more tightly and developing missing ancillary tools. We will devise a framework to present the entirety of our scientific capabilities to users as components in standardized workflows and guide them to frame complex scientific questions in a manner that our facilities will help them answer. In the long term, we expect this framework to help accelerate the process of elucidating protein function based on its genomic sequence, which is a grand challenge in molecular biology. We also expect this work to attract users from new research areas, particularly those funded by the Office of Biological and Environmental Research. We can therefore pursue new funding to expand our effort.

APPROACH:

The major challenge in this project is to design a generic framework that can be applicable to a broad range of scientific problems. To accomplish this, we will take on several test cases, so that we can determine how to generalize the application of available methods from the specific needs that we encounter. These test cases are selected based on the potential interest from funding agencies, so that there will be meaningful scientific output regardless of our progress with the generic framework. Experimentally, we will start from the solution scattering technique, where there have already been efforts in the field to integrate computation and modeling to help explain the functional mechanism of the overall system from static and/or partial structures. Other experimental techniques will be added as the overall framework takes shape.

TECHNICAL PROGRESS AND RESULTS:

This is the first year of this project. Our first priority is to assemble a team with both experimental and computational expertise, including two postdoctoral researchers. Dr. Estella Yee arrived in June and took on the majority of the work on protein production and characterization. The second postdoc is expected to arrive in March 2022 to perform computational work.

Scientifically, we have selected our first two test cases as discussed above: i). a heme-binding protein that is proposed to behave as a heme chaperone in plants; and ii). a guanosine triphosphate (GTP)-regulated zinc (Zn)-transferase protein that delivers its metal cofactor to Zn-dependent client proteins in a multi-step process. Since a crystal structure already exists for the heme-binding protein, we first studied how the protein structure might differ in solution under various chemical conditions, as well as due to point mutation in the sequence, so as to infer its function. Some data are summarized in Figure 1. For the GTPase, we have been working on optimizing protein production.

Protein production is a rate-limiting step for many structural biology projects. We have been discussing how our generalized workflow needs to include selection criteria in order to maximize the prospects of having a sufficient quantity of purified proteins for structural characterization and avoid wasting our limited resources. For solution scattering, we have drafted a "decision tree" that helps to determine the data collection and analysis strategy based on sample behavior.

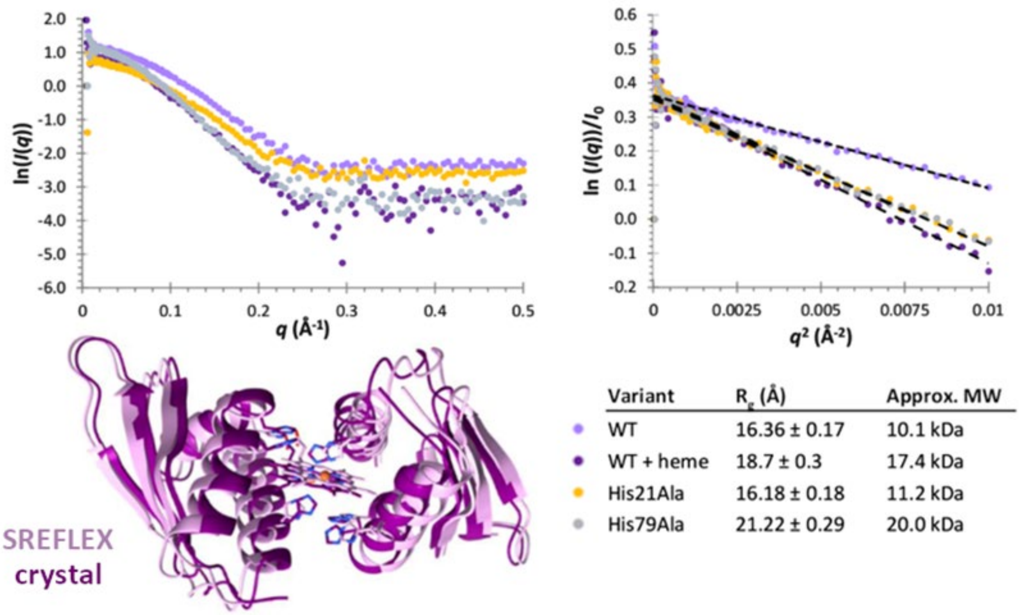


Figure 1. (Top Left) Scattering profiles and (Top Right) Guinier plots of heme-binding protein (HBP) samples: wild-type with/without heme and HBP variants with substituted His residues. Guinier region is linear and shows minimal aggregation. (Lower Left) Model calculation using SREFLEX, scattering data, and the Protein Data Bank file of the dimer illustrates minute deviations from the crystal structure. (Lower Right) Calculated Rg values and molecular weights confirm that the wild-type is monomeric in solution and dimeric with added heme. Curiously, the His79Ala variant is dimeric in solution.

Since both test cases involve metalloproteins, information on the interactions between the metal ions and the proteins adds to the overall understanding of the systems' functions. We have pursued several other experimental methods to supplement the structural information from the measurements at NSLS-II. X-ray absorption spectroscopy (XAS) is sensitive to the local electronic structure of metal ions and is helpful for studying the coordination environment of the metal ions in our test cases. Since there is no support for biological XAS at NSLS-II, we have applied for beam time and conducted measurements at the Stanford Synchrotron Radiation Lightsource. We also started a discussion with the Inner-Shell Spectroscopy beamline to bring back this capability to NSLS-II. We reactivated a benchtop Raman spectroscopy system in the Building 745 structural biology lab, with minimal investment in staff effort and material costs to bring this system online and up-to-date. We have also pursued electron spin resonance (ESR) measurements at Cornell University through the Center for AdvanCed ESR Technology facilities to complement studies of the metal environments and ligation states.

MILESTONES:

Year 1 (June 2021-May 2022): start of project; hiring postdocs; significant progress on test cases 1 and 2; selection of test cases 3 and 4

Year 2 (June 2022-May 2023): significant progress on all test cases; clear framework for the structural characterization workflow

Year 3 (June 2023-May 2024): implementation of workflow in a user-accessible portal; demonstration of workflow with a user project.

Quantum Techniques for Advanced Atmospheric Lidar

LDRD # 21-039

T. Tsang

PURPOSE:

The first stage of the project is to improve the resolution and signal-to-noise of the atmospheric lidar by developing a time-gated single-photon detection technique with the capability to reach high spatial resolution that is of interest to climate science. The second stage of the lidar development will incorporate a technique to enhance the low photon flux of the return lidar signal, such as Stevens Institute of Technology's proprietary quantum parametric mode sorting (QPMS), using nonlinear optics to improve the resolution and signal-to-noise of the atmospheric lidar. The third stage is to fully explore the quantum lidar technique (Q-LIDAR) using polarization entangled pair photon sources.

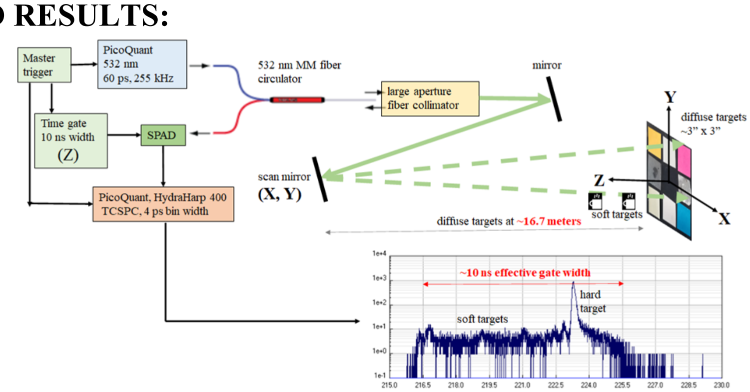
The project is synergistic with two ongoing thrust areas in the Instrumentation Division (IO): free-space quantum networking and high-resolution time-gated lidar, both of which involve single-photon detectors sensitive to quantum properties of the light field. This project will increase Brookhaven National Laboratory (BNL) collaboration with other climate science research centers, attract additional support from other funding agents, strengthen BNL Quantum Information Science and Technology infrastructure, and advance BNL quantum sensing capability.

APPROACH:

We established that 532 nm rather than near-infrared (NIR) lidar is better suited for atmospheric science studies. We initiated the classical lidar capability by building an independent 532 nm high resolution lidar setup in IO, using time-gated single-photon sensitive detection. We quantified the signal enhancement and noise limitation of quantum vs. classical lidar. We seek an approach other than QPMS to address the low photon lidar return flux and to improve the resolution and signal-to-noise of the atmospheric lidar signal. We will fully explore the Q-LIDAR technique using entangled photons and will demonstrate Q-LIDAR quantum correlations by performing quantum sensing on the lidar return signal.

TECHNICAL PROGRESS AND RESULTS:

We completed the construction and characterization of a coaxial two-dimensional (2D) (x, y) scanned 532 nm time-correlated single-photon counting Lidar ranging in the laboratory, see Figure 1. Photons returned from targets of various diffuse scattering strengths at different standoff distances are captured by a time gated Single-Photon Avalanche Diode (SPAD) (z). A 3D (x,y,z) spatial scatter map can be reconstructed. The



	TDC bin width	Lidar range	resolution	pulse energy	single photon Lidar range
Lab.	8 ps	78.6 meters	1.2 cm	2.5 nJ	35 to ~50 meters
Atmospheric	1024 ps	10.1 km	1.5 m	*1 µJ*	*14 to 20 km*

Figure 1. A coaxial 2D scanned 532 nm TCSP lidar arrangement and the lidar ranging capability in the laboratory and the projection to the atmospheric study.

lidar return signal falls off rapidly in proportion to the square of the object's distance. It is established that with the current laboratory instruments, a single-photon signal can be captured at a lidar ranging distance of up to 50 meters. With a higher energy/pulse laser, we anticipate lidar ranging up to 20 kilometers can be achieved in atmospheric science research.

We have quantified the results of a 532 nm time-correlated single photon counting (aka TCSPC) lidar ranging system and improved the capability towards a cloud chamber test run for the Environmental and Climate Sciences Department. We have evaluated a spontaneous parametric down-conversion (SPDC) polarization entangled single-photon pulse paired source - verifying that SPDC is an extremely weak ~10^{-11} linear optical process without a threshold (Figure 2) and one of its quantum characteristics, where no time coincidence detection of single-photons can be observed after a beam splitter. Although there is a small non-degenerate behavior on the signal/idler, where a spectral shift with pump power was observed that could lead to a non-maximally entangled state, we confirmed the complete absence of a multiphoton state in the signal/idler at various pump powers by capturing single photons on a photon-number resolving photodetector.

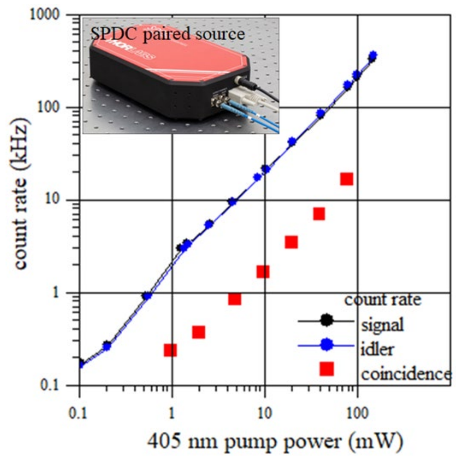


Figure 2. Count rate dependence on pump power of a SPDC polarization entangled single-photon paired source.

MILESTONES:

FY22: Guided by the experience on the evaluation of a SPDC paired source, we aim to develop a high repetition rate polarized entangled photon pair source at 532 nm that would better suit atmospheric science studies by down converting a MHz repetition rate, 60 ps pulse width, 355 nm laser to the 532 nm lidar signal (Figure 3). In contrast to a conventional point-by-point scanning lidar, we will explore the flash lidar concept where the entire field of view is illuminated with a wide diverging laser beam in a single pulse. The depth information is extracted from time-of-flight lidar return photons alongside 2D diffuse scatter images, all captured on a 2D image sensor LinoSPAD2 (Advanced Quantum Architecture Lab [known as AQUA], E. Charbon). This 2D SPAD sensor is single-photon sensitive, with a resolution of 50 ps, time-gate width of 5 ns, individually addressable to the pixel level, and 64 ps single-photon timing resolution.

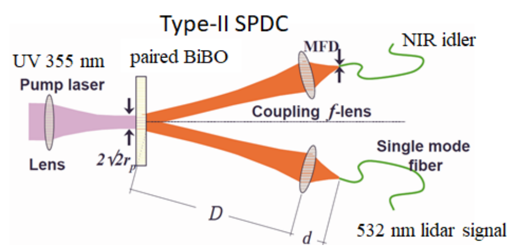


Figure 3. Arrangement of a 532 nm entangled photon source.

FY23: We will use QPMS or similar nonlinear optical techniques to improve the resolution and signal-to-noise of the atmospheric lidar return signals. We will fully explore the Q-LIDAR technique using entangled photon sources: quantum correlations of the input photons and quantum sensing for signal detection - joint interference detection of entangled signal lidar return photons with the retained idler photons.

Towards the Realization of an Electron-Ion Collider (EIC) Detector Design

LDRD # 21-041

A. Deshpande, T. Ullrich

PURPOSE:

This project provides scientific as well as technical support towards the realization of an Electron-Ion Collider (EIC) detector design. This includes defining and refining detector requirements and developing matching detector concepts. It entails strong participation and leadership in the preparation of a detector proposal and ultimately, scientific input into the detector's technical design. It aims at realizing a strong Brookhaven National Laboratory (BNL) leadership role in a future EIC collaboration to serve as a seed for future research funds from the Department of Energy (DOE) and to ensure that substantial portions of the detector construction come to BNL.

APPROACH:

BNL personnel were instrumental in all efforts leading to the launch of the EIC project. Since 2009, a Physics Department EIC Science Task Force, in collaboration with the BNL Nuclear Theory Group, has very successfully pushed the science case for this facility, producing software required to generate rigorous, quantitative simulations of physics processes for Quantum Chromodynamics phenomena in the EIC's kinematically accessible regime and defining the machine and detector requirements for key measurements. The group was essential to the production of the EIC White Paper that is now widely accepted as the definitive instrument for the scientific case for an EIC by providing key arguments for its realization. Members are advocating for the EIC in seminars and talks at national and international conferences and organizing workshops on various topics related to the EIC. Results of their studies are frequently published in peer-reviewed journals and conference proceedings.

In 2019, the DOE began to inform the two competing labs, BNL and Thomas Jefferson National Accelerator Facility (JLab), about a more aggressive timeline towards site selection. Critical Decision 0 (CD-0) was awarded in December 2019 and the selection of BNL as the site of the EIC was announced in January 2020. It was therefore necessary to adjust these efforts to the revised timeline and address the new challenges associated with it. This led to the pursuit of this project.

Given the success and substantial experience of the scientists involved, this project builds on previous achievements and works towards securing leading roles in the experimental EIC collaboration, detector design, and subsequent detector construction. It focuses on two main efforts: (i) the realization of the "Yellow Report" and (ii) the design of a new EIC detector within a newly-formed collaboration. The workforce funded by this project plays a leading role in many aspects of these efforts. It includes Dr. Alexander Kiselev, Dr. Brian Page, Dr. Salvatore Fazio (until March 2021), and Dr. Zhoudunming Tu (since July 2021).

TECHNICAL PROGRESS AND RESULTS:

In December 2019, the EIC User Group engaged in a massive effort to advance the state and detail of the documented physics studies and especially detector concepts in preparation for the EIC. The results of this initiative were planned to be released in a report (Yellow Report) in early Spring 2021. In FY21, most of the scientific staff funded by this project were fully engaged in this effort. S. Fazio was a convener of the Physics Working Group on Exclusive Processes, B.

Page was convener of the Jets and Heavy Flavor Physics Working Group, A. Kiselev was convener of the Magnet and Integration Working Group, and the Principal Investigator (PI), together with Rolf Ent (JLab), led and organized the Yellow Report effort. All members of the group were instrumental in many of the studies and in writing the final report. The report was released in March 2021 (arXiv:2103.05419). It comprises 902 Pages, 1780 references, and 675 figures, from 414 authors. This document presents a comprehensive path towards building a detector that meets all the requirements for conducting the physics program expressed in the Nuclear Science Advisory Committee Long Range Plan and the National Academy of Sciences Report on EIC Science, as mandated by DOE. The group was instrumental in almost all aspects of this effort, organizing, conducting simulations and studies, and writing and editing.

In March 2021, BNL and JLab jointly issued a Call for Collaboration Proposals for Detectors at the EIC (https://www.bnl.gov/eic/cfc.php), with proposals due by December 1, 2021. In response to that call, three proto-collaborations have formed. The largest, ATHENA, was established at the end of March. The members supported by this project form the backbone of BNL's membership in the ATHENA collaboration. The planned general-purpose detector is built around a new 3 T magnet to be located in the official EIC Interaction Region (IR), IP6. All scientists funded by this project hold (and held) leadership positions in the collaboration, focusing almost exclusively on efforts related to the detector proposal. A. Kiselev was a member of the Steering Committee that ran the effort during the first months. He later developed the ring-imaging Cherenkov (RICH) detector in the backward hemisphere (pfRICH) and pursues R&D on the development of Large Area Picosecond Photodetector (known as LAPPD) photosensors that are vital for all particle identification detectors in ATHENA. The PI (Ullrich) is a member of the proposal committee, the global design and integration group, and, since July, a member of the Executive Board. He was a member of the ATHENA charter committee and is the BNL representative on the ATHENA Institutional Board. Brian Page is a convener of the Jet, Heavy-Flavor, Electroweak, and Beyond the Standard Model Physics working group. In parallel, he evaluated the effects of beam crossing angle and other beam effects on the physics measurements. The method he developed is now being used by ATHENA to simulate and correct for realistic beam conditions. Page initiated this effort, formed and organized a task force, and provided key technical notes on the subject. Z. Tu is responsible for the evaluation and feasibility study on one of the most critical measurements in e+A, namely that of diffractive, exclusive vector meson production in ATHENA. All three contributed substantially to the proposal and to the subsequent review talks. They strengthened the visibility of BNL in ATHENA and helped to design a detector that is meeting all physics requirements. Independent of the detailed outcome of the proposal process, the members funded by this project will play key roles in the construction and operation of a future EIC detector.

This one-year long project had defined three milestones of which two are met: (i) realization and release of the Yellow Report and (ii) formation of and membership in a collaboration, design of a comprehensive general-purpose EIC detector, and creation and submission of the proposal for it. The third milestone entailed engaging in the design and construction of forward calorimeters for an EIC detector. This idea has been dropped in favor of involvement in a barrel time-of-flight detector based on alternating current-low-gain avalanche detector (known as AC-LGAD) sensors that is better matched to the group's and BNL's strength.

MILESTONES:
None. This project completed in FY21.

A Path Forward to Retain BNL's Leadership in Electron-Ion Collider (EIC) Science

LDRD # 21-042

E. Aschenauer

PURPOSE:

This project provides the scientific guidance needed to move the Electron-Ion Collider (EIC) through the Critical Decision (CD) phases. In addition, the project provides critical support for EIC community building. For that, Brookhaven National Laboratory (BNL) needs to have scientists, who are fully dedicated to the EIC and provide help in fully integrating the EIC science in the accelerator, Interaction Region (IR), and detector design. This project builds on the successful approach in past years to have a dedicated group for the EIC, which ultimately led BNL to be selected as the site for the EIC. This project proposed to follow the same strategy to secure Department of Energy (DOE) research funding for the EIC.

APPROACH:

On January 9, 2020, DOE announced the selection of BNL as the site for a planned major new nuclear physics research facility, the EIC. This announcement followed DOE's earlier approval of "mission need" (known as CD-0 on December 19, 2019). This approval enabled work to begin on R&D and the conceptual design for this one-of-a-kind next-generation collider, and allowed the EIC to move to the next phase, CD-1. The project is moving forward very rapidly. Unfortunately, the announcement of CD-0 was not paired with DOE research funding nor are scientific collaborations yet formed to support the EIC project scientifically. Therefore, the only way to address these points requires BNL to continue to invest discretionary funding for the EIC. This project addresses several critical points:

- It is part of a capture strategy for future EIC research funding from DOE. The same strategy, i.e., having an agile vibrant group of young scientists developing the EIC science case was critical to the success of BNL becoming the EIC site.
- The unique scientific scope of this project has become even more critical after the EIC received CD-0. The rapid path of the EIC project to the CD-1 Independent Project Review (IPR) review (01/2020) and the path to the CD-2 IPR review has been laid out.
- The scientists, Dr. J. Adam, Dr. A. Jentsch, and Dr. Z. Zhang, supported by this project are deeply involved in providing the important scientific validation on ongoing IR design due to the design impacts on the acceptance and hence physics program support provided by the common/ancillary IR detectors. This work is the basis for several scientific publications.

TECHNICAL PROGRESS AND RESULTS:

J. Adam focused his EIC efforts on the realization of a full Geant4 model for conceptual studies of the luminosity monitor and low-Q^2 tagger. Luminosity measurement is based on detecting high-energy forward bremsstrahlung photons from Bethe-Heitler scattering. The luminosity monitor provides two independent methods to detect these bremsstrahlung photons, one to address instantaneous collider performance, the other optimized for precision. The Geant4 model of the luminosity monitor contains all the essential components. Studies with the model were being performed to address the acceptance as a function of photon energy for specific cases of geometry layout and for possible construction of the conversion layer. Current results indicate the feasibility of both methods of luminosity measurement. The low-Q^2 tagger is aimed at detecting electrons scattered at very small angles, outside the acceptance of the central detector. The tagger is placed beside the outgoing electron beam pipe at a distance from the central detector. J. Adam developed a complete Geant4 model to study the acceptance of the tagger as a function of Q^2 for different possible positions of the tagger detector. The model includes a complete layout of the outgoing beam magnets between the central detector and the tagger and a simplistic model of the tagger detector itself, which counts scattered electrons which hit its volume. Results from referring simulations were used to demonstrate the feasibility of reconstructing the Q^2 in the tagger with the required precision.

In 2020, A. Jentsch became a sub-convener for the Far-Forward Detector Working Group for the EIC Yellow Report. Besides helping lead that group to a successful completion of its Yellow Report tasks, he also helped turn many of the studies carried out for the report into published scientific papers. In more recent analysis work, he has been the primary analyst for a study of the extraction of free nucleon structure in tagged Deep Inelastic Scattering with e+d collisions at the EIC, which was also published. Besides his work for the Yellow Report, he also contributed to the refinement of the design for the first EIC IR and contributed studies to the EIC Conceptual Design Report (CDR). Dr. Jentsch has helped all three proto-collaborations in their efforts to simulate the IRs both for IP-6 and IP-8. Additionally, A. Jentsch has been a prime participant in the efforts of eRD24, performing detector simulations to help refine requirements for the development of an alternating current-coupled low-gain avalanche detector (aka LGAD) sensor for use in a Roman Pot detector system, and for other applications in the far-forward region. Finally, he has been instrumental in validating the pre-conceptual design for the second IR far-forward hadron lattice, providing detailed particle acceptance studies and ideas for optimization of detectors.

Z. Zhang has completed the basic IR design of the EIC's Compton polarimeter in the storage ring for the electron beam in IR6. The polarimeter will be located in the forward direction of IP6. He studied the acceptance of the scattered photon and the recoil electron as well as the time dependence of the polarization's statistical uncertainty. He also investigated the requirement for the magnets between the laser interaction point and the photon detector. In parallel, Z. Zhang worked on measurements of photoproduced diffractive di-jets in ep collisions and the emerging detector requirement for an EIC detector. He also investigated various "tunes" of the Pythia8 event generator for ep collisions at EIC energy. All of his work has been essential to the different EIC proto-collaborations in the proposal process.

E.C. Aschenauer shepherds the experimental program, together with Rolf Ent, as part of the EIC project. The work done as part of this project was critical for this; it allowed us to ensure the IR design at IP-6 fulfills the scientific requirements and that the knowledge gained from this was readily transferred to design a complementary IR at IP-8. During the Yellow Report process as the convener for the detector complementarity working group, the detector and IR complementarity was especially critical to make the case for two detectors and IRs regions at the EIC. She is also the main contact to the accelerator experts to ensure the science requirements are well-integrated into the EIC accelerator design and are not jeopardized by technical decisions. The simulation tools and deep understanding of the interplay between the accelerator design, the far forward and backward experimental equipment along the IR (+/-40 m), and the methods to measure polarization with the EIC scientific program are critical for the next phases of the EIC and broadening the research directions of the EIC.

MILESTONES:

July FY22: Provide scientific guidance for the lepton polarimeter and the experimental equipment along the beam line, together with the EIC collaboration, to ensure that the design level required for CD-2 can be achieved.

Accelerating State Preparation in Quantum Field Theory (QFT) Calculations on a Universal Quantum Computer

LDRD # 21-043

T. Izubuchi, C. Lehner

PURPOSE:

This project aims to optimize the preparation of quantum states to control uncertainties of simulations of simple Quantum Field Theories (QFTs) on the first available universal quantum computers in the noisy intermediate-scale quantum (NISQ) era. By collecting QFT, especially Lattice Field Theory and Quantum Information Science (QIS) expertise, we are establishing a QIS application community at Brookhaven National Laboratory, which conducts a broad research program.

APPROACH:

To study methods to prepare quantum states, which is one of the profound problems in applications of Quantum Computing for QFT that will theoretically predict the physical observables as the matrix elements and their further time evolution, the efficiency of the length of the quantum circuit is a critical factor due to the decoherence and fidelity properties of practical quantum computers and also for sampling the relevant potentially large Hilbert space.

In FY21, a project led by Gumaro Rendon, improved state preparation techniques that use the quantum phase estimation algorithm. Also a project, led by Yutta Kikuchi, designed quantum algorithms to simulate QFT and demonstrated the methods by using the Schwinger model; he also studied quantum chaos/information scrambling from the viewpoint of quantum computing.

TECHNICAL PROGRESS AND RESULTS:

In an earlier stage of the project, we prepared the ground state via an adiabatic process in the simplest toy model of Quantum Chromodynamics (QCD), the 1+1 dimension Quantum Electrodynamics (QED) or Schwinger model, and successfully produced the correct results within the current QIS simulator (IBM Qiskit and Quspin). This method worked for the Schwinger model with an external electric field, which has the notorious sign problem, just like QCD with the chemical potential.

This year, we provided an improvement to the quantum phase estimation algorithm (QPEA) inspired by classical windowing methods for spectral density estimation. From this improved circuit, shown in Figure 1, we obtained an upper bound in the cost that implies a cubic improvement with respect to the algorithm's error rate (Figure 2). Numerical evaluation of the costs also demonstrates an improvement. Moreover, with similar techniques, we detailed an iterative projective measurement method for ground state preparation that gives an exponential improvement over previous bounds using QPEA. Numerical tests that confirm the expected scaling behavior were also obtained. For these numerical tests, we used a Lattice Thirring model as a testing ground. Using well-known perturbation theory results, we also showed how to better estimate the cost scaling with respect to the state error instead of the evolution operator error.

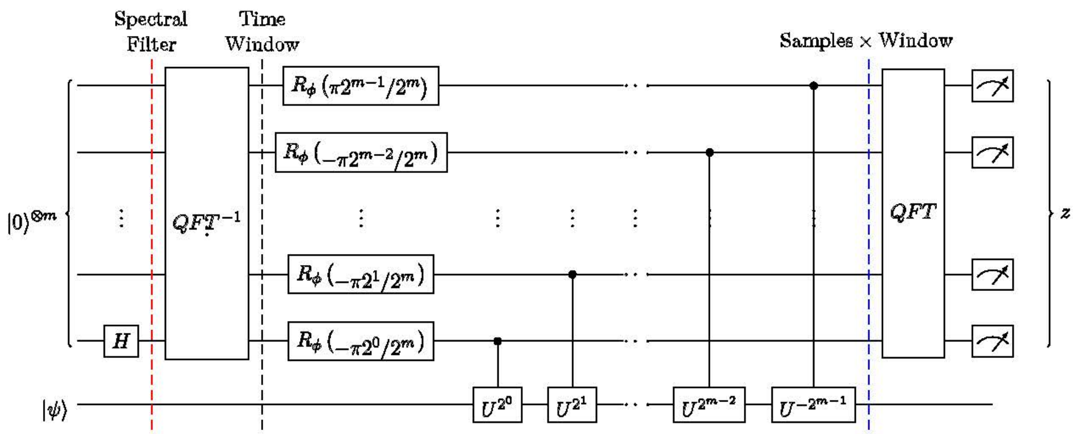


Figure 1. Quantum circuit to implement the improved m-qubit phase estimation algorithm. U is the unit time evolution exp(2πiλH). The red dashed line marks the samples of the filter function stored in the ancillary register and the black dashed line the samples of the corresponding window function.

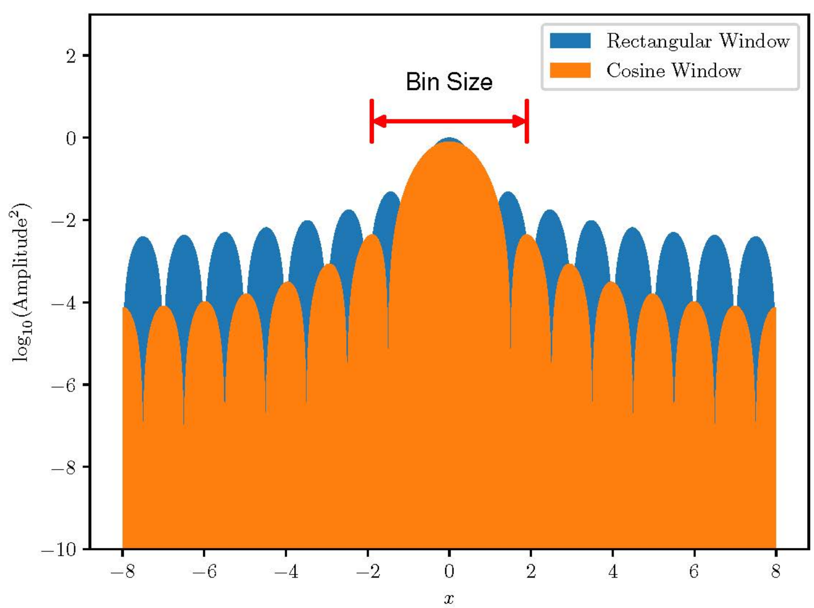


Figure 2. The cosine tapering windows to filter quantum states with (shifted) energy apart from zero are shown in the orange curve, which has a wider main lobe but provides a higher peak. This modification introduced a cubic improvement in gate complexity scaling with respect to the error rate. We also showed the effects of this window when the phase estimation algorithm is repurposed for quantum state preparation. Simultaneously, we showed that using repeated blunted filter operations was more efficient than performing a single sharper filter operation. The improvements were exponential of inverse error and inaccuracy of the input quantum state, while linear scaling with respect to the inverse of the energy gap remains.

The exploration of other windows, their optimization with respect to other metrics, as well as the exploration of possible hybrid quantum-classical approaches is left to future research.

MILESTONES:
None. The project completed in FY21.

DEDUCE: Differentiated Evaluation to Decrease Uncertainty in Computational Experiment

LDRD #21-044

B-J. Yoon

PURPOSE:

This project aims to address a fundamental problem that pervades a wide range of science and engineering disciplines: the need to design optimal strategies and robust computational campaigns for complex systems in the presence of substantial uncertainty. Specifically, the primary objective of this project is to develop capabilities to optimally steer computational campaigns to maximize the expected "return-on-computational-investment (ROCI)" and to enable objective-based uncertainty quantification and optimal experimental design for the latest machine learning/artificial intelligence models, which can serve as effective surrogates for complex real-world systems. Our hypothesis is that the proposed framework will significantly enhance the ROCI for various objective-driven scientific problems that involve uncertain complex systems, as one may find in drug discovery, atmospheric science, materials science, or complex energy systems.

APPROACH:

To achieve the aforementioned project goals, we formulate an optimization framework that incorporates the computational cost, constraints, fidelity/accuracy, as well as the statistical relations among the multiple stages that comprise a scientific discovery/screening pipeline/workflow. This allows us to identify the optimal policy for steering the pipeline/workflow to optimize the throughput and enhance the predictive outcomes in the presence of practical constraints and uncertainties present in the models and data. Our proposed strategies can lead to robust and effective solutions for various scientific and engineering problems that require optimal operation/design for complex systems under uncertainty.

TECHNICAL PROGRESS AND RESULTS:

During the previous reporting period, we have obtained preliminary yet promising results relevant to the overall project goals.

First, in collaboration with members of the ATOM (Accelerating Therapeutics for Opportunities in Medicine) Consortium, we have developed a multi-objective latent space optimization method that can be used to significantly enhance the sampling efficiency of generative models used for molecular design. The proposed method has been shown to effectively shift the latent space through iterative retraining of a generative model (e.g., a variational auto-encoder). The efficacy of the top candidate molecules generated by our model have been computationally tested – via docking models and molecular dynamics simulations to predict the binding affinity against the desired target – by our collaborators in the ATOM Consortium. These preliminary validation results showed that the predicted molecules are highly effective. We are currently considering experimental validation of the top molecules.

Furthermore, we have developed a novel method for optimal computational campaign for molecular screening. The work was inspired by the COVID-19 drug screening pipeline, IMPECCABLE, developed by some of our team members in collaboration with other Department of Energy National Laboratories. Results have shown that our proposed method can reduce the computational cost of a four stage screening pipeline by up to 50% virtually, without any degradation in the average screening accuracy. The preprint of our work is available at: https://arxiv.org/abs/2109.11683. We are finalizing the manuscript for submission to a leading journal in data science.

MILESTONES:

- Develop optimal computational campaign algorithms for molecular screening
- Develop a multi-objective latent space optimization method for models used for generative molecular design (GMD)
- Explore the use of deep reinforcement learning to learn optimal policy for steering a computational high-throughput screening pipeline
- Validate molecules suggested by the optimized GMD
- Disseminate the results to the research community via journal/conference publications by the end of the project.

The Study of the Nucleon Structure at Jefferson Lab and the Future Electron-Ion Collider (EIC)

LDRD # 21-045S

H. Gao

PURPOSE:

To reliably extract the Transverse Momentum Dependent Parton Distribution Functions (TMD-PDFs or just TMDs) from the Semi-Inclusive Deep Inelastic Scattering (SIDIS) process, Quantum Electrodynamics (QED) radiation effects must be considered. At higher energies (12 GeV) relevant to Jefferson Lab and especially to the Electron-Ion Collider (EIC) to be built, the phase space for photon radiation becomes larger. Hence, at higher energies accounting for all radiated photons with the existing conventional approach becomes more challenging. One needs to study various approaches to get a good handle on systematic issues associated with radiative effects.

APPROACH:

Liu et al. [1] recently proposed a new factorized framework for SIDIS reaction, which simultaneously treats QED and Quantum Chromodynamics (QCD) effects on the same footing. Instead of treating the QED radiation as a correction to the Born process, they unify the QED and QCD contributions to the lepton-nucleon scattering cross-section in a consistent factorization formalism. Our goal is to develop a code to implement the new factorized approach for SIDIS to compute the Radiative Correction (RC) and compare the RC from the factorized approach with the conventional approach from Akushevich and Ilyichev [2]. This comparison will provide an understanding of systematic uncertainty due to RC, which is one of the most important sources of uncertainty in extractions of TMDs.

TECHNICAL PROGRESS AND RESULTS:

We have implemented a new factorized approach to compute the RC for the unpolarized SIDIS $H(e,e'\pi^+)X$ reaction and compared it with the result from the conventional approach for an unpolarized target. In our analysis, the structure functions are taken from the JAM3D20 global analysis [3]. The radiative correction is given by the ratio of the six-fold differential cross-section ($d^6\sigma/dx_B dy dz dP_T^2 d\Phi_h d\Phi_S$) without radiative effects to the differential cross-section with radiative effects. The RC ratios computed for Jefferson Lab and EIC kinematics are presented in Table 1.

$\sqrt{s}$ (GeV)	x_B	Q^2 (GeV^2)	z	RC ratio
Jefferson Lab Kinematics				
3.2	0.32	2.3	0.55	1.06
4.9	0.48	8	0.375	1.08
6.7	0.48	15	0.375	1.09
EIC Kinematics				
140	0.01	9	0.5	1.08
140	0.01	25	0.5	1.10
140	0.01	100	0.5	1.10

Table 1. Comparison of RC between factorized and conventional approaches at different Jefferson and EIC kinematics. RC from the factorized approach is 6-10% larger compared to the conventional approach.

For the given kinematics, we studied the variation of RC with respect to a hadron transverse momentum P_T and found that RC ratios were stable for P_T below 1 GeV. We observed 6-10% difference in RC between the two approaches. This difference especially at lower energy might be explained by the contribution of the radiative tail to RC in the conventional approach, which has not been implemented yet.

MILESTONES:
The following are our milestones for fiscal year 2022:

- Estimate the effect of RC in the extraction of Sivers and Collins asymmetry in the case of a polarized target
- Estimate the exclusive tail contribution in the conventional approach, which has a significant contribution [4] but due to the limited knowledge of exclusive structure function, it has not been added in our analysis yet
- Finish writing a paper and submit it to a journal for publication.

References:

[1] Tianbo Liu, W. Melnitchouk, Jian-Wei Qiu and N. Sato. "Factorized approach to radiative corrections for inelastic lepton-hadron collision", 2021

[2] Igor Akushevich and Alexander Ilyichev. "Lowest order QED radiative effects in polarized SIDIS", *Physical Review D*, 100 (3), Aug 2019.

[3] J. Cammarota, *et al*. "Origin of single transverse-spin asymmetries in high-energy collision". *Phys. Rev. D,* 102:054002, Sep 2020

[4] I. Akushevich, A. Ilyichev, and M. Osipenko. "Lowest order QED radiative correction to five-fold differential cross-section of hadron leptoproduction". *Physical Letters B*, 672 (1).

Progress Toward a Resilient, Lower Carbon Electric Grid via Improved Forecasting

LDRD # 21-046
A. Marschilok

PURPOSE:
There is an urgent national interest and regional need to develop a more resilient, lower carbon electric grid. However, there are multifaceted challenges to utilities and energy providers in today's environment. Without appropriate mitigation, the impact of hazardous weather events can be severe. Integration of renewables and energy storage, including vehicle to grid concepts, provides significant opportunity yet added complexity for design of a greener future grid.

The New York Climate Leadership and Community Protection Act (CLCPA) set out a bold vision of a Statewide energy paradigm that will result in a more sustainable society for today's citizens and those yet to come by 2050 – with the goal of a totally carbon-free grid by 2040. With a four seasons climate, population density ranging from dense urban to rural, and aging infrastructure in nationally critical economic centers, New York State and the Northeast region encompass a diverse landscape with unique electric grid challenges.

There is a need for improved forecasting capability at the requisite timescale and resolution to serve energy providers. This includes forecasting of weather events and forecasting of modern consumer energy use cases, such as vehicle to grid energy storage. These issues are of particular interest to Con Edison (ConEd), operator of one of the world's largest energy delivery systems, who provides energy for ten million New York residents.

APPROACH:
Under this program, targeted forecasting projects of mutual interest to Brookhaven National Laboratory's (BNL) Interdisciplinary Science Department and ConEd will be pursued. This project also aligns with New York State's Clean Energy Climate Protection Act Goals and could facilitate future funding from the New York State Energy Research and Development Authority (NYSERDA). Finally, this project aligns with Department of Energy (DOE) interests, including the current cross DOE (Office of Science, Office of Energy Efficiency and Renewable Energy, Office of Electricity, and Office of Technology Transitions) Energy Storage Grand Challenge.

TECHNICAL PROGRESS AND RESULTS:
This project started in August 2021.

Con Edison and BNL hosted a strategic summit, titled "Envisioning a Sustainable NY State Electric Grid" on November 15 and 16, 2021. This summit was a major step toward incorporating the intellectual and investigative capabilities of BNL into a wider collaboration with the entities that will have a role to play in meeting CLCPA goals.
Over 200 participants attended the two-day virtual summit, with representation from utilities, national laboratory and university scientists, government, federal and state funding agencies, engineering firms, environmental justice advocates, and minority- and women-owned businesses.

The meeting was structured as a series of plenary sessions, topical breakout sessions, and panel discussions. The vision for the New York State electric grid was the topic for the opening plenary session on the first day. Challenges and future opportunities for a sustainable electric grid was the topic for the second session. Energy equity was the theme for day two, including a panel discussion on Building an Equitable Energy Future.

The participants displayed recognition of the importance of the topic, with high enthusiasm and commitment to move forward with solutions. The breakout sessions outlined seventeen actionable project opportunities to advance the goals of decarbonization and energy equity and ultimately realize the vision of a totally carbon-free Northeast grid by 2040.

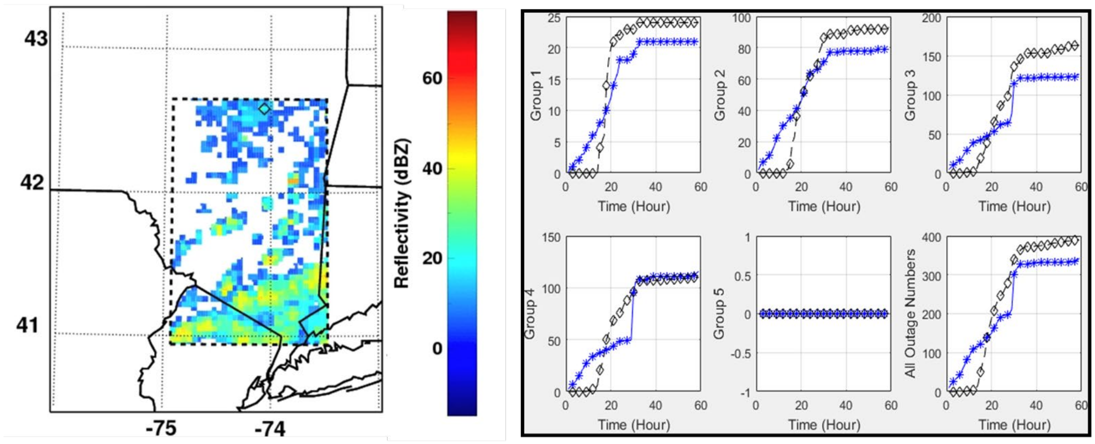


Figure 1. (Left) Time series of storm activity and outage data within ORU utility service area. (Right) Failure rate model calculated using median weather data, with outage prediction for five areas of the utility service territory.

A weather forecasting study is underway, as a collaborative effort of the Interdisciplinary Science Department and the Climate and Environmental Sciences Department. This study is developing a data-driven dynamic, granular, and multi-day power outage forecasting (up to 72 hours lead time) tool (Figure 1, left) by leveraging BNL expertise in atmospheric science, power grid engineering, machine learning applications, and probabilistic risk assessment. This work builds on the models, algorithms, and tools developed by the BNL team under a NYSERDA-funded project, jointly performed with Orange and Rockland Utilities (ORU) and Central Hudson Gas and Electric (CHG&E), that developed a tool for nowcasting of weather-related damage to the power grid.

A failure rate model (Figure 1, right) was calculated using median weather data in a small area, where the total number of outages exceeds 400 and most of the outages are overhead cable-related. The outage prediction was performed for five areas of the utility service territory with hourly weather condition data and the utility's component inventory as inputs and hourly evolution of overhead outages in different areas as outputs. The development of Bayesian outage prediction using pseudo-weather forecasts is underway. The sensitivity of assumptions/choices in the development of the model is under investigation so that uncertainty in the model can be assessed.

MILESTONES:

- Hosted joint Con Edison/BNL joint strategic summit on 15-16, November 2021
- Assembled a BNL project team on weather forecasting; biweekly project meetings continue
- Provided an update on initial weather forecasting results as part of the Con Edison/BNL joint strategic summit discussion
- Continued discussions to establish a Con Edison – BNL Cooperative Research and Development Agreement.