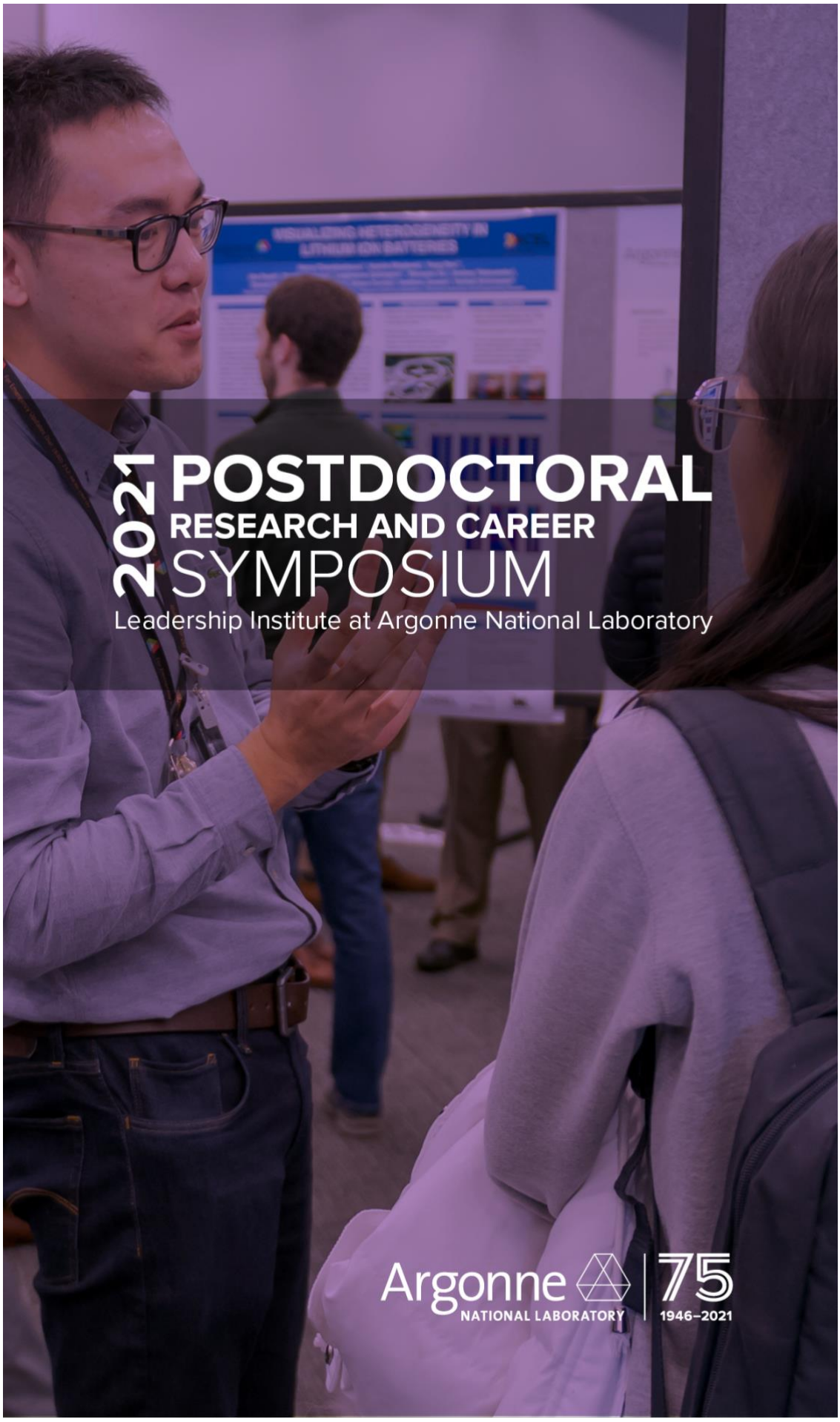
2021
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Argonne National Laboratory Postdoctoral Research and Career Symposium

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2021 Postdoctoral Research and Career Symposium
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Agenda

Note: All times are Central Standard Time

Day 1 – Thursday, November 4, 2021

09:00 AM Event Opening and Welcome Remarks
09:15 AM Keynote Address by Dr. Linda Horton
10:00 AM Research Presentations #1
12:00 PM Lunch Break
01:00 PM Networking with Companies #1
03:00 PM Career Panel – DOE/National Lab track
04:00 PM Close of Day 1 Sessions

Day 2 – Friday, November 5, 2021

09:00 AM Career Panel – Academia/R&D Research Track
10:00 AM Research Presentations #2
12:00 PM Lunch Break
01:00 PM Networking with Companies #2
03:00 PM Career Panel – Nontraditional Industry track
04:00 PM Award Presentations and Closing Remarks
04:15 PM Trivia Social
04:30 PM End of Day 2 Sessions

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Keynote Address

Dr. Linda L. Horton, Associate Director of Science for Basic Energy Sciences,
Department of Energy

Dr. Linda L. Horton is the Associate Director of Science for Basic Energy Sciences (BES) in the Department of Energy (DOE) Office of Science (SC). With an annual budget of more than $2 billion, BES is the nation's leading supporter of fundamental research in materials sciences, chemistry, geosciences, and aspects of biosciences. BES is also a major supporter of scientific user facilities, including the nation's premier x-ray light sources, neutron scattering facilities, and nanoscale science research centers. These facilities serve over 16,000 users annually, and they provide the tools for the preparation and examination of materials and the study of their physical and chemical properties and transformations.

Previously, Dr. Horton was the Director for the Materials Sciences and Engineering (MSE) Division in BES. Since joining DOE in 2009, Dr. Horton has been extensively involved in interdepartmental coordination activities, including serving as the DOE-SC management lead for batteries and energy storage, the SC Early Career Research Program, and is the past co-chair for the OSTP-led Materials Genome Initiative. Her BES roles have included interim leadership of the management team for the newly established Energy Frontier Research Centers. She has served as the materials lead for international activities that included workshops with the European Union on energy storage and critical materials. As part of the BES communication strategy, she had lead responsibility for the “Science Serving the Nation” brochure. Since 2016, she led the BES teams responsible for 7 strategic “basic research needs” workshops/roundtables and the associated brochures and reports.

Prior to joining BES, Dr. Horton was the Director of the Center for Nanophase Materials Sciences at Oak Ridge National Laboratory, one of the five DOE national user facilities for nanoscale science research. Her personal research emphasized applications of electron microscopy to materials science problems including investigations of the effects of ion implantation and neutron irradiation and studies of the growth and characterization of diamond thin films. She is a Fellow of ASM International and has served on several national and international advisory committees. In professional service, she has been an officer of the Materials Research Society and of the

Microscopy Society of America, Trustee of ASM International, and Vice-Chairman of the DOE Basic Energy Sciences Advisory Committee. Her Ph.D. in Materials Science is from the University of Virginia.

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Welcome and Closing Remarks Speaker

Stephen K. Streiffer, Interim Deputy Laboratory Director for Science, Associate Laboratory Director, Photon Sciences, and Director of the Advanced Photon Source

Stephen Streiffer is the Interim Deputy Laboratory Director for Science, Associate Laboratory Director for Photon Sciences and Director of the Advanced Photon Source at Argonne. The Photon Sciences directorate consists of the X-ray Science, Accelerator Systems, and Advanced Photon Source Engineering Support divisions, which comprise the Advanced Photon Source (APS) and the Argonne Accelerator Institute.

The APS is the brightest source of high-energy X-rays in the Western Hemisphere and is used to study the structures of materials and processes at the atomic scale. It is also the largest scientific user facility in the North America, with more than 3,500 users visiting each year.

He has also served as interim director of Argonne's Center for Nanoscale Materials, a national user facility that provides capabilities explicitly tailored to the creation and characterization of new functional materials on the nanoscale. The center's portfolio includes research on electronic and magnetic materials and devices, nanobio interfaces, nanofabrication, nanophotonics, theory and modeling, and X-ray microscopy.

Dr. Streiffer's scientific expertise is in nanostructured complex oxides and in structural characterization of materials particularly using transmission electron microscopy and X-ray scattering techniques.

Overarching themes in his research program include the development of novel concepts for integration of oxide heterostructures, establishing a fundamental understanding of polar interfaces, and exploring how these interfaces may be manipulated to influence electronic and chemical function.

His active research projects focus on utilizing *in situ* synchrotron X-ray methods to probe chemical vapor deposition of complex oxides as well as phase transformations and nanoscale size effects in ferroic thin films. He is also currently involved in *in situ* synchrotron X-ray studies of the synthesis of InGaN heterostructures as part of an effort to expand the basic understanding of materials for energy-efficient solid-state lighting. He has authored or co-authored more than 150 scientific publications and holds one patent.

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Career Panelists

Careers in National Laboratories/DOE

Giselle Sandi, Deputy Division Director, Chemical Sciences and Engineering
Argonne National Laboratory

Dr. Sandi has nearly two decades of experience in the national laboratory complex. Her research interests include nuclear forensics, nanoscale engineering and materials for energy storage. Sandi came to Argonne in 2020 from Rush University Medical College where she was the Director of the Office of Rush Mentoring Programs from 2013 to 2020, supporting the successful transition of junior faculty to independent investigators. She also was associate professor in the Department of Microbial Pathogens and Immunity at Rush University Medical College. Prior to her time at Rush, Sandi served as Argonne's Women in Science and Engineering Program Initiator in 2012. In that capacity, she provided support for the success of women in science and pushed for gender equity. From 2007 to 2011, Sandi served as the Argonne Postdoctoral Programs Coordinator in the Division of Educational Affairs and received the UChicago Argonne, LLC, Board of Governors Pinnacle of Education Award for contributions and leadership in establishing and managing Argonne's postdoctoral program. Sandi first joined Argonne as a postdoctoral fellow in 1994 where she conducted fundamental research in the areas of energy storage, materials for hydrogen storage, electrocatalytic membranes, nuclear forensics, sensor development and nanoscale engineering. She was also an adjunct professor in the Department of Chemical and Environmental Engineering at the Illinois Institute of Technology. Dr. Sandi is the recipient of the 2004 Luminary Award for Excellence in Science, Engineering, and Leadership (granted by the Hispanic Engineer National Achievement Awards Conference) and the 2019 Lifetime Achievement Award in Education, Great Minds in STEM. She is also the recipient of the 2019 organizational leadership award granted by the Group of Women in Medicine in Science of the American Association of Medical Colleges. Dr. Sandi holds a PhD in chemistry from Northern Illinois University and has 89 scientific publications, seven book chapters and three U.S. patents.

Ashley Head, Staff Scientist, Center for Functional Nanomaterials
Brookhaven National Laboratory

Ashley Head received her B.S. degree in chemistry from James Madison University in 2005. In 2011, she graduated from the University of Arizona with a PhD in physical chemistry while working with Dennis Lichtenberger. She studied gas phase photoelectron spectroscopy and managed a small instrument facility as a graduate student. She moved to Lund University as a postdoc from 2012 to 2015 and worked in a synchrotron user facility known as Max Lab at the time but has since been upgraded to MAX IV. Working with Joachim Schnadt in the Physics Department, Ashley studied atomic layer deposition mechanism of metal oxides with ambient pressure X-ray photoelectron spectroscopy (APXPS). She continued APXPS studies of metal oxides in the Chemistry Division at Lawrence Berkeley National Laboratory from 2015 to 2017, studying the interactions between nerve agent simulants and gas mask filter materials. Since 2018, Ashley has been a staff scientist at the Center for Functional Nanomaterials at Brookhaven National Laboratory. She comanages an APXPS instrument and several IR spectrometers. Her research focuses on the catalytic activity of metal oxides and metal organic frameworks.

Anibal Boscoboinik, Staff Scientist, Center for Functional Nanomaterials
Brookhaven National Laboratory

Anibal Boscoboinik is a materials scientist at the Center for Functional Nanomaterials at Brookhaven National Laboratory. He specializes in the synthesis of nanostructured surfaces with novel properties, and their study through a variety of techniques, including X-ray photoelectron spectroscopy and infrared reflection absorption spectroscopy. He received his PhD from the University of Wisconsin-Milwaukee and did postdoctoral studies through an Alexander von Humboldt fellowship at the Fritz-Haber Institute of the Max-Planck Society in Berlin, Germany.

Paulina Rychenkova, Business Development Executive, Science and Technology Partnerships and Outreach, Argonne National Laboratory

Paulina is a Business Development Executive in the Science and Technology Partnerships and Outreach directorate. She supports researchers in computational science, AI/ML, physics and high energy physics. Paulina also leads the Argonne in Chicago program. Prior to joining Argonne in 2018, Paulina worked as a venture investor in early- and growth-stage technology companies for Charles River Ventures, The Carlyle Group, and the BlueCross BlueShield Venture Fund. She started her professional career as a consultant in McKinsey & Company's Boston office. Paulina holds a Ph.D. in Theoretical Physics from the University of Cambridge and a B.A. in Physics from Hope College (Mich.).

Paul Albertus, Associate Director of the Maryland Energy Innovation Institute and Assistant Professor of Chemical and Biomolecular Engineering
University of Maryland, College Park
(previously) Program Director, ARPA-E, US DOE

Dr. Paul Albertus holds a B.S.E. in Chemical Engineering from the University of Michigan, and a Ph.D. in Chemical and Biomolecular Engineering from the University of California, Berkeley, where he studied Electrochemical Engineering. Dr. Albertus then spent five years at the Bosch Research and Technology Center in Palo Alto, CA, as a Senior Research Engineer, where he continued research on electrochemical systems, including Li/Oxygen and flow batteries. In 2014 Dr. Albertus joined the Advanced Research Projects Agency – Energy, where he led the development of two programs (IONICS and DAYS) focused on energy storage for both portable and stationary applications, and managed electrochemical projects across a range of ARPA-E programs. During his time at ARPA-E Dr. Albertus led the initiation of $96M in new projects and additional funding for existing projects. Dr. Albertus transitioned to academic roles at the University of Maryland, College Park, in 2019, and currently serves as Associate Director of the Maryland Energy Innovation Institute and Assistant Professor of Chemical and Biomolecular Engineering. His research group focuses on mathematical modeling, experimental characterization, and innovations for electrochemical technologies, including lithium metal and solid state batteries. He is the author of over 20 journal and book chapter publications, and holds over 15 patents.

Traditional Academia and R&D

Kiran Sasikumar, Research Scientist, Avant-garde Materials Simulation, Germany

Kiran Sasikumar received his Ph.D. in Materials Science and Engineering from Rensselaer Polytechnic Institute (NY, USA) in 2014. As a computational materials scientist his research interests span crystal structure prediction, thermodynamics, nucleation and phase change, heat transfer, and reaction kinetics in nanomaterials. He is currently a research scientist and scientific software developer at Avant-garde Materials Simulation (Germany), where he develops tools for extensive sampling of potential energy landscapes and improve the accuracy of organic crystal structure prediction (CSP). Further, he uses specialized CSP workflows for virtual polymorph screening, integrated with structure solution from experimental powder diffraction data, in order to support small molecule solid-form selection during early stage drug development research at pharmaceutical companies. Before joining Avant-garde Materials Simulation, he was a postdoctoral researcher at the Center for Nanoscale Materials in Argonne National Laboratory. At Argonne, he worked on investigating materials interaction at nanoscale interfaces and surfaces using a wide variety of tools, including coherent diffractive imaging, molecular dynamics, and continuum finite element simulations.

Matt Smylie, Assistant Professor of Physics, Hofstra University

Dr. Matt Smylie received his Ph.D. in Physics at the University of Notre Dame. Following this, he was a postdoctoral researcher in MSD at Argonne, then an ND Energy Postdoctoral Fellow at Notre Dame. He started a tenure-track position at Hofstra University in the Department of Physics and Astronomy in 2018, where he has built a low-temperature research lab. His research interests include topological and ferromagnetic superconductors.

Sarah Phan-Budd, Associate Professor of Physics, Winona State University

I originally trained as an experimental particle physicist, obtaining my PhD from the University of Illinois Urbana-Champaign in 2008. My graduate thesis was on the discovery of single-top quark production at CDF. I did a short stint as a visiting assistant professor at the University of Redlands before taking a postdoctoral position at Argonne from 2009-20012. At Argonne, I studied neutrino oscillation and was a member of the NOvA and MINOS collaborations. In 2012, I was hired as a tenure track faculty member at Winona State University, where I was granted tenure in 2017. In addition to particle physics, I am also interested in physics education research, analysis of large datasets, equity and inclusion in physics and the development of our new general engineering program at Winona State.

Santanu Chaudhuri, Professor of Materials Engineering, University of Illinois at Chicago
Director of Manufacturing Science and Engineering, Argonne National Laboratory

Dr. Santanu Chaudhuri is the Director of Manufacturing Science and Engineering in the Argonne National Laboratory. He is also a Professor of Materials Engineering at University of Illinois - Chicago with a courtesy appointment in the Chemical Engineering and Electrical & Computer Engineering Departments. Dr. Chaudhuri moved to Argonne and UIC in 2017 after serving as Associate Director in the Applied Research Institute in the University of Illinois at Urbana-Champaign College of Engineering. Dr. Chaudhuri's academic research group, *Accelerated Materials Research Laboratory* (AMRL webite: http://amrl.uic.edu), is working on advancing first-principles theory and molecular dynamics methods for developing multiscale science approach for structure-property-processing relations for materials design and manufacturing processes. Dr. Chaudhuri's team develops atomistic and mesoscale simulation tools for high-performance computing environment. Use of materials databases, high-throughput DFT, molecular dynamics, reaction engineering, and computational workflow to inform machine learning models are currently the focus of multiple ongoing projects in the group.

Badri Narayanan, Assistant Professor of Mechanical Engineering
University of Louisville

Dr. Badri Narayanan is an Assistant Professor of Mechanical Engineering at University of Louisville (since August 2018), where he leads the Predictive Materials Modeling Laboratory (PMML). Previously, he has held appointments at Argonne National Laboratory as Assistant Materials Scientist (2016-2018), and postdoctoral associate (2014-2016). His group is interested in utilizing multi-scale materials modeling, data-science, and machine learning to accelerate discovery/design of functional materials and interfaces relevant for applications in energy storage, neuromorphic computing, catalysis and nano-electronics. He is currently leading multi-year research projects on solid-state batteries, quantum materials, and electrochemical interfaces funded by Department of Energy and Lam Research Inc. He is the recipient of Ralph. E. Powe Junior Faculty Enhancement award from Oak Ridge Associated Universities (2020) and Early Career High Impact Science Achievement Award from the National Energy Research Scientific Computing Center (2017). Dr. Narayanan earned Ph.D. in Materials Science from Colorado School of Mines, USA in 2013, a Master's degree in Materials Engineering from Indian Institute of Science in 2008, and a Bachelor's degree in Metallurgical and Materials Engineering from National Institute of Technology Karnataka, India in 2006.

Non-traditional Careers

Aaron Fluitt, Senior Manager, Initiative Development, Argonne National Lab

Aaron serves as Senior Manager, Initiative Development, at Argonne National Laboratory. In this role, he leads program development activities and develops new science and technology initiatives in energy and global security, through engagement of key internal and external stakeholders, collaborators, and partners. He joined Argonne in 2017 from the Boston Consulting Group (BCG), where he worked with senior leaders of client organizations to develop and implement strategies for growth, innovation, and efficiency. He earned his Ph.D. in the Institute for Molecular Engineering at the University of Chicago. He also holds an M.S. from the University of Wisconsin-Madison and a B.S. with highest distinction from the University of Nebraska-Lincoln, both in chemical engineering.

Aeraj Haque, Senior Patent Agent/Patent Law Clerk, Foley & Lardner LLP

Aeraj U. Haque, Ph.D. is a patent agent with Foley & Lardner LLP. He offers an interdisciplinary background in mechanical, agricultural, and biological engineering, having a broad knowledge of business development, entrepreneurship, patent analysis, patent drafting, and IP due diligence. He is extensively experienced in biomedical diagnostic technologies, agricultural technologies, biosensors, nanotechnology, detectors, microfluidics, battery technologies, medical devices, automotive technologies, including emission systems and power generation. Dr. Haque is a member of the Mechanical & Electromechanical Technologies Practice. Prior to joining Foley, Dr. Haque has held positions as patent technical advisor and patent technical specialist with an international law firm. In 2016, Dr. Haque co-founded and assumed the position of chief technologist at Argonne National Laboratory. In 2012, Dr. Haque co-founded and served as CTO of Site-Diagnostics, a veterinary diagnostics company. Dr. Haque earned his doctorate in agricultural and biological bioengineering from Purdue University (Ph.D., 2010), where he also completed his master's degree in mechanical engineering (M.S., 2006). Dr. Haque received his undergraduate degree in mechanical engineering from NED University (B.S., 2002).

Jenny Morber, Freelance Science Writer and Journalist

Jenny Morber holds a B.S. and Ph.D. in materials science and engineering from the Georgia Institute of Technology where her research focused on developing magnetic nanomaterials for applications in localized cancer therapies. She now is a professional freelance science writer and journalist based out of the Pacific Northwest. Dr. Morber has written about science and the people who do it for academic publications, science magazines, children's magazines, and women's magazines, spanning a diverse array of subjects. Her recent work includes an investigative report on scientists forced out or sidelined under the Trump administration, an exploration into the size of a blue whale's anus, and an article on naming hurricanes for Scholastic Math magazine.

Joseph Barforoush, Co-founder and Chief Technology Officer, Avium LLC

Dr. Barforoush is a graduate of the Leonard group at the University of Kansas with extensive experience in the development of catalysts and novel electrochemical reactors. He is PI on Avium's NSF SBIR/STTR Phase I/II awards, and he was Entrepreneurial Lead on a National NSF I-Corps grant to determine customer interest in Avium distributed hydrogen generators (water electrolyzers).

James Dolan, Science Communicator, King's College, University of Cambridge, UK

James' scientific research lies at the intersection of nanophotonics and soft matter physics. He investigates ways to use the (directed) self-assembly of liquid crystals and block copolymers to create two- and three-dimensional optical metasurfaces and metamaterials with dynamically reconfigurable optical properties. With respect to science communication, James is interested in how scientists' conceptions of science—what it is, how it works, and what it's for—affect how and why they communicate their science, and in reconciling the lived experience of scientist-communicators with the scholarly literature on science communication. In particular, he focuses on science communication with policy makers, working in collaboration with the Centre for Science and Policy (CSaP); and, together with students in Cambridge and collaborators at Brunel University London, investigates improv comedy as a novel public engagement technique.

David Nackashi, Chief Executive Officer, Protochips

Trained as an Electrical Engineer, David is the CEO and co-founded Protochips after struggling with gathering data during his graduate research. He and his colleagues often wished they would visualize processes at a very small scale, taking advantage of electron microscopy. He cofounded Protochips which commercialized MEMS technology, revolutionizing in situ microscopy. His passion today is applying modern machine vision technologies to the field in order to greatly increase the quality and throughput of research.

David obtained a bachelors degree in Electrical Engineering from the Georgia Institute of Technology, and a Masters and Ph.D. in Electrical Engineering from North Carolina State University. He worked for Alcatel Telecom early in his career and cofounded Protochips shortly after completing his graduate work.

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MATERIALS SCIENCE

1
Conductivity of Nanoporous Indium Oxide Derived from Sequential Infiltration Synthesis

Aaron D. Taggart[1], Nari Jeon[1], Vepa Rozyyev[2], Evguenia Karapetrova[3], Nestor J. Zaluzec[4], Ruben Z. Waldman[5]

[1]Materials Science Division, Argonne National Laboratory, Lemont, Illinois 60439
[2]Pritzker School of Molecular Engineering, The University of Chicago, Chicago, Illinois 60637
[3]Advanced Photon Source, Argonne National Laboratory, Lemont, Illinois 60439
[4]Photon Sciences, Argonne National Laboratory, Lemont, Illinois 60439
[5]Chemical Sciences and Engineering, Argonne National Laboratory, Lemont, Illinois 60439

Sequential infiltration synthesis (SIS) is derivative of atomic layer deposition (ALD) that expands the morphological palate of materials to enable an even greater variety of applications. During SIS, a vapor phase precursor reversibly and selectively adducts with a polymer matrix. By controlling the polymer moieties, precursor chemistry, and reaction parameters such as time, temperature, and pressure, the process can yield precise deposition and high growth rates (equivalent to > 30 nm/cycle) in complex matrices that are 100s of nanometers thick. In our work, we leverage those parameters to deposit nano-porous In_2O_3 electrodes for water remediation. Electrified water treatment has the potential to greatly improve energy and cost efficiency over current methods. SIS has been used previously to create well-defined nano-porous structures, yet there has not been a report of an electrode developed *via* SIS. Our work describes how amorphous $InOH_x$ clusters deposited by SIS combine to form an interconnected and electrically conductive network of crystalline In_2O_3. Changes in optical parameters, probed by spectroscopic ellipsometry, reveal that the oxygen annealing burns off the polymer and leads to partial densification of the film. XRD and TEM further demonstrate the development of a crystalline network of In_2O_3 with grain sizes of approximately 20 nm. Additional annealing in forming gas yields resistivity values on the order of 10^{-1} Ω*cm. Aside from the annealing conditions, the microstructure of the In_2O_3 network also influences electrical properties. Using SIS to control the volume fraction of In_2O_3 in PMMA, annealed samples were prepared with porosity ranging from 30-80% In_2O_3. As the amount of In_2O_3 increases, Hall measurements reveal that the resistivity decreases and the mobility increases, which suggests that the prevalence of charge conduction pathways within the In_2O_3 film can be controlled. We demonstrate nano-porous In_2O_3 films with tunable electrical properties and further highlight the opportunity to precisely control properties of In_2O_3 for applications such as water remediation.

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In Situ Pyroelectric Calorimetry of Atomic Layer Deposition Reactions

Ashley R. Bielinski[1], Emily A. Sprague-Klein[2], Brian T. Phelan[2], Alex B. F. Martinson[1]

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[2]Chemical Sciences and Engineering Division, Argonne National Laboratory, Lemont, IL 60439

Atomic layer deposition (ALD) is a thin film deposition technique based on sequential self-limiting surface reactions between precursor vapor and a substrate. It is prized in both research and industry for applications requiring precise control of film thickness and composition and in applications requiring conformal coating of 3D nano or microstructures. The deposition process, and film nucleation in particular, are dependent on the selected precursor molecules, deposition conditions, and the substrate surface. However, ALD processes are typically empirically optimized without detailed understanding of the nucleation and reaction pathways.

Pyroelectric calorimetry for ALD was developed as an *in situ* technique for calibrated measurements of the time resolved heat generation from ALD reactions. We built pyroelectric detectors with thermal resolution of 0.1 μJ/cm^2 and temporal resolution of 50 ns. A thermal and electric model of the calorimeter was developed from the calorimeter's response to laser calibration. We demonstrated ALD calorimetry by measuring the reaction of trimethylaluminum on a hydroxylated alumina surface, which produced 96 μJ/cm^2. [1] Calorimetry will provide insight into the kinetics and mechanisms of ALD reactions enabling emerging applications such as selective defect passivation.

References

[1] Bielinski, A.R.; Sprague-Klein, E.A.; Phelan, B.T.; and Martinson, A.B.F.; *Chem. Mater.*, **33**, 15, 6176–6185 (2021).

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Low-Cobalt, High-Nickel Li-Ion Cathodes for Solid State Batteries: A Computational Exploration

Vuk Brajuskovic[1]

[1]Materials Science Division, Argonne National Laboratory, Lemont, IL 60439

The behavior of Magnetic nanostructures has long been of interest to the field of condensed matter physics. A fundamental element to understanding and controlling this behavior is the understanding and control of the underlying energy landscapes. This work has focused on the study of the energy landscape in nanoscale ferromagnetic Archimedean spirals. Nanoscale ferromagnetic Archimedean spirals show the potential for demonstrating curvature induced magnetic phenomena such as the curvature induced exchange driven Dzyaloshinskii-Morya Interaction (DMI) and curvature induced magnetic anisotropy. However, to date experimental observation of these curvature induced phenomena has been limited. This work has used the MuMax micromagnetic modeling software to show that nanoscale ferromagnetic spirals are promising candidates for the observation of curvature induced exchange driven DMI and curvature induced anisotropy.

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Low-Cobalt, High-Nickel Li-Ion Cathodes for Solid State Batteries: A Computational Exploration

Joshua J. Gabriel[1], **Juan Garcia**[2], **Noah H. Paulson**[1], **John J. Low**[3], **Marius Stan**[1], **and Hakim Iddir**[2]

[1]Applied Materials Division, Argonne National Laboratory, Lemont, IL 60439
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High-nickel cathodes such as Lithium Nickel Oxide (LNO) and Lithium Nickel Manganese Cobalt Oxide (NMC) are of interest for solid-state batteries that are important for long-range electrical vehicles. However, these materials are susceptible to large changes in the *c* lattice constant of the atomic structure, which can cause volumetric strain fracture during high state of charge (low Li content). The presentation includes recent results and ongoing efforts using innovative machine learning accelerated density functional theory (DFT) calculations to understand the effect of phase transitions in high-nickel cathodes. It also includes a discussion of the impact of dopants that substitute Li-ion and transition metal sites in the cathode atomic structure on mitigating the volumetric strain issue.

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Site-Selective Atomic Layer Deposition On Rutile TiO_2 via Selective Hydration

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Atomic layer deposition (ALD) is a commonly utilized technique for precise growth of monolayers on different substrates via self-limiting surface reactions. Despite the widespread use of ALD, few attempts have been made to extend the technique for more selective growth. With site selective ALD, materials with electronic defects could be repaired or healed by use of this technique. One pathway by which to achieve site selective ALD is to identify defects in which water will adsorb more strongly to so that the desorption temperature can be leveraged to control growth. Here we evaluate the feasibility of this strategy that enables site-selective ALD is investigated amongst four rutile TiO_2 facets and their most prevalent step edges which represent a common defect. First principles simulations were used to evaluate the adsorption free energies for molecular and dissociative adsorption of H_2O on the various substates. We predict that hydration selectivity is possible on the 110 and 001 step edges and further computationally evaluate three metalorganic ALD precursors for their compatibility with the selective hydration strategy.

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Coherent-Enhanced Dark Field Imaging for Structural Heterogeneity in Materials

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The greatly enhanced coherent X-ray flux enabled by fourth-generation facilities like upcoming APS-U will open the possibility of new ways to characterize crystalline materials when combined with established methods like lens-based dark-field X-ray microscopy (DFXM). The geometric and topological signatures of lattice heterogeneities in such materials are encoded in the Bragg-diffracted coherent wavefields which subsequently pass through a lens system and are then incident upon an area detector. These measured intensity distributions correspond to the structural information of the material in real/reciprocal phase space, when recorded between the back focal plane of the lens (Fourier-space image) and the conventional image plane (real-space). We describe initial defocus-regime simulations of such “fractionally” propagated coherent Bragg diffraction as a means to understand how to enhance signatures of the crystal defect fields in a crystalline sample. Our preliminary full-field simulation results support the feasibility of such “coherence-enhanced DFXM” for the characterization of defects by exploiting the enhanced contrast. This will ultimately provide a route for querying irreversible structural dynamics (and any coupled physics thereof) within crystalline materials, that currently eludes lensless imaging methods such as Bragg coherent diffraction imaging (BCDI).

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In Situ Lithium Ion Doping of Poly(ethylene oxide) by Reaction with Metallic Li

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Poly(ethylene oxide) (PEO) is the most popular polymer for solid-state lithium metal battery electrolytes. While many studies focus on increasing PEO ionic conductivity through doping with lithium salts, little work has addressed using PEO and Li directly to generate Li^+-conducting species in situ. Here, electrochemical impedance spectroscopy shows the ionic conductivity of PEO thin films increases up to three orders of magnitude (from 10^{-7} S cm^{-1} to 10^{-4} S cm^{-1}) when contacted with Li at elevated temperature. This is due to the reduction of ether bonds, which produces lithium alkoxides that are responsible for Li^+ transport. Density functional theory analysis confirms this mechanism as thermodynamically favorable. X-ray photoelectron spectroscopy also shows the presence of organolithium species which are responsible for propagating reactions with PEO. Preliminary work with free-standing PEO electrolytes and Li electrodes shows promising results for in situ doping at larger scale. This work clarifies the underlying mechanisms of Li-polymer electrolyte reactions and presents new pathways for in situ Li^+-doping of polymer electrolytes.

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Fermi surface topology of Kagome lattice superconductor CsV_3Sb_5 explored through de-Haas van Alphen oscillations

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Topological materials with unique quantum mechanical properties such as topological insulators and topological superconductors are at the research frontier in condensed matter physics. Among these quantum materials, the recently discovered Kagome lattice CsV_3Sb_5 presents a wide variety of intriguing properties such as superconductivity, charge density wave, and a non-trivial electronic topology. We have investigated its normal-state electronic properties via magnetic torque measurements in fields up to 35 Tesla. Analyzing the de-Haas van Alphen oscillations with H // c direction, seven distinct frequencies are identified: F_α = 27 T, F_β = 98 T, $F\gamma$ = 223 T, F_η = 711 T, F_ξ = 848 T, $F_{\tilde{\delta}}$ = 1570 T, F_ϑ = 2000 T. Using the Lifshitz-Kosevich equation to fit the experimental data, the effective masses are obtained with $m_\alpha^* = 0.13(5)m_0$, $m_\beta^* = 0.24(6)m_0$, $m_\gamma^* = 0.46(2)m_0$, $m_\eta^* = 0.54(7)m_0$, $m_\xi^* = 0.63(8)m_0$, $m_{\tilde{\delta}}^* = 0.35(3)m_0$, $m_\vartheta^* = 0.27(5)m_0$ (m_0 is the free electron mass). By constructing the Landau fan diagram for each band, we extract the Berry phase, which is non-trivial for the α, β and γ bands. These experimental findings are consistent with the theoretical predictions which that CsV_3Sb_5 is a potential platform to explore topological superconductivity.

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Effects of Salt Aggregation in Perfluoroether Electrolytes

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Electrolytes comprised of polymers mixed with salts have great potential for enabling the use of Li metal anodes in batteries for increased safety. Ionic conductivity is one of the key performance metrics of these polymer electrolytes and achieving high room-temperature conductivity remains a challenge to date. For a bottom-up design of the polymer electrolytes, we must first understand how the structure of polyelectrolytes on a molecular level determines its properties. Here, we use classical molecular dynamics to study the solvation structure and ion diffusion in electrolytes composed of a short-chain perfluoroether with LiFSI or LiTFSI salts. Density functional theory is also used to provide some insights into the structures and energies of the salt interactions with the perfluoroether. We observe the formation of aggregates of salts in the fluorinated systems even at low salt concentrations. The fluorine-fluorine attraction in the solvent is the governing factor for creating the salt aggregates. The aggregates' size and lifetime change with concentration and anion. These simulations provide an insight into the structure and dynamics of perfluoroether based electrolytes that can be used to improve Li-ion batteries.

ENERGY

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Multi-cycle LES of Motored Sandia DISI Engine Using Nek5000

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Commercial software have limitations in the predictive capabilities and massive parallelization in internal combustion engine (ICE) simulations. By leveraging Argonne's massively parallel computing platform, Theta, our group has used an open source high-order spectral element code, Nek5000, to perform multi-cycle large-eddy simulations (LES) of internal combustion engines. This code has demonstrated high scalability on a variety of leadership computing platforms with minimal numerical errors. Current work has focused on performing multi-cycle LES of motored Sandia direct injection spark ignition (DISI) engine which features complex geometries including pent-roof, spark-plug and piston surface during compression and expansion strokes. The high-fidelity simulation data will be used to understand the dynamics of the in-cylinder flow and investigate the cycle-to-cycle variabilities. It enables us to improve wall-modeling techniques and develop data-driven models for future exascale platforms.

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Multi-cycle LES of Motored Sandia DISI Engine Using Nek5000

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A novel open-source chemistry model for OpenFOAM is introduced to speed-up the reactive computational fluid dynamics (CFD) simulations using finite-rate chemistry. First, a dynamic load balancing model called DLBFoam is introduced to balance the chemistry load during runtime in parallel simulations. In addition, the solution of the cell-based chemistry problem is improved by utilizing an analytical Jacobian using an open-source library called pyJac and improved linear algebra and matrix operations. Combined effect of these models yields a speed-up factor of 200 for a high-fidelity large-eddy simulation spray combustion case compared to the standard OpenFOAM implementation without compromising the solution accuracy.

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Projection-based Methods for Spectral Analysis of Data without Temporal Information

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Extracting coherent structures and physically important features from complex systems has become the subject of great interest in fluid dynamics, motivated and enabled by a variety of modal analysis techniques. The availability of spatiotemporally resolved computational and experimental data enables the use of techniques such as dynamic mode decomposition and spectral proper orthogonal decomposition to isolate and analyze structures that correspond to specific temporal frequencies. However, in many scenarios time-resolved data remains challenging to acquire and store. Here, we explore how spectral content may be recovered from spatially-resolved data in the absence of any temporal information. This is achieved by analyzing the spectral content of reduced-order models identified from projecting the governing equations onto a subspace identified from the original data via proper orthogonal decomposition, through either a pseudospectral analysis of the identified linearized system or by considering the spectral proper orthogonal decomposition of time-resolved data generated by evolving nonlinear reduced-order models. We explore the method performance for example problems, ranging from systems with a single dominant frequency to those with broadband frequency content. We find that the method can identify spectral content even when the identified reduced-order models have limited accuracy in predicting the time-evolution of the system dynamics.

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Development of Predictive Ignition Models for Spark-Ignition (SI) Engine Applications

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Carbon dioxide emission reduction to curb the effects of climate change on the world's economy, security, and ecosystems, is at the forefront of research. In the United States, the transportation sector is the largest emitter of CO_2 with light-duty vehicles (LDVs) contributing the most. While the electrification of LDVs promises significant CO_2 reductions, improvements in SI engine efficiency are still necessary. Ignition is key for reliable combustion, especially in strenuous operating conditions that promise engine efficiency gains but require advanced ignition system design. The development of predictive ignition models is critical to the success of such efforts. Beyond LDVs, ignition has applications in jet engines, hybrid vehicles, and off-road engines. In this work, an advanced ignition model is developed and coupled with combustion models for computational engine research. The ignition model utilizes a local velocity-based algorithm to track the movement of the spark channel and deposit energy. The ignition model is then equipped with additional capabilities to model the secondary electrical circuit and predict the occurrence of short-circuits, blowouts, and restrikes. Computational results highlighting the development and operation of the ignition model in engine and non-engine geometries are compared to experimental results. Finally, future direction for this research is discussed.

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Vehicle-cycle and life-cycle analysis of Class 6 vocational trucks: Conventional, hybrid and electric

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Road-based freight transport is a major contributor to greenhouse gas (GHG) and pollutant emissions within the US transportation sector. This has led to increased interest and development of energy-efficient technologies and alternative powertrains for freight vehicles, driven by the push for newer, more stringent environmental regulations. Medium-duty trucks constitute a key component of this sector, with their importance having increased during the CoVID-19 pandemic with the increasing direct-to-consumer delivery of goods. Here, we conduct life-cycle analysis of Class 6 pickup-and-delivery (PnD) trucks across three powertrains: an internal combustion engine (ICE, diesel), a hybrid powertrain with an ICE as its primary power source, and a battery-electric. Energy use and GHG emissions are calculated for all trucks over their vehicle-cycle (vehicle manufacturing, fluids, and end-of-life) and fuel-cycle (well-to-wheels) using Argonne's GREET® (Greenhouse gases, Regulated Emissions, and Energy use in Technologies) model. The analysis identifies major contributors to GHG emissions for each PnD truck, with a focus on steps needed to further reduce their environmental impacts. Our results show the relevance of material composition and the nature of energy source on vehicle-cycle and fuel-cycle effects respectively, regardless of the propulsion technology employed.

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Non-linear Increase in the Octane Rating for Alcohol Biofuel Blends at MON like Conditions

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Research and motor octane numbers (RON and MON) do not adequately predict the reactivity of fuels in lean advanced compression ignition (ACI) conditions. Homogeneous charge compression ignition (HCCI) fuel ratings at high-temperature equivalent to MON on the CFR octane engine have correlated well with fuel reactivity on modern ACI and multi-mode engines. Therefore, this study was aimed to identify the reactivity blending characteristics of biofuel blends in lean high-temperature ACI combustion. The base gasoline of RD587, before ethanol addition, (RD587BOB) was selected as a base fuel and blended with 10 high-octane biofuels. The experiments were performed at high-temperature (HT: 600 rpm, 1.0 bar, 150°C) conditions using an instrumented Cooperative Fuel Research (CFR) octane engine. The HCCI octane number of several mid-level blends (20-30 v%) were found to be higher than the base fuel (RD587BOB) and the neat high octane biofuels, indicating a "hyper-boosting effect". For some of the bio-blends, synergistic non-linear blending was observed and the characteristics were a function of combustion mode. This work shows that with minor modifications, the widely distributed CFR octane engine can be used to rate fuels for future lean high efficiency ACI engines.

CHEMICAL SCIENCES

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Molecular Cobalt Catalysts for Electrochemical CO_2 Reduction: Exploring the Effect of Second-Sphere Functionalities on Catalysis

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The world's reliance on fossil fuels has led to increasing atmospheric CO_2 concentrations associated with climate change and ocean acidification. With more effective catalysts, CO_2 could instead be recycled back into energy-rich fuels or commodity chemicals to close the carbon cycle. Carefully designed ligands can dictate the geometry, stability, and reactivity of the metal active site for CO_2 activation and conversion. Recently our group has reported two molecular Cobalt catalysts with redox active bipyridine framework for hydrogen evolution.[1] The two Cobalt catalysts systems are only different by one amino group, which makes one an open "macrocyclic" catalyst and the other as a closed macrocyclic system. In this work, the "open" Co(II) complex has been repurposed to use it as a catalyst for electrocatalytic CO_2 conversion to value-added products. Analogous Co(II) complexes have been synthesized with introducing amine, pyridine, methyl pyridine etc. as second-coordination sphere functional groups and the catalytic activities of them have been assessed through electrochemical studies.

References

[1] Kohler L.; Niklas, J.; Johnson, R.C.; Zeller, M.; Polektov, O.G.; and Karen Mulfort, *Inorg. Chem.*, **58**, 2, 1697–1709 (2019).

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Nature Inspired Small Molecule Redoxmer For Redox Flow Batteries

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Renewable energy sources such as solar and wind energy are making substantial inroads into the global energy supply as environment-friendly alternatives. To accommodate those renewable resources, grid-scale energy storage technologies have drawn enormous amounts of interest, with redox flow batteries (RFBs) among the leaders in performance. RFBs uses liquid solutions of redox-active chemicals (redoxmers) as energy storage media, rather than the solid-state electrode materials found in conventional batteries. Due to the liquid nature, RFBs feature remarkable flexibilities, including decoupled energy and power that is highly desired for adapting various scales of energy storage requirements. Non-aqueous RFB systems are extensively studied to achieve high voltage and high energy dense batteries. To that end, developing redoxmers with low molecular weight, high solubility in non-aqueous solvents and high stability are actively pursed. Design of redox-active materials based on natural products allows for the development of sustainable and potentially environment-friendly redoxmers for RFBs. Here, we describe the development of a small molecule bipolar redoxmer based on a natural product. We show that a simple modification of the redoxmer leads to high stability in galvanostatic H-cell cycling and high solubility in non-aqueous solvents. The identification and molecular engineering of non-aqueous compatible natural product based redoxmers represent a promising strategy for developing sustainable redoxmers for RFBs.

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Dramatic Conformer-Dependent Reactivity of Acetaldehyde Oxide Criegee Intermediate with Dimethylamine via a 1,2-Insertion Mechanism

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The reactivity of carbonyl oxides has previously been shown to exhibit strong conformer and substituent dependencies. Multiplexed photoionization mass spectrometry experiments (298 K, 4 Torr) and high-level theoretical calculations demonstrate that the reaction of acetaldehyde oxide (CH_3CHOO) with dimethyl amine (DMA) is highly conformer dependent and leads to the formation of an amine-functionalized hydroperoxide product via a 1,2-insertion mechanism. The rate coefficient of DMA with *anti*-CH_3CHOO is predicted to exceed that for the reaction with *syn*-CH_3CHOO by a factor of ~34,000, which is attributed to submerged barrier (*syn*) vs. barrierless (*anti*) mechanisms for energetically downhill reactions.

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The Importance of Surface Oxygen for Lithiation and Morphology Evolution During Calcination of High-Nickel NMC Cathodes

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Nanoscale morphology has a direct impact on the performance of materials for electrochemical energy storage. Despite this importance, little is known about the evolution of primary particle morphology nor its effect on chemical pathways during synthesis. In this study, operando characterization is combined with atomic-scale and continuum simulations to clarify the relationship between morphology of cathode primary particles and their lithiation during calcination of nickel-rich (NMC-811). This combined approach reveals a key role for surface oxygen adsorption in facilitating the lithiation reaction by promoting metal diffusion and oxidation, and simultaneously providing surface sites for lithium insertion. Furthermore, oxygen surface termination is shown to increase the activation energy for sintering, leading to smaller primary particle sizes at intermediate temperatures. Smaller particles provide both shorter diffusion lengths for lithium incorporation and increased surface site density for lithium insertion. These insights provide a foundation for more tailored syntheses of cathode materials with optimized performance characteristics.

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Electrochemical and Solvation Properties of a Perfluoroalkoxyaluminate-Calcium Salt

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Calcium-ion batteries (CIBs), like other multivalent systems, are promising alternatives to current lithium-ion technology as they hold advantages of high theoretic energy density, comparable reductive potential, and pose less safety concerns. Particularly useful to CIBs is the ability to use a solid anode. Unfortunately, the field of viable robust electrolytes that display room temperature electrochemical plating and stripping remains small. This is partially due to the ease of calcium surface passivation. Critical to understanding reactivity at the anode is the solvent-ion interactions in solution.[1-3] In this work, a new calcium tetrakis(perfluoro-tert-butoxy) aluminate salt (CaTPFA) was developed and has shown reversible electrochemical plating at room temperature. Computational analysis and X-ray spectroscopy was used to probe solvent-salt coordination to better understand interactions that help and hinder electrolyte activity.

References

[1] Hahn, N. T.; Driscoll, D. M.; Yu, Z.; Sterbinsky, G. E.; Cheng, L.; Balasubramanian, M.; Zavadil, K. R.; *ACS Appl. Energy Mater.*, **3**, 8437–8447 (2020).
[2] Connell, J. G.; Zorko, M.; Agarwal, G.; Yang, M.; Liao, C.; Assary, R. S.; Strmcnik, D.; Markovic, N. M.; *ACS Appl. Mater. Interfaces*, **12**, 36137–36147 (2020).

[3] Lau, K.-C.; Seguin, T. J.; Carino, E. V.; Hahn, N. T.; Connell, J. G.; Ingram, B. J.; Persson, K. A.; Zavadil, K. R.; Liao, C.; *J. Electrochem. Soc.*, **166**, A1510–A1519 (2019).

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Theoretical Investigation on CO_2 Conversion to Ethanol by Clusters Formed from Atomically Dispersed Copper

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Direct electrochemical conversion of CO_2 to ethanol offers a promising strategy to lower CO_2 emissions while storing energy from renewable electricity. However, the high selectivity for producing ethanol using current electrocatalyst is rather difficult. Our recent study reported that a synthesized carbon-supported copper (Cu) catalyst achieves a single-product Faradaic efficiency (FE) of 91% at -0.7 onset potential as low as -0.4 V for ethanol. Furthermore, operando X-ray absorption spectroscopy was carried out and identified a transformation from atomically dispersed Cu atoms to Cu_n clusters during the reaction. Therefore, density functional theory (DFT) calculations were carried out to study the reaction mechanism and reveal the active site structures. Using the experimentally measured X-ray absorption near edge structure (XANES) as a guidance, we qualitatively reveal the structures of the pre, in-situ, and post catalyst with structure analysis of the theoretical XANES simulations, which confirms the transformation of the active site structure during the reaction. Based on the mechanistic study, we also evaluated the limiting potential of CO_2 reduction as -0.41 V and identified the rate determining step as C-C bond formation. Our computational results agree well with experimental investigation and validate our previous findings, which also provides guidance for future catalyst development.

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Orientation of Thiocyanate Anions (SCN^-) at Positively Charged Air/aqueous Interface

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The organization of anions at air/aqueous interfaces plays significant roles in various chemical, environmental, and biological processes. The interfacial behavior of background anions has a significant impact on the extraction efficiency and selectivity of metal ions in liquid-liquid extraction (LLE), for instance, the presence of thiocyanate (SCN^-) anions preferentially extract heavier lanthanides over lighter lanthanides with quaternary amine extractants. However, their molecular-scale mechanism is not well understood. Here, we use phase-sensitive vibrational sum frequency generation spectroscopy (PS-VSFG) measurements and molecular dynamics simulation study to gain insight into the interfacial distribution of thiocyanate (SCN^-) anions in the presence of positively charged 1,2-dipalmitoyl-3-trimethylammonium-propane (DPTAP) monolayer. The conventional VSFG measurement shows an increase in CN-stretch band intensity with increasing bulk SCN^- concentration. But the intensity decreases when the bulk concentration is greater than 1 M. The imaginary part of the second-order nonlinear susceptibility (Im $\chi^{(2)}$) spectra obtained from PS-VSFG measurements shows a positive and negative band at concentration 1.5 M or higher, suggesting for the upward and downward orientations of anions. The complementary molecular dynamics simulation results show a broad orientational distribution of anions at 2 M compared to its distribution at lower concentrations. These results demonstrate the formation of antiparallel structures or ion-pairs of SCN^- anions having opposite

orientations at the air/aqueous interface. This study highlights the importance of investigating the fundamental interfacial behavior of background anions on a molecular scale to better understand the LLE of rare earth elements.

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Screening Multivalent Electrolyte Salt Additives for Stabilizing Silicon Anodes for Lithium-ion Batteries

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Over the past few decades, silicon anodes have gained a lot of attention for lithium-ion batteries due to their high energy density compared to conventional graphite electrodes. However, their implementation in electric vehicles has seen limited success due to volume expansion that results in the loss of electric contact and reactivity of the lithium silicides (such as Si^{4-}) with electrolyte components that results in poor cycling performance. Novel electrolyte salt additives containing multivalent cations (such as Mg and Ca) were recently found to stabilize silicon through the formation of ternary Li-Mi-Si phases that were less reactive with electrolyte constituents. Here, we present our recent efforts in extending this chemistry to screening novel multivalent cations with known ternary phases and characterizing their structure by high resolution synchrotron X-ray diffraction and elemental analysis as well as evaluation of their electrochemical performance.

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Computational Investigation of Catalytically Active Sites for a Supported Organovanadium Catalyst

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Supported organometallic catalysts (SOMC) are receiving increasing attention for the development of unique combinations of homogeneous and heterogeneous catalysts. A crucial consideration for SOMC is the nonuniformity of the active sites. Standard spectroscopic techniques, such as X-ray Absorption Near Edge Structure (XANES) spectroscopy, reflect the nature of the most populated sites, which are often intrinsically structurally distinct from the most active catalytic sites. With computational models, often, only a few representative structures are used to depict catalytic active sites on a surface even though there are numerous observable factors of surface heterogeneity that contribute to the kinetically favorable active species. To showcase the importance of modeling surface heterogeneity and its effect on catalytic activity, density functional theory (DFT) computational models of a series of potential active sites for a surface organovanadium-(III) catalyst for the reaction pathways are applied in combination with kinetic Monte Carlo (kMC) simulations. Also, an integrated computational and experimental strategy is developed to determine both primary (vanadium hydride interactions) and secondary (second-shell coordination) bonding interactions within the XANES pre-edge region using a rigorous calibration with time dependent DFT. This work will further elucidate computational techniques regarding modeling characterization and XANES simulations for SOMC.

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Combined Experimental and First-Principles Determination of the Solvation Structure of Ru Dyes

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Transition metal complexes feature synthetically-tunable photophysical and photochemical properties, making them attractive components in dye-sensitized solar/photoelectrosynthesis cells. However, the physiochemical properties of the cell emerge from the interaction between the dye and the remaining components, such as the solvent and the solid-state support. Hence, it is important to characterize the nature of these interactions in both the ground and photoexcited states of the dye, under experimental conditions. Toward this goal, we have studied the aqueous solvation structures of two prototypical Ru-polypyridyl dyes, $[Ru(bpy)_3]^{2+}$ and N3 (N3 = *cis*-bis(isothiocyanato)bis(2,2′-bipyridyl-4,4′-dicarboxylato)ruthenium(II)), via high-energy X-ray pair distribution function (PDF) measurements, conducted at the Advanced Photon Source. Analysis of the solvent background-subtracted PDF data, combined with atomistic molecular dynamics simulations, allows us to experimentally-determine the solvation structure of the Ru dyes in their ground states with sub-Ångstrom resolution. We anticipate that application of the modelling approach developed herein to photoelectrochemical cells, in combination with time-resolved X-ray scattering techniques, will ultimately provide a detailed picture of the structural dynamics of dye-sensitized solar energy conversion technologies *in operando*.

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Dimeric Redoxmers that Self-Report Flow Battery State of Health

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Increased energy storage capacity is needed for the growing renewables market. Typically, large-format batteries are proposed such as redox flow batteries (RFBs). In particular, nonaqueous RFBs have wide potential windows, enable the use of soluble organic redox-active molecules (redoxmers), and allow high energy densities. However, small molecule redoxmers are subject to crossover and capacity fade that affects the state of health of the battery. A common strategy is to engineer scaffolds with multiple redox units such as oligomers and polymers. Here, we examine the radical speciation and degradation products of dimeric redoxmers based on a diglycolamide linker attached to a fluorescent self-reporting anolyte (negative-charge storing) redoxmer.[1] When electrochemically charged, the dimers only exist as the doubly-charged form, and this dianion is extremely stable with > 2 month lifetime. A gradual decay reaction forms a pigment species with strong and distinct absorbance, fluorescent, and cyclic voltammetry that allows easy monitoring. We used this pigment to study the battery state of health *in situ*. In a battery cycling experiment, aliquots were periodically removed and tested. The pigment was easily observed by fluorimetry at very low concentration and indicated degradation could be detected before capacity fade in cycling experiments.

References
[1] Robertson, L. A.; Shkrob, I. A.; Agarwal, G.; Zhao, Y.; Yu, Z.; Assary, R. S.; Cheng, L.; Moore, J. S.; Zhang, L.; *ACS Energy Lett.*, **5**, 3062–3068 (2020).

ENGINEERING

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Towards the Development of Novel, Fluidically Pattern-Reconfigurable, Textile-Based Antenna Arrays for 5G/6G-Wearable Applications

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In this work, I will present the first pattern-reconfigurable textile antenna array using distilled and sea waters as fluidic phase shifters. This system will enable the powering of devices health monitoring devices. The sources of transmitting power will be from antennas/antenna arrays with beamforming modality for efficient wireless power transfer. This antenna array will use water-based actuators to steer the antenna's beam to efficiently collect ambient RF power. Various methods of beam steering like Rotman's lens [1], Butler matrices [1,2] have been developed before, and other methods include the use of fluidics [3] like water for the same purpose, but they were not implemented into clothing structures. The proposed antenna resonates at 1.1393 GHz and 2.0423 GHz. When using 4 quarter-cylindrical channels of fluidics like distilled and sea waters, the frequencies don't drastically change. However, the amplitude of the realized gain shifts from 1 dB to 15 dB and the phase, from -145 degrees to 139 degrees in the elevational plane. This antenna array will be able to harvest RF signals coming from multiple angles of arrival for efficient harvesting. These results can be used as guidelines to the next generation of on-clothing textile antennas/antenna arrays for 5G/6G wearable applications.

References
[1] Laxmikant, S.; Raut, H.D.; Malekar, R.; Kumar, S.; *Inventive Communication and Computational Technologies*, 589-595, (2021).
[2] Karimbu, V.A.; Rahim, M.K.A.R.; Khawaja, B.A.; Murad, N.A.; Gajibo, M.M.; *IEEE Access* (2020).
[3] Babakhani, Behrouz, B.; Sharma, S.K.; *Multifunctional Antennas and Arrays for Wireless Communication Systems*, 122-171 (2021).

ENVIRONMENTAL SCIENCE

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Effects of Land Use Change on the Hydrologic Cycle and Atmosphere of Central Argentina

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Since the 1970s, land cover in central Argentina has shifted away from perennial crops and grasses towards annual crops, largely soy. In this work we use observations and modeling to understand how this shift in land use has affected the sub-surface, surface and atmospheric fluxes of moisture and energy. We analyze the flux tower data from a paired site at Marcos Juarez in central Argentina during the period of the RELAMPAGO field campaign (2018-2019). When compared to perennial alfalfa, the observations over soy show lower evapotranspiration and specific humidity, higher sensible heat, higher outgoing shortwave radiation and soil temperature. Water table depth is shallower below the soy than the alfalfa sites. We use the Noah-MP land surface model calibrated at both soy and alfalfa sites. Long-term simulation of the calibrated model suggests that ~95% of precipitation is evaporated in the alfalfa site with negligible recharge and runoff. In the case of soy, ET is about 68% of precipitation, leaving nearly 28% for recharge and 4% for runoff. Observed increases in streamflow and decreases in water table depth over time are likely linked to shifts in land cover. Furthermore, a 250% increase in Bowen ratio (from 0.2 to 0.7) was found.

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The Biofuel Crop Switchgrass (*Panicum virgatum*) Genotype x Environment Interactions and Soil Bacterial Community Associations

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As the world transitions towards a more sustainable energy economy, switchgrass (*Panicum virgatum*) has been selected as a potential biofuel crop due to its high productivity and large genetic diversity allowing it to grow in marginal lands and across a wide range of climates. It is also a deep-rooting native perennial species that can improve soil health in degraded agricultural areas. This research aims to better understand the ecological and environmental dynamics resulting from the large-scale production of switchgrass in an agricultural setting. Thirty 6x6m stands of switchgrass (consisting of 5 replicate plots of 6 cultivars) were planted in a randomized complete block design at three sites across a latitudinal gradient (IL, MO, and TX). Differences in phenology and productivity between cultivars were measured over a 3-year period and analyzed using mixed effects models to explain the effects of site-specific climate variables on harvest yield. Additionally, soil cores were taken to measure carbon and nitrogen content, and to extract DNA for 16S rRNA sequencing to determine bacterial community structure and reveal cultivar specific associations with certain microbial taxa. This presentation will highlight preliminary results from our study and explore important next steps and future directions of switchgrass research.

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CUP of TEA: the Community Understanding Platform of Terrestrial Exchange with the Atmosphere

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The land play an important role in regulating climate via exchanges of gases (e.g., water and CO_2) and energy with the atmosphere. Measurements of land-atmosphere exchanges are performed across the globes, ultimately generating multiple databases that can be used to inform Earth System Models (ESMs). To do so, there is a need for cross-disciplinary (across Sciences performed at different scales, and across experimentalists and modelers) frameworks and platforms. This approach allows merging information between ESMs and databases dynamically, to keep analysis updated with new databases and models in real time. We are creating such a framework, with results communicated on a web platform named "CUP of TEA" (the Community Understanding Platform of Terrestrial Exchange with the Atmosphere, https://c-u-p-of-t-e-a.github.io/Home/), hosted on GitHub, with analysis performed on global databases and local land-atmosphere exchanges measurements we are taking at Argonne. Our framework consists of investigating apparent responses of land-atmosphere observed and modeled fluxes (e.g., carbon and water fluxes) at various scales (e.g., soil and ecosystem respiration) to environmental variables (e.g., soil temperature and moisture). The outcome of this project will be to better understand local and global spatiotemporal heterogeneity and scaling of land properties and land-atmosphere exchanges.

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Projection-based Methods for Spectral Analysis of Data without Temporal Information

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Extracting coherent structures and physically important features from complex systems has become the subject of great interest in fluid dynamics, motivated and enabled by a variety of modal analysis techniques. The availability of spatiotemporally resolved computational and experimental data enables the use of techniques such as dynamic mode decomposition and spectral proper orthogonal decomposition to isolate and analyze structures that correspond to specific temporal frequencies. However, in many scenarios time-resolved data remains challenging to acquire and store. Here, we explore how spectral content may be recovered from spatially-resolved data in the absence of any temporal information. This is achieved by analyzing the spectral content of reduced-order models identified from projecting the governing equations onto a subspace identified from the original data via proper orthogonal decomposition, through either a pseudospectral analysis of the identified linearized system or by considering the spectral proper orthogonal decomposition of time-resolved data generated by evolving nonlinear reduced-order models. We explore the method performance for example problems, ranging from systems with a single dominant frequency to those with broadband frequency content. We find that the method can identify spectral content even when the identified reduced-order models have limited accuracy in predicting the time-evolution of the system dynamics.

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Multi-model Ensemble Predictions of Extreme Drought based on Climate Change in the United States

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Extreme drought conditions are responsible for significant socioeconomic impacts due to extensive crop loss and wildfire. Previously, precipitation was a key indicator of drought. However, in a warming world, mean precipitation is decreasing and precipitation extremes are increasing, shifting key contributors of drought to temperature and relative humidity. This work applies statistical methods to quantify future extremes with Vapor Pressure Deficit (VPD). VPD is a measure of evaporative demand as moisture moves from the surface into the atmosphere under warmer conditions. Data for this work was produced with the Weather Research and Forecasting (WRF) model. Initial and boundary conditions for the model were supplied by CCSM4, GFDL-ESM2G, and HadGEM2-ES Global Climate Models (GCM) and based on the 8.5 Representative Concentration Pathway. The WRF model was downscaled to a 12 km resolution for three time frames: 1995-2004, 2045-2054, and 2085-2094, and Generalize Extreme Value (GEV) theory was applied. Grid dependent stationary and non-stationary GEV were calculated and assessed with the likelihood ratio test. Results show VPD and VPD return levels are increasing across the United States throughout the 21st century. Regional drought extremes are concentrated in the Midwest and Western U.S., which will likely exacerbate exhaustive water issues and wildfire risks.

MATHEMATICS & COMPUTER SCIENCE

4

Formalizing the generalization-forgetting trade-off in continual learning

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One of the critical features of an intelligent system is to continually execute tasks in a real-world environment. As a new task is revealed, we seek to efficiently adapt to a new task (improve generalization) and, in the process of generalization, we seek to remember the previous tasks (minimize catastrophic forgetting). Consequentially, there are two key challenges that must be modelled: catastrophic forgetting and generalization. Despite promising methodological advancements, there is a lack of a theoretical approach that enable analysis of these challenges. In this talk, we discuss a new theoretical approach for meta continual learning where we mathematically model the learning dynamics in the context of CL through dynamic programming. We discuss the broad applicability of our approach and show that, with this framework, we can easily derive approaches to study and address the key challenges in the CL setting.

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Multi-Fidelity Reinforcement Learning with Control Variates

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In many computational science and engineering applications, simulators with different levels of fidelity are often designed to trade-off between computational cost and accuracy. Typically, a high-fidelity simulator describes the real-world system with the highest accuracy, yet it incurs a much higher computational expense compared to a potentially inaccurate low-fidelity simulator. In this work, we study the reinforcement learning (RL) problem in presence of multiple simulators with different levels of fidelity for a target control task. A multi-fidelity estimator which exploits the cross-correlations between the low- and high- fidelity returns is proposed to reduce the variance in the estimation of the state-action value function. The proposed estimator, which is based on the method of control variates, is used to design a multi-fidelity Monte Carlo RL (MFMCRL) algorithm that improves the learning efficiency of the agent in the high-fidelity simulator. The impacts of variance reduction on policy evaluation and policy improvement are theoretically analyzed from a statistical standpoint. Our theoretical analysis and numerical experiments demonstrate that for a finite budget of high-fidelity data samples, our proposed multi-fidelity RL algorithm significantly outperforms the single-fidelity RL baseline which only uses the high-fidelity simulator in terms of policy evaluation accuracy and policy performance.

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Anticipating Frequency Excursions in Power Grid with Bayesian Decision Theory

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Disturbances in power systems, such as a generator trip, affect the frequency and voltage of the power grid. A framework that enables anticipating potentially dangerous frequency excursions (as signs of the disturbance start to reach sensors) is crucial to allow the operator more time to maintain or improve the electrical system's reliability. We propose statistical models for anticipating disturbances in the frequency response at several locations simultaneously, and we quantify the risk of a frequency excursion. After probabilistic forecasts of the power grid frequency are obtained, a Bayesian decision framework is developed to issue real-time event recommendations based on actions that minimize the risk over some loss functions predefined by stakeholders. A case study with real-time monitored data from the FNET/GridEye system shows that the traditional autoregressive models underestimate the uncertainty, leading to high chances of missing a rapid frequency drop. On the other hand, this case study reveals the value of including spatial dependence in the model. The proposed spatiotemporal model better captures the uncertainty and significantly increases the probability of identifying large frequency excursions. Results show that the spatiotemporal model can significantly reduce risks for the system operator as it consistently indicates lower costs than the ones from classical models for a wide range of loss functions.

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Improving Scientific Data Analysis with Functional Approximations

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When analyzing data sets produced by scientific simulations, it is often necessary to use discrete data points to approximate smoothly-varying quantities like derivatives, integrals, and normal vectors. These approximations can possess different levels of accuracy, can propagate errors when combined, and may even be impossible to compute for certain data set formats. Our research investigates the use of functional approximations for scientific data sets, which is a method for creating a continuous function that acts as a model for the data set. Using our functional approximation, derivatives, integrals, interpolated values, and more can be computed quickly and are exact relative to the model function. In this paradigm, scientific data sets are converted to a functional form before any analysis is done, which introduces a small but controllable error. Then, the initial data set may be discarded, and all subsequent data analysis is done relative to the model (surrogate) function without incurring further, compounding errors. This short presentation will give an overview of functional approximation with B-splines, give an example of its application to research in neutrino physics, and discuss ongoing work to improve the methodology.

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Polynomial Depth Quantum Circuits For Time Evolution Of Heisenberg Models Using The Yang-Baxter Equation

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Quantum time dynamics (QTD) is considered a promising problem for quantum supremacy[1-2] in near-term quantum computers. However the quantum circuits for QTD grows with increasing time sumulation. This study[3] focuses on simulating the time dynamics of the 1-D integrable spin chains with nearest neighbors' interactions. We show how the Yang-Baxter[4-5] equation can be exploited to compress a quantum circuit. With this compression scheme, the depth of the quantum circuit becomes independent of time step or step size and only depends on the number of spins. The compressed circuit scales quadratically with system size, which allows for the simulations of time dynamics of very large 1-D spin chains. In addition, each time steps of the simulation can run independently in parallel.

The Heisenberg model is a statistical mechanical model used to study the magnetic system critical points and phase transitions. In this model spins of the magnetic systems are treated quantum mechanically. The Hamiltonian of the model having only spin-spin interaction can be written as

$$\hat{H} = -\sum_{\alpha}\{J_{\alpha}\sum_{i=1}^{N-1}\sigma_i^{\alpha}\otimes\sigma_{i+1}^{\alpha}\}$$

where, α sums over $\{x, y, z\}$, the coupling parameters J_{α} denotes the exchange interaction between nearest neighbour spins along the α-direction, σ_i^{α} is the α-Pauli operator on i_{th} spin. Yang-Baxter equation is considered a master equation in integrable models in statistical mechanics and quantum field theory. Algebriacally it can be defined using action, $\hat{R}$, on a $V\otimes V\otimes V$, where V is a complex vector space. The action follows the equation as

$$(\hat{R} \otimes 1)(1 \otimes \hat{R})(\hat{R} \otimes 1) = (1 \otimes \hat{R})(\hat{R} \otimes 1)(1 \otimes \hat{R})$$

This unique form of commutation allows to prove the integrability of many lattice models in statistical mechanics. In this study we have utilized this identity for time dynamics operators for subclasses of 1D-Heisenberg model. As a result it allowed us to compress the quantum circuit to a polynomial depths, which only depend on system size.

References

[1] Harrow A.; Montanaro A.; *Nature*, **549**, 203-209, (2017).
[2] Zhong H.S. *et al*, *Science*, **370**, 1460-1463 (2020).
[3] Gulania, S.; Alexeev, Y.B.; Peng, Y.A.; Govind, N.; in preparation.
[4] *Exactly Solved Models in Statistical Mechanics*, R.J Baxter, Academic Press (1989).
[5] *Beautiful Models*, Bill Sutherland, World Scientific (2007).

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Customized Monte Carlo Tree Search for LLVM/Polly's Composable Loop Optimization Transformations

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Polly is the LLVM project's polyhedral loop nest optimizer. Recent user-directed loop transformation pragmas were proposed based on LLVM/Clang and Polly. The search space exposed by the transformation pragmas is a tree, wherein each node represents a specific combination of loop transformations that can be applied to the code resulting from the parent node's loop transformations. To find the best combination of these loop transformations, we have developed a search algorithm based on Monte Carlo tree search (MCTS). The algorithm consists of two phases: exploring loop transformations at different depths of the tree to identify promising regions in the tree search space and exploiting those regions by performing a local search. Moreover, a restart mechanism is used to avoid the MCTS getting trapped in a local solution. The best and worst solutions are transferred from the previous phases of the restarts to leverage the search history.
We compare our approach with breadth-first, beam, global greedy, and random search methods using PolyBench kernels and ECP proxy applications. Experimental results show that our MCTS algorithm finds pragma combinations with a speedup of 2.3x over Polly's heuristic optimizations on average.

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Rapid Assessment of Wind Energy Potential in Urban Areas: Deriving Physics-informed Data-driven Low Order Models from High Fidelity Simulations

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Distributed wind turbines are being deployed as a sustainable energy source in urban and suburban areas. Detailed, high-resolution atmospheric surface layer simulations allow for the optimization of the design and operation of wind turbines and wind farms. However, due to the high computational cost associated with the simulation of high-Reynolds number atmospheric boundary layer flows, these

approaches are impractical for the rapid/"real-time" assessment of wind energy potential. Our current study focuses on the development of a fast low order model capable to predict the complex flow pattern at the wake of buildings. Simulations results from high-Reynolds number wind flows around simple cuboids representing buildings were used to generalize analytical models by means of a new physics-informed machine learning model. Additionally, a data-driven correction function was proposed to tackle challenges associated with the prediction of the wind acceleration around buildings caused by the formation of a horseshoe vortex which simple analytical models from the literature fail to predict. The performance of the proposed physics-informed machine learning model was compared against that of a solely data-driven low order model, showing that the constrains associated to the physics-informed approach were beneficial when those models are applied in pilot field scale campaigns.

DATA SCIENCE AND LEARNING

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A data-driven and probabilistic approach for data integration, calibration, and analysis in physics-based models of cellular processes

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Physics-based dynamical modeling provides new investigative capabilities into the mechanisms of cellular processes. We investigate novel apoptosis-necroptosis regulatory crosstalk mechanisms through development and calibration (via Bayesian inference) of dynamical models of apoptotic and necroptotic signaling. Parameter robustness analysis of competing hypothetical regulatory mechanisms revealed novel crosstalk interactions i*n silico* that were subsequently corroborated *in vitro*. Model calibration of dynamical models, however, typically requires quantitative direct measurements that cellular experiments less commonly provide. Recent efforts to address the dearth of quantitative direct measurements in cellular biology have aimed to substitute nonquantitative measurements with varying degrees of success. These approaches introduce *ad hoc* assumptions and/or impair Bayesian inference methods, resulting in calibration biases that obscure model interpretation and compromise model accuracy and certainty. To address these limitations, we introduce an unbiased data-driven and probabilistic methodology that leverages nonquantitative and indirect measurements in the calibration of dynamical model of cell death. The approach employs Bayesian inference methods to estimate the contributions of nonquantitative data to mechanistic model accuracy. While far more nonquantitative measurements are needed to compensate for quantitative measurements, these measurements can have diverse sources. The data-driven representation of measurement also provides additional potential insight into the mechanistic/dynamic predictors of nonquantitative phenotypes.

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CRISPRAct: A Method for Machine Learning-Assisted Design of Secure Biosystems

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As new technologies increase availability and decrease the cost of genome modification, it becomes increasingly important to ensure that methods of containing modified organisms likewise become cheaper, easier to implement, and more effective in order to ensure safety of researchers, environment, and the general public.
We are developing a CRISPR/Cas "self-destruct" mechanism to address these needs. The mechanism utilizes a self-targeting gRNA that is inactive in a laboratory environment, but activates to induce a double-stranded break in the host genome when the cell undergoes physiological changes in response to non-laboratory environments. We are working to import the system from E. coli to other prokaryotes of interest, including Pseudomonas, Shigella, and Salmonella.
In order to identify Cas9-targetable sites likely to display this desired behavior, we have developed a machine learning model, CRISPRAct, that predicts gRNA activity using gRNA sequence and genomic context under various physiological conditions. CRISPRAct combines through polynomial regression a genomic natural language processing transformer model and a conventional neural network model to predict the log2 fold change in cell population as a proxy for gRNA activity. On a set of 71,228 E. coli gRNAs, CRISPRAct had a Mean Absolute Error of 0.62 and Spearman Correlation Coefficient of 46.8%.

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Distilling Prevalent Modes of Protein-Protein Association via Machine Learning

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In approximately 30% of human cancers, mutations identified in the RAS protein are implicated in uncontrolled cell division and growth via over-activation of a molecular pathway. But RAS is essentially "undruggable" due to lack of viable drug binding sites. Hence identifying alternative drug intervention opportunities in the cellular environment of RAS have been of interest. One such opportunity is exploration of membrane bound RAS-RAS complexes because this complex is hypothesized to be important in the activation of the pathway. Attempts have been made to understand how RAS associate with each other but these involve predetermined RAS-RAS interfaces informed by human intuition that risk being biased and limited in diversity. To overcome this drawback, we have developed an automated machine learning based framework that is applied to understand the prevalent modes of RAS-RAS association without resorting to predetermined interfaces. The framework works on several coarse-grained molecular dynamics simulations where two RAS came in contact while being bound to a complex membrane of varied compositions. The framework identifies the prevalent protein-protein conformations in an unsupervised manner. This involves performing nonlinear dimensionality reduction by training autoencoders at scale followed by clustering analysis in the low dimensional space informed by a physics based quality metric to determine the optimal number of clusters. The framework processed ~1 million molecular dynamics frames involving ~1 terabyte of data and distilled out three prevalent modes of RAS-RAS association.

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Accelerated Sampling Techniques for Lattice Gauge Theory

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We describe recent developments in reducing the computational cost of Markov Chain Monte Carlo techniques for simulations in lattice gauge theory. By training a series of neural networks that generalize the traditional update step, we are able to generate independent configurations more efficiently than current methods.

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Throughput-oriented and Accuracy-aware DNN Training with Mixed Precision

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Deep neural networks (DNNs) have transformed the field of artificial intelligence and achieved extraordinary success in cognitive tasks such as image and speech recognition. Meanwhile, the training of large DNNs is commonly compute- and memory-intensive, which has inspired optimizations for a new training approach for this application. Among them, mixed-precision is a typical and widely used technique to accelerate DNN training and reduce memory requirements. However, applying mixed precision to all involved operations in DNN training is not appropriate due to its inherent overhead, and the use of mixed precision in some operations not only does not improve overall throughput but can even hurt model accuracy. Furthermore, due to the differences in hardware architecture, such as GPUs of different generations or from different vendors, it is tough to take full advantage of the mixed-precision performance by deploying a fixed approach across all hardware. In this work, we introduce the throughput-oriented and accuracy-aware framework which automatically determines the optimization strategy of mixed precision, and we also take the hardware utilization and the rate of convergence in the design scope. We evaluate our framework on three GPUs, and the results show that our framework can perform better than other state-of-the-art approaches.

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Dataflow Architectures to Accelerate ML Workloads

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Machine Learning(ML) workloads depend on huge amounts of data, compute, and effective mapping on the underlying architectures to exploit their full potential. So far, GPUs have played an important role in keeping up with this increased demand. However, huge data movement costs, resulting in higher power requirements make GPUs non-ideal moving forward. To keep up with Moore's law as well as address this increase of compute and data demand, computer architects are moving towards design of domain-specific architectures to accelerate ML workloads.

AI accelerator is a high-performance parallel architecture that is specifically designed for the efficient processing of AI workloads like neural networks. Dataflow architectures are coming up as the preferred choice for many of these accelerators where data movement is optimized by exploiting data reuse and efficient mapping of ML workloads to these architectures.
Though the dataflow model is the common underlying guiding principle for these architectures, they differ in their approach resulting in performance tradeoffs. We study these tradeoffs and performance implications for various ML workloads in context of science applications of interest that align with DOE mission. We present a brief overview of the various AI testbeds at ALCF and an overview of the preliminary performance studies.

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Online Learning of Turbulence Closure Models with SmartSim

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Large eddy simulation (LES), which requires modeling of the sub-grid stress (SGS) tensor, can offer a compromise between accuracy and efficiency of numerical computations of turbulence flows. Data-driven approaches, such as neural networks (NN), have recently emerged and present encouraging results for improved predictive capacity over traditional models. However, these NN models must be trained on instantaneous high-fidelity turbulent data. While this is feasible for smaller computations, learning from complex flows at higher Reynolds number requires multi-terabyte databases. The work presented offers a solution to this limitation by performing online learning, wherein the NN model is trained concurrently with the flow simulation producing the data thus eliminating the need to store large training datasets on disk. The flow solver PHASTA is combined with a data parallel machine learning algorithm through the SmartSim software package, which enables the deployment of a distributed in-memory database and provides API that facilitate data streaming between the applications and database. This talk covers the implementation of the proposed software infrastructure on the Theta system, its performance at increasingly larger scale, and its application to online learning of a SGS NN model from forced homogeneous isotropic turbulence flow.

PHYSICS & HIGH ENERGY PHYSICS

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Observation of the WWW Production in p-p Collision at √s = 13 TeV with the ATLAS Detector

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WWW production was recently observed by the ATLAS collaboration using the full Run II data set with an integrated luminosity of 139 fb^{-1} at √s = 13 TeV. These first observation and cross-section measurements are presented. Measurements are performed in two final states. Events with two same-sign electrons or muons in association with two jets as well as events with three charged leptons with no same flavor opposite sign lepton pairs are selected. Machine learning techniques are used to improve the signal sensitivity by training different kinematic variables separately for each channel. Triboson

WWW production is observed with a significance of 8.2σ, where the expectation is 5.4σ. The inclusive WWW production cross-section is measured to be 850 ± 100 (stat.) ± 80 (syst.) fb.

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Model-Independent Searches for New Physics in Multi-Body Invariant Masses

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Model-independent searches for physics beyond the Standard Model typically focus on invariant masses of two objects (jets, leptons or photons). In this study we explore opportunities for similar model-agnostic searches in multi-body invariant masses. In particular, we focus on the situations when new physics can be observed in a model-independent way in three- and four-body invariant masses of jets and leptons. Such searches may have good prospects in finding new physics in the situations when two-body invariant masses, that have been extensively explored at collider experiments in the past, cannot provide sufficient signatures for experimental observations.

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Extraction Of Transverse Momentum Distributions Up To N3LL From Drell-Yan Data

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One of the main aims of hadronic physics is to describe the internal structure of the nucleon in terms of its constituents, quarks and gluons. A lot of information has been collected over the past forty years concerning the distribution of partons in one dimension, encoded in the well–known collinear Parton Distribution Functions (PDFs). In the last years, we are extending the study to the distribution of partons in full three–dimensional momentum space, encoded in the so–called Transverse Momentum Distributions (TMDs).
We present an extraction of unpolarized Transverse-Momentum Dependent (TMD) Parton Distribution Functions (PDFs) based on a fit on data from Drell-Yan processes in different experiments and kinematic ranges, including in particular LHC experiments. The analysis is performed in the TMD factorization framework and reaches the unprecedented perturbative accuracy of Next–to–Next–to–Next–to–Leading Logarithm (N3LL).
We obtained a very good description of both the shape and the normalization of the data without introducing normalization coefficients as it was done in the literature before.

NANOSCIENCE AND TECHNOLOGY

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Deep Learning Crystallographic Information from Electron Diffraction Images

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Ongoing development in electron detector technology, in the recent years, has enabled a new age of electron microscopy which facilitates easy accessibility of multi-dimensional data. The 4D Camera at Lawrence Berkeley National Laboratory, for example, is capable of generating enormous amounts of data (estimated ~ 5 PBs/year) due to the development of high-speed electron detector technology. To make analysis of 4D Camera and other 4D-STEM (4D- Scanning Transmission Electron Microscope) data easy and accessible, tools such as the robust open-source python-based analysis package py4DSTEM have been developed. These tools are effective in many instances, however, there is a need for image analysis using cutting-edge machine learning (AI/ML) techniques to complement the existing relatively slow, complex, and hyperparameter-dependent pipeline. To this end, we propose the design of a fully automated AI/ML python-based pipeline which is fast, automated, and robust against experimental error and background noise. In this project, state-of-the-art deep learning techniques are developed, designed, tested, and deployed to the traditional data analysis pipeline of 4D STEM measurements in an aim to retrieve useful crystallographic information, such as crystalline Bragg vector mapping, strain maps and orientation maps for unknown polycrystalline samples.

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Fully Automated Nanoscale to Atomistic Structure from Theory and X-Ray Spectroscopy Experiments

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In complex photocatalytic reactions involved in the generation of solar fuels, it is critical to extract the oxidation state and atomic configuration of intermediate first-row transition metal molecules to understand the photocatalytic reaction mechanism and optimize the catalytic rate and total catalytic yield. X-ray Transient Absorption spectroscopy can be used to perform in-situ mechanistic studies, but theoretical insight requires searching a vast structural space, where it is critical to not only match experimental data but also minimize other quantities such as the energy to ensure that the structures are physically plausible and realizable. In response, we have extended our previously developed FANTASTX code to include full support for x-ray spectroscopy simulations. FANTASTX is a multi-objective evolutionary algorithm which performs structure search using genetic algorithm and basin hopping methods, yielding the structural candidate(s) which best matches both provided experimental data and computationally measured structural quantities. In photocatalytic reactions, the situation is further complicated by the simultaneous presence of multiple molecular species. To search such a multi-molecular structural space more efficiently, we have further extended FANTASTX by incorporating structural fingerprinting and clustering methods, enabling the identification of

fundamentally different molecular candidates which can uniformly prioritized through a novel cluster-driven evolutionary algorithm approach.

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Utilizing colloidal materials chemistry to explore condensed phases for next-generation solid state electrolytes

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I employ a topotactic method called cation exchange to produce semiconductor nanocrystals (NCs) in novel morphologies, compositions, and crystallographic phases. My dissertation research focused on the understanding of the physical properties and phase transitions of these new nanomaterials prepared by cation exchange. With the goal of fostering fruitful collaborations with fellow postdocs through scientific discourse at this symposium in mind, I shall discuss some results on superionic conductors realized through colloidal chemistry which could benefit from electrochemical measurements to prove their suitability in solid state battery applications. I shall also describe my attempts at the realization of computational plasmonic framework to quantitatively investigate the near-field interactions in arrays consisting of plasmonic nanoparticles.

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Manipulating topology in tailored artificial graphene nanoribbons

D.J. Trainer[1], S. Srinivasan[1], B.L. Fisher[1], Y. Zhang[2], C.R. Pfeiffer[1], S.-W. Hla[1,3], P. Darancet[1], N. P. Guisinger[1]

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Topological phases of matter give rise to exotic physics that can be leveraged for next generation quantum computation[1-3] and spintronic devices[4,5]. Thus, the search for topological phases and the quantum states that they exhibit have become the subject of a massive research effort in condensed matter physics. Topologically protected states have been produced in a variety of systems, including artificial lattices[6-9], graphene nanoribbons (GNRs)[10,11] and bismuth bilayers[12,13]. Despite these advances, the real-time manipulation of individual topological states and their relative coupling, a necessary feature for the realization of topological qubits, remains elusive. Guided by first-principles calculations, we spatially manipulate robust, zero-dimensional topological states by altering the topological invariants of quasi-one-dimensional artificial graphene nanostructures. This is achieved by positioning carbon monoxide molecules on a copper surface to confine its surface state electrons into artificial atoms positioned to emulate the low-energy electronic structure of graphene derivatives. Ultimately, we demonstrate control over the coupling between adjacent topological states that are finely engineered and simulate complex Hamiltonians. Our atomic synthesis gives access to nanoribbon geometries beyond the current reach of synthetic chemistry, and thus provides an ideal platform for the design and study of novel topological and quantum states of matter.

References

[1] Fu, L.; & Kane, C. L; *Physical Review Letters* **100**, 096407 (2008).
[2] Nadj-Perge, S. *et al.*; *Science* **346**, 602-607 (2014).

[3] Nadj-Perge, S.; Drozdov, I. K.; Bernevig, B. A.; Yazdani, A.; *Physical Review B*, **88**, 020407 (2013).
[4] Bernevig, B. A.; Hughes, T. L.; Zhang, S.-C.; *Science* **314**, 1757-1761 (2006).
[5] König, M. *et al.*; *Science* **318**, 766-770 (2007).
[6] Freeney, S. E.; van den Broeke, J. J.; Harsveld van der Veen, A. J. J.; Swart, I. & Morais Smith, C.; *Physical Review Letters* **124**, 236404 (2020).
[7] Kempkes, S. N. *et al.*; *Nature Materials* **18**, 1292-1297 (2019).
[8] Gardenier, T. S. *et al.*; *ACS Nano* **14**, 13638-13644 (2020).
[9] Yan, L. & Liljeroth, P.; *Advances in Physics: X* **4**, 1651672 (2019).
[10] Rizzo, D.J.; Veber, G.; Cao, T. *et al.*; *Nature* **560**, 204–208 (2018).
[11] Gröning, O.; Wang, S.; Yao, X. *et al.*; *Nature* **560**, 209–213 (2018).
[12] Drozdov, I.; Alexandradinata, A.; Jeon, S. *et al.*; *Nature Phys* **10**, 664–669 (2014).
[13] Jäck, B.; Xie, Y.; Li, J.; Jeon, S.; Bernevig, B.A.; Yazdani, A.; *Science* **364**, 1255-1259 (2019).

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Trapping and manipulating single-electron qubits on solid neon in a hybrid circuit quantum electrodynamics architecture

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The promise of quantum computing has driven a persistent quest for new qubit platforms with long coherence, fast operation, and large scalability. Electrons, ubiquitous elementary particles of nonzero charge, spin, and mass, have commonly been perceived as paradigmatic local quantum information carriers. Despite superior controllability and configurability, their practical performance as qubits via either motional or spin states depends critically on their material environment. Here we report our experimental realization of a new qubit platform based upon isolated single electrons trapped on an ultraclean solid neon surface in vacuum. By integrating an electron trap in a circuit quantum electrodynamics architecture, we achieve strong coupling between the motional states of a single electron and a single microwave photon in an on-chip superconducting resonator. Qubit gate operations and dispersive readout are implemented to measure the energy relaxation time T1 of 15μs and phase coherence time T2 over 200ns. These results indicate that the electron-on-solid-neon qubit already performs near the state of the art as a charge qubit.

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Unsupervised Machine Learning for Spatio-Temporal Characterization of Ultrafast Electron Microscopy Datasets

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Advancements in microscopy techniques have made material investigations at nanometer-picosecond spatio-temporal resolutions possible. This work details the use of a machine learning based approach to extract quantitative information regarding the motion of features as captured by an ultrafast electron microscope (UEM). UEM is an emerging technique that uses pulsed electron beams to image structural dynamics at nm-ps resolutions. This spatio-temporal characteristic of a UEM dataset is one of the main challenges encountered during its analysis. Classical computer vision techniques for characterizing motion between image frames are parametric, and hence require manual supervision. In this work, a U-net type convolutional neural network is designed to take a pair

of UEM images as input and generate the optical flow at each pixel as output. A custom loss function is defined, consisting of a photometric loss term and a gradient loss term. Additionally, the uncertainty associated with the estimate at each pixel is quantified using the Monte-Carlo Dropout method. The performance of an OpenCV implementation of a dense optical flow algorithm is chosen as the baseline, and it is shown that the model proposed in the current work outperforms the baseline method.

NUCLEAR SCIENCES AND ENGINEERING

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tRAPID: A Hybrid Code for Time-Dependent Neutron Transport Calculations

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Current methods for time-dependent neutron kinetics calculations for nuclear systems (e.g., nuclear reactors) are extremely computer-intensive. The deterministic neutron transport codes need to rely on significant approximations as the phase space variables and timescales of the Linear Boltzmann Equation (LBE) span several orders of magnitude. Monte Carlo codes, on the other hand, do not entail any major approximation, but their statistical convergence can be extremely slow, especially when calculating detailed space-and-time dependent distributions. tRAPID is a hybrid code based on the Multi-stage Response-function Transport (MRT) methodology, that decouples the transport problem into a series of independent physics-based stages. For each of the stages, response functions are pre-calculated for various problem-relevant parameters (e.g., control rod positions). tRAPID utilizes the Transient Fission Matrix (TFM) approach for solving the LBE. After pre-calculating the TFM coefficients, the algorithm utilizes them to solve reactor kinetics problems in real-time retaining Monte Carlo-level accuracy. The tRAPID algorithm has been developed at Virginia Tech and experimentally validated using control rod insertion experiments performed at the Jožef Stefan Institute TRIGA Mark-II reactor (Ljubljana, Slovenia). tRAPID is capable of accurately calculating time-dependent 3-D fission neutron sources in seconds to minutes, a fraction of the time required to standard transport codes.

SYNCHROTRON RESEARCH

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de Gennes Narrowing-like Phenomenon and Nano-ripple Velocity Measurement in Self-Organized Ion-Beam Nanopatterning

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Real-time coherent Grazing-Incidence Small-Angle X-ray Scattering was used to investigate the kinetics and the fluctuation dynamics during ion beam nano-patterning of silicon surface at room temperature. Initially flat silicon samples at room temperature were bombarded by a broad collimated

beam of 1keV Ar+ and Kr+ ions at 65° polar angle, leading to the amorphization of the ion-irradiated surfaces and the spontaneous formation of nanoscale ripples. The temporal evolution of the average X-ray scattering intensity shows the evolution of kinetics, while the fluctuation dynamics can be investigated by correlation of X-ray speckles. The surface behavior at early times can be explained within a linear theory framework. The transition away from the linear theory behavior is observed in the dynamics since the intensity correlation function quickly evolves into a compressed exponential decay on length scales corresponding to the peak ripple wavelength and a stretched exponential decay on shorter length scales. The correlation times for silicon nano-patterning are maximum at the ripple wavelengths while they are smaller at other wavelengths. This has notable similarities and differences with the phenomenon of de Gennes narrowing observed in a wide range of soft materials. Overall, such dynamic behavior is found to be consistent with the simulations based on nonlinear growth models. Following the formation of self-organized nano-ripples, they move across the surface. While homodyne XPCS is not sensitive to velocities, but because of the gradient of ion flux across the sample, we were able to measure the ripple velocity gradients by cross-correlating X-ray speckles and tracking their movements *in situ*.

References

[1] Myint, P.; Ludwig, K.F.; Wiegart, L.; Zhang, Y.; Fluerasu, A.; Zhang, X.; Headrick, R.L.N.; *Phys. Rev. B*, 103, 195423 (2021).

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