Plenary and Prize Session

* Invited Paper

SESSION I1.01: Plenary and Prize Session
Session Chairs: Peter Gehring, Young Lee, Katie Weigandt and Stephen Wilson
Monday Morning, June 6, 2022
UMC Conference Room 235

8:00 AM WELCOME AND INSTRUCTIONS

8:15 AM *I1.01.01
CLIFFORD G. SHULL PRIZE WINNER: Tailoring Instruments to the Science and the Source: 35 Years at the NCNR Dan A. Neumann; National Institute of Standards and Technology, United States

TBA

9:10 AM *I1.01.02
PLENARY: The Dark Energy of Quantum Materials Laura Greene; Florida State University, United States

I'll give an overview of the MagLab, why unconventional superconductors remain one of the greatest unsolved questions in physics of our day.

9:45 AM BREAK

SESSION I2.01: Plenary and Prize Session
Session Chairs: Peter Gehring, Young Lee, Katie Weigandt and Stephen Wilson
Tuesday Morning, June 7, 2022
UMC Conference Room 235

8:15 AM FELLOWS ANNOUNCEMENT

8:30 AM *I2.01.01
SCIENCE PRIZE WINNER: Magnons are not Forever Martin Mourigal; Georgia Institute of Technology, United States

TBA

9:10 AM *I2.01.02
PLENARY: SANS Contrast Variation Experiments on Multi-Component Biological Complexes: What's the Big Deal? Susan Krueger; National Institute of Standards and Technology, United States

Small-angle neutron scattering (SANS) contrast variation experiments have been providing key information for researchers studying multi-component biological complexes in aqueous solution for more than four decades. In spite of numerous success stories over the years, many are still unfamiliar with the technique and how it can be applied to their research. Even those who have heard about it might be wondering if it is worth the effort. After all, multiple measurements in solvents with differing amounts of D2O and H2O are implicitly required and deuterium labeling of one or more of the components might be necessary. The big deal is that SANS contrast variation experiments can provide unique information about the individual components as they exist in the complex. This information could be the crucial link to solving the solution structure of the complex and gaining insight into its function.
This talk will provide an overview of SANS from biological macromolecules in solution, with a focus on contrast variation from protein-nucleic acid, protein-protein, protein-lipid and proteindetergent complexes. Contrast variation will also be discussed in the context of proteins in complex environments, such as during freezing or separation chromatography and under high pressure. If time permits, an overview of current software developments at the NIST Center for Neutron Research in support of SANS contrast variation experiments will be presented.

9:45 AM BREAK

SESSION I3.01: Plenary and Prize Session
Session Chairs: Peter Gehring, Young Lee, Katie Weigandt and Stephen Wilson
Wednesday Morning, June 8, 2022
UMC Conference Room 235

8:30 AM *I3.01.01
SUSTAINED RESEARCH PRIZE WINNER: Neutrons, Biological Membranes, and Future Directions John Katsaras; Oak Ridge National Laboratory, United States

Biological cells are bound by plasma membranes (PMs) – supramolecular assemblies of lipids and proteins that separate a cell's interior from its external environment. Although the existence of cells has been known since the mid-19th century, it was not until the early part of the 20th century that the underlying structure of the PM began to emerge. It then took another 50 years for Singer and Nicolson to propose the now well-known "fluid mosaic" model of the PM. I will describe neutron and x-ray scattering studies that have addressed longstanding questions in membrane biology and mention that without lipids there would be no mRNA vaccines for COVID-19. I will conclude with the future use of lipid bilayers as platforms for the next-generation of neuromorphic computers and as novel therapeutic targets for brain diseases.

9:10 AM *I3.01.02
PLENARY: Neutron Scattering from Exotic Magnetic Ground States Bruce D. Gaulin; McMaster University, Canada

The last two decades has seen great interest and activity focussed on new magnetic materials with exotic ground states, made possible by either geometrical frustration or quantum fluctuations or both. Neutron scattering has been critical to understanding these materials and their ground states, but these studies have not been without their challenges. In part these arise as conventional long range order is not the norm, and disordered, spin liquid-like states tend to distribute scattering across momentum and energy space. In part, these challenges arise as the materials of interest are typically new quantum materials, which are available, at least initially, only as powders or small single crystals. I will illustrate some of the successes that neutron scattering has had in this area, drawing from my own group's experience in pyrochlore and other quantum magnets, and will discuss how these challenges are helping to motivate the design of new neutron spectrometers.

9:45 AM BREAK

SESSION I4.01: Plenary and Prize Session
Session Chairs: Peter Gehring, Young Lee, Katie Weigandt and Stephen Wilson
Thursday Morning, June 9, 2022
UMC Conference Room 235

8:15 AM *I4.01.01
OUTSTANDING STUDENT RESEARCH PRIZE WINNER: Magnetic Phase Transitions and Spin-Wave Dynamics in $Y_{1-x}La_xTiO_3$ and $Y_{1-y}Ca_yTiO_3$ Sajna Hameed; University of Minnesota Twin Cities, United States

The perovskite titanate class of materials encompasses a wide variety of phenomena, including magnetic phase transitions, insulator-metal transitions, and superconductivity. In this talk, I will present our findings from neutron scattering and complementary measurements of the magnetically-ordered ground states and spin dynamics of the chemically-substituted Mott insulator $Y_{1-x}La_xTiO_3$ and of hole-doped $Y_{1-y}Ca_yTiO_3$. The former system exhibits an intriguing transition from ferromagnetic to antiferromagnetic ground states, whereas the latter material first exhibits a transition to a paramagnetic state at y 0.2 prior to undergoing an insulator-metal transition at y 0.35 [1,2,3,4].
[1] S. Hameed *et al.*, Phys. Rev. Materials **5**, 125003 (2021)
[2] S. Hameed *et al.*, Phys. Rev. B **104**, 024410 (2021)
[3] S. Hameed *et al.*, Phys. Rev. B **104**, 045112 (2021); Phys. Rev. B **105**, 159902(E) (2022)
[4] S. Hameed *et al.*, manuscript in preparation (2022).
Work funded by the US Department of Energy through the University of Minnesota Center for Quantum Materials, under grant number DE-SC-0016371.

9:10 AM *I4.01.02
PLEANARY: Neutrons for Clean Bioenergy Jeremy Smith; University of Tennessee/Oak Ridge National Laboratory, United States

Bioenergy, the largest single source of renewable energy in the US, is derived from recently living organisms (biomass) used to produce transportation fuels, heat, electricity, and bioproducts that can replace petrochemicals. Second-generation bioenergy has the potential to greatly reduce our greenhouse gas emissions. However, the complex three-dimensional structure of biomass makes it recalcitrant to deconstruction by enzymes or chemical catalysts. To try to overcome this recalcitrance biomass is commonly pretreated but hitherto with varying success. Understanding the molecular-level driving forces holding biomass together and altering its structure on pretreatment has facilitated the rational design of effective methods for deconstruction. Recent neutron scattering, polymer theory and computational work has revealed common solvation thermodynamic phenomena underlying biomass assembly and deconstruction, together with the mechanisms by which solvents can deconstruct cellulose. Green, inexpensive solvents are presented with close to 100% efficiency in solubilizing biomass.

Langan, P et al .*Green Chemistry* 16 1 (2014) 63-68.

Petridis, L et al. *Nature Reviews Chemistry* 2 11 (2018) 382-389.

Pingali, S.V. et al. *Proceedings of the National Academy of Sciences* 117 29 (2020) 16776-16781.

9:40 AM POSTER AWARD ANNOUNCEMENT

9:45 AM BREAK

Advances in Neutron Facilities, Instrumentation and Software A

* Invited Paper

SESSION A1.01: Facilities
Session Chair: John Ankner
Monday Morning, June 6, 2022
UMC West Ballroom 208

10:15 AM *A1.01.01
Development of Neutron Scattering Facilities at the McMaster Nuclear Reactor Patrick Clancy; McMaster University, Canada

The McMaster Nuclear Reactor (MNR) is a 5 MW open-pool reactor located on the campus of McMaster University in Hamilton, Ontario. Following the closure of the NRU Reactor at Chalk River in 2018, the MNR has become Canada's only source of neutron beams for materials research. This has led to a major national effort to revitalize the neutron scattering facilities at the MNR, with a planned investment of ~$24M over the next five years. The MNR is currently home to two neutron scattering beamlines: the McMaster Alignment Diffractometer (MAD), a general purpose triple-axis spectrometer, and MacSANS, the McMaster Small Angle Neutron Scattering facility. In this talk I will discuss the current status of MAD and MacSANS, and describe plans for an expanded suite of instruments which includes a neutron powder diffractometer, a neutron reflectometer, and a neutron residual stress scanner.

10:45 AM A1.01.02
Reactor Institute Delft 2.0 Jeroen Plomp; Delft University of Technology, Netherlands

The Reactor Institute Delft (RID) is the only research facility in the Netherlands that uses neutron- and positron beam line instruments for academic research. The 2.3 MW research reactor not only facilitates beam-line instruments that offer the unique opportunities of this special radiation but operates irradiation facilities where isotopes can be produced for heath research or element analyses as well. The field of application is very broad and varies from Energy to Health and even cultural heritage.

New instruments and the implementation of a cold source (end 2022) and user program will broaden the scientific footprint of the reactor.

Instruments available are: Neutron Imaging (thermal and cold), Neutron Powder Diffraction, Small Angle Neutron Scattering, Neutron reflectometry, Neutron Depth profiling, Posh-Pals, Posh-2D ACAR, Irradiation facilities, Instrument Neutron Activation Analysis

We would like to present an overview of the latest developments and discuss the strengths and weaknesses of a relatively small university facility.

11:00 AM A1.01.03
New Guide System and Upgraded Instrument Suite at HFIR after Beryllium Reflector Change Georg Ehlers, Amy Jones, Michael C. Hoffmann, Lowell Crow, Franz X. Gallmeier, Thomas Huegle, Matthew J. Frost, Cassie S. Sabens, Kenneth C. Littrell, Richard M. Ibberson, Lisa DeBeer-Schmitt, Garrett E. Granroth, Sai Venkatesh Pingali, Hassina Z. Bilheux, Yuxuan Zhang, Andrii Y. Kovalevskyi, Travis J. Williams, Adam Aczel and Matthias D. Frontzek; Oak Ridge National Laboratory, United States

Planning is underway for a replacement of the permanent Beryllium reflector at the High Flux Isotope Reactor (HFIR) at Oak Ridge National Laboratory (ORNL) in 2025. This renewal involves a complete removal of most scattering instruments and thus provides an opportunity to upgrade the neutron guides and the instruments. In this contribution we give a high-level overview of the new guide designs and some of the instrument upgrades foreseen at this time.

11:15 AM A1.01.04
Neutron Beamline Shielding Studies for the HFIR Beryllium Reflector Replacement Project Kyle Grammer and Wei Lu; Oak Ridge National Laboratory, United States

The HFIR beryllium reflector replacement (HBRR) project includes both upgrades to thermal beam room instruments as well as a redesigned cold neutron guide network. The HBRR provides an opportunity for upgrades to existing shielding infrastructure in the thermal beam room and requires new shielding design studies for the cold guide hall (CGH). There will also be an extension to the CGH and an expansion from 4 neutron guides to 6. The current status of HBRR shielding analysis for both the thermal beam room and the CGH will be discussed. Efforts for the thermal beam room include verification of existing shielding at HB2, studying the post-HBRR configuration of HB2 and HB3, and the addition of a permanent shielding wall between HB2 and HB3 to replace temporary shielding. The CGH shielding studies involve developing shielding plans for new instruments, such as the Imaging station on NB4, as well as validating new components of existing instruments, including the shielding for new velocity selectors and studying the optimal placement of secondary beamline shutters.

11:30 AM A1.01.05
Future of Development Beamlines at the High Flux Isotope Reactor after the upcoming HFIR Beryllium Reflector Replacement Lowell Crow; Oak Ridge National Laboratory, United States

With the HFIR Beryllium Reflector Replacement (HBRR) approaching in 2025, the present development instruments, like many of the user instruments, will be moved or modified. The revised instrument at the HB-2D position in the beam room will allow for continuing polarized neutron development with a more intense beam and a shorter wavelength than the present 4.25 Angstrom instrument. It will also provide a beam for detector development, to replace the present function of the CG1A instrument in the guide hall. There will be an opportunity to build an interim instrument on the NB2 cold guide for cold neutron Larmor technique development. These plans will be discussed within the context of the overall HBRR effort.

11:45 AM A1.01.06
The Cold Source Upgrade Project at the NIST Center for Neutron Research Rodrigo Vilaseca, Daniel Adler, Donald Pierce, Brian J. Kirby and Dan A. Neumann; NIST, United States

The NIST Center for Neutron Research will upgrade its large liquid hydrogen cold neutron source with a liquid deuterium cold source that will double the number of neutrons at long wavelengths. This evolution will include the replacement of three neutron guides and associated upgrades to several instruments including the cold neutron imaging station, the alpha-gamma facility, and the cold triple axis instrument - SPINS. In addition, the NCNR will take advantage of the opportunity to replace the Neutron Spin Echo spectrometer with one based on superconducting precession coils that promises to perform 10x better than the current instrument. This work will require an 11-month shutdown planned for 2023.

12:00 PM A1.01.07
Preliminary Neutronics Design of a Second Target Station at the ORNL's SNS Igor Remec, Franz X. Gallmeier, Kristel Ghoos, Tucker McClanahan, Thomas Miller, Kumar Mohindroo, Wouter de Wet and Lukas Zavorka; Oak Ridge National Laboratory, United States

The Oak Ridge National laboratory (ORNL) currently operates the High Flux Isotope Reactor (HFIR) and the Spallation Neutron Source (SNS), two of the world's most advanced neutron scattering research facilities. HFIR has been in operation for 57 years, and SNS celebrated 15-th anniversary of neutron production in 2021. To keep the research capabilities on the cutting edge several projects are currently in progress. At the HFIR the upgrade and optimization of the cold neutron guide hall and isotope production facilities is underway which will increase capabilities for production of important isotopes such as Pu-238, improve the performance of the current instruments, and provide space to for new instruments. The exchange of beryllium reflector is also in preparation. At the SNS a few remaining un-occupied beamlines at the First Target Station (FTS) are being constructed and equipped with new instruments. The SNS is also undertaking the Proton Power Upgrade (PPU) Project that will double the accelerator proton beam power to 2.8 MW. The PPU will increase the beam power delivered to FTS and provide proton beam to the new addition - the Second Target Station (STS). The long-planned fourth-generation neutron source, the STS, received the DOE Critical Decision 1 (CD-1) approval in November 2020.

The STS will operate as accelerator driven pulsed neutron source with 700 kW of the proton beam power delivered in short, less than 1 μs long pulses, with 15 Hz repetition rate. The core of the STS is rotating water-cooled tungsten target which feeds neutrons into two coupled cryogenic moderators filled with liquid hydrogen, surrounded with water pre-moderators and beryllium reflector. The STS will be specifically designed to provide exceptionally bright beams of cold neutrons that will enable simultaneously probing of the structure and function of complex materials across broader length and time scales and will complement the existing capabilities of the FTS and HFIR.

The high neutron beam brightness is obtained by specifically optimizing the moderators for operation with pure para-hydrogen, by creating tight coupling of target and moderators, and by using small size neutron beams. The cylindrical moderator, which will serve 16 beamlines, has four rectangular extraction ports with the viewed areas of 30 mm × 30 mm size, while the novel one-dimensional "tube" moderator has three circular viewed areas with 30 mm diameter and will serve six beamlines.

The early completion of the STS project is projected in 2032 and will include eight instruments ready for the operation. The instruments were selected based on the recommendation of a committee of 22 national and international experts. We will report on the current status of the preliminary neutronics design of the STS. The emphasis will be on the moderator design and on the characteristics of the neutron beams delivered to the instruments.

SESSION A1.02: Software: Instrumentation
Session Chair: Paul Butler
Monday Afternoon, June 6, 2022
UMC West Ballroom 208

2:00 PM *A1.02.01
Material Decomposition for Hyperspectral Neutron Tomography Charles A. Bouman[1], Gregery Buzzard, T[1], Mohammad Samin Nur Chowdhury[1], Singanallur Venkatakrishnan[2] and Hassina Z. Bilheux[2]; [1]Purdue University, United States; [2]Oak Ridge National Laboratory, United States

Hyperspectral neutron tomography has the potential to reveal important material features that are not easily determined using other sensing modalities. In this talk, we present recent progress in the development of algorithms for the separation of materials and the associated tomographic reconstruction from neutron imaging data. Our methods are based on the automatic separation of neutron absorption spectra using a combination of space-domain constraints and non-negative matrix factorization. We then integrate the material decomposition with a model-based iterative reconstruction (MBIR) algorithm that performs fast, sparse-view reconstruction and that acounts for measurement noise and prior knowledge of image structure. We also describe the publicly-available, open-source SVMBIR software package, which enables real-time reconstructions from current spectral-detector technology. We demonstrate our method using both synthetic and real data.

2:30 PM A1.02.02
McStas2CAD: A Python-based Software Package for Scattering Instrument Concept Geometry Conversion Matthew J. Frost and Lee Robertson; Oak Ridge National Laboratory, United States

Most neutron scattering instrument development begins with a ray tracing simulation to understand the potential performance of a proposed design at a source. A majority of the proposed scattering instrumentation at Oak Ridge National Laboratory has used the McStas ray-tracing software package to meet this need. This can include incident beam optics, energy selectors, and detectors, amongst others. Once the conceptual design process has completed in McStas, the next step is to begin mechanical and radiological design. As modern instrumentation designs have grown more complex it has become a

challenge to efficiently connect the proposed conceptual design to the mechanical design process. This extends the time it takes to check feasibility of the design and ultimately delivery of the project. In addition, the need to input the geometry manually can become a source of human error. McStas2CAD is a software package written in Python3 using the NumPy module and has been developed to make the jump from scattering concept to detailed mechanical/radiological design easier and less prone to error. This is done with universally used CAD formats (IGES and STEP) with simple shapes that can be directly referenced in the detailed model.

2:45 PM A1.02.03
Towards a User-friendly Workflow for Monte Carlo Neutron Scattering Simulations Fahima Islam, Garrett E. Granroth, Jiao Lin and Thomas Huegle; Neutron Scattering Division, United States

The analysis of neutron scattering experiments is complex due to the factors such as scattering from sample environments, multiple scattering and the varied instrument resolution contributions. We are in the process of developing a new workflow that is used for performing simulations of neutron scattering for various systems with different instrument configurations. The current way of performing the simulation involves numerous manual steps and requires an expert user. The goal of this development is to provide a workflow that is faster and more user friendly. In this work, we will focus on the automatic calculation of the incident beam spectra (up to the sample) for ARCS, GPSANS and SNAP at ORNL to cover monochromatic and time of flight instruments. These calculations are in McStas and take advantage of the parallelization features in McStas 3.x. which supports CPU multicore (mpi directives) as well as heterogeneous CPU/GPU (OpenAcc) acceleration. We have scripted the process of creating an executable of a McStas simulation starting from the McStas instrument file, to run in heterogenous CPU/GPU environment. Specifically the c file is generated on local machines and then transferred to a NVIDIA DGX A100 box. Here it is then compiled and executed using an Nvidia provided HPC development Docker image which contains the necessary libraries and compilers to enable the parallelization. By using this GPU configuration we have achieved a speed up of four to five orders of magnitude over single-threaded CPU implementations, making it possible to run an incident beam simulation of equivalent statistics as a beam measurement in a time commensurate with that measurement. The incoming beam simulation will be cataloged and stored and can thus be fed into a McVine simulation for detailed simulation of the sample scattering. Speeding up the incident beam simulation will allow users to do these simulations in real time. This work is sponsored by the Lab Directors' Research and Development Fund of Oak Ridge National Laboratory.

3:00 PM A1.02.04
Tally Components in McStas Thomas Huegle, Matthew J. Frost, Garrett E. Granroth and Lee Robertson; Oak Ridge National Laboratory, United States

A new class of McStas components records the details of each neutron interaction with guide surfaces. The resulting data can be refined using large-scale data analysis tools (e.g. python's pandas package). This enables a detailed study of e.g. the reflectivity of the guide coating required on each surface of each guide segment for ideal neutron transport of guide designs, especially of those following non-trivial geometries. Other applications include the investigation of outliers in phase space, correlation between certain guide surfaces, and the significance of individual guide surfaces.
In this talk, we will discuss inner workings of the components themselves and provide examples of how they were used to optimize several beam line designs at ORNL.
This work was sponsored by the User Facility Division of Basic Energy Sciences of the US DOE.

SESSION A1.03: Software: Data Analysis and Modeling
Session Chair: Charles Bouman
Monday Afternoon, June 6, 2022
UMC West Ballroom 208

3:45 PM A1.03.01
Browser Based Visualization of Large 3D Datasets using NVIDIA IndeX Evan Carlin[1], Kevin Bruhwiler[1], Robert Nagler[1], David Bruhwiler[1], Christina Hoffmann[2], Andrei T. Savici[2], Zachary Morgan[2], Matthew Tucker[2], Alexander Kuhn[3], Jörg Mensmann[3], Peter Messmer[3], Marc Nienhaus[3], Steffen Roemer[3] and Dragos Tatulea[3]; [1]RadiaSoft LLC, United States; [2]Oak Ridge National Laboratory, United States; [3]NVIDIA, Germany

Neutron scattering has undergone significant technological development through large area detectors with concurrent enhancements in neutron transport and electronic functionality. This allows one to collect neutron scattering data in event space tracking detector pixel location, time and associated metadata of every event, which in turn are used to display multidimensional space with meaningful parameters. A result of volumetric data collection are data sets in the range of MB to GB that span vast areas of time-space-energy volume. Conventional methods of displaying static line cuts or planes of interest are insufficient to help visualize and disentangle the complexity of signals in a given data set. This leads to the need of using cutting edge visualization capabilities for large neutron scattering data files in multidimensional space.

NVIDIA's IndeX technology [1] is a 3D visualization tool that harnesses the compute power of NVIDIA GPUs. It is the basis for ongoing development of cloud based visualization capabilities, being delivered in the near term to neutron beamline scientists at Oak Ridge National Laboratory. The IndeX 3D volumetric visualization framework is being used via the HTML5 client viewer and the Jupyter notebook interface. Present capabilities will be demonstrated [2]. Future plans include integration with browser-based visualization tools under development at ORNL, as well as the Sirepo scientific gateway [3], enabling interactive exploration of 3D datasets in multiple scientific disciplines.
[1] IndeX home page, https://developer.nvidia.com/nvidia-index
[2] K. Bruhwiler et al., IPAC Proceedings, TUPAB413 (2021), https://accelconf.web.cern.ch/ipac2021/papers/tupab413.pdf
[3] Sirepo scientific gateway, https://sirepo.com

4:00 PM A1.03.02
Addressing the Resource Problem Through Community. SasView: An "Open, Collaborative, Community Development" Platform for Small Angle Scattering Paul D. Butler[1,2,3]; [1]National Institute of Standards and Technology, United States; [2]University of Delaware, United States; [3]The University of Tennessee, Knoxville, United States

As the needs for advanced computational tools become increasingly critical for the extraction of new scientific knowledge from scattering data, the need for increasingly sophisticated tools has become a bottleneck despite relatively large (compared to the past) investments in software and the "cyberinfrastructure revolution." Here we posit that this problem is not going away, fundamentally because analysis needs are, and will remain, effectively infinite, while resources will always be finite, and suggest a potential path forward.

SasView is a SAS data analysis package that originated out of the NSF funded DANSE project and has evolved into an international collaborative effort that is experimenting with ways of providing robust and sustainable analysis software in this new cyberaware age. In this presentation we will not focus on the software and its features per se, but rather on how it is trying to deliver those features. We will start by defining the current scope of SasView followed by a brief history of the project. We will present the variety of help/support/education and outreach resources and the development infrastructures put together over time. We will take a particular look at how each word in "open, collaborative, community development" is currently addressed, and discuss how we got to where we are, the lessons learned along the way and the challenges faced, then and now.

Besides helping to optimize the use of limited resources by not ending up with excessive duplication, this type of community approach has the potential of being a way to self select the areas of most important need on which to focus the finite resources. While this "experiment" remains a work in progress, we propose that this model, no doubt with some adjustments, should at the very least be an important part of any holistic plan for providing scientific software tools in the future, particularly for data emanating out of large scale facilities.

4:15 PM A1.03.03
Event Based Data Analysis and Visualization in Neutron Spectroscopy Andrei T. Savici[1], Igor Zaliznyak[2], Garrett E. Granroth[1] and Ovidiu Garlea[1]; [1]Oak Ridge National Laboratory, United States; [2]Brookhaven National Laboratory, United States

Data analysis and visualization for single crystal neutron spectroscopy experiments have been historically performed using data pre-histogrammed in energy transfer. To get the physical meaning of the data in terms of the dynamic structure factor, each histogram bin, for each detector, is transformed to energy and momentum transfer. For further analysis and visualization, the data is re-histogrammed in energy-momentum space, such that every pixel contains the average of the contributing histogram bins [1,2]. The main deficiencies of such existing approach are (i) the large amount of memory resources needed to store the histogram bins with zero neutron counts, and (ii) the difficulty of error propagation in re-histogramming, which resulted in the absence of weighting of contributions with different statistical significance. Based on the lessons learned from single crystal diffraction [3,4], we have developed a different approach, where the histogramming to energy-momentum space is based on neutron detection events and accounts for their statistical significance. The algorithm is implemented in the Mantid software [5] and provides options for processing and correcting both upolarized and polarized data. Here, we will emphasize the experience we gained with implementing straight forward python scripting based data processing and visualization based on the event approach, and plans for a future graphical user interface.

[1] R. T. Azuah et al., Journal of Research of the National Institute of Standards and Technology 114, 341 (2009)
[2] R.A. Ewings et al., Nuclear Instruments and Methods in Physics Research Section A 834, 132 (2016)
[3] T. Michels-Clark et al., J. Appl. Crystalography 49, 497 (2016)
[4] A. T. Savici et al., in preparation
[5] O. Arnold et al., Nuclear Instruments and Methods in Physics Research Section A 764, 156 (2014)

This research used resources at the Spallation Neutron Source, a DOE Office of Science User Facility operated by the Oak Ridge National Laboratory.

4:30 PM A1.03.04
Do We Need an ISO Standard for the Reduction of Time-of-Flight Powder Diffraction Data? Malcolm Guthrie; Oak Ridge National Laboratory, United States

ISO standards are a method to define the best way of doing "something" as agreed upon by an international panel of experts. Once defined, an ISO standard therefore provides a guarantee that the product has been created according to established practice and that the "consumer" can trust in a defined level of quality.

Although overall approaches to the reduction of time-of-flight (TOF) powder diffraction data are commonly accepted, there is a lot of detail in these and, correspondingly, a rather large spectrum of different philosophies in application. Consequently, data collected from the same sample, at different facilities, from different instruments at the same facility, or even from the same instrument on different occasions may not be generated consistently. Given the significant cost, both financial and in terms of effort of generating neutron TOF data there may be significant value in establishing an agreed way to convert raw neutron counts to meaningful scientific data. Further impetus comes from a growing international recognition of the importance of FAIR principles for data management and stewardship that explicitly mandate established and reproducible workflows.

In this presentation, in addition to asking the question if an ISO standard is needed, I will ask what an ISO standard for TOF data reduction might look like. It would certainly be wide ranging. It might, for example, span physical details such as what calibrants should be used, how accurately and reproducibly must they be installed and how long they should be collected. It might also cover decisions made in the software workflows: data limits and binning, parameters and methodologies for generation of vanadium corrections, corrections for experimental backgrounds and attenuation from both sample and sample environments. Lastly, it should also address requirements for reproducibility, ensuring that all necessary metadata, relating to the reduction, are captured sufficiently to allow an independent party to achieve the same result from the same dataset.

4:45 PM A1.03.05
Hyperspectral Neutron CT with Material Decomposition Thilo Balke[1,2], Alexander Long[1], Sven C. Vogel[1], Brendt Wohlberg[1] and Charles Bouman[2]; [1]Los Alamos National Laboratory, United States; [2]Purdue University, United States

Energy-resolved neutron imaging is an advanced neutron radiography technique capable of non-destructively extracting spatial isotopic information within a sample. In combination with uniquely characteristic isotopic neutron cross-section spectra, isotopic areal densities can be determined on a per-pixel basis, thus resulting in a set of areal density images for each isotope present in the sample. By performing measurements over several rotational views, an isotope-decomposed 3D computed tomography is possible.

However, this objective is made difficult by having very low per-pixel neutron counts even after long scan times, significant background counts, and the non-linear effects of the finite neutron pulse width (moderator broadening).

We demonstrate a model-based approach based on Poisson noise modeling that incorporates modeling of the background, moderator broadening and exposure variations. This approach allows for quantitatively accurate isotopic density estimates for both neutron time-of-flight radiography as well as tomography which is demonstrated in our experimental results collected for nuclear fuel samples on the FP-5 beamline at LANSCE.

5:00 PM A1.03.06

Simulation of Inelastic Neutron Scattering Spectra for Direct and Indirect-Geometry Instruments with AbINS Adam Jackson and Sanghamitra Mukhopadhyay; Science and Technology Facilities Council, UK, United Kingdom

Inelastic neutron scattering (INS) is a form of vibrational spectroscopy. The technique is used routinely at neutron facilities in a variety of scattering configurations: some parameters are constrained by the nature of the incident neutrons and instrument geometry, while others may be adjusted by varying chopper speeds, swapping crystalline monochromators etc. It is helpful to assign peaks to movements of relevant atoms, functional groups, molecular confirmations etc.; *ab initio* atomistic calculation packages can generate sufficiently accurate reference information, but typically only provide tools for plotting a simple phonon density-of-states (DOS).

In the incoherent approximation, a DOS-like spectrum may be computed from harmonic phonon calculations, accounting for energy-momentum relations, Debye-Waller intensity effects and generating multi-phonon contributions in an analytic powder-averaging approximation. For hydrogenous materials, the incoherent approximation is generally effective and multi-phonon terms are likely to be significant. Finally instrumental broadening is considered; this is generally treated as an energy-dependent function. (A more exact treatment would involve 4-D ellipsoids described by covariance matrices.)

We present recent developments in the AbINS tool, which resides in the open-source Mantid framework and so brings simulation to the same environment as experimental reduction and analysis. These include efficient calculation of multi-phonon spectra, support for a variety of instruments and sampling from Phonopy force constants datasets. By simulating resolution effects it is possible to improve planning of INS measurements (and use of beamtime) by selection of appropriate instruments and parameters.

1 K. Dymkowski, S. F. Parker, F. Fernandez-Alonso and S. Mukhopadhyay, *Phys. B (Amsterdam, Neth.)*, 2018, **551**, 443–448.
2. O. Arnold et al. *Nucl. Instrum. Methods Phys. Res., Sect. A*, 2014, **764**, 156–166.
3. A. Togo and I. Tanaka, *Scr. Mater.*, 2015, **108**, 1–5.

5:15 PM A1.03.07
Generic Calibration Workflow for Time-of-Flight Instruments Yuanpeng Zhang; Oak Ridge National Laboratory, United States

A generic workflow for calibrating time-of-flight (TOF) instruments has been developed. Three major steps are involved in the workflow - automatic grouping of pixels, cross correlation, and calibration. At the stage of grouping, an unsupervised machine learning based clustering algorithm is introduced to group pixels 'next to' each other, in terms of peak properties. By grouping those similar pixels, the following cross correlation can be conducted more reliably, through which the statistics could be accumulated to further guarantee a reliable calibration against the nominal peak positions of standard sample (e.g., diamond). The calibration workflow has been successfully applied to NOMAD and POWGEN diffractometers at Spallation Neutron Source, Oak Ridge National Laboratory. Multiple diagnostics were generated to demonstrate the robustness of the calibration workflow. It is worth noting that by applying this calibration workflow to replace the initially used pixel-by-pixel calibration routine on POWGEN diffractometer, the measurement time needed for diamond calibrant could be successfully reduced from ~10 hrs to ~3 hrs. Such a generic calibration workflow is expected to be generally applicable to TOF instruments which are seeking for the proper TOF-to-d transformation.

SESSION AP1.04: Poster Session I: Advances in Neutron Facilities, Instrumentation and Software I
Session Chair: Leland Harringer
Monday Afternoon, June 6, 2022
5:30 PM - 7:30 PM
UMC Center Ballroom 210

AP1.04.01
200 W Ultra-narrow Band Pump Laser Diode System Aleksandr Ryasnyanskiy, Vadim Smirnov, Oleksiy Mokhun and Alexei Glebov; OptiGrate Corp., United States

Polarized ^{3}He has been shown to be advantageous when used as an analyzer for polarized neutron scattering, an area of physics research that is growing quickly. Spin-exchange optical pumping (SEOP) of ^{3}He is an important method in several areas of physics, including neutron spin filters for neutron scattering and fundamental neutron physics. The unique advantages of using polarized ^{3}He as neutron spin filters, such as broadband and wide angular acceptance of neutron beams, have made it widely used in most neutron facilities.

In most cases hyperpolarization of the noble gas nuclei is achieved by spin exchange with rubidium atoms that have been optically pumped into one electronic spin state with the use of circularly polarized light from a high-power diode laser system. Rb has been the alkali metal of choice due to large spin-exchange cross sections, relatively high vapor pressures, and availability of high-power pump lasers that can effectively excite its *D*-line.
The technology of volume Bragg gratings (VBGs) recorded in photo-thermo-refractive glass allows narrowing of the linewidth of laser diodes from several nanometers down to tens of picometeres. The ultra-narrow laser emission together, high signal-to-noise ratio and high-power capability of laser diodes allows efficient hyperpolarization of ^{3}He.
We present the first results for 200 W narrowband high-power laser diode systems for pumping of Rb atoms. The system not only produces up to 200 W of narrowband light, but also has a unique capability of tuning the emitting wavelength in a range of 300-400 pm. The system is also capable to deliver 770 nm for K pumping as well as several different wavelengths for hybrid pumping (e.g., 795 nm/770 nm) with individual control of each wavelength position. Using VBGs for laser resonator longitudinal mode selection allowed us to narrow the linewidth of individual laser diode bars down to 20 pm.
In addition to the wavelength tuning, independent control of individual laser channels allows continuous tuning of the laser linewidth in the range of 20-100 pm. When considering the linewidth of the diode laser it is necessary to take in to account the amount of free running lasing suppression. It determines what percentage of light is confined within the narrow band. Current progress in VBG technologies makes it possible to fabricate VBGs with high uniformities that results in laser modules with up to 20-25 dB noise suppression. It means that more than 90% of light is confined within 20-50 pm band. The high power, linewidth and wavelength tunability, polarization control makes the developed pump laser diode system an ideal tool for neutron science applications.

AP1.04.02
Optimization of the Guide Coating for the Manta Incident Beamline Garrett E. Granroth, Adam Aczel, Travis J. Williams and Thomas Huegle; Oak Ridge National Laboratory, United States

Plans are under way to upgrade the cold triple axis capabilites at the High Flux Isotope Reactor at Oak Ridge National Laboratory. The first step is to provide an updated incident beam with worldclass flux on sample that can be used by a traditional single detector backend or a multi analyzer backend when it is developed. The geometry of the guide was optimized using very high index (m=7) coatings so the coating did not influence the guide shape. Here we report the subsequent task of reducing the guide coating on each surface until a performance impact is noticed. This optimization process provides the minimum coating values that can be used in each section. Mcstas simulations were used with modified guide components that recorded not only the probability packet, but also the angle of reflection off the guide surface for each neutron event in each surface These events were then converted to a maximum m value and histogrammed for each surface. Therefore the m value could be set to a level just above the largest m value observed. Subsequent optimizations decreased this m value further so that some neutrons would not be reflected by some of the surfaces. This was done while monitoring the flux on the sample to see if an impact was observed. The idea is that the neutrons at highest m for all regions upstream of the last few are filtered out later in the guide system. The results of this optimization for both the horizontal and vertical directions will be presented. The aformention histograms were particulalry useful in understanding which surfaces to target for coating reduction.

This work was sponsored by the User Facility Division of Basic Energy Sciences of the US DOE.

AP1.04.03
CHESS: A Direct Geometry Chopper Spectrometer Optimised to Study Small Scatterers Gabriele Sala[1], Martin Mourigal[2], Olivier Delaire[3], Yang Zhang[4], Nicholas Butch[5], Raphael P. Hermann[1], Michael E. Manley[1], Andrew D. Christianson[1], Matthew Stone[1], Thomas Huegle[1], Dante Quirinale[1] and Jiao Lin[1]; [1]Oak Ridge National Laboratory, United States; [2]Georgia Institute of Technology, Georgia; [3]Duke University, United States; [4]University of Illinois at Urbana-Champaign, United States; [5]National Institute of Standards and Technology, United States

CHESS is a direct geometry neutron spectrometer designed to detect and analyze weak signals intrinsic to small cross-sections (*e.g.,* small mass, small magnetic moments or neutron absorbing materials) or dynamical phenomena (*e.g.,* atomic and magnetic dynamics in complex crystals, nonlinear or hybrid modes). The ability to simultaneously measure dynamic processes over a wide energy range for very small samples will make CHESS the spectrometer of choice for the initial exploration of new materials. The broad dynamic range will also be well matched to measurements of relaxation processes and excitations in soft and biological matter. The 15 Hz repetition rate of STS enables use of multiple incident energies within a single source pulse, greatly expanding the information gained in a single experiment. An essential feature of CHESS is the capability for polarization analysis to separate nuclear from magnetic scattering or coherent from incoherent scattering in hydrogenous materials, and better understanding spin-anisotropic correlations. This instrument will employ advanced sample environments such as high-pressure cells, dilution refrigerators, high field cryo-magnets and polarization devices, as well as combinations of these, to solve problems at the forefront of materials research.

AP1.04.04
MCViNE and Preliminary Design of STS Instruments Jiao Lin[1], Thomas Huegle[2], Matthew J. Frost[2], Gabriele Sala[1], Alexandru Stoica[2], Ovidiu Garlea[2], Hassina Z. Bilheux[2], Yaohua Liu[1], Changwoo Do[2], Shuo Qian[1], Barry Winn[2], Eugene Mamontov[2], Georg Ehlers[2], Stuart Calder[2], Joe Paddison[2], Fahima Islam[2], Huibo Cao[2], Ke An[2] and Leighton Coates[1]; [1]Oak Ridge National Lab, United States; [2]Oak Ridge National Laboratory, United States

The Second Target Station (STS) at the Spallation Neutron Source and its instrument system is being designed at Oak Ridge. The preliminary design of twelve instrument concepts were intensively developed and advanced during the instrument selection process that lasted nearly a year. These developments were in part facilitated by MCViNE, an open source, object-oriented Monte Carlo neutron ray-tracing simulation software package, which was used in simulation of optical designs and/or scientific outputs for a variety of the STS instrument concepts, including diffractometers, spectrometers, and multi-modal instruments. MCViNE was also employed to study the guide misalignment effects for selected instrument concepts. This presentation will review the recent development of the MCViNE software framework and its applications for STS instruments.

AP1.04.05
CUPI2D: Complex, Unique and Powerful Imaging Instrument for Dynamics Adrian Brugger[1], Hassina Z. Bilheux[2] and Jiao Lin[3]; [1]Columbia University, United States; [2]Neutron Scattering Division, United States; [3]Oak Ridge National Lab, United States

The Oak Ridge National Laboratory is planning to build a Second Target Station (STS) at the Spallation Neutron Source, which will host a suite of novel neutron scattering instruments. Among the first 8 instruments will be the state-of-the-art neutron imaging beamline, named the Complex, Unique and Powerful Imaging Instrument for Dynamics (CUPI^{2}D). CUPI^{2}D is designed for a broad range of applications that require length and time scales that expand from angstrom to micrometer, and min to hr, respectively. The beamline will be a world-class cold neutron instrument with the combination of Bragg edge imaging (BEI) and neutron grating interferometry (nGI). It will be positioned on a cylindrical moderator, which offers a high brightness of cold neutrons with a short pulse width, allowing for a ~0.3% wavelength resolution needed for BEI at a short ~30 m distance from the moderator, which accommodates a wide wavelength range of ~8 Å, ideal for nGI. The high flux (40 times higher than the First Target Station VENUS instrument for cold neutrons), wide wavelength range, and simultaneous (when needed) BEI and nGI capabilities will enable a broad range of scientific applications such as energy storage and conversion (batteries, fuel cells), materials science and engineering (additive manufacturing, superalloys, archaeometry), nuclear materials (novel cladding materials and nuclear fuel, moderators), cementitious materials, biology/medical/dental applications (regenerative medicine, cancer detection & morphology), and life sciences (plant nutrients in soil). This presentation gives an overview of the transformative scientific applications that will be possible at CUPI^{2}D including, for example, understanding chemical processes in Li-ion batteries and advanced energy storage materials which will enable future renewable energy grid; detecting defects and porosities in additive manufactured materials to advance 3D printing techniques; mapping strain evolution in engineering materials that are critical for renewing national infrastructure; illuminating fractures in nuclear cladding materials that are crucial in future nuclear energy technologies; and providing insights to soil-plant water dynamics for enhancing agriculture yield. Also included are a preliminary conceptual design and desired performance of the beamline.

Our co-author list is longer than the upper limit of 25 co-authors. Hence we include the full list of authors here: Adrian Brügger, Hassina Bilheux, Jiao Lin, George Nelson, Andrew Kiss, Jonathan Morris, Matthew Connolly, Ale Long, Anton Tremsin, Andrea Strzelec, Mark Anderson, Bob Agasie, Charles Finney, Martin Wissink, Mija Hubber, Roland Pellenq, Claire White, Brent Heuser, Aaron Craft, Jason Harp, Chuting Tan, Kathryn Morris, Ann Junghans, Sanna Sevanto, Jeffrey Warren, Fernando Esteban Florez, Ale Biris, Maria Cekanova, Nikolay Kardjilov, Burkhard Schillinger, Matthew Frost, Sven Vogel

AP1.04.06
The Larmor Phase Correction of MIEZE Fankang Li and Georg Ehlers; Oak Ridge National Laboratory, United States

Modulation of Intensity Emerging from Zero Effort (MIEZE) is a neutron resonant spin echo technique which allows one to measure the time correlation functions in materials by modulating the neutron beam using radio-frequency (RF) neutron spin flippers. The technique avoids neutron spin manipulation

between the sample and the detector, and thus could find applications in cases where the sample depolarizes the neutron beam. However, the finite sample size creates a variance in the neutron path length between the locations where scattering and detection happens, which causes the aberrations in Larmor phase. Such aberrations greatly limit the contrast in the intensity modulation in particular towards long correlation times or large scattering angles. We propose two approaches to correct such aberrations, which will enable us to extend those detection limits to longer times and larger angles. One approach involves the physical tilting of the RF flippers in the primary spectrometer with respect to the beam direction [1] and the other approach involves two magnetic Wollaston prisms in addition to the two RF flippers [2]. Both approaches can shape the wave front of the intensity modulation at the sample position to compensate for the path variance from the sample and the detector. The simulation results indicate that both approaches enable one to operate a MIEZE instrument at much increased RF frequencies, thus improving the effective energy resolution of the technique.

[1] R. Dadisman, G. Ehlers and F. Li, Modulation of intensity emerging from zero effort (MIEZE) with extended Fourier time at large scattering angle, Review of Scientific Instruments 93, 013301 (2022);
[2] F. Li, The linear phase correction of modulation of intensity emerging from zero effort (MIEZE) with magnetic Wollaston prisms, accepted by J. Appl. Crystal. 2022

AP1.04.07
Commissioning the New MacSANS Small Angle Neutron Scattering Instrument at the Canadian Neutron Beam Laboratory at the McMaster Nuclear Reactor Devin Burke[1], Patrick Clancy[1], Zin Tun[2] and Bruce D. Gaulin[1]; [1]McMaster University, Canada; [2]TVB Associates Inc., Canada

MacSANS is a new 24 m small angle neutron scattering instrument nearing completion in the subterranean neutron beam hall at the 5 MW McMaster Nuclear Reactor. I will describe the design and expected performance of the new SANS instrument, including a description of the new Mirrortron 100 x 100 cm^2 2D neutron detection system and sample stage capabilities. MacSANS will be operational in 2023 and will be supporting a robust user program addressing important questions related to nanostructure in a broad range of materials, including soft and biomaterials, quantum materials and energy materials.

AP1.04.08
Design of a Multi-Analyzer Neutron Scattering Spectrometer Using Ray-Tracing Monte-Carlo Simulations Adit Desai[1], Martin Mourigal[1], Garrett E. Granroth[2], Travis J. Williams[2], Adam Aczel[2] and Barry Winn[2]; [1]Georgia Institute of Technology, United States; [2]Oak Ridge National Laboratory, United States

Condensed matter physics was revolutionized by neutron spectroscopy when the triple-axis spectrometer was invented in 1956 and became a Nobel award-winning invention. By using neutron spectroscopy to measure the atomic and magnetic motion of atoms, information about phonons, spin-waves, and other quasiparticle dispersion curves can be developed. Since its creation, the triple axis spectrometer has been greatly improved, and the latest development in spectroscopy is the use of a multiplexing analyzer system, seen at other laboratories such as the Swiss Neutron Source's instrument CAMEA [1]. The novel technology seen in CAMEA is the ability to measure several energies at once. By using 8 energy "stations," each of which are a series of five analyzer panels set to analyze a different energy, CAMEA is able to greatly improve upon efficiency of the traditional triple-axis. Oak Ridge National Laboratory aims to utilize this technology, along with further developments, to create the latest development in triple-axis spectrometry known as the Multi-Analyzer Neutron Triple Axis (MANTA). An optimal way to design a neutron scattering instrument without exorbitant expense is to use Monto-Carlo simulation programs, such as McStas. Through this program various techniques have been used to determine the effectiveness of the design. By using various basic samples meant to model liquid Helium-4 or a basic photon dispersion, has been discovered that CAMEA was designed to maximize flux efficiency, so as a result, the energy resolution is not ideal. Indeed, a distortion effect of the resolution has been discovered, which could potentially result in analysis issues. As such, exploration into improving resolution at a minimal cost to flux has been explored. One major technique utilized thus far includes increasing station count from 8 to 12, and to 15. By increasing station count and tightening energy widths, resolution is improved as there are more energy "bins" in the same energy space. These techniques, along with other developments, aim to make MANTA a top-of-the-line instrument in neutron spectroscopy.

AP1.04.09
Virtual Learning Resources for Education and Training in Neutron Scattering Patrick Clancy[1], Greg Van Gastel[1], Symphony Huang[1], Evan Smith[1], Yijia Zi[1], Taryn McMillan[1], Zahra Yamani[2], Drew Marquardt[3], Young-June Kim[4], Maikel Rheinstadter[1] and Bruce D. Gaulin[1]; [1]McMaster University, Canada; [2]Canadian Nuclear Laboratories, Canada; [3]University of Windsor, Canada; [4]University of Toronto, Canada

The education and training of new neutron scatterers is an essential activity, but one which can be fraught with logistical challenges. Hands on training opportunities are often limited by the availability of neutron beamtime and the resources required to bring groups of students to a major neutron source. Such challenges have only been exacerbated by the ongoing COVID-19 pandemic. In this poster we will describe recent efforts to develop virtual learning resources for education and training in neutron scattering. In particular, we have created a series of virtual lab experiments which are targeted at upper year undergraduate students. These experiments can be individually incorporated into an upper year undergraduate lab course, or combined to form a one-semester practical introduction to neutron scattering. We have also developed two new virtual instruments that simulate neutron scattering beamlines at the McMaster Nuclear Reactor. This includes virtual versions of the McMaster Alignment Diffractometer (MAD) and the McMaster Small Angle Neutron Scattering facility (MacSANS).

**This project was supported by eCampusOntario and the Ontario Virtual Learning Strategy program*

AP1.04.10
Neutron Velocity Selector Design and Impact on Simulation William T. Higgins, Thomas Huegle, Kenneth C. Littrell, Franz X. Gallmeier and Georg Ehlers; Oak Ridge National Laboratory, United States

Modeling neutron instruments with McStas is facilitated greatly using templates and components both officially distributed and institution specific. Neutron instruments are subject to the quality of individual components, from the static and easily- simplified, such as guide components, to the multifaced and rotating, such as velocity selectors. To provide a monochrome beam of low-velocity neutrons (λ = 0 - 50 Å) most velocity selectors may use a bladed rotor, but a similar system can be achieved through a series of notched discs acting as choppers. Rotor-type and disk-type velocity selectors can be simulated in tens of lines of similar code. Both provide a monochromatic beam with the central wavelength specified by the user, within a subset of the range 0 - 50 Å, depending on selector geometry.
We compare the officially distributed rotor-type *Selector* component to one simulating a disk-type velocity selector intended to accurately model the component installed at the GPSANS instrument at ORNL. Both components provide monochromated beams having the desired central wavelength and comparable integrated flux, but the peak intensity and bandwidth differ. Disk-type spectra show lower peak intensity and wider bandwidth, indicating a better representation of GPSANS.
One feature of velocity selector components is the ability to introduce horizontal tilt in alignment to the beam center to vary the width of the distribution

around the central wavelength to shift the balance between flux and resolution. McStas simulations, however, show the central wavelength shifting to longer wavelengths in response to tilt, without the expected effect on resolution. Tilt induces a linear change of central wavelength in both, though magnitude differs. We discuss the simulation of these components, their accuracy, and address their divergence from expected behavior.

SESSION A2.05: Instrumentation: Hard Matter
Session Chair: Ken Herwig
Tuesday Morning, June 7, 2022
UMC West Ballroom 208

10:15 AM *A2.05.01
BIFROST: A Multiplexing Indirect Geometry Time-of-Flight Spectrometer for Extreme Environments Rasmus Toft-Petersen[1,2], Liam Whitelegg[2], Bjørn C. Hauback[3], Philippe Bourges[4], Christof Niedermayer[5], Henrik M. Ronnow[6], Kim Lefmann[7] and Niels B. Christensen[1]; [1]Technical University of Denmark, Denmark; [2]European Spallation Source, Sweden; [3]Institute for Energy Technology, Norway; [4]Laboratoire Léon Brillouin, France; [5]Paul Scherrer Institut, Switzerland; [6]Swiss Federal Institute of Technology Lausanne, Switzerland; [7]Niels Bohr Institute, Denmark

Inelastic neutron scattering is currently the only technique available to probe low-energy collective excitations in materials with sub-meV resolution. The technique has been instrumental in making key discoveries in a broad range of materials, such as superconductors, multiferroic materials and quantum magnets. However, the applicability of the technique is limited by the inherently weak signals produced in a measurement. When either magnetic moments, samples or merely the scattering cross sections become very small, experiments quickly become infeasible. In addition, since mapping of significant portions of (Q, ω)-space can take up to one week, parametric studies of entire dispersion relations as a function of temperature, pressure and magnetic field is restricted to the most forgiving samples. In overcoming these limitations, the triple axis community has improved the efficiency of triple-axis spectrometers by constructing multiplexing analyser arrays covering large intervals of scattering angles in the horizontal plane. The CAMEA spectrometer at the PSI in Switzerland, the BAMBUS back-end for PANDA at MLZ in Munich as well as the MACS spectrometer at NIST are all instruments that have successfully demonstrated the large gains of multiplexing on continuous neutron sources.

BIFROST, currently under construction at the European Spallation Source (ESS), employs a CAMEA-like variant of the multiplexing backend in conjunction with an indirect geometry time-of-flight (ToF) front end. Making full use of the long neutron pulses of the ESS, the primary spectrometer enables an unprecendented polychromatic sample flux exceeding 10^{10} n/s/cm^2 at 5 MW accelerator power, with a bandwidth of 1.7 Å. Due to the long primary flight path (162 meters), this can be achieved whilst retaining a primary spectrometer resolution $\Delta E_i/E_i$ of 4 %, common in cold neutron spectroscopy. The multiplexing backend consists of 9 Q-channels, each containing 5 fixed analysers probing a scattered neutron energy range of 2.7 to 5.0 meV. The analysers utilize the graphite crystal mosaicity combined with position sensitive neutron detectors to gain continuous scattering angle sensitivity. Small samples and position sensitive detection combine with a large analyser scattering geometry to narrow the acceptance energy band of a single pixel, resulting in a back-end resolution considerably better than on a classical TAS setup. By tuning the primary spectrometer resolution to match the backend, an elastic line resolution better than 30 μeV can be achieved at 2.7 meV, whilst retaining a respectable flux above 10^8 n/s/cm^2 at 2 MW.

The BIFROST design is thus optimized for small samples and for fast, high resolution mapping of a single scattering plane, making the instrument ideal for extreme environments. Harsher restrictions on sample sizes in pressure cells can be tolerated, due to the large flux. High field cryomagnets does not reduce the measured spatial angle, and the long primary flight path suppresses spurions from multiple scattering processes in the sample environment. I will present the design of BIFROST in general, focusing on the most novel aspects, and put the analyser design in perspective to the latest instrumental developments on continuous sources. I will conclude by discussing some of the science cases that the instrument could address, and new avenues for experimental investigation.

10:45 AM A2.05.02
Update on the Cold Spectrometer Project, PoLAR, at the NCNR Leland W. Harriger[1], Stephen D. Wilson[2], Jeffrey Lynn[1], Dan A. Neumann[1], Jeremy Cook[1], Donald Pierce[1] and nancy hadad[1]; [1]National Institute of Standards and Technology, United States; [2]University of California, Santa Barbara, United States

The NCNR is currently designing a new multi-axis cold neutron crystal spectrometer, PoLAR, to replace the SPINS cold triple axis on the NG-5 beamline. The performance of nearly all devices in the incident beam are coupled to the resultant flux shaping associated with the ballistic transport of neutrons along the 37m elliptic supermirror guide . These devices include a double focusing monochromator, velocity selector, V-cavity supermirror polarizer, resolution masks, and fast neutron filter. Each device was carefully modeled independently before integrating into the overall PoLAR Monte-Carlo design. The full suite of instrument parameters was then globally optimized to maximize the sample flux and the design of each device fine-tuned. In all cases, this careful analysis successfully leveraged the complicated flux profile to improve the performance of each device, often on par or above their performance level on a straight guide. This talk will outline the key design elements of each device operating within the incident beam, the strategy employed to optimize these elements, and there expected performance on the PoLAR instrument.

11:00 AM A2.05.03
Progress on the Design of Centaur, the Small- and Wide-Angle Neutron Scattering Diffractometer/Spectrometer at the Second Target Station of SNS Shuo Qian; Oak Ridge National Laboratory, United States

Centaur has been selected as one of the first eight instruments to be built in the Second Target Station of SNS at Oak Ridge National Laboratory. As a work-horse SANS instrument, it provides capabilities exceeding a traditional single-purpose instrument. The high flux, broad wavelength band, and large angular detector coverage provide simultaneous small/wide-angle neutron scattering and diffraction capability with a large dynamic range, thus enabling measurement of length scales across interatomic distances (diffraction) to hundreds of nanometers (small-angle scattering). Additionally-with the help of a high-speed Fermi chopper-it can be used as a direct geometry spectrometer for probing the dynamics of large length-scale structures.

We are designing Centaur to fill a capability gap in the current instruments at ORNL. Notably, the WANS and diffraction capability will be unique among SANS instruments in the United States. Centaur will enable in situ/operando experiments on materials with hierarchical architecture. This includes kinetic and/or out-of-equilibrium studies of phenomena with seconds time-resolution in many areas of materials research including soft matter, polymer science, geology, biology, and quantum condensed matter. The spectrometer mode extends the momentum transfer and energy transfer to a lower Q than those typically covered by other spectrometers and provides a unique capability for inelastic SANS experiments. Additionally, beam polarization will enable

detailed structural and dynamical investigations of magnetic materials and quantum materials.

In this presentation, we will present the progress on the preliminary design of the Centaur instrument. We invite the community to provide feedback and suggestions for the design and to envision future experiments to take advantage of its full capability.

11:15 AM A2.05.04
An Update on PIONEER, a Single-Crystal Neutron Diffractometer at the Second Target Station Yaohua Liu and Peter Torres; Oak Ridge National Laboratory, United States

PIONEER is a high Q-resolution single-crystal neutron diffractometer with the half-polarization capability at the Second Target Station (STS). It will enable scientists to study tiny crystals (0.001 mm^3) and ultra-thin films (10 nm) comparable to those typically used for x-ray studies but not feasible at existing neutron diffractometers. PIONEER will provide a variety of sample environments, including high/low temperatures, high magnetic fields, high pressures, and will cover a wide range of scientific fields, such as energy materials, quantum materials, and minerals physics. Here, we will report the recent instrument development, mainly on beam collimation and transportation, to achieve a high-flux, high-homogeneity, and truly horizontal beam and a low background to study small volume samples. To do this, PIONEER will use two Montel mirrors, also known as nested KB mirrors. Each Montel mirror has two elliptical mirrors arranged perpendicular and side-by-side to transport neutrons. We have performed Monte Carlo ray-tracing simulations and considered the gravity effect to investigate the impact of mirror geometries on beam profiles at the sample position. We have found a competing effect between the thermal neutron transport efficiency and the cold neutron homogeneity. We have also studied kinked beamlines, where the two Montel mirrors tilt differently. A kinked beamline helps reduce the background by moving the sample out of the direct line-of-sight to the moderator and provides an additional knob to optimize the brilliance transfer efficiency and beam homogeneity, which offers an optimal solution for PIONEER.

This research used resources of the Spallation Neutron Source Second Target Station Project at Oak Ridge National Laboratory (ORNL). ORNL is managed by UT-Battelle LLC for DOE's Office of Science, the single largest supporter of basic research in the physical sciences in the United States.

11:30 AM A2.05.05
Development of the Multi-Analyzer Neutron Triple Axis (MANTA) Spectrometer at ORNL Travis J. Williams[1], Garrett E. Granroth[1], Adam Aczel[1], Barry Winn[1], Adit Desai[2], Marcus Daum[2] and Martin Mourigal[2]; [1]Oak Ridge National Laboratory, United States; [2]Georgia Institute of Technology, United States

The Multi-Analyzer Neutron Triple Axis (MANTA) Spectrometer is a cold-neutron triple axis instrument currently in development as part of the planned upgrade of the HFIR instrument suite. This will replace the current CTAX instrument at HFIR and will boast a factor of 50 improvement in the incident neutron flux, positioning MANTA as a world-class cold-neutron instrument. This is achieved through a better position on the cold neutron guide, as well as newly-optimized neutron optics through rigorous McStas simulations of the neutron guide. Work is continuing on other instrument components, including a neutron velocity selector, monochromator and upgraded sample table. The project scope is also planned to include a dedicated 15T vertical field magnet to increase the sample environment capabilities of the instrument over the current CTAX instrument and an incident beam v-cavity for polarization analysis. The existing CTAX back-end will be re-located to MANTA but will be modified to include a v-cavity for the scattered beam, as well as space to accommodate Wollaston prisms and spherical neutron polarimetry options. Finally, work is ongoing to design a multiplexing analyzer-detector backend as part of future instrument upgrades, which includes McStas simulations of phonon dispersions to validate the design criteria.

11:45 AM A2.05.06
Upgrade of the BT-8 Diffractometer for Stress and Texture Thomas Gnaupel-Herold[1], Justin Milner[2] and Ed Binkley[1]; [1]NIST, United States; [2]NASA GRC, United States

The upgrade of the BT-8 diffractometer is nearing completion. As of December 2021, procurement, component fabrication and assembly are > 95% complete, and all old equipment has been removed. The installation of new components is >50% complete, and substantial progress is expected in the coming months. The new instrument features two double focusing monochromators: one of the single wavelength type (Si(400) or Si(311)) and a novel triple wavelength composite device for simultaneous Si(311)/(400)/(422) diffraction. The new detector is an array of high pressure (10 bar 3He) tube detectors with an effective area of 25 cm (w) x 38 cm (h). Spatially resolved measurements with multiple diffraction peaks are optimized through apertures with curved slots (conical slits) specific for a given combination of material and wavelength. A conventional single opening aperture is available as well. Two stages for sample positioning and orientation are installed: Small to medium weight samples <20kg are mounted on a large phi-chi goniometer with XYX-table. The goniometer eliminates the need for sample remounting and the table can be used as sample changer for high-throughput texture measurements. Large samples up to 200 kg weight can measured using the base XYZ-table after removing the top goniometer. The sample environment consists of two multi-axial straining devices aimed at the study of multi-axial deformation in sheet metal.

12:00 PM A2.05.07
Polychromatic Multiplexing Stress-Strain Diffractometer Sean Fayfar[1], Jay T. Cremer[2] and Boris Khaykovich[1]; [1]Massachusetts Institute of Technology, United States; [2]Adelphi Technology, Inc., United States

The development of new materials for advancing technologies such as electric cars and clean energy production requires improvements in characterization techniques, especially with in-situ measurements during synthesis. Neutron scattering techniques have been gaining popularity with the development of new sources and capabilities in smaller facilities. However, these small sources lack sufficient neutron flux for many applications. We present our development of an efficient stress-strain diffractometer intended to be optimized for smaller research reactors such as the MIT Reactor and laboratory-based neutron generators but also suitable for large sources like SNS. The diffractometer will utilize a polychromatic neutron beam to significantly increase the flux at the sample, and a series of bent perfect crystals will be used as analyzers rather than monochromators. The silicon crystal analyzers are relatively transparent to neutrons, which has the benefit of multiplexing capabilities with analyzers placed in subsequent order. Multiplexing is also accomplished by placing analyzers at $2\theta_s=\pm 90°$, which has the capability to measure several components of the stress tensor at once. We will present neutron ray-tracing simulation results for this instrument configuration and report on the current state of the construction of a prototype instrument at the MIT Reactor.

We acknowledge useful discussions with A. Stoica. This work is supported by the U.S. Department of Energy (DOE), Office of Science, under a Small Business Technology Transfer (STTR) Grant, Grant No. DE-SC0020555.

12:15 PM A2.05.08
Concept for a Hybrid Neutron Diffraction/ Small Angle Scattering Instrument for Nuclear Energy Applications Kenneth C. Littrell and Georg Ehlers; Oak Ridge National Laboratory, United States

Neutron diffraction (ND) and small-angle scattering (SANS) can provide valuable information about materials and processes in many parts of the nuclear energy ecosystem. The sensitivity of neutrons to magnetism and isotope differences—especially the high sensitivity to and ability to differentiate between isotopes of hydrogen—make neutrons particularly valuable as a probe. Furthermore, the low absorption of neutrons in many materials, including some heavy elements and structural or refractory materials, allow neutrons to be used to measure radiologically shielded samples and samples in complicated, structurally robust environments that simulate nuclear reactor operating or fuel processing conditions. Examples of systems where neutrons have provided valuable information include radiation damage, thermal aging, and hydride evolution and their repair in reactor pressure vessel steels, fuel and fuel cladding materials and other reactor structural materials and waste storage materials, solution structure of solvent extraction systems for fuel reprocessing, and mineral formation and corrosion in waste storage tanks. However, the publication-focused competitive process with reviewers mostly from outside the community together with the needs for more complicated sample environment and sample handling and transportation procedures limits the access to neutron scattering techniques to this community through general scientific user programs.

The proposed reactor pressure vessel replacement to extend the life and enhance the utility of the Oak Ridge National Laboratory High Flux Isotope Reactor offers a unique opportunity to make a hybrid ND/SANS Oak Ridge Nuclear Energy Research Instrument to specifically serve this community's need. As a part of this upgrade plan, the existing neutron scattering instruments or their successors would be moved outside the reactor's regulatory boundary into a new thermal guide hall with lower background and fewer special constraints, potentially allowing for significant gains in instrument performance. At the same time, this will free up two beam tubes inside the reactor beam room for other applications, one of which could be used for this instrument. Being inside the reactor's regulatory envelope, this instrument would benefit the nuclear energy community by convenience of transfer between irradiation facilities, proposed hot-cell prep labs, and the instrument together with a good infrastructure for the safe handling of radioactive samples and special materials. This presentation will provide a sketch of what such an instrument might look like and what sample environments would be needed, with examples of measurement on relevant system that have been used to develop the initial specifications.

SESSION A3.06: Instrumentation: Soft Matter and Imaging
Session Chair: John Barker
Wednesday Morning, June 8, 2022
UMC West Ballroom 208

10:15 AM *A3.06.01
Performance Upgrades to the BT-2 Neutron Imaging Facility Jacob M. LaManna[1], Michael Cyrus Daugherty[1,2], Youngju Kim[1,2], Eli Baltic[1], Daniel S. Hussey[1] and David Jacobson[1]; [1]National Institute of Standards and Technology, United States; [2]University of Maryland, United States

The BT-2 Neutron Imaging Facility was commissioned in 2006 and has been the imaging workhorse for the user program at the NIST Center for Neutron Research (NCNR). With changing research topics and the addition of the simultaneous neutron and X-ray tomography (NeXT) capabilities in 2015, significant upgrades are required to improve the performance of the instrument and modernize it. Experiments aimed at investigating dynamic phenomena require ever increasing temporal resolution. To reduce the required image acquisition time the primary collimator will be expanded from 2 cm to 3 cm for about a 3x increase in neutron flux. A new neutron shutter will be installed to reduce the streaming field inside the instrument. The NeXT system will be upgraded with new motorized motion axes for faster, more accurate and repeatable positioning and the X-ray tube will be upgraded from 90 keV to 225 keV peak energy. The higher peak energy of the tube will allow the X-rays to penetrate more metal than currently possible allowing the use of larger pressure vessels and sample environments. The new tube also offers a 3 µm spot size for improved resolution and higher flux when imaging fast is required for dynamic experiments. New detectors have been developed and fabricated to improve image quality and reduce tomography reconstruction artifacts. The new detectors reduce component misalignments and allows the camera rotation to be fine-tuned to reduce detector tilt. The rotation capability also allows the cameras with rectangular image chips to be rapidly swapped between portrait and landscape modes for easy tailoring of the field-of-view to the sample size. This talk will give an overview of all current upgrades in progress for BT-2 and the timeline for installation during the 2023 NCNR cold source upgrade.

10:45 AM A3.06.02
INFER: Dark-field Tomography of Hierarchical Structures Daniel S. Hussey[1], Caitlyn M. Wolf[1], Youngju Kim[2], Sarah M. Robinson[1], Michael Cyrus Daugherty[2], Ryan P. Murphy[1], Paul A. Kienzle[1], Nikolai N. Klimov[1], Michael G. Huber[1], Peter N. Bajcsy[1], David Jacobson[1], Jacob M. LaManna[1] and Katie M. Weigandt[1]; [1]National Institute of Standards and Technology, United States; [2]University of Maryland, United States

Hierarchical structures – electrodes, geology, concrete, bones, additively manufactured materials – pose a challenge to characterization since length scales of interest span from the sub-nm to the meter (or kilometer in geology). The dark field contrast of a neutron grating far field interferometer (FFI) provides a means to measure the pair correlation function in the range 1-104 nm averaged over voxels that are about 50 µm. NIST is working to realize a prototype instrument that is capable of measuring dark field tomograms at ~100 auto correlation lengths within a day of measurement to enable the study of hierarchical structures that (slowly) evolve. With a FFI one can tune the period of the interference pattern by orders of magnitude, so that the sample position (and hence resolution) is the same for a broad range of autocorrelation lengths. Our first-generation phase gratings are providing good visibility, over 60%. To realize the full FFI tunability, we have developed a neutron source grating whose period can be adjusted on the fly. Recent advances in camera and lens technology enable measurement times of a monochromatic beam ($\Delta\lambda / \lambda \sim 15\%$) of about 2 s. Applying dose reduction tomographic reconstruction algorithms, it is anticipated that 90 projections will be sufficient for a high-quality reconstruction. We have been studying phantom objects in two dimensions to quantify the measurement quality. Assuming that the NCNR recovers operation, we will also present early measurements of three-dimensional phantoms.

11:00 AM A3.06.03
Upgrade of the Neutron Spin Echo Spectrometer at the NIST Center for Neutron Research Antonio Faraone[1], Norman Wagner[2], Michihiro Nagao[1,3,2], Christoph Brocker[1,3], Nicholas C. Maliszewskyj[1], Michael Monkenbusch[4], Olaf Holderer[4], Tadeusz Kozielewski[4] and Dan A. Neumann[1]; [1]NIST Center for Neutron Research, United States; [2]University of Delaware, United States; [3]University of Maryland, United States; [4]Jülich Centre for Neutron Science, Germany

Neutron Spin Echo (NSE) spectroscopy measures the dynamics of materials on longest time scales among neutron spectroscopic techniques. Currently instruments in the US routinely reach Fourier times of 100 ns. However, recent advances in the optimization of the precession field significantly increase the field integral homogeneity and therefore the maximum Fourier time, as already implemented on IN-15[1)] at Institut Laue-Langevin and J-NSE-Phoenix[2)] at Heinz Maier-Leibnitz Zentrum. The University of Delaware, in collaboration with the National Institute of Standards and Technology Center

for Neutron Research (NCNR), has received funding from the National Science Foundation through the Mid-scale Research Inflastructure-1 program to acquire, assemble and commission a new NSE spectrometer employing optimally designed superconducting precession coils developed for the J-NSE-Phoenix, increasing the maximum Fourier time 2.5x. The installation of the new instrument is planned for 2023 during an outage of the NCNR to install a new D_2 cold source, which will provide about a factor 2 increase in the neutron flux at long wavelength regions. Taking advantage of the new design, the increased flux provided by the new cold source, and a number of instrument elements optimized for long wavelength operation, a Fourier time of 300 ns should be achieved routinely, with the possibility of reaching 700 ns for strongly scattering samples.
As the planned upgrade extends the longest accessible time, the upgraded instrument will expand the opportunities for the studies various materials systems especially in soft matter fields.

References:
1) B. Farago, P. Falus, I. Hoffman, M. Gradzielski, F. Thomas, C. Gomez, Neutron News. 26, 15 (2015).
2) S. Pasini, O. Holderer, T. Kozielewski, D. Richter, M. Monkenbusch, Review of Scientific Instruments 90, 043107 (2019).

11:15 AM A3.06.04
The Quite Intense Kinetics Reflectometer (QIKR) at the Spallation Neutron Source (SNS) Second Target Station (STS) John F. Ankner, Danielle Wilson, Rudy Thermer, Scott Dixon and Zeke Salazar; Oak Ridge National Laboratory, United States

QIKR will be a general-purpose, horizontal-sample-surface reflectometer. By exploiting the brilliance and broad wavelength bandwidth of the STS, QIKR will collect specular and off-specular reflectivity data faster than the best existing such machines. Utilizing pulse skipping, it will often be possible to collect complete specular reflectivity curves using a single instrument setting, enabling “cinematic” operation, wherein the user turns on the instrument and “films” the sample. Samples in time-dependent environments (e.g. temperature, electrochemical, magnetic, or undergoing chemical alteration) will be observed in real time, in the most favorable cases with frame rates as fast as 1 Hz. Cinematic data acquisition promises to make time-dependent measurements routine, with time resolution specified during post-experiment data analysis. This capability will be deployed to observe such processes as *in situ* polymer diffusion, battery electrode charge-discharge cycles, magnetic and other hysteresis loops in real time, and membrane protein insertion into lipid layers. We will present the current instrument design concept and solicit user input for QIKR, which is expected to begin operation early in the next decade.

11:30 AM A3.06.05
Recent Advances at the Cold Neutron Imaging Instrument at High Flux Isotope Reactor Yuxuan Zhang[1], Hassina Z. Bilheux[1], Erik Stringfellow[1], Jean Bilheux[1], Jonathan Smith[1], Les Butler[2], Kyungmin Ham[2], Wieslaw Stryjewski[2] and Michael Vincent[2]; [1]Oak Ridge National Laboratory, United States; [2]Louisiana State University, United States

Neutron imaging is a non-destructive technique that can spatially resolve internal features/structures in bulk materials. Given the unique interaction of neutrons with the matter, it provides high penetration in metals and high sensitivity to light elements (H, Li, etc.). Therefore, neutron imaging provides unique complementary contrast to X-ray imaging in many research fields. In this presentation, recent advances in improving the spatial resolution from ~100 mm to 20-30 mm at the cold neutron imaging instrument (CG-1D) at the High Flux Isotope Reactor (HFIR) will be discussed. A few scientific examples of how such improvement would further facilitate material characterizations will be presented. Additionally, the effort has been focused on implementing a Talbot-Lau grating interferometer at CG-1D, which will enable simultaneous access to three imaging modalities: attenuation, differential phase contrast, and scattering. Recent progress will be presented.

Acknowledgments
This research was sponsored by the Scientific User Facilities Division, Office of Basic Energy Sciences, U.S. DOE. This research used resources at the Spallation Neutron Source and the High Flux Isotope Reactor, U.S. Department of Energy (DOE) Office of Science User Facilities operated by the Oak Ridge National Laboratory.

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11:45 AM A3.06.06
Angle-encoding Radiography with Neutrons Sam McKay[1,1,1], Fankang Li[2], David V. Baxter[1,1,1] and Roger Pynn[1,1,1]; [1]Indiana University, United States; [2]Oak Ridge National Laboratory, United States

We introduce the technique of *angle-encoding radiography* (AER), a neutron imaging technique that uses polarized neutrons to surpass the usual *geometric resolution* $R = L_s / (L_d / D)$ of a neutron imaging beamline, where L_s and L_d are the distances from sample to detector and source to detector, respectively, and D is the source aperture size. We show that the fundamental resolution of AER is primarily dependent on the wavelength uncertainty.
The technique is performed using *Larmor labelling*, in which the neutron trajectory is labeled via its spin; AER generates a sinusoidal intensity modulation as a function of the neutron propagation angle. The frequency of this modulation depends on the magnetic field experienced by the neutron. By measuring at several different modulation frequencies, a Fourier decomposition of the sample’s transmission function can be created within each detector pixel.
We show both analytically and using McStas ray-tracing simulations that one can implement one-dimensional AER using suitably focused *magnetic Wollaston prisms*, birefringent neutron polarization devices based on superconducting technology [1]. We discuss the transmission function reconstruction procedure and generalizations of the technique to two and three-dimensional transmission functions.
We compare and contrast this technique to the related techniques of neutron grating interferometry and spin echo small angle neutron scattering (SESANS).

[1] F. Li et al. *Review of Scientific Instruments*, (2014)

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12:00 PM A3.06.07
Dynamic Microfluidic Modulation of Neutrons and x-rays Ryan P. Murphy, Sarah M. Robinson, Jacob M. LaManna, Caitlyn M. Wolf, Youngju Kim, Michael Cyrus Daugherty, Michael G. Huber, Peter N. Bajcsy, David Jacobson, Paul A. Kienzle, Katie M. Weigandt, Daniel S. Hussey and Nikolai N.

Klimov; National Institute of Standards and Technology, United States

Efficient spatial modulation of visible light revolutionized computational imaging methods, such as super resolution microscopy and ghost imaging. Meanwhile, the spatial modulation of neutrons and x-rays remains an ongoing challenge, mostly due to the inherently high transmission through most materials. Here, we propose a new approach referred to as dynamic microfluidic modulation, which spatially modulates the periodicity and transmission of incident neutron and x-ray beams. An efficient microfluidic modulator requires a robust and scalable method of actuation, as well as precision flow control of a fluid that strongly absorbs neutrons and x-rays. We demonstrate one such microfluidic approach by combining high-speed nanofluidic jetting, spontaneous capillary transport, and nanofabricated silicon substrates. The scalability and generality of this approach is demonstrated by individually actuating hundreds of high aspect ratio microchannels. Currently, the spatial modulation of neutrons and x-rays spans from several micrometers (channel scale) up to several centimeters (substrate scale) with nanometer precision. Details of the operating principles, preliminary neutron and x-ray measurements, and the cycle throughput are presented and discussed. Lastly, we demonstrate the applicability of this approach by creating a reconfigurable source grating as part of a neutron interferometric microscope (INFER) under development at NIST.

12:15 PM A3.06.08
The VENUS iMaging Beamline Construction Project at the Spallation Neutron Source Hassina Z. Bilheux, Tommy Thomasson, Aaron Hanks, Amy Byrd, Amy Jones, Harley Skorpenske, Erik Stringfellow, Bill McHargue, Irina Popova, Franz X. Gallmeier, Jean Bilheux, Ryan Mangus, Scott Keener and George Rennich; Oak Ridge National Laboratory, United States

The Oak Ridge National Laboratory is building the neutron-imaging instrument named VENUS (**VE**rsatile **N**eutron Imaging Instr**U**ment at the **S**pallation Neutron Source) that will uniquely utilize the Spallation Neutron Source (SNS) intrinsic time-of-flight (TOF) capabilities to measure and characterize large-size and complex systems. VENUS is optimized for the measurement of micro-scale structures utilizing both radiography (2D) and computed tomography (3D). The broad range of science enabled at VENUS comprises energy materials, materials science and engineering, biology, geosciences, etc. The beam is optimized for time-of-flight (TOF) imaging techniques, i.e., Bragg edge and resonance imaging, although it can also function in a similar fashion as a reactor-based imaging beamline.
Installed on beamline 10, VENUS views a decoupled poisoned H_2 moderator and can utilize a large wavelength range of neutrons between ~ 0.001 (this wavelength is limited by the time resolution of the beamline, not by the capability of the neutron source) and 10 Å. The flightpath from source to detector is at a maximum distance of 25 m. The beamline can be broken down into three main sections: i) the front-end/upstream section, ii) the instrument cave, and iii) the radiological materials area (RMA) and control hutch. The front-end section is surrounded by high density concrete shielding and are composed of (from upstream to downstream) a core vessel insert (CVI), a shutter insert (SI) housing the fixed aperture, variable apertures, with the largest aperture diameter yielding the highest flux, followed by choppers, beam collimators and filters. The instrument cave houses flight tubes, sample stages and sample environment, along with detectors that are followed by a get-lost-tube and beam stop. The RMA and control hutch, respectively utilized for sample preparation instrument operations, are located outside of the shielded instrument areas.
Neutrons from the moderator pass through apertures of various sizes, selected by the user, a set of collimators, then through tapered flight tubes (equipped with beam scrapers), and finally through a sample to be detected by either a micro-channel plate (MCP) detector, a charge-coupled device (CCD) system, or a scientific Complementary Metal Oxide Semiconductor (sCMOS) detector. These detectors will be mounted on a translation stage to facilitate the change of detectors during routine operations of the instrument. The MCP and sCMOS detectors can be triggered using the accelerator timing signal. A timing delay is also necessary to account for the time it takes neutrons to travel from the source to the detector. Between components, neutrons are transported in flight tubes that can be under vacuum or He-filled.
VENUS is currently under construction with an anticipated early completion date of January 2024. This presentation provides an update of the construction project and examples of scientific research, demonstrated at the SNS SNAP beamline, that will be performed at the instrument.

A portion of this research used resources at the Spallation Neutron Source, a DOE Office of Science User Facility operated by the Oak Ridge National Laboratory.

SESSION A3.07: Instrumentation: Sample Environment
Session Chair: Sergiy Gladchenko
Wednesday Afternoon, June 8, 2022
UMC West Ballroom 208

2:00 PM *A3.07.01
Advances in High-Pressure Neutron Scattering at Oak Ridge National Laboratory Mary-Ellen Donnelly; Oak Ridge National Laboratory, United States

Pressure is a powerful tool that can enable the formation of exotic states of matter and the synthesis of novel materials, tune the parameter space of a materials phase diagram and can replicate the conditions within the Earth and other planetary bodies. The application of pressure is commonly coupled with *in situ* methods such as x-ray scattering and infrared/Raman spectroscopy. One *in situ* technique where application of pressure is of extreme interest is neutron scattering as the neutron's scattering properties can allow for the determination of light elements within materials, the study of magnetic properties and comprehensive studies of the atomic vibrations. The weak interaction of the neutron, however, creates an issue in the ability to study samples with *in situ* neutron scattering at high pressure as large volumes are required which generally limit pressure regime available.

At Oak Ridge National Laboratory's two neutron sources – the High Flux Isotope Reactor (HFIR) and Spallation Neutron Source (SNS) – a large variety of extreme environments for *in situ* high-pressure studies is available ranging from large volume gas cells to small volume diamond anvil cells. As part of this talk, I will give a general overview of the current developments in our high-pressure sample environments across the instrument suites. This will be followed by a more in-depth description of some of these current developments. One of these will be the extension of the 'workhorse' of high-pressure neutron scattering – the Paris-Edinburgh press – to the diffraction and spectroscopy beamlines of WAND^2 and ARCS at the HFIR and SNS, respectively. The first of these enables the ability to study materials above 10 GPa the HFIR, whilst the latter allows for the determination of the phonon density of states under high pressure. The other focus topic will be recent developments in diamond anvil cell technology and analysis. These now allow for studies at megabar pressures and techniques are also becoming adapted to studies on current materials of interest such as superhydrides.

This research used resources at the High Flux Isotope Reactor and the Spallation Neutron Source, both DOE Office of Science User Facilities operated by the Oak Ridge National Laboratory.

2:30 PM A3.07.02
A Pre-Conceptual Design of a 20 – 25 T Vertical-Field Split Magnet for Neutron Scattering Mark Bird[1], Scott Bole[1], Ken Herwig[2], Dylan Kolb-Bond[1] and Jack Toth[1]; [1]NHMFL - FSU, United States; [2]ORNL, United States

In recent years the fields available from solenoid magnets for condensed matter physics and nuclear magnet resonance have risen from 22 T to 32 T and from 23.5 T to 28.2 T, respectively due to the use of the high temperature superconducting (HTS) materials. The fields available from split magnet suitable for neutron scattering have been fixed at ~15 T for ~20 years.

A magnet development team at the National High Magnetic Field Laboratory has undertaken a pre-conceptual design of a vertical field split magnet suitable for neutron scattering that is based on the HTS magnet technology used in the MagLab's 32 T all-superconducting magnet. The mid-plane of the magnet was specified to include 8 equally spaced scattering ports. Six are to be 25° wide with two being 26.5° each. The split is 12 mm on axis and includes a +/- 5° vertical take-of angle. We assume the sample will be a right circular cylinder 10 mm tall with diameter also 10 mm. Two port shapes were considered: elliptical and rectangular, both of which allow viewing of the entire sample.

Design work focused on structural analysis of the mid-plane spacer that provides the scattering space and supports that electromagnetic clamping force between the two halves of the magnet and optimization of the HTS coils to maximize field while maintaining limits on peak mechanical strain, fraction of critical current, etc. The coil lengths were also optimized to distribute the clamping pressure on the mid-plane spacer to minimize its thickness.

Results indicate the elliptical ports result in lower stress in the mid-plane spacer than the rectangular ones do which allows the spacer to be thinner which, in turn results in higher field. The rectangular ports allow larger solid angle for neutron scattering but at a price of 2 T lower central field. The results of the design process are presented.

2:45 PM A3.07.03
RheoSurfR – Neutron Reflectivity-Rheology Sample Environment for Studying Soft Matter, Biology, and Materials Processing at Air-Liquid and Liquid-Liquid Interfaces Benjamin R. Thompson[1], Mason Keresty[2], Hannah Nevel[2], Richard Dombrowski[2] and Norman Wagner[1,2]; [1]University of Delaware, United States; [2]STF Technologies LLC, United States

Researchers amongst the scientific and industrial communities have an unmet need of the ability to analyze a broad range of soft matter i.e., polymers, complex fluids, colloids, and biological materials i.e., hydrogels, proteins, and lung surfactants, across a range of processing or biologically relevant conditions. A current commonly utilized method consists of a Langmuir-Pockels trough, along with neutron reflectivity. However, this ignores the issue of the mixed flow fields present during interfacial deformations on the trough, of which decoupling the contributions of compression and shear is a considerable challenge.[1] Separating the compressional and shear components when deforming an interface is a topical area of research, with a radial trough developed by Pepicelli *et al.* in 2017 which demonstrated interfacial deformations in a pure compressional regime.[2] Our work further expands on this by using a novel instrument that can *independently* control compression *and* shear. It can be used standalone in a research lab and, importantly, can be placed onto a beamline to allow for simultaneous measurement of surface moduli and microstructure *via* neutron reflectivity. In this talk, I will describe the current progress made with this sample environment, as well as show some of the instrument capabilities. This work is funded under a DOE STTR Award: DE-SC0020534.
[1] Jaensson, N.; Vermant, J. Tensiometry and Rheology of Complex Interfaces. *Curr. Opin. Colloid Interface Sci.* **2018**, *37*, 136–150.
[2] Pepicelli, M.; Verwijlen, T.; Tervoort, T. A.; Vermant, J. Characterization and Modelling of Langmuir Interfaces with Finite Elasticity. *Soft Matter* **2017**, *13* (35), 5977–5990.

3:00 PM A3.07.04
4D Rheo-SANS: A Novel Sample Environment for Measuring Structure-Property Relationships in Soft Matter and Biological Materials Nathan Alexander[1], Jonathan Kopf[2], Benjamin R. Thompson[1], Richard Dombrowski[2] and Norman Wagner[1,2]; [1]University of Delaware, United States; [2]STF Technologies LLC, United States

Small Angle Neutron Scattering under flow, along with "ultra" and "very" SANS variants, has proven to be a powerful tool for the development of structure-property relationships in complex fluids and soft matter. Designing soft materials for specific functionalities is critical to a wide variety of scientific disciplines, industrial processes, and consumer products. To obtain a desired material performance or process often requires a complex process with specific thermal and shear history to create a specific nano/microstructure with particular functionality. Some important biological processes are also affected or even triggered by a shear flow. Some practical examples range from the conductive inks necessary in battery and solar cell manufacturing to food products such as ketchup and mayonnaise, to shampoos and liquid detergents, paper coatings, and tissue engineering and the stability of biopharmaceuticals during processing and delivery. Therefore, better instruments are required to quantitatively measure the shear deformation of the microstructure using SANS techniques simultaneously with accurate measurements of the mechanical responses under well-defined flow conditions. Importantly, the symmetry of the material breaks under shear and the scattering patterns from three independent shear planes (flow-velocity gradient, flow-vorticity, and velocity gradient-vorticity planes) are required to fully characterize the material structure. However, the current level of instrumentation lacks the capability of studying all three planes concurrent with rheological properties.
In this work, under the auspices of a DOE STTR Program *DE-SC0019595*, a novel sample environment is developed to measure the full 3D structure with enhanced time resolution, thereby creating a 4D measurement. Designed to be a commercially available standard instrument for use on beamlines world-wide, Large- and small-amplitude oscillation and steady shear measurements are performed on a range of complex fluids spanning typical material behaviors. The range of applicability for quantifying the microstructure under well-defined shear flow histories along with the range of measurable mechanical responses are presented along with some example measurements.

3:15 PM A3.07.05
Rapid and Controllable Cooling of High-Temperature Neutron Furnace Yue Xiao[1], Chien-Hua Chen[1], Patryk Radyjowski[1], Max Demydovych[1], Chad Burkholder[1] and Rebecca A. Mills[2]; [1]Advanced Cooling Technologies, Inc, United States; [2]Oak Ridge National Laboratory, United States

The development of neutron science has resulted in the increased need for high-temperature furnaces for neutron scattering experiments. However, the common Institut Laue-Langevin (ILL) type furnaces suffer from hours long cooling times due to ineffective radiation heat transfer at lower temperatures. To resolve this issue, we designed a rapid cooling system using helium (He) gas circulation inside the neutron furnace. Particularly, the He gas is introduced within the radiation shielding and cools both sample and radiation shields directly via He sensible heat capacity. As a result, the cooling time from 400 °C to 100 °C of 11 min has been demonstrated in our prototype furnace. This is an improvement over >6 h for radiation-only cooling and ~1 h for bulk He gas circulation outside the radiation shields. With our approach, the cooling rate can also be controlled by adjusting the He flow rate. A near-constant cooling rate (0.3 °C/s) has been demonstrated. Transient CFD simulations are also performed and the results show an excellent agreement to the

experimental data.

SESSION AP3.08: Poster Session II: Advances in Neutron Facilities, Instrumentation and Software II
Session Chair: Hassina Bilheux
Wednesday Afternoon, June 8, 2022
5:30 PM - 7:30 PM
UMC Center Ballroom 210

AP3.08.01
The Levitation Suite at ORNL Dante Quirinale; Oak Ridge National Laboratory, United States

Performing scattering experiments at high temperatures can lead to complications, such as difficulties in selecting a crucible, containment failure, or large furnace background contributions. To counter these difficulties and expand the science possible on the beamline, the Neutron Sciences Directorate at ORNL has been developing a suite of levitation sample environments. Here will be presented an overview of current capabilities, updates on developing containerless processing chambers, and new capabilities coming online over the next year.

AP3.08.02
McStas Component Development at the Spallation Neutron Source and High Flux Isotope Reactor Matthew J. Frost, Garrett E. Granroth, Thomas Huegle and Lee Robertson; Oak Ridge National Laboratory, United States

The HFIR Beryllium Reflector Replacement (HBRR) Project and Second Target Station (STS) projects at Oak Ridge National Laboratory have provided inspiration to develop, optimize and integrate new optical components into the next generation of scattering instrument concepts. Of note is the testing and design of true ellipsoidal mirrors, long elliptical reflectors, and octagonal cross-section guides. In addition, new methods have been devised by which to quantify performance in order to optimize designs for a new generation of scattering instrumentation. A summary of these developments and their applications will be presented.

AP3.08.03
Maximizing Detector Count Rate and q-Range on Pin-Hole SANS Instruments at the NCNR John G. Barker; National Institute of Standards and Technology, United States

Calculations and measurements show that the detector count rate at fixed q-resolution and over a fixed q-range can be increased considerably by simultaneously increasing the overall size scale of several instrument parameters such as apertures, separation distances, beam stops and detectors. In addition, the typical q-range can also be extended by adding larger angle detectors. The design and calculated performance of different detector concepts such as picture frames or curtains are presented. Practical methods for enhancing count rates and q-range on existing SANS instruments at the NCNR are described.

AP3.08.04
Overview of the Polarized 3He Program at the Oak Ridge National Laboratory Chenyang Jiang; Oak Ridge National Laboratory, United States

Polarized neutron scattering is an indispensable technique in modern neutron scattering, which takes advantage of the unique property of the neutron spin and its interaction with matter. To date, every major neutron scattering facility in the world is committed to providing polarized neutron capability to general users. Nuclear spin-polarized ^{3}He gas has large spin dependent neutron absorption cross sections, and polarized 3He based neutron spin filters have been widely used to polarize or analyze neutron beams worldwide. Compared to other commonly used neutron polarizing techniques like Heusler crystals and polarizing supermirrors, polarized 3He has several unique features: First, it can work over a wide neutron wavelength/energy range. Second, it can accept large divergent neutron beams while not changing the beam divergence. Third, it can be used as a high-efficiency neutron spin flipper by flipping the 3He polarization through nuclear magnetic resonance (NMR). Because of these advantages, over the last decade the Oak Ridge National Laboratory (ORNL) has invested many resources in the development of polarized 3He systems based on spin-exchange optical pumping (SEOP). In particular, great efforts have been made in developing in situ polarized 3He systems, with which the 3He polarization will be kept stable throughout the whole experiment on the beamline where it is deployed. Different sized in situ systems have been built to suit the needs of each individual neutron instrument that has requested polarized 3He. We will show the design, construction and performance of these systems.

AP3.08.05
Real-Time Control and Feedback of Hyperspectral Neutron Computed Tomography at the Spallation Neutron Source Shimin Tang[1], Mohammad Samin Nur Chowdhury[2], Diyu Yang[2], Singanallur Venkatakrishnan[1], Charles Bouman[2], Gregery Buzzard, T[2] and Hassina Z. Bilheux[1]; [1]Oak Ridge National Laboratory, United States; [2]Purdue University, United States

Hyperspectral Computed Tomography (CT) systems are vital tools to characterize materials in 3D across a large range of length scales. Such systems are also starting to be widely investigated in the context of neutron CT. However, in order to obtain high quality reconstructions of an object using conventional algorithms and workflows, the measurements require sufficient projection angles, good signal-to-noise ratio (SNR) and abundant wavelength coverage, which leads to the total scanning time to be extremely long (several days for only one hyperspectral CT scan). Because most of the current imaging systems have no real-time feedback in terms of reconstruction quality, , strong artefacts present in the reconstructed object are detected after the experimental measurements are completed.

In order to optimize, and hopefully reduce, the scanning time of a hyperspectral neutron imaging system, real-time CT reconstruction is developed as a projection data set is collected on the Spallation Neutron Source (SNS) data storage server. Then, feedback is provided to the user by means of algorithms that compare two sequential reconstructed data sets. The scan can either be stopped when the new scan does not provide additional information, or it can send commands to the beamline controls to re-orient the sample to improve SNR and increase measurement diversity. This system uses advanced model-based tomographic reconstruction algorithms that utilize powerful computing resources while producing low SNR hyperspectral 3D reconstructions. Specifically, we are working to accomplish three goals:

- Update the SNS SNAP beamline stages and hardware to monitor reconstruction results and allow interactions between the computation and the

instrument
- Design advanced algorithms that can compute the current reconstruction as the data is measured, evaluate the reconstruction quality via diverse metrics which will include but not be limited to Mean Square Error (MSE), Structure Similarity Index (SSIM), and Noise Power Spectrum (NPS)
- Design a decision tool based on a machine learning model that will be trained with the above multiple metrices to obtain the most optimized reconstruction. Using this novel tool, users can make decisions based on reconstructed data in a much faster and more accurate manner

This project lays the foundations for an artificial intelligence (AI)-driven neutron CT system that will allow users to perform high-efficient hyperspectral CT and better time management of imaging experiments at the Spallation Neutron Source.

A portion of this research used resources at the Spallation Neutron Source, a DOE Office of Science User Facility operated by the Oak Ridge National Laboratory.

AP3.08.06
Science-Driven Optimization of Neutron Instrumentation From Source to Detector Christoph U. Wildgruber, Hugh O'Neill, Volker S. Urban, Shuo Qian, Serena Chen and Ken Herwig; ORNL, United States

Advances in neutron instrumentation and optics have provided opportunities to use neutron scattering to study structures and dynamics of biological systems under physiologically relevant conditions. Demand to improve the performance of neutron optical components and to use available neutrons more efficiently is driving the design of neutron instrumentation into a high dimensional parameter search space. Even though computer simulations leveraging software packages like Monte Carlo N-Particle Transport (MCNP) and McStas may assist in estimating the performance of a given neutron optical configuration for specific scientific problems, the same brute force approach faces practical limitations in searching for the best neutron optical configuration in new instrument design. Here we propose a novel automated workflow by integrating McStas with a global optimization algorithm, differential evolution, to efficiently explore the high dimensional parameter space. We demonstrate this approach using a small-angle neutron scattering instrument concept with 48 parameters that is proposed to be built at the future Second Target Station at the Spallation Neutron Source of ORNL. The robustness of this workflow was evaluated by tasking the instrument concept to distinguish different conformational states of the same protein.

AP3.08.07
Bragg Edge Energy Calibration for Time-of-Flight Neutron Spectroscopy Daniel M. Pajerowski; Oak Ridge National Laboratory, United States

Calibration of instrumentation is essential to time-of-flight neutron spectroscopy. At times, these calibrations may have large, covariant parameter spaces. To get a neutron velocity (incident energy) with a high precision, distance travelled and time elapsed must be known with a high precision, and difficult to quantify uncertainties may be present in detector/monitor positions and clocking. I will present an investigation that use a standard material, with a precisely determined d-spacing, to see how well neutron energy (velocity) may be determined by measurement of a Bragg edge using a conventional time-of-flight neutron spectrometer.

AP3.08.08
Larmor a Larmor Labeling TOF SANS Instrument Jeroen Plomp[1], Ad van Well[1], Robert Dalgliesh[2] and Catherine Pappas[1]; [1]Delft University of Technology, Netherlands; [2]ISIS, United Kingdom

LARMOR is a neutron scattering instrument at the ISIS neutron source in the UK that combines a competitive Time-Of-Flight SANS instrument with polarized neutron capabilities. The instrument basis with the (polarized) SANS mode has been developed by ISIS and is in operation since summer 2015. Within this design the instrument has the flexibility in space, magnetic footprint and electronics to accommodate a rich flavour of Larmor labelling techniques, that have been developed within a Dutch national grand contribution and significantly increase its functionality.

The construction project closed summer 2019 and the instrument is currently operating in a user program. The modes that are presently tested and operational are: SANS, Polarized SANS, Diffraction, Spin Echo SANS (SESANS), Spin Echo Modulated SANS (SEMSANS), Time Of Flight Larmor precession (TOFLAR), Modulated Intensity by Zero Effort (MIEZE) and High Resolution Larmor Diffraction (HRLD). Some of these different techniques may be combined, like SANS and SEMSANS, simultaneously and will allow to investigate phenomena from nanometre to micrometre scales. Another example would be Diffraction and SANS to, for instance, investigate phase transitions in steel. The flexibility of the instrument now allows new components like Superconducting magnetic Wollaston prism for neutron spin encoding developed in Indiana University Bloomington, USA.

The Larmor labelling components have all been developed and produced at Delft University of Technology, which together with the University of Groningen and Eindhoven has launched a scientific program to exploit the capabilities of the instrument.

We would like to present an overview of the development of the Larmor labelling features of this instrument, including the strengths and weaknesses. As well as an overview of the broad scope of science that has been delivered in its first years of operation.

AP3.08.09
Update on the Extended Q-Range Small-Angle Scattering Diffractometer at the SNS Gergely Nagy, Changwoo Do, Carrie Y. Gao and William Heller; Oak Ridge National Laboratory, United States

The status of the EQ-SANS diffractometer at the Spallation Neutron Source of the Oak Ridge National Laboratory will be presented. Recent upgrades in the sample environment area, in the available sample environment options, in the instrument control software and the data reduction protocol renders this instrument more versatile and more accessible to the user community. A redesign of the sample environment area enabled a wider range of sample environment equipment to be deployed. Many of the new ones are primarily of interest to the soft matter and biological sciences user community, the instrument can now accommodate sample environments for studies of magnetic materials and alloys. New equipment developed for the EQ-SANS instrument include a tensile stage combined with a designated oven – allowing the study of deformation of soft materials over a wide temperature range; a rotatable sliding plate shear cell – allowing the time-resolved characterization of deformed polymeric materials in different planes relative to the neutron beam; a Peltier-block system – allowing sample temperature control in the -10 C ~ 130 C range with improved precision. The instrument can also host Rheo-SANS experiments in conventional bob-cup geometries. A stopped-flow system is also under commissioning. The instrument control software was upgraded to the EPICS platform, which resulted in improved instrument stability, ease of sample equipment control and remote access. This change, coupled with the recently implemented drtsans data reduction package, largely improves the user experience especially for new users.
This research used resources at the Spallation Neutron Source, a DOE Office of Science User Facility operated by the Oak Ridge National Laboratory.
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SESSION A4.09: Neutron Devices and Ancillary Equipment
Session Chairs: Mary-Ellen Donnelly and Fankang Li
Thursday Morning, June 9, 2022
UMC West Ballroom 208

10:15 AM *A4.09.01
Status of the Second Target Station Project Ken Herwig; Oak Ridge National Laboratory, United States

The Second Target Station (STS) of the Spallation Neutron Source at the Oak Ridge National Laboratory will be a high brightness source optimized for the production of cold neutrons. The high brightness of cold neutrons will enable time-resolved measurements of kinetic processes and provide more intense neutron beams to study smaller samples of newly discovered materials or materials under extreme environmental conditions. The STS pulse rate of 15 Hz will enable simultaneous measurement of hierarchical architectures across a broad range of length scales. The project received Critical Decision – 1 approval in November 2020 and has now entered the design phase. A recent major focus has been selection of the instruments that will be constructed as the initial STS instrument suite. This talk will review the status and plans for the STS Project, highlight review recent progress, and present an overview of the instruments selected for construction.

10:45 AM A4.09.02
Correcting Divergent Beam Aberrations in a Neutron Resonance Spin Echo (NRSE) instrument Stephen J. Kuhn[1], Sam McKay[1], Fankang Li[2], Eric B. Dees[1], Jiazhou Shen[1] and Roger Pynn[1,2]; [1]Indiana University Bloomington, United States; [2]Oak Ridge National Laboratory, United States

Neutron Spin Echo (NSE) is a high-energy-resolution technique using large static fields. A potential alternative is Neutron Resonance Spin Echo (NRSE) which uses four radio-frequency (RF) flippers and has the potential to increase the resolution while reducing the overall length of the instrument. Several decades of advancements have produced rf flippers capable of performance comparable to static-field coils. However, a remaining limitation of NRSE compared to NSE is the difficulty in correcting Larmor phase aberrations across the beam. These aberrations arise because neutrons traveling at different angles take different times to travel between RF flippers. While this effect cancels in the absence of scattering from a sample, it does not cancel for finite scattering angles.
We have constructed a small static-field coil that produces a field profile to correct the Larmor phase of scattered neutrons on a transverse-field NRSE instrument. In this coil, a neutron on a divergent path will travel through a higher field integral and, consequently, obtain a Larmor phase that compensates for its path. We simulate that installing three of these correction coils in each arm of the NRSE instrument allows energy transfers to be distinguishable from elastic scattering even with a finite scattering angle and sample size, enabling a more useful measurement. The coil uses high-temperature superconducting films as magnetic screens and curved iron pieces to shape the field integral. Use of this correction coil broadens the potential applications of an NRSE instrument.
Supported by DOE STTR grant DE-SC0021482

11:00 AM A4.09.04
The Strange Invisibility of Cold Neutrons in Highly Neutron Absorbing B4C—Towards a Novel Family of Neutron Optics Malik Maaza[1,2]; [1]University of South Africa, South Africa; [2]iThemba LABS-National Research Foundation of South Africa, South Africa

Conjugating several quantum mechanics phenomena, namely; wave-particle duality, quantum tunnelling & resonance among others, this contribution reports on the equivalent of the Bormann enhanced X-rays transparency for thermal and cold neutrons through one of the most notorious high neutron absorbing media; B4C. The observed peculiar neutron wave-packet invisibitity /transparency through this notorious neutrons absorber of B4C, was validated using high neutron absorbing Boron consisting of 20% and 80% of 10B and 11B respectively with an average neutron absorption cross section of sa =767.8 barns. To validate experimentally such a phenomenon, an artificial crystal consisting of (B4C/Ti) multi-layered nanostructure in the so called Zeldovich- Vinogradov geometry was considered. The experimental first diffraction Bragg peak with a reflectivity as high as 73.5% and several corresponding Kiessig fringes were observed in support of the stationary configuration of the neutron wave packets within multilayered structure comprising B4C and Ti (optically transparent layer). This type of High absorbing-transparent neutron materials based artificial crystals could open the perspective to a new family of neutron optics in line with high flux neutron sources.

Honoring the Memory of Prof. B. Pardo‡, Prof. H. Rauch‡ , Prof. J. Rossat-Mignod‡ & Prof. F. Abeles‡

11:15 AM A4.09.05
Development of an Enhanced Solid-State Neutron Detector Hank Thurston[1,2,3] and Elias Garratt[1,1]; [1]Michigan State University, United States; [2]Hillsdale College, United States; [3]Trinary Capital, LLC, United States

Diamond is a favorable material for neutron detection, particularly in high radiation environments, due to its high atomic density, relatively large neutron cross section, and radiation hardness. Diamond is a wide band gap semiconductor (5.45 eV), allowing for the production of solid-state neutron detectors. Solid state detectors have been proving their worth in the detection of charged particles for decades, and more recently in the detection of intermediate and fast neutrons. Advantages of these devices include lower bias voltage requirements, smaller form factor, greater end-user simplicity, and faster response times. Diamond based solid state detectors combine the advantages of solid state detectors with the radiation hardness and large neutron cross section of diamond, making such a device well-suited for development for use into the fast and ultrafast neutron energy regimes.

In this work, the authors develop an enhanced solid-state diamond neutron detector by engineering and exploiting the Schottky barrier created at the metallic readout contacts. Its unique architecture offers improved current-voltage properties over contemporary diamond neutron detectors, which are typically limited to the fast regime and below. Like contemporary designs, this detector relies on detection via a neutron capture or inelastic process, typically the 12C(n, α)9Be pathway. By careful material selection, we can create a metallic contact layer which will form a suitable Schottky barrier for optimal dark current performance, provide adequate electronic readout, and serve as a neutron activation layer enhancing the overall efficiency of the

detector by between 2 and 5 percent beyond what semiconducting diamond can do on its own.

The architecture of this detector is expected to reduce the form factor from contemporary high energy neutron detectors by nine orders of magnitude (from cubic meters to cubic millimeters) with only a two order of magnitude reduction in gross efficiency. The small form factor of the detector will provide desirable capability to nuclear scientists, including, but not limited to, unparalleled spatial resolution and self-vetoing. The device will provide neutron detection capability to nuclear experiments which may otherwise be unable to take advantage of larger scintillator detectors, radiation-hard beamline monitoring capabilities across a wider range of neutron (or charged particle) energies, among other application such as nuclear medicine and port screening.

Detector design has been aided through GEANT 4 and the GEANT Crystal Object Code (GECO) package, SRIM 2013, and bespoke code. Device construction has been facilitated through Michigan State University, Fraunhofer USA, and Hillsdale College. Testing will be undertaken at the Los Alamos Neutron Science and Oakridge National Laboratory during 2022.

11:30 AM A4.09.06
A Superconducting Device for Widening the Effective Angle in Quasi-Elastic Spin-Echo Neutron Scattering Experiments Eric B. Dees[1], Robert Dalgliesh[2], Steven R. Parnell[3], Stephen J. Kuhn[1], Fankang Li[4], Sam McKay[1], Jiazhou Shen[1] and Roger Pynn[1,4]; [1]Indiana University, United States; [2]Rutherford Appleton Lab, United Kingdom; [3]TU Delft, Netherlands; [4]Oak Ridge National Laboratory, United States

For neutron spin echo (NSE) experiments where neutrons are scattered through a wide range of angles, it is desirable to have a given change in neutron energy result in the same Larmor precession phase, independent of the scattering angle. While this result can be achieved using configurations such as that now implemented at WASP at the ILL, such a solution is expensive. For this reason, we have experimented with a lower cost, more compact, and modular solution, potentially allowing an increase in neutron intensity at the sample position, and flexibility within typical beamline experimental space constraints. The device we have built employs a magnetic field perpendicular to the neutron scattering plane. This field is produced by circulating current through high-temperature superconducting (HTS) wires, wound about the soft iron pole pieces of a window-frame magnet. The magnetic field is surrounded on four sides by HTS films, which serve to increase the homogeneity of the field and allow Larmor precession to be started and stopped at precise locations. By altering the shape of iron pole pieces, we can tailor the magnetic field profiles such that the neutron path-integral is near-independent of scattering angle. The maximum field integral of our prototype device is expected to be ~0.04 Tm, covering a range of scattering angles of ~5°. A wider range of scattering angles may be covered by using multiple devices or a single device can be positioned at a scattering angle of interest.

Funding: DOE STTR grant # DE-SC0018453

11:45 AM A4.09.07
Low Temperature Goniometer for Neutron Research. Sergiy Gladchenko; National Institute of Standards and Technology, United States

Elastic and inelastic neutron scattering experiments on single crystals require precise alignment of spin directions or crystallographic axes with respect to the neutron beam. Low and, especially, ultra-low (mK) temperature experiments (with and without application of magnetic fields) require placing the crystal inside a cryostat vacuum chamber using several thermal shield layers eliminating the possibility of visual observation. The cooling and warming of the cryostat are slow processes, so realigning or changing the crystal position is a highly time-consuming procedure. Some experiments will not be possible due to restrictions of precisely changing crystal position between different orientations, considering the method previously described before. To address these issues, we have developed a system that allows for the in situ alignment and reorientation of the sample inside the cryostat, allowing for changing the crystal orientation to the neutron beam and/or magnetic field during neutron experiments without disrupting the experimental conditions.

12:00 PM A4.09.09
High Resolution Larmor Diffraction at Oak Ridge National Laboratory Kaleb Burrage[1], Masaaki Matsuda[1], Jaime A. Fernandez-Baca[1], Chengjie Mao[2], Olivier Delaire[2] and Fankang Li[1]; [1]Oak Ridge National Laboratory, United States; [2]Duke University, United States

The inverse relationship between resolution and useable flux for neutron beams creates limitations in data collection from conventional neutron scattering techniques to determine accurate sample mosaics and inherent sample quality. The Larmor precession of neutron spin, however, provides a method of ultra-high resolution measurements by encoding the neutron momentum change into small shifts in the total Larmor phase. This can be used to measure crystal mosaics, thermal expansion, and relative lattice distortion with a resolution beyond the conventional neutron diffractometer. The high achievable resolution ensures beneficial diffraction capabilities for material and other scientific studies while also making the best use from all neutrons, even in large or divergent beams. In this study, we will report a newly developed technique using magnetic Wollaston prisms to investigate the crystal mosaic spread of samples on the HB-1 polarized triple axis spectrometer at the High Flux Isotope Reactor (HFIR), Oak Ridge National Laboratory, TN. Such technique would allow us to characterize the mosaic of the sample over a large volume simultaneously while minimizing the contribution from the instrument.

12:15 PM A4.09.10
In-Situ ^{3}He polarization for JCNS instrumentation Earl Babcock; Juelich Centre for Neutron Science, Germany

An in-situ polarization analyzer has been in operation for the MARIA magnetic reflectometer. Here it provides a continual polarization and a wide angle of ±6° for a cold neutron beam that is fully decoupled for the sample environment conditions and magnetic field. This device has been used as the basis for additional polarizer devices, using what has been learned to develop improved devices. A polarizer for thermal-energy beams, such as on the planned TOPAS spectrometer, has been completed and tested on POLI for 0.89 Å neutrons. Consequently two new polarizers for neutron polarization and analysis have been constructed specifically for the POLI hot-beam diffractometer. Further a compact device based on a 50% reduced scale of the MARIA polarizer is in testing for KWS1 (SANS) as is make a very high angular coverage, ±17°, device based on a compensated solenoid geometry for KWS2 (SANS). The status of the various projects and device performance including scientific examples where available will be presented.

Hard Condensed Matter

* Invited Paper

SESSION B1.01: Magnetism and Topological Band Structures
Session Chair: Martin Greven
Monday Morning, June 6, 2022
UMC East Ballroom 212

10:15 AM *B1.01.01
Neutron Diffraction Studies on the Magnetic Properties of $YMn_6Sn_{6-x}Ge_x$ Rebecca L. Dally[1], Peter Siegfried[2,2], Hari Bhandari[2], David Jones[2], Dina Michel[2,2], Madhav Ghimire[3], Lekhanath Poudel[1], Markus Bleuel[1], Jeffrey W. Lynn[1], Igor Mazin[2,2] and Nirmal Ghimire[2,2]; [1]National Institute of Standards and Technology, United States; [2]George Mason University, United States; [3]Tribhuvan University, Nepal

The importance of neutron diffraction to elucidate magnetic structures is indisputable. Particularly, in systems with coupled degrees of freedom (*e.g.* lattice, spin, and charge), neutron diffraction can detect changes to the magnetic structure which, in conjunction with ex-situ charaterization methods, can shed new light on phase transitions. This concept is notably evident in our studies on YMn_6Sn_6, where the observation of the topological Hall effect was enigmatic until the magnetic structure coinciding with it was understood. The *B-T* magnetic phase diagram for an in-plane field contains many incommensurate and commensurate structures including the transverse conical spiral, which at elevated temperatures, leads to a fluctuation driven mechanism for the topological Hall effect. Understanding the magnetic structure phase diagram was also pivotal in the interpretation of magnetotransport data which identified two Fermi surface topology changes attributed to Lifshitz transitions. To further study the coupled electronic and magnetic properties of YMn_6Sn_6, additional neutron studies on the isoelectronically substituted $YMn_6Sn_{6-x}Ge_x$ structure have been performed. With $x \approx 2$, the topological Hall effect is notably absent in addition to changes in the magnetic phase diagram.

10:45 AM B1.01.02
Weyl Mediated Helical Magnetism in NdAlSi and NdAlGe Jonathan Gaudet[1,2,3], Hung-Yu Yang[4], Santu Baidya[5], Baozhu Lu[6], Guangyong Xu[1], Yang Zhao[1,2], Jose A. Rodriguez-Rivera[1,2], Christina Hoffmann[7], Lisa DeBeer-Schmitt[7], Adam Aczel[7], David Graf[8], Darius Torchinsky[6], Predrag Nikolic[9,3], David Vanderbilt[5], Tafti Fazel[4] and Collin Broholm[3,1]; [1]National Institute of Standards and Technology, United States; [2]University of Maryland, United States; [3]Johns Hopkins University, United States; [4]Boston College, United States; [5]Rutgers University, United States; [6]Temple University, United States; [7]Oak Ridge National Laboratory, United States; [8]National High Magnetic Field Laboratory, United States; [9]George Mason University, United States

Emergent relativistic quasiparticles in Weyl semimetals are the source of exotic electronic properties such as surface Fermi arcs, the anomalous Hall effect, and negative magnetoresistance, all observed in real materials. Whereas these phenomena highlight the effect of Weyl fermions on the electronic transport properties, less is known about what collective phenomena they may support. Here, we report two Weyl semimetals, NdAlSi and NdAlGe, that offer an example. Using neutron diffraction and small-angle neutron scattering, we found a long-wavelength stripe helical magnetic order in these materials whose periodicity is linked to the nesting vector between two topologically non-trivial Fermi pockets, which we characterize using density functional theory and quantum oscillation measurements. We further show the chiral transverse component of the spin structure is promoted by bond-oriented Dzyaloshinskii-Moriya interactions associated with Weyl exchange processes. Our work provides a rare example of Weyl fermions driving collective magnetism.

11:00 AM B1.01.03
Field-Induced Fan-like Magnetic Orders in Topological $EuIn_2As_2$ Studied by Single-Crystal Neutron Diffraction Simon X. Riberolles[1], Thaís Victa Trevisan[1,2], Brinda Kuthanazhi[1,2], Feng Ye[3], D. C. Johnston[1,2], Sergey L. Bud'ko[1,2], Paul C. Canfield[1,2], R. J. McQueeney[1,2], Peter P. Orth[1,2] and Benjamin G. Ueland[1]; [1]Ames Laboratory, United States; [2]Iowa State University of Science and Technology, United States; [3]Oak Ridge National Laboratory, United States

We recently demonstrated that hexagonal $EuIn_2As_2$ exhibits a zero-field low-symmetry helical antiferromagnetic order that makes the compound a stoichiometric magnetic topological-crystalline axion insulator[1]. We further predicted that the topological properties of the surface states can be tuned by the direction and strength of an applied magnetic field that modifies the magnetic order. Here we present in-field single-crystal neutron diffraction results measured on $EuIn_2As_2$ using the CORELLI diffractometer at the SNS ORNL facility. With increased field applied in the basal plane, our results show the stabilization of an intermediate field-induced order preceding the fully field-polarized state that prevails at large fields. We find the intermediate state to be a coplanar helifan that progressively evolves into a fan structure[2]. We discuss whether the helifan and fan orders give rise to exotic topology in this material.

This work was supported by the Center for Advancement of Topological Semimetals, an Energy Frontier Research Center funded by the U.S. DOE Office of Science, Office of BES, through Ames Laboratory under its Contract No. DE-AC02-07CH11358. Contributions also comes from work performed at the Ames Laboratory supported by the U.S. Department of Energy, Office of BES, Division of Materials Sciences and Engineering under the previously mentioned Contract. This research uses resources at the Spallation Neutron Source, a DOE Office of Science User Facility operated by the Oak Ridge National Laboratory.

11:15 AM B1.01.04
Single Pair of Weyl Points in a Time-Reversal Symmetry Broken Semi-Metal Keith Taddei[1], Li Yin[1], Duminda Sanjeewa[2], Yu Li[3], Jie Xing[4], Clarina dela Cruz[1], Daniel Phelan[3], Athena Sefat[1] and David Parker[1]; [1]Oak Ridge National Laboratory, United States; [2]MURR, United States; [3]Argonne National Laboratory, United States; [4]University of South Carolina, United States

We are in an intriguing era of condensed matter physics where much of our efforts are focused on the search for solutions to relativistic quantum mechanical wave-equations in solid state materials. However, whereas originally these exotic particles were born of 'beautiful' simplifications of the Dirac equation, in materials they require a careful tuning of numerous factors to achieve - and even then, rarely is the ideal situation found leaving some ambiguity to the titular quasiparticles' observation. For instance, in the search for the massless chiral Weyl particle, most systems have numerous Weyl points and other trivial bands near the Fermi energy adding higher order interactions and obscuring the desired physics in all but ARPES measurements. Here we discuss the discovery of a Weyl semi-metal with a more ideal set of conditions – a single pair of Weyl points generated by broken time reversal symmetry which live close to the Fermi Energy. Using a combination of neutron scattering, density functional theory and careful transport measurements we elucidate the relevant symmetries, their implications for the band structure and show the resultant transport properties suggesting the clear presence of

topological physics.

11:30 AM B1.01.05
Spin Dynamics in the Antiferromagnetic Topological Insulator $MnBi_4Te_7$ Bing Li[1,2], Simon X. Riberolles[1,2], Daniel M. Pajerowski[3], J.-Q. Yan[3] and R. J. McQueeney[1,2]; [1]Ames Laboratory, United States; [2]Iowa State University of Science and Technology, United States; [3]Oak Ridge National Laboratory, United States

$MnBi_{2+2n}Te_{4+3n}$ are promising topological insulators (TI) where the natural intergrowth of magnetic layers and TI layers provides a unique platform for studying the interplay between magnetism and topological electronic states. Here we present results from our inelastic neutron scattering study on single crystals $MnBi_4Te_7$. The spin waves at base temperature can be described by a Heisenberg model with intra-layer interactions and the single-ion anisotropy. Through comparison to the spin waves in $MnBi_2Te_4$, we discuss the changes in the intra- and inter-layer magnetic interactions and the single-ion anisotropy across the Mn-Bi-Te series. At higher temperatures, $MnBi_4Te_7$ shows quasielastic scattering at magnetic peak positions below and above Neel temperature T_N, which indicates the existence of two-dimensional magnetic correlations. We extract the correlation lengths and the damping parameters at various temperatures and discuss the influence on magnetism due to the reduced dimensions.

This work is supported by the U.S. Department of Energy, Office of Basic Energy Sciences, Division of Materials Sciences and Engineering. Ames Laboratory is operated for the U.S. Department of Energy by Iowa State University under Contract No. DE-AC02-07CH11358. This research used resources at the Spallation Neutron Source, a DOE Office of Science User Facility operated by the Oak Ridge National Laboratory.

11:45 AM B1.01.06
Topological Magnons in a Honeycomb Lattice Magnet $CoTiO_3$ Bo Yuan[1,2], Matthew Stone[3], Guo-Jiun Shu[4], Fangcheng Chou[5], Patrick Clancy[6] and Young-June Kim[1]; [1]University of Toronto, Canada; [2]Max Planck Institute for the Structure and Dynamics of Matter, Germany; [3]Oak Ridge National Laboratory, United States; [4]National Taipei University of Technology, Taiwan; [5]National Taiwan University, Taiwan; [6]McMaster University, Canada

Recent experimental and theoretical interests in the thermal Hall effect in various magnetic insulators have put a spotlight on the role magnons play in heat transport. In particular, magnons with topologically protected band structures, topological magnons, have been drawing much attention. In this talk, we report our inelastic neutron scattering investigation of topological magnons in a honeycomb lattice XY magnet $CoTiO_3$, which features ferromagnetic layers of Co^{2+}, stacked antiferromagnetically along the c axis. Its energy-momentum resolved magnon spectrum is broadly consistent with a simple XXZ Hamiltonian with a Dirac-cone-like dispersion at the K-point [1,2]. However, a recent high-resolution inelastic neutron scattering measurement revealed important details of the magnon dispersion, namely a magnon gap at the Brillouin zone center as well as at the Dirac point. We found that the magnon at the Dirac point is fully gapped indicating anisotropic interactions beyond the XXZ model are required to explain the origin of the zone-center gap and the Dirac gap. The form of these anisotropic interactions can be further constrained with the observed magnetic field dependence of the magnon excitation spectrum.

[1] Bo Yuan, et al. Phys. Rev. X 10, 011062 (2020).
[2] M. Elliot, et al. Nat. Comm. 12, 3936 (2021).

12:00 PM B1.01.07
Gapless Dirac magnons in $CrCl_3$ Despina Louca[1], John Schneeloch[1], Yu Tao[1], Yongqiang Cheng[2] and Luke Daemen[2]; [1]University of Virginia, United States; [2]Oak Ridge National Laboratory, United States

Bosonic Dirac materials are testbeds for dissipationless spin-based electronics. In the quasi two-dimensional honeycomb lattice of CrX_3 (X=Cl, Br, I), Dirac magnons have been predicted at the crossing of acoustical and optical spin waves, analogous to Dirac fermions in graphene. Here we show that, distinct from $CrBr_3$and CrI_3, gapless Dirac magnons are present in bulk $CrCl_3$, with inelastic neutron scattering intensity at low temperatures approaching zero at the Dirac K point. Upon warming, magnon-magnon interactions induce strong renormalization and decreased lifetimes, with a 25 % softening of the upper magnon branch intensity from 5 to 50 K, though magnon features persist well above T_N. Moreover, an unusual negative thermal expansion (NTE) of the a-axis lattice constant and anomalous phonon behavior are observed below 50 K, indicating magnetoelastic and spin-phonon coupling arising from an increase in the in-plane spin correlations that begins tens of Kelvin above T_N.

SESSION B1.02: Disorder Inhomogeneity and Strong Correlations
Session Chair: Olivier Delaire
Monday Afternoon, June 6, 2022
UMC East Ballroom 212

1:45 PM *B1.02.01
Correlated Structural Inhomogeneity in Oxide Superconductors Martin Greven; University of Minnesota, United States

Superconductivity has been a major research topic for more than a century, yet in many important materials this macroscopic quantum phenomenon remains poorly understood. We have uncovered that superconductivity emerges in an unusual, yet remarkably universal manner upon cooling in three well-known families of complex oxides – strontium titanate, strontium ruthenate, and the cuprates – for which the origin of superconductivity is thought to differ [1]. From complementary neutron and x-ray scattering measurements, we subsequently uncovered evidence that this universal electronic behavior is caused by intrinsic correlated structural inhomogeneity that must be inherent to the oxides' perovskite-based crystal structures [2]. The prevalence of such inhomogeneity has far-reaching implications for the interpretation of electronic properties of perovskites in general, including thin films and heterostructures. In the case of the cuprates, this constitutes a pivotal part of a robust phenomenological model that comprehensively captures hitherto elusive properties of the normal and superconducting states [3]. In the case of strontium titanate, these insights motivated a systematic study of plastically-deformed crystals that led to the discovery of remarkable superconductivity and ferroelectricity enhancements associated with the self-organization of dislocations into periodic structures, as evidenced by neutron and x-ray scattering [4].

[1] D. Pelc *et al*., Nat. Commun. **10**, 2729 (2019)
[2] D. Pelc *et al*., arXiv:2103.05482

[3] D. Pelc *et al*., Sci. Adv. **5**, eaau4538 (2019); Phys. Rev. B **102**, 075114 (2020)
[4] S. Hameed *et al.*, Nat. Mater. **21**, 54 (2022)

The work at the University of Minnesota was funded by the US Department of Energy through the University of Minnesota Center for Quantum Materials, under grant number DE-SC-0016371.

2:15 PM B1.02.02
Large Change of Interlayer Vibrational Coupling with Stacking in $Mo_{1-x}W_xTe_2$ John Schneeloch[1], Yu Tao[1], Jaime A. Fernandez-Baca[2], Guangyong Xu[3] and Despina Louca[1]; [1]University of Virginia, United States; [2]Oak Ridge National Laboratory, United States; [3]National Institute of Standards and Technology, United States

Stacking variations in quasi-2D materials can have an important influence on material properties, such as changing interlayer magnetic interactions or the topology of the electronic band structure. Via inelastic neutron scattering, we find evidence that *elastic* properties can change as well. $Mo_{1-x}W_xTe_2$ is material where three differently-stacked phases are conveniently accessible by temperature changes: 1T', T_d*, and the reported Weyl semimetal phase T_d. The transitions proceed via layer sliding, and the corresponding interlayer shear mode (ISM) is relevant not just for the stacking energetics, but for understanding the relationship between the Weyl physics and structural changes. However, the interlayer interactions of $Mo_{1-x}W_xTe_2$ are not well understood, with wide variation in computed properties. In fact, the weakness of interlayer van der Waals interactions in quasi-2D materials makes computation difficult, and even for a simple material like graphite, the stacking energetics remain unclear. We report inelastic neutron scattering of the ISM in a $Mo_{0.91}W_{0.09}Te_2$ crystal. The ISM energies are generally consistent with the linear chain model (LCM), as expected given the weak interlayer interaction, though there are some discrepancies from predicted intensities. However, the interlayer force constants K_x in the T_d* and 1T' phases are substantially weaker than that of T_d, at 75(3)% and 83(3)%, respectively. Considering that the relative positioning of atoms in neighboring layers is approximately the same regardless of overall stacking, our results suggest that longer-range influences, such as stacking-induced electronic band structure changes, may be responsible for the substantial change in the interlayer vibrational coupling and, by extension, the C_{55} elastic constant.

2:30 PM B1.02.03
Role of Magnetic Defects and Defect-Engineering of Magnetic Topological Insulators Farhan Islam[1,2], Daniel M. Pajerowski[3], jiaqiang yan[3], R. J. McQueeney[1,2] and David Vaknin[1,2]; [1]Iowa State University of Science and Technology, United States; [2]Ames Laboratory, United States; [3]Oak Ridge National Laboratory, United States

Magnetic defects play an important, but poorly understood, role in magnetic topological insulators (TIs). Topological surface transport and bulk magnetic properties are controlled by magnetic defects in Bi_2Se_3-based dilute ferromagnetic (FM) TIs and $MnBi_2Te_4$ (MBT)-based antiferromagnetic (AFM) TIs. Despite its nascent ferromagnetism, our inelastic neutron scattering data show that Mn defects in Sb_2Te_3 form strong AFM dimer singlets within a quintuple block. This super-exchange coupling occurs via Mn-Te-Mn linear bonds and is identical to the AFM coupling between anti-site defects and the main Mn layer in MBT, establishing a common set of interactions in the two materials classes. We also find that FM order in $(Sb_{1-x}Mn_x)_2Te_3$ is driven by long-range correlations between magnetic defects in adjacent quintuple blocks across the van der Waals gap. In addition to providing answers to long-standing questions about the evolution of FM order in dilute TI, these results also show that the evolution of global magnetic order from AFM to FM in Sb-substituted MBT is controlled by defect engineering of the inter-block coupling.

Ames Laboratory is supported by the U.S. Department of Energy, BES under Contract No. DE-AC02-07CH11358. The Spallation Neutron Source is a DOE Office of Science User Facility operated by Oak Ridge National Laboratory.

2:45 PM B1.02.04
Diffuse Neutron and X-Ray Scattering from Inorganic Halide Perovskites Matthew Krogstad[1], Alex Rettie[2], Stephan Rosenkranz[1], Duck Young Chung[1], Mercouri Kanatzidis[3], Feng Ye[4], Yaohua Liu[4], Xing He[5], Tyson L. Lanigan-Atkins[5], Olivier Delaire[5] and Raymond Osborn[1]; [1]Argonne National Laboratory, United States; [2]University College London, United Kingdom; [3]Northwestern University, United States; [4]Oak Ridge National Laboratory, United States; [5]Duke University, United States

Halide perovskites have enjoyed significant attention over the past several years due to their intriguing optoelectronic properties, both in hybrid organic-inorganic and pure inorganic forms. In both cases, the long carrier lifetimes underpinning these properties appear to be strongly coupled to local structure. Diffuse neutron and x-ray scattering measurements on the inorganic halide perovskites $CsPbBr_3$, $CsSnBr_3$, $CsPbCl_3$, and $Cs_2AgBiBr_6$ show clear similarities in diffuse scattering, all showing a network of rods indicative of two-dimensional correlations in their high-temperature phases. Neutron scattering measurements from CORELLI show that this scattering is quasi-static, consistent with an overdamped two-dimensional phonon model. This scattering can be modelled via correlated tilts in the halide octahedron networks, with the octahedral tilt systems defining the crystal structure at low temperatures persisting at higher temperatures in a short-range form. The commonality of this feature suggests that short-range order is a defining part of the physics in these systems.

This work was supported by the US DOE, Office of Science, Basic Energy Science, Materials Sciences and Engineering Division.

3:00 PM B1.02.05
Impact of Site-Specific Cation Disorder on the Magnetic Structure Formation and Evolution in Entropy-Stabilized Perovskite Oxides Nathan D. Arndt[1], Brianna Musico[2], Keon Sahebkar[1], Qiang Zhang[3], Alessandro Mazza[3], Veerle Keppens[2], T. Z. Ward[3] and Ryan Need[1]; [1]University of Florida, United States; [2]The University of Tennessee, Knoxville, United States; [3]Oak Ridge National Laboratory, United States

Large configurational disorder created in entropy-stabilized single-phase materials leads to functional properties that deviate from traditional alloy mixing rules. Recently, entropy-stabilization has been applied to complex transition metal oxides and adds to the already rich composition-structure-property phase space found in this class of materials. Previous work on entropy-stabilized oxides (ESOs) has already shown the formation of unique and useful properties such as: colossal dielectric constant, enhanced magnetic exchange coupling, and mixed phase magnetic structures. In this talk, we present bulk magnetometry and neutron diffraction results connecting site-specific disorder to the magnetic structure formed in ESO perovskites, with an emphasis on how long-range magnetic order forms despite large disorder of the cation sublattice and magnetic exchange interactions. We collected temperature-dependent elastic scattering measurements on three samples at SNS's POWGEN instrument. Each of the samples had five elements equally alloyed on one or more cation sites, specifically: $(5A)MnO_3$ –A-site disorder, $La(5B)O_3$ – B-site disorder, and $(5A)(5B)O_3$ – both A- and B-site disorder. Our magnetometry data show that direct disruption of the B-site network leads to a massive broadening of the transition temperature, while alloying the A-site retains a sharp transition despite the presence of exchange bias suggesting a mixed phase magnetic structure as was previously proposed for B-site alloying. Neutron diffraction shows clear long-range antiferromagnetic (AFM) order in the A-site alloyed sample, and only weak or suppressed AFM in samples with B-site disorder. Fitted diffraction data support these results and provide additional insight into the magnetic structure of entropy-stabilized perovskites,

the phases present, their correlation lengths, and how phase fractions change as a function of temperature.

3:15 PM B1.02.06
The Nature of Local Dynamic Order in $CH_3NH_3PbI_3$ Tyler C. Sterling[1], Nicholas Weadock[2,2], Ballal Ahammed[3], Elif Ertekin[3], Michael Toney[2,2] and Dmitry Reznik[1,2]; [1]University of Colorado, Boulder, United States; [2]University of Colorado Boulder, United States; [3]University of Illinois at Urbana-Champaign, United States

The hybrid organic-inorganic halide perovskite $CH_3NH_3PbI_3$ (MAPI) is a promising high-efficiency solar-energy converting material. Still, after over a decade of research, the lattice dynamics in MAPI are only poorly understood. There is evidence for structural distortions and local dynamical order at room temperature, but scattering experiments on MAPI are difficult to interpret: e.g. there is substantial diffuse scattering intensity that forms a rod-like structure in reciprocal space. The lattice dynamics could have important effects on carrier properties, but without further knowledge of the microscopic dynamics, little else can be said. Molecular-dynamics (MD) simulations are a useful complement to experiment, allowing us to directly probe the microscopic dynamics in the crystal. We developed a method to calculate neutron and X-ray scattering spectra from MD trajectories and found exceptional agreement between our calculations and experiments. Convinced that the dynamics in MD accurately represent the real crystal, we looked at the simulated real-space dynamics to identify the origin of the diffuse rods. We find that the nominally cubic structure contains fluctuating, two-dimensional disks of tetragonal-like distortions. The disks are about 3 nm in diameter and live for a few picoseconds. They consist of rotated domains of PbI_6 octahedra that extend in the PbI plane perpendicular to the rotation axis. The disks exist in all PbI planes in the crystal, consistent with the observed cubic Bragg peaks in MAPI at room temperature.

This work was supported by the DOE, Office of Basic Energy Sciences, Office of Science, under Contract No. DE-SC0006939

SESSION B1.03: Novel Magnetic Structures and Excitations
Session Chair: Rebecca Dally
Monday Afternoon, June 6, 2022
UMC East Ballroom 212

4:00 PM *B1.03.01
Coexisting Singlet and Ordered Spins in a Complex Quasi-2D magnet Cu3B2O6 Bo Yuan[1], Kemp Plumb[2], Matthew Stone[3], Yiming Qiu[4], Nicholas Butch[4], Guangyong Xu[4], Patrick Clancy[5] and Young-June Kim[6]; [1]Max Planck Institute for the Structure and Dynamics of Matter, Germany; [2]Brown University, United States; [3]Oak Ridge National Laboratory, United States; [4]National Institute of Standards and Technology, United States; [5]McMaster University, Canada; [6]University of Toronto, Canada

The concept of quasiparticles has given physicists a powerful framework to study strongly interacting many-body systems, which can be made more tractable by being reformulated using weakly interacting quasiparticles. Such a description can often provide a unifying description of two seemingly different many-body systems. Understanding the low energy dynamics can be reduced to understanding the energy-momentum relation of quasiparticles and their statistical properties. With an almost endless array of model magnetic materials with distinct symmetry, structure and chemical composition, a plethora of magnetic phases have been discovered, producing a rich catalog of quasi-particles ranging from magnons to spinons. While a system with interacting quasi-particles systems have been investigated, the interaction between two different species of magnetic quasi-particles has not been explored much as this requires the extremely rare scenario where two ground states co-exist in the same material.

In this talk, I will propose a quasi-2D magnet, Cu3B2O6, as a rare material candidate which potentially realizes such physics. Despite extensive studies, simple questions concerning its ground state (such as whether the system is ordered), and its excitations remained entirely unanswered due to many conflicting results. Using elastic and inelastic neutron scattering, we provided conclusive evidence for the existence of antiferromagnetic order in Cu3B2O6. However, as I will highlight in the talk, the order observed here is of a very unconventional type as the high energy magnetic excitations are not of a spin.wave type but instead, take the characteristics of triplet excitations from a singlet ground states. In conjunction with earlier results using NMR and muSR, I will argue that Cu3B2O6 is a unique system consisting of both singlet and ordered spins, making it an ideal platform to study interactions between the interactions between magnons and triplet excitations.

4:30 PM B1.03.02
Longitudinal Magnon Decay and Renormalization in $Ba_2FeSi_2O_7$ Seunghwan Do[1], Hao Zhang[2], Travis J. Williams[1], Tao Hong[1], Ovidiu Garlea[1], Jose A. Rodriguez-Rivera[3], Tae-Hwan Jang[4], Sang-Wook Cheong[5], Jae-Hoon Park[4], Cristian Batista[2] and Andrew D. Christianson[1]; [1]Oak Ridge National Laboratory, United States; [2]The University of Tennessee, Knoxville, United States; [3]NIST Center For Neutron Research, United States; [4]MPPHC-CPM, Max Planck POSTECH/Korea Research Initiative, Korea (the Republic of); [5]Rutgers, The State University of New Jersey, United States

One of the strongest signatures of collective quantum behavior is the spontaneous quasiparticle decay in interacting bosonic systems. In quantum magnets this many body effect impacts the lifetime and energy of spin wave modes and is typically enhanced near a quantum critical point. Here we study how mode interactions impact the S=1 easy-plane quantum magnet, $Ba_2FeSi_2O_7$. Inelastic neutron scattering measurements of the spin excitations show that $Ba_2FeSi_2O_7$ is a quasi-two-dimensional square lattice antiferromagnet with large single-ion anisotropy (D>5J). These measurements further reveal a rich spectrum consisting of two transverse modes and a longitudinal mode. The longitudinal mode is observed throughout the Brillouin zone and near zone center the mode is broadened far beyond instrumental resolution indicating the importance of magnon interactions and the concomitant mode decay. To understand this decay process, we generalize the standard spin-wave theory and model the inelastic neutron scattering data. This shows that the longitudinal mode is not only subject to strong decay, but that the mode interactions also substantially renormalize the energy of the mode. The mode renormalization has important consequences--only by accounting for this renormalization is the material found to match the experimental result that $Ba_2FeSi_2O_7$ in on the antiferromagnetically ordered side of the quantum critical point.

This research was supported by the U.S. Department of Energy, Office of Science, Basic Energy Sciences, Materials Sciences and Engineering Division.

4:45 PM B1.03.03
Magnetic Structures and Dynamics in CuMnAs and Related Cu_2Sb-type Antiferromagnets Daniel Shoemaker; University of Illinois at Urbana-Champaign, United States

Recent demonstrations have shown that it may be possible the change the orientation of magnetic ordering (the Néel vector) of antiferromagnetic materials by application of electrical currents, even though the currents are not spin-polarized and the material has zero net magnetic moment. The candidate materials belong to centrosymmetric space groups and have magnetic cations in non-centrosymmetric sites. Key materials include tetragonal CuMnAs and tetragonal Mn_2Au. However, this seemingly modest proposal is complicated by strong magnetostrictive interactions in CuMnAs and related materials that share the Cu_2Sb structure type, such as Fe_2As and Mn_2As. We will present structural neutron diffraction data that reveal a rich evolution of phases in CuMnAs around room temperature, which complicates electrical writing experiments that require large applied currents and their associated temperature excursions. Furthermore, the symmetry of these materials makes extracting exchange interactions from inelastic scattering challenging. Understanding the fundamental phenomena and engineering devices from these materials requires continued neutron scattering to understand magnetic phase evolution and domain structures.

5:00 PM B1.03.04
Bootstrapped Dimensional Crossover of a Spin Density Wave in Layered Nickelate Anjana M. Samarakoon[1], Joerg Strempfer[1], Feng Ye[2], Yiming Qiu[3], Stephan Rosenkranz[1], Michael Norman[1], John Mitchell[1] and Daniel Phelan[1]; [1]Argonne National Laboratory, United States; [2]Oak Ridge National Laboratory, United States; [3]National Institute of Standards and Technology, United States

Pr4Ni3O10 is a metallic, trilayer, Ruddlesden-Popper oxide that possesses an anomalous metal-metal transition (MMT) at a temperature of ~ 158 K. Below the MMT, Pr4Ni3O10 develops a three-dimensionally correlated charge density wave similar to its cousin La4Ni3O10, along with a spin-density wave (SDW) that is weakly correlated along the c-axis. The SDW is evidenced by diffusive rods along [0,0,l] which were observed via neutron diffraction. We have successfully modeled these rods using a numerical simulation of short-range SDW stacking. However, unlike La4Ni3O10, as the temperature is further lowered below ~ 30 K, the local magnetic fields created by the spin density wave induce magnetic moments on the Pr3+ cations. Exchange interactions couple the Pr and Ni moments which drives the spin density wave to cross over into a three-dimensionally correlated magnetic order that is commensurate along the c-axis but incommensurate and locked to the charge density wave-vector in the basal plane. We have developed a model for the low-temperature state and an interaction-based understanding of hysteretic effects observed as an imprint of the Pr-ordering on the magnetic Ni sublattice which is present upon heating out of the ground state. This work was supported by the U.S. Department of Energy, Office of Science, Basic Energy Sciences, Materials Sciences, and Engineering Division.

5:15 PM B1.03.05
Chemically-Induced Magnetic Dead Shells in Superparamagnetic Ni Nanoparticles from Polarized Small-Angle Neutron Scattering Bhaskar Das[1], Joseph Batley[1], Kathryn L. Krycka[2], Julie A. Borchers[2], Patrick Quarterman[2], Caroline Korostynski[1], My Nguyen[1], Ishita Kamboj[1], Eray Aydil[3] and Chris Leighton[1]; [1]University of Minnesota, United States; [2]National Institute of Standards and Technology, United States; [3]New York University, United States

Advances in the synthesis and characterization of magnetic nanoparticles (NPs) have yielded great gains in the understanding of their complex magnetic behaviour, with implications for numerous applications. Recent work using Ni NPs as a model soft ferromagnetic (FM) system achieved the first *quantitative* understanding of the superparamagnetic blocking temperature-particle diameter relationship [1]. This hinged, however, on the assumption of an FM NP volume lower than the chemical volume due to a non-FM surface dead shell, as indirectly deduced from magnetometry. Here, we completely determine both the chemical and magnetic internal structure of such Ni NPs, *via* a suite of unpolarized, half-polarized, and fully-polarized small-angle neutron scattering (SANS) measurements and analyses, coupled with X-ray diffraction and magnetometry. The postulated magnetic dead shell is not only detected but conclusively identified as a Ni phosphide, derived from the trioctylphosphine that is widely used in hot-injection NP syntheses. The shell thickness is controllable *via* synthesis temperature, falling to as little as 0.5 nm at 170 °C. Polarized SANS further reveals bulk-like ferromagnetism in the Ni core, both in terms of bulk-like saturation magnetization at low temperatures, and a temperature-dependent magnetic scattering length density in close agreement with magnetometry. These observations quantitatively support the assumptions made in successful prior modelling of superparamagnetism [1]. In totality, these findings [2] advance the understanding of synthesis-structure-property relationships in metallic magnetic NPs, point to a route to ligand-free stabilization (using phosphide shells), and highlight the power of polarized SANS measurement and analysis capabilities for magnetic NP science and technology.

Work supported by the University of Minnesota DOE Center for Quantum Materials.

[1] J.T. Batley, M. Nguyen, I. Kamboj, C. Korostynski, E.S. Aydil and C. Leighton, *Chem. Mater.* 32, 6494 (2020).
[2] B. Das, J.T. Batley, K.L. Krycka, J. Borchers, P. Quarterman, C. Korostynski, M. Nguyen, I. Kamboj, E.S. Aydil and C. Leighton, in preparation (2022).

SESSION B2.04: Frustrated Magnetism
Session Chair: Kemp Plumb
Tuesday Morning, June 7, 2022
UMC East Ballroom 212

10:15 AM *B2.04.01
Neutron Scattering Studies of Rare-earth-based Quantum Spin Liquid Candidates Sara Haravifard; Duke University, United States

The quantum spin liquid (QSL) state is an exotic state of matter featuring a high degree of entanglement and lack of long-range magnetic order in the zero-temperature limit. Recently, rare-earth-based triangular lattice antiferromagnets have garnered significant interest as possible QSL candidates, however, the presence of chemical disorder in real-world compounds has made directly measuring the Hamiltonian parameters challenging. To further elucidate role of chemical disorder and to explore phase diagram of these materials, we present neutron scattering and thermodynamics measurements of a numbers of raer-earth-based triangular antiferromagnets. We use key observations to motivate an exploration of the field- and parameter-dependent phase diagram, providing an expanded view of the available magnetic states. More broadly, our approach demonstrates a means of pursuing QSL candidates where Hamiltonian parameters might otherwise be obscured by chemical disorder.

10:45 AM B2.04.02
Evolution of Field-Induced and Impurity-Induced Magnetic Order in the Quantum Spin Liquid Candidate $NaYbSe_2$ Ganesh Pokharel, Soren Bear and Stephen D. Wilson; University of California, Santa Barbara, United States

Two-dimensional triangular-lattices of rare-earth ions with effective spin-1/2 local moments are perfect candidates for the investigation of the physics of magnetic frustration in the presence of strong quantum fluctuations. The existence of natively fluctuating magnetic ground states manipulate are of interest due to their potential applications in data storage and quantum computing devices, and a key means of understanding these quantum spin liquid states is to perturb the state and induce long-range magnetic order. Here we present the evolution of long-range order magnetic order in the quantum spin liquid candidate $NaYbSe_2$ by the application of both an external magnetic field and through the introduction of non-magnetic Lu impurities. The substitution of Lu^{3+}-ions in the triangular lattice network should dilute the competing interactions, between Yb^{3+}-ions, responsible for the QSL state and induce magnetic order at finite temperature. The nature of nearby field-driven magnetic order and its ability to be stabilized via impurity substitution will be discussed.

11:00 AM B2.04.03
Anomalous Crystalline Electric Field Excitation in Triangular Lattice Cerium Materials Mitchell Bordelon[1,2], Brenden Ortiz[2], Pritam Bhattacharyya[3], Lorenzo Posthuma[2], Ganesh Pokharel[2], Paul Sarte[2], Thorben Petersen[3], Mohamed Eldeeb[3], Garrett E. Granroth[4], Xiaoling Wang[2], Mark Sherwin[2], Clarina dela Cruz[4], Ulrich Roessler[3], Liviu Hozoi[3], Martin Mourigal[5], Stuart Calder[4], Craig M. Brown[6], Daniel M. Pajerowski[4], Arnab Banerjee[4], Douglas L. Abernathy[4], Eric Bauer[1], Priscila Rosa[1] and Stephen D. Wilson[2]; [1]Los Alamos National Laboratory, United States; [2]University of California, Santa Barbara, United States; [3]Institute for Theoretical Solid State Physics, Germany; [4]Oak Ridge National Laboratory, United States; [5]Georgia Institute of Technology, United States; [6]National Institute of Standards and Technology, United States

In recent years, the frustrated triangular lattice materials $ALnX_2$ (A = alkali; Ln = lanthanide; X = chalcogenide) have emerged as promising hosts for exotic dynamically disordered magnetic ground states. The materials within this family with trivalent lanthanides containing small effective spin 1/2 moments are expected to be of particular interest, as their strong quantum fluctuations may further engender unconventional magnetism. These small moments are generated in odd integer total angular momentum Ln ions with crystalline electric field (CEF) interactions significantly separating the ground state Kramers doublet from its excited states. Variations in chemistry across the $ALnX_2$ compounds is expected to result in differing CEF schemes, which in turn interconnect with changes to lattice parameters and overall strength of magnetic interactions to promote a diverse set of magnetic ground states. It remains an outstanding challenge to understand the role of each ion on instigating long range ordered or quantum disordered states.

Here, we report the systematic investigation of five triangular lattice materials, $KCeO_2$ and $RbCeX_2$ (X = O, S, Se, Te), with a range of CEF potentials. Despite indications that all of these materials exhibit long range magnetic order, we uncover an unusual extra CEF excitation that questions our base understanding of the local properties of Ce in the $ACeX_2$ materials. Trivalent Ce resides in a local D_{3d} environment that can maximally split the J = 5/2 manifold into three Kramers doublets. However, we observe three sharp excitations out of the ground state Kramers doublet via high energy inelastic neutron scattering, indicating that there is one extra CEF mode in these materials. This anomalous mode is resolution limited, resides above the phonon background cutoff in $KCeO_2$ and $RbCeO_2$, and correspondingly shifts in energy with the nominal J = 5/2 Kramers doublets as the local ionic potential varies through all five $ACeX_2$ materials.

The appearance of this single extra mode is striking, as thus far it has been unexplainable by conventional phonon coupling, chemical disorder, or multiple Ce environments. Our high energy inelastic neutron scattering data presents the question of whether there are additional local degrees of freedom at play in the D_{3d} environment or the $ACeX_2$ materials as a whole. To further parametrize this anomalous mode, we have begun investigations of trivalent Ce in the $CeLi_3Pn_2$ (Pn = Sb, Bi) materials with a D_{3d} environment to help elucidate if the extra mode is endemic to all D_{3d} trivalent Ce.

11:15 AM *B2.04.04
Relaxation Dynamics in Spin Ice Ho2Ti2O7 Yishu Wang[1], Timothy R. Reeder[1], Yoshitomo Karaki[2], Jonas Kindervater[1], Thomas Halloran[1], Nicholas C. Maliszewskyj[3], Yiming Qiu[3], Jose A. Rodriguez-Rivera[3], Sergiy Gladchenko[3], Seyed Koohpayeh[1], Satoru Nakatsuji[4] and Collin Broholm[1]; [1]Johns Hopkins University, United States; [2]University of Ryukyus, Japan; [3]NIST Center for Neutron Research, United States; [4]The University of Tokyo, Japan

Spin ice represents a realization of classical spin liquid that hosts fractional excitation known as “magnetic monopoles”. In this talk, I will describe a time-resolved neutron scattering technique that we developed to visualize monopole motion in the momentum space under the perturbation of a magnetic field. Employing a pump-probe method with microsecond time resolution, we have tracked neutrons elastically scattered from the (HHL)-plane in response to hundreds-Oe-range step changes in a field applied along the out-of-plane <110> direction. Opposite time-dependence has been documented for the scattering intensity in diffusive and Bragg scattering, which satisfies the total moment sum-rule and reveals the acutely diverging relaxation time from 10^{-3}s to over 10^{4}s upon cooling from 1.3K to 0.6K. Further corroborated by the broad-band magnetic susceptibility to extend our probed time scale to over ten decades, we clearly discover a thermal crossover between two distinct relaxation processes, namely, monopole motion through the spin-ice vacuum at low temperatures, and reorientation of spin dipoles at elevated temperatures. I will also discuss the ongoing time-resolved neutron scattering experiment with an *in-plane* perturbation field to further explore the monopole dynamics.

11:45 AM B2.04.05
Real Space and Time Dynamics of Heisenberg Antiferromagnet $KCuF_3$ Measured by Neutron Scattering Allen Scheie[1], Pontus Laurell[2], Bella Lake[3], Stephen Nagler[1], Matthew Stone[1], Jean-Sebastian Caux[4] and Alan Tennant[2,1]; [1]Oak Ridge National Laboratory, United States; [2]The University of Tennessee, Knoxville, United States; [3]Helmholtz-Zentrum Berlin, Germany; [4]University of Amsterdam, Netherlands

Van Hove proposed in 1954 that inelastic magnetic neutron scattering could be Fourier transformed into a real-space picture of spin dynamics as a function of time. However, this technique appears to have been untested nearly 70 years later. Here we apply this transformation to experimental scattering on the 1D Heisenberg spin chain $KCuF_3$. Via comparisons with theoretical models, we show that real space spin correlations provide a sharp experimental distinction between classical and quantum behavior, reveal a quasi-periodic oscillating state analogous to time crystals, show a “quantum wake” of bounded quantum entanglement spreading, and at high temperatures give a direct experimental view of fractionalized spinon quasiparticles. We propose this technique for understanding the dynamics of highly entangled systems without coherent magnons, as well as probing decoherence at well-defined temperatures.

12:00 PM B2.04.06
Magnetic Structure of Single Crystalline Barlowite in an Applied Magnetic Field Aaron T. Breidenbach[1,2], Rebecca Smaha[1,2], Wei He[1,2], Adam Aczel[3], Jeffrey Lynn[4,5] and Young Lee[1,1,2]; [1]Stanford University, United States; [2]SLAC National Accelerator Laboratory, United States; [3]Oak Ridge National Laboratory, United States; [4]NIST Center for Neutron Research, United States; [5]The University of Tennessee, Knoxville, United States

Quantum magnets display exotic phases that may be strongly influenced by small differences in structure and composition. The quantum spin liquid (QSL) is an unusual magnetic ground state, characterized by long-range quantum entanglement of the spins but a lack of long-range magnetic order down to T=0K. This is believed to be possible in geometrically frustrated spin-1/2 systems, so materials with a kagome arrangement of Cu^{2+} ions are prime candidates. We find that when the bonds of the kagome lattice are modulated with a periodic pattern, new quantum ground states emerge. Specifically, crystalline barlowite ($Cu_4(OH)_6FBr$)—the parent compound of a QSL candidate, Zn-barlowite—demonstrates the delicate interplay between singlet states

and spin order on the spin-1/2 kagome lattice. Our previous studies on barlowite indicate that different synthesis pathways will result in subtle differences that change the zero-field ground state. We denote these two different forms of barlowite as 1 and 2. Barlowite 1 is highly magnetically frustrated but undergoes an orthorhombic distortion that allows it to magnetically order at low temperature (~15K). Meanwhile, barlowite 2 maintains hexagonal symmetry at low temperature and exhibits two magnetic transitions (T_{N1}~10K and T_{N2}~6K) accompanied by a highly anisotropic magnetic structure. Elastic neutron scattering in zero applied field, along with simulations and susceptibility measurements, suggest that the partial ordering of interlayer spins induces a pinwheel valence bond solid ground state between the two magnetic transitions. This is followed by full ordering of all spins into an interesting q=0 pinwheel magnetic order at T_{N2}~6K [1].
This subtle difference in the ground states of the two forms of barlowite is striking, especially as their room temperature nuclear structures are identical to within experimental uncertainty. Thus, small external perturbations may substantially affect the ground state behavior of barlowite. To this end, we employed elastic neutron scattering to measure the magnetic structure of single crystals of barlowite 2 in large (8T) out-of-plane magnetic field. We contrast this with the zero-field magnetic structure determined previously. This comparison reveals that the energy scales separating ground states in this family are very small and sheds light on which interactions are important in stabilizing the disparate ground states of the two barlowite variants.
References:
[1] R. Smaha et al., *npj Quantum Mater*. **5**, 23, 2020.

12:15 PM B2.04.07
Dynamical Ground State in the XY Pyrochlore Yb_2GaSbO_7 Adam Aczel[1], Paul Sarte[2], Joe Paddison[1], Christopher Wiebe[3], Brenden Ortiz[2], K.H. Hong[4], Mitchell Bordelon[2], Dalmau Reig-i-Plessis[5], Matthew Stone[1], Stuart Calder[1], Daniel M. Pajerowski[1], Lucile Mangin-Thro[6], Yiming Qiu[7], Paul Attfield[4], Stephen D. Wilson[2], Christopher Stock[4], Haidong Zhou[8], Alannah Hallas[5], Eun Sang Choi[9] and Minseong Lee[10]; [1]Oak Ridge National Laboratory, United States; [2]University of California, Santa Barbara, United States; [3]University of Winnipeg, Canada; [4]University of Edinburgh, United Kingdom; [5]University of British Columbia, Canada; [6]ILL, France; [7]National Institute of Standards and Technology, United States; [8]The University of Tennessee, Knoxville, United States; [9]National High Magnetic Field Laboratory, United States; [10]Los Alamos National Laboratory, United States

The magnetic ground state of the pyrochlore Yb_2GaSbO_7 has remained an enigma for nearly a decade. Here I will discuss our magnetic characterization of Yb_2GaSbO_7 aimed at addressing this open question. Through DC and AC magnetic susceptibility, heat capacity, and neutron scattering experiments, we observe evidence for a dynamical ground state that makes Yb_2GaSbO_7 a promising candidate for disorder-induced spin-liquid behavior. This state is quite fragile, being tuned to a splayed ferromagnet in a modest magnetic field of 1.5 T.

SESSION B2.05: Spin Glass and Complex Magnetic Structures
Session Chair: Martin Mourigal
Tuesday Afternoon, June 7, 2022
UMC East Ballroom 212

2:00 PM *B2.05.01
Highly Tunable Magnetic Phases in Transition Metal Dichalcogenide $Fe_{1/3+\delta}NbS_2$ Shan Wu[1,2], Zhijun Xu[3], Shannon Haley[2,1], Sophie Weber[2,1], Eran Maniv[2,1], Yiming Qiu[3], Adam Aczel[4], Jeffrey Neaton[2,1], James Analytis[1,2] and Robert Birgeneau[1,2]; [1]University of California, Berkeley, United States; [2]Lawrence Berkeley National Laboratory, United States; [3]NIST Center for Neutron Research, United States; [4]Oak Ridge National Laboratory, United States

Layered magnetic van der Waals (vdW) materials have attracted tremendous interest, leading to fundamental studies of novel vdW physical properties as well as exploration of potential spintronic applications. The wide flexibility of two-dimensional atomic samples makes possible the efficient manipulation of magnetic states through external perturbations. In bulk crystals, high pressure or significant chemical substitution are usually used to modulate the magnetic states via controlling the interlayer exchange coupling. Magnetic defects are typically considered to inhibit long-range magnetism due to the concomitant disorder. In this talk, I will discuss our discovery of novel behavior of magnetic defect-tuned magnetism in the Fe intercalated transition metal dichalcogenide (TMDC) antiferromagnet [1], $Fe_{1/3+\delta}NbS_2$, which also possess highly relevant, fascinating spintronics properties [2]. $Fe_{1/3+\delta}NbS_2$ belongs to a large class of intercalated TMDCs, M_xTA_2 family (M = 3d transition metal; T = Nb, Ta; A = S, Se). This family has a crystal structure with a non-centrosymmetric space group and a bi-layer triangular arrangement of the intercalated atoms. Unlike most members, $Fe_{1/3+\delta}NbS_2$ displays antiferromagnetic ordering at the Néel transition temperature T_N ~ 45 K and strong easy-axis anisotropy. Interestingly, both current-induced resistive switching [3] and large exchange bias effects [4] have been reported in bulk crystals below T_N. Moreover, these intriguing properties are susceptible to the Fe intercalation ratios. We performed comprehensive single-crystal neutron diffraction experiments on under-intercalated (~ -0.01), stoichiometric, and over-intercalated (~ 0.01) samples. Surprisingly, we observe highly tunable magnetic long-ranged ordered states as the Fe ratio is varied from under-intercalated to over-intercalated samples, that is from Fe vacancies to Fe interstitials. The under- and over-intercalated samples reveal distinct AFM stripe and zig-zag orders, associated with wave vectors k_1=(0.5,0,0) and k_2=(0.25,0.5,0), respectively. The stoichiometric sample shows two successive magnetic phase transitions associated with these two wave vectors with an unusual rise-and-fall feature in the intensities connected to k_1. These results can be attributed to the nearly degenerate energies for the two spin structures; this conjecture is strongly supported by our theoretical calculations. The rapid change of the magnetic states underlies the sensitive switching behaviors, providing crucial insights on the magnetic ground states that form the basis for understanding the interesting spintronic behavior. Our results show the first example of highly flexible tuning in the intercalated TMDC family, providing a new avenue for defect-induced controllability in magnetic van der Waals systems.
[1] S. Wu et al. arXiv: 2106.01341 (2021)
[2] Nair, N. L. et al. Nature Materials volume 19, page 153–157(2019).
[3] E Maniv et al. Science Advances 7, 2 (2021)
[4] E. Maniv, et al. Nature Physics 17, 525-530 (2021).
This work is funded by the U.S. Department of Energy, Office of Science, Office of Basic Energy Sciences, Materials Sciences and Engineering Division under Contract No. DE-AC02-05-CH11231 within the Quantum Materials Program (KC2202).

2:30 PM B2.05.02
Inelastic Neutron Scattering Study of the Anisotropic Spin Glass Fe2TiO5 Yu Li[1], P. G. LaBarre[2], Daniel M. Pajerowski[3], A. P. Ramirez[2], Stephan Rosenkranz[1] and Daniel Phelan[1]; [1]Argonne National Laboratory, United States; [2]Univ. California Santa Cruz, United States; [3]Oak Ridge National Laboratory, United States

Fe2TiO5 stands as an outlier amongst other spin-glass systems due to its anisotropic freezing response in susceptibility, which is observed only along the c-

axis and not in the basal plane, despite the fact that it contains Fe3+ which are isotropic due to a half-filled d-shell. Our recent diffuse neutron scattering experiment [1] demonstrated the importance of nano-scale regions of antiferromagnetic order in which spins order (anti)parallel to the a-axis. Indeed, the spin-glass behavior may arise from the freezing of the surfboards' transverse spin fluctuations. To further understand this, we have performed an inelastic neutron scattering study of the spin fluctuations in Fe2TiO5 single crystals and investigated the temperature- and energy- dependence of the low energy excitation spectra. Non-dispersive spin fluctuations out of the surfboard ordering wave vectors were observed, suggesting strong local exchange interactions. By fitting the data to a damped oscillator, these spin fluctuations exhibit typical freezing behavior as seen conventional spin glasses and have remnant fluctuations at base temperature. Furthermore, we observed additional spin fluctuations arising at a new set of wave vectors in the reciprocal space below 2 meV. These results unveil the complex magnetic interactions in Fe2TiO5 and shed light on understanding its unique behavior from a new perspective.

2:45 PM B2.05.03
Freezing of a Disorder Induced Spin Liquid with Strong Quantum Fluctuations Xiao Hu[1], Daniel M. Pajerowski[2], Depei Zhang[2], Andrey Podlesnyak[2], Yiming Qiu[3], Qing Huang[4], Haidong Zhou[4], Israel Klich[1], Alexander I. Kolesnikov[2], Matthew Stone[2] and Seung-Hun Lee[1]; [1]University of Virginia, United States; [2]Oak Ridge National Laboratory, United States; [3]National Institute of Standards and Technology, United States; [4]The University of Tennessee, Knoxville, United States

Sr2CuTe0.5W0.5O6 is a square-lattice magnet with superexchange between S=1/2 Cu2+ spins mediated by randomly distributed Te and W ions. Here, using sub-K temperature and 20 μeV energy resolution neutron scattering experiments we show that this system transits from a gapless disorder-induced spin liquid to a new quantum state below Tf=1.7(1) K, exhibiting a weak frozen moment of <S>/S∼0.1 and low energy dynamic susceptibility, χ"(ω), linear in energy which is surprising for such weak freezing in this highly fluctuating quantum regime.

3:00 PM B2.05.05
Competing Magnetic Ground States of $NaCo_2(SeO_3)_2(OH)$: A New Sawtooth Structure with Co^{2+} $S = 3/2$ Duminda Sanjeewa[1], Ovidiu Garlea[2] and Keith Taddei[2]; [1]University of Missouri, United States; [2]Oak Ridge National Laboratory, United States

Synthesis and characterization of complex competing magnetic interaction systems are a crucial part of the development of theoretical models that can predict the emergent physics in condensed matter systems. The sawtooth-chain compound $NaCo_2(SeO_3)_2(OH)$ is an excellent example of such a complex system with competing magnetic interactions. Sawtooth-type $NaCo_2(SeO_3)_2(OH)$ compound was synthesized as single crystals using a low-temperature hydrothermal method. X-ray single crystal structure analysis reveals that the material crystallizes in the orthorhombic space group of *Pnma* (no. 62). Its structure exhibits a one-dimensional chain of corner-sharing isosceles triangles that are made from two crystallographically different Co^{2+} sites (Co(1) and Co(2)). These chains run along the *b*-axis and are interconnected via SeO_3^{2-} groups to form a three-dimensional structure. The temperature dependent magnetization data show a ferromagnetic-like (FM) transition at 11 K (T_1) followed by an antiferromagnetic (AFM) transition at 3.8 K (T_2). Neutron-powder diffraction measurements reveal that at 11 K only Co(2) site orders magnetically, forming ferromagnetic zigzag chains along the *b*-axis. Below 3.8 K, both Co(1) and Co(2) sites order with the Co(1) moment pointing along the sawtooth chain plane while the Co(2) moment points away from the sawtooth chain plane. These chains rotate along the *a*-axis creating an overall AFM structure. Inelastic neutron scattering measurements reveal crystal field excitations that are consistent with the presence of a spin-orbital entangled pseudo-spin state $J_{eff}=1/2$ for the Co^{2+} ions. Low-energy spin-wave excitations are observed below the second magnetic transition. The analysis of powder excitation spectrum suggests a complex exchange interaction pattern that goes beyond a Heisenberg Hamiltonian model with nearest neighbor couplings. Our results demonstrate the richness of the magnetic properties of sawtooth-type structures and encourage the study of similar structures with different oxyanion groups.

SESSION B3.06: Phonons and Lattice Dynamics
Session Chair: Dmitry Reznik
Wednesday Morning, June 8, 2022
UMC East Ballroom 212

10:15 AM B3.06.01
Inelastic Neutron Scattering Measurements of New Spectral Features from Nonlinear Phonon Interactions Brent Fultz[1], Yang Shen[1], Claire N. Saunders[1], Camille Bernal[1], Michael E. Manley[2] and Vladimir Ladygin[1]; [1]California Institute of Technology, United States; [2]Oak Ridge National Laboratory, United States

At high temperatures, the normal modes of vibration in a crystal begin to interact with each other and lose independence. This is an effect of "anharmonicity," which alters, and can dominate, thermophysical properties such as thermal expansion, thermal conductivity, and the temperature dependence of elastic constants. Many-body theory, as developed within perturbation theory of phonon-phonon interactions, is effective for analyzing phonon shifts and broadenings when the anharmonicity is not too large. Large anharmonicity with nonlinear phonon interactions requires approaches beyond many-body theory.
When anharmonicity is significant, phonon behavior can sometimes be interpreted with the physics of nonlinear systems. In a nonlinear medium, the output ψ_o from an input vibrational amplitude, ψ_i, goes as $\psi_o = \psi_i + \eta\psi_i^2$. When this medium is driven by two frequencies, e.g., $\psi_i = \exp(-i\omega_1 t)+\exp(-i\omega_2 t)$, the output ψ_o has spectral components at additional frequencies $\omega_1+\omega_2$ and $\omega_1-\omega_2$, called "intermodulation sidebands." (This is intermodulation distortion in nonlinear amplifiers.) Intermodulation phonon sidebands were recently observed by inelastic neutron scattering in NaBr at 300 K. [1] DOI: 10.1103/PhysRevLett.125.085504. [2] DOI: 10.1103/PhysRevB.103.134302. In a crystal of NaBr, the lower sideband is an intrinsic localized mode (ILM), previously observed by Manley. The newly discovered upper sideband completes the picture.
Intermodulation phonon sidebands require strong nonlinear interactions (large η above) of at least two phonon modes. Because the interacting phonon branches are mostly dispersionless in NaBr, we developed a new explanation of the shapes and intensities of phonon intermodulation sidebands by adapting the theory of sidebands from the input-output theory of quantum optics. We replaced the noiseless laser with a thermally excited phonon and solved the Heisenberg-Langevin equations of motion to obtain susceptibility functions for inelastic neutron scattering. The sideband excitations are predicted to be new quasiparticles like those from the coupling between a laser and a mode of vibration of a cavity.
Effects of anharmonicity grow as temperature drives the amplitude of vibrations. With increasing numbers of phonons in the vibrational modes, there will be a transition from quantized phonons into classical dynamics. This transition offers new possibilities for nonlinear dynamics that have been studied in mechanics on nonlinear systems, such as bifurcations of oscillator amplitudes. At higher *T*, NaBr showed additional spectral features, and phonon spectral weight moved between different regions of momentum and energy. This effect is under study, as are its contributions to thermodynamic functions.
This work is supported by DOE BES award No. DE-FG02-03ER46055.

10:30 AM B3.06.02
Structural Fluctuations, Complex Ground-States and Strongly Anharmonic Phonons in Metal Halide Perovskites Olivier Delaire[1], Xing He[1], Tyson L. Lanigan-Atkins[1], Matthew Krogstad[2], Mayanak K. Gupta[1], Chengjie Mao[1], Daniel M. Pajerowski[3], Douglas L. Abernathy[3], Feng Ye[3], Tao Hong[3], Songxue Chi[3], Yaohua Liu[3], Guangyong Xu[4], Zhijun Xu[4], Stephan Rosenkranz[2] and Raymond Osborn[2]; [1]Duke University, United States; [2]Argonne National Laboratory, United States; [3]Oak Ridge National Laboratory, United States; [4]National Institute of Standards and Technology, United States

Inorganic metal halide perovskites (MHPs) and double-perovskite variants are attracting significant attention for their excellent performance in photovoltaic, optoelectronic and radiation detection devices, as well as potential for thermoelectric applications. MHPs are known to exhibit a soft lattice with large atomic fluctuations. While crucial to understand electron-phonon and phonon-phonon couplings, the spatiotemporal correlations of these fluctuations remain largely unknown. We discuss these correlations based on comprehensive neutron and x-ray scattering measurements, complemented with first-principles simulations augmented with machine learning. Our inelastic neutron scattering (INS) and diffuse scattering measurements revealed complex potential energy surfaces, resulting in multiple soft phonons that promote 2D dynamic fluctuations of cooperative halide octahedra tilts, as revealed by extensive diffuse scattering rods [1,2]. The short-ranged correlations are not static. Their dynamics were studied with INS measurements, which revealed pervasive overdamped phonon spectra and quasielastic signatures. Further, competition between lattice instabilities can yield complex ground states. Based on ab-initio molecular dynamics simulations, we use machine learning to train neural net based surrogate interatomic potentials, from which we performed large-scale simulations (>10,000 atoms) that reproduce diffuse scattering and strongly anharmonic phonon spectral functions. These results provide new insights into the atomic structure and fluctuations in MHPs, critical to understand the unusual electron-phonon and phonon-phonon couplings that underpin their optoelectronic and thermal properties.

[1] T. Lanigan-Atkins*, X. He*, M. J. Krogstad, D. M. Pajerowski, D. L. Abernathy, Guangyong NMN Xu, Zhijun Xu, D.-Y. Chung, M. G. Kanatzidis, S. Rosenkranz, R. Osborn, and O. Delaire, "Two-dimensional overdamped fluctuations of soft perovskite lattice in $CsPbBr_3$", Nature Materials 20, 977–983 (2021)
[2] X. He, M. Krogstad, M. K Gupta, T. Lanigan-Atkins, C. Mao, Feng Ye, Y, Liu, T. Hong, S. Chi, H. Wei, J. Huang, S. Rosenkranz, R. Osborn, and O. Delaire, "Multiple lattice instabilities and complex ground state in $Cs_2AgBiBr_6$", [arXiv:2112.04717].

10:45 AM B3.06.03
Mutual Spin-Phonon Driving Effects and Phonon Eigenvector Renormalization in NiO Qiyang Sun[1], Bin Wei[1,2], Yaokun Su[1], Hillary Smith[3], Jiao Lin[4], Douglas L. Abernathy[4] and Chen Li[1,1]; [1]University of California, Riverside, United States; [2]3Henan Key Laboratory of Materials on Deep-Earth Engineering, School of Materials Science and Engineering, China; [3]Swarthmore College, United States; [4]Oak Ridge National Laboratory, United States

The physics of mutual interaction of phonon quasiparticles with electronic spin degrees of freedom, leading to unusual transport phenomena of spin and heat, has been a subject of continuing interests for decades. Despite its pivotal role in transport processes, the effect of spin-phonon coupling on the phonon system, especially acoustic phonon properties, has so far been elusive. Unusual spin and phonon transport phenomena, including phonon pumping of spin current and pumping of phonon by magnetization dynamics, suggest phonon propagation can greatly affect the transport of spin and vice versa. Using inelastic neutron scattering and first-principles calculations, anomalous scattering spectral intensity from acoustic phonons was identified in the exemplary collinear antiferromagnetic nickel (II) oxide, unveiling strong spin-lattice correlations that renormalize the polarization of acoustic phonon. A clear magnetic scattering signature of the measured neutron scattering intensity from acoustic phonons is demonstrated by its momentum transfer and temperature dependences. The anomalous scattering intensity is successfully modeled with a modified magneto-vibrational scattering cross section, suggesting the presence of spin precession driven by phonon. The renormalization of phonon eigenvector is indicated by the observed "geometry-forbidden" neutron scattering intensity from transverse acoustic phonon. Importantly, the eigenvector renormalization cannot be explained by magnetostriction but instead, it could result from the coupling between phonon and local magnetization of ions. Our results provide a new approach to identify and quantify strong spin-phonon interactions, shedding lights on engineering functional spin-caloritronic materials through these interactions.

11:00 AM B3.06.04
Role of Anharmonicity in the High-temperature Thermodynamics of Chromium Camille Bernal[1], Hillary Smith[2], Claire N. Saunders[1], Dennis S. Kim[3], Douglas L. Abernathy[4] and Brent Fultz[1]; [1]California Institute of Technology, United States; [2]Swarthmore College, United States; [3]Massachusetts Institute of Technology, United States; [4]Oak Ridge National Laboratory, United States

Phonon densities of states (DOS) of a powder sample of bcc chromium were measured from 6 to 1493 K using the Wide-Angle Chopper Spectrometer (ARCS) at Oak Ridge National Laboratory. Density functional theory (DFT) calculations with both a quasi-harmonic (QH) and an anharmonic (AH) approximation were also performed at 330, 1000, and 1500 K and compared to the experimental results.

At high temperatures, phonon features in DOS below 32 meV soften substantially (decrease in energy). The magnitude of this energy shift was quantified by fitting our ARCS DOS to a Born-von Kármán model with a genetic optimization algorithm to determine phonon dispersion relationships at each temperature. A softening of almost 17% is found in the high transverse branch between 330 and 1500 K, while the low transverse branch shows a more modest softening of 9%. QH calculations cannot account for this behavior and predict a larger shift in the low transverse branch than the high transverse. Our AH calculations, performed with the stochastic temperature-dependent effective potential (sTDEP) package, capture the observed behavior of the phonon branches.

Using the connection between vibrational densities of states and thermodynamics, we also compute the entropy of chromium. Excellent agreement is observed between our measured vibrational entropy and calorimetric entropy up to 600K. An additional component contributes to the entropy at higher temperatures, although phonons remain dominant. Our QH and AH thermodynamic predictions exhibit similar behavior. (This is not necessarily unusual, see https://doi.org/10.1073/pnas.1707745115.) When we account for the temperature-dependence of the electronic states and electron-phonon coupling, we see this thermodynamic agreement extends to 1500 K. Broader implications of the purely anharmonic contributions in Cr will be discussed.

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11:15 AM B3.06.05
Flattening of the Acoustic Phonon Branches in the Clathrate $Ba_8Ga_{16}Ge_{30}$ Susmita Roy[1], Tyler C. Sterling[1], Dan Parshall[1], Eric Toberer[2], Mogens Christensen[3], Devashibhai T. Adroja[4] and Dmitry Reznik[1,5]; [1]UNIVERSITY OF COLORADO BOULDER, United States; [2]Colorado School of Mines, United States; [3]University of Aarhus, Denmark; [4]ISIS Facility, STF, Rutherford Appleton Laboratory, United Kingdom; [5]Center for Experiments on Quantum Materials, University of Colorado Boulder,, United States

Thermoelectric materials directly convert waste heat into electricity which could be used as an important alternative power source to meet the challenge of

a globally sustainable energy solution in near future. Generally, a high performance thermoelectric material must have low thermal conductivity, high electrical conductivity, and high value of Seebeck coefficient. The challenge is to achieve high electrical conductivity as well as low thermal conductivity in a particular material as the electronic thermal conductivity is directly related to the electrical conductivity via Widemann Franz law. The only way is to reduce lattice thermal conductivity. This kind of concept can be realized in Phonon-Glass Electron Crystals (PGEC) materials where both glass-like low phonon thermal conductivity and crystal-like high electrical conductivity simultaneously exist. Research focused on $Ba_8Ga_{16}Ge_{30}$ (BGG), a clathrate which has a guest Ba atom trapped inside Ga/Ge cage structures, has
found avoided crossings between acoustic phonons and the at guest atom modes. The avoided
crossings are predicted of being the source of the low lattice thermal conductivity in $Ba_8Ga_{16}Ge_{30}$. Ga/Ge site disorder with Ga and Ge exchanging places in different unit cells has also been reported. We used time-of-flight neutron scattering to measure the complete phonon spectrum in a large single crystal of BGG and compared these results with predictions of density func-
tional theory. Calculations assuming the structure where Ga/Ge atoms occupy their nominal sites i.e. ordered configuration, as well as a disordered configuration showed that the latter agrees much better with the experimental data. Disorder strongly reduces phonon group velocities despite nearly identical masses of Ga and Ge, which might be the source for low thermal conductivity that plays a crucial role in efficient thermoelectrics. Our work directs a new path towards optimizing thermoelectrics.

11:30 AM B3.06.06
Atomic Tunneling in BaTiS3 Raphael P. Hermann, Michael E. Manley, Duncan H. Moseley, Daniel M. Pajerowski, Barry Winn and Eugene Mamontov; Oak Ridge National Laboratory, United States

Inelastic neutron scattering measurements of a double-well local structure and associated two-level excitation present in BaTiS3 reveal that atomic tunneling is the most likely source of the ultralow glass-like thermal conductivity in $BaTiS_3$ [1]. This are the first observations of translational atomic tunneling other than hydrogen using neutrons in a crystal. The tunneling behavior was observed at the HYSPEC, CNCS, and BASIS spectrometers of the Spallation Neutron Source. It appears as a local mode with ~0.5 meV energy, with modulated intensity, which suggests more complex behavior than isolated double-wells. The tunneling mode survives up to room-temperature. Such two-level systems are known in glasses where they impact low temperature thermal transport, but this is first time when tunneling has sufficiently high frequency to disrupt thermal transport over a large temperature range in a crystal. This mechanism suggests an alternative route to controlling thermal transport in functional materials.

We gratefully acknowledge the collaboration with our coauthors in the teams of Prof. J. Ravichandran at USC and Prof. A. Minnich at Caltech, with Katherine Page at UTK and with Ahmet Alatas at APS/ANL. Neutron work supported by the US Department of Energy (DOE), Office of Science, Office of Basic Energy Sciences (BES), Materials Sciences and Engineering Division. This research used resources at the Spallation Neutron Source and at the Advanced Photon Source, facilities supported by DOE, BES, Scientific User Facilities Division.

[1] B. Sun, S. Niu, R. P. Hermann, et al., High frequency atomic tunneling yields ultralow and glass-like thermal conductivity in chalcogenide single crystals, *Nat. Commum.* **11**, 6039 (2020).

11:45 AM B3.06.07
Investigation of the Thermophysical Properties of Cuprite by Inelastic Neutron Scattering and Machine Learning Calculations Claire N. Saunders[1], Vladimir Ladygin[1], Dennis S. Kim[1], Olle Hellman[2], Hillary Smith[3], Camille Bernal[1] and Brent Fultz[1]; [1]California Institute of Technology, United States; [2]Linköping University, Sweden; [3]Swarthmore College, United States

In previous studies of cuprite (Cu_2O), both quasiharmonic (QH) and anharmonic (AH) models successfully predicted the small anomalous thermal expansion behavior. However, AH calculations explain the measured temperature-dependent phonon behavior better than the QH approximation. The cancellation of errors in the thermal trends of phonons in the QH theory allows for good predictions of thermal expansion but reinforces the need to look deeper at the physics of thermal expansion and other thermophysical properties.

While calculations with high-order force constants were more effective than low-order in predicting the observed temperature-dependent phonon behavior, none of the calculations sufficiently accounted for an observed diffuse intensity in the measured inelastic neutron scattering (INS) phonon spectrum. This study presents INS single crystal data from the Wide Angle Chopper Spectrometer (ARCS) at the Oak Ridge National Laboratory Spallation Neutron Source. Data collected at 700K and 900K was post-processed using Mantid, reduced with an original multiphonon background correction for polyatomic crystals, and folded back into a single irreducible wedge in the first Brillouin zone to amplify the spectral intensities of the data. This workflow provided the 4D scattering function S(Q, E) [2, 3].

Classical molecular dynamics with machine learning interatomic potentials models will be compared with perturbative anharmonic calculations and experimental data to model the diffuse spectral intensity [4, 5]. We propose a local dynamics model to explain this diffuse inelastic scattering, analogous to oscillator phase noise. We will discuss the prevalence of phase noise in other systems, including NaBr and Si, and the potential implications.

This work is supported by DOE BES award No. DE-FG02-03ER46055.

[1] C.N. Saunders, et al. in review (2022)
[2] O. Arnold, et al. Nucl. Instrum. Methods Phys. Res. A, **764**, 156–166 (2014).
[3] Mantid: Manipulation and Analysis Toolkit for Instrument Data.; Mantid Project. http://dx.doi.org/10.5286/SOFTWARE/MANTID (2013)
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[5] O. Hellman, et al., Phys. Rev. B **87**, 104111 (2013)

12:00 PM B3.06.08
Phason Dominated Thermal Transport in Fresnoite Michael E. Manley[1], Andrew May[1], Barry Winn[1], Douglas L. Abernathy[1], Raffi Sahul[2] and Raphael P. Hermann[1]; [1]Oak Ridge National Laboratory, United States; [2]Amphenol Corporation, United States

Fast-propagating waves in the phase of incommensurate structures, called phasons, have long been argued to enhance thermal transport. Although supersonic phason velocities have been observed, the lifetimes, from which mean free paths can be determined, have not been resolved. Using inelastic neutron scattering and thermal conductivity measurements, we establish that phasons in piezoelectric mineral fresnoite make a major contribution to thermal conductivity by propagating with higher group velocities and longer mean free paths than phonons. The phason contribution to thermal conductivity is maximum near room temperature, where it is the single largest contributing degree of freedom.

SESSION B3.07: Orbital Physics and Beyond Dipolar Magnetism
Session Chair: Benjamin Ueland
Wednesday Afternoon, June 8, 2022
UMC East Ballroom 212

2:00 PM B3.07.01
Anisotropic Spin Wave Excitations in a Multiferroic $BiFeO_3$ Masaaki Matsuda[1], Depei Zhang[1], Sachith Dissanayake[1], Barry Winn[1], Toshimitsu Ito[2] and Randy Fishman[1]; [1]Oak Ridge National Laboratory, United States; [2]AIST, Japan

Polarized inelastic neutron scattering experiments have been performed to elucidate the anisotropic behavior of the low energy spin wave excitations in a multiferroic $BiFeO_3$, which shows a cycloidal spin structure below 640 K. With the neutron polarization analysis for single magnetic domained crystals, magnetic excitation modes in and out of the cycloidal plane below 6 meV were separated successfully. The overall feature of the experimental result was reproduced reasonably well using the linear spin wave theory. The low-energy magnon density of states consist of several magnon modes, including the two anisotropic modes, Φ and Ψ modes, distributed in and out of the cycloidal plane, respectively, which were previously observed using optical spectroscopies.

2:15 PM B3.07.02
Magnetism and Symmetry Lowering in the $5d^1$ Double Perovskite Ba_2NaOsO_6 Probed with Polarized Neutron Diffraction and Total Scattering Stuart Calder, Yan Wu, Jue Liu and Jiaqiang Yan; Oak Ridge National Laboratory, United States

Materials with 5d ions have been intensely studied due to the emergence of various exotic quantum phenomena [1]. These properties emerge from the delicate balance of large spin-orbit coupling, large crystal field splitting and increased orbital overlap in 5d ions. The enhancement of quantum effects with reduced moments has led to much focus on 5d iridates with a d^5 electronic configuration and spin-orbit entangled J_{eff}=1/2 ground state. Conversely $5d^1$ materials with S=1/2 offers a natural route to enhanced quantum behavior, but the rarity of such a valence state has limited experimental studies. One exception is the double perovskite material Ba_2NaOsO_6 where the Os ion has a d^1 electronic configuration. Initial observations of non-trivial behavior have fostered several experimental and theoretical investigations, with recent predications of spin-orbit dimers, Weyl nodal loops, orbital ordering and nematic phases [2,3]. Despite the interest in Ba_2NaOsO_6 fundamental questions remain unanswered. For example, the magnetic ground state is debated between ferromagnetic or canted antiferromagnetic and the presence of a structural distortion is debated between local or long ranged [4,5,6]. Here, we reveal key new insights through a comprehensive neutron scattering investigation using polarized neutron diffraction, that is highly sensitive to small magnetic signals [7], and total scattering, which can disentangle local and long-range structural phase transitions. The nature of the magnetic ground state is determined and intriguingly the presence of oxygen magnetism is directly extracted from the polarization analysis along with the 5d magnetism. The neutron total scattering and Bragg diffraction measurements reveal a crystallographic symmetry lowering from cubic which impacts the local oxygen octahedra and offers routes to nematic or orbital ordering. We discuss our results on $Ba_2NaOsO6$ in the context of proposed multipolar and orbital order phase diagrams for d^1 double perovskites.

[1] Witczak-Krempa et al., Annu. Rev. Condens. Matter Phys. 5, 57–82 (2014).
[2] Romhányi et al., Phys. Rev. Lett. 118, 217202 (2017).
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[6] Willa et al., Phys. Rev. B 100, 041105(R) (2019).
[7] Kibalin et al, Phys. Rev. Research 1, 033100 (2019).

2:30 PM B3.07.03
Dual Orbital Degeneracy Lifting in a Strongly Correlated Electron System Emil Bozin[1], Robert J. Koch[1], Ryan Sinclair[2], Marshall McDonnell[3], Runze Yu[1], Milinda Abeykoon[1], Simon Billinge[1], Alexei Tsvelik[1], Matthew Tucker[3], Haidong Zhou[2] and Weiguo Yin[1]; [1]Brookhaven National Laboratory, United States; [2]The University of Tennessee, Knoxville, United States; [3]Oak Ridge National Laboratory, United States

The local structure of $NaTiSi_2O_6$ is examined across its Ti-dimerization orbital-assisted Peierls transition at 210 K. Combined neutron and X-ray atomic pair distribution function approach evidences local symmetry breaking preexisting far above the transition. The analysis unravels that, on warming, the dimers evolve into a short range orbital degeneracy lifted (ODL) state of dual orbital character, persisting up to at least 490 K. The ODL state is correlated over the length scale spanning ~6 sites of the Ti zigzag chains. Results imply that the ODL phenomenology, previously seen in weakly correlated materials exhibiting metal-insulator transitions, extends to strongly correlated electron systems [1].

[1] R.J. Koch et al, Phys. Rev. Lett. **126**, 186402 (2021)

Acknowledgment:
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2:45 PM B3.07.04
Lattice and Magnetic Dynamics in YVO_3 Mott Insulator Studied by Neutron Scattering and First-Principles Calculations Yu Tao[1], Douglas L. Abernathy[2], Tianran Chen[3], Taner Yildirim[3,4], Jiaqiang Yan[2,5], Jianshi Zhou[6], John Goodenough[6] and Despina Louca[1]; [1]University of Virginia, United States; [2]Oak Ridge National Laboratory, United States; [3]National Institute of Standards and Technology, United States; [4]University of Pennsylvania, United States; [5]The University of Tennessee, Knoxville, United States; [6]The University of Texas at Austin, United States

The Mott insulator YVO_3 with $T_N = 118$ K is revisited to explore the role of spin, lattice and orbital correlations across the multiple structural and magnetic transitions observed as a function of temperature. Upon cooling, the crystal structure changes from orthorhombic to monoclinic at 200 K, and back to orthorhombic at 77 K, followed by magnetic transitions. From the paramagnetic high temperature phase, C-type ordering is first observed at 118 K, followed by a G-type spin re-orientation transition at 77 K. The dynamics of the transitions were investigated via inelastic neutron scattering and first principles calculations. An overall good agreement between the neutron data and calculated spectra was observed. From the magnon density of states, the magnetic exchange constants were deduced to be $J_{ab} = J_c = -5.8$ meV in the G-type spin phase, and $J_{ab} = -3.8$ meV, $J_c = 7.6$ meV at 80 K and $J_{ab} = -3.0$ meV, $J_c = 6.0$ meV at 100 K in the C-type spin phase. Paramagnetic scattering was observed in the spin ordered phases, well below the C-type transition temperature, that continuously increased above the transition. Fluctuations in the temperature dependence of the phonon density of states were observed between 50 and 80 K as well, coinciding with the G-type to C-type transition. These fluctuations are attributed to optical oxygen modes above 40 meV, from first principles calculations. In contrast, little change in the phonon spectra is observed across T_N.

3:00 PM B3.07.05
The Detection of Magneto-Electric Multipoles with Spherical Neutron Polarimetry: CuO and $LiMnPO_4$ Jian Rui Soh[1,2], Andrea Urru[3], Paola Forino[1], Rasmus Toft-Petersen[2], NICOLA SPALDIN[3] and Henrik M. Ronnow[1]; [1]EPFL, Switzerland; [2]Technical University of Denmark, Denmark; [3]ETH Zurich, Switzerland

The fundamental interaction between the neutron dipolar field and the magnetisation density surrounding the scattering ion, lies at the heart of magnetic neutron diffraction. However, if the ion resides in an environment which breaks both time and spatial inversion symmetry, the current formalism for magnetic diffraction does not fully account for all the possible scattering mechanisms arising from the asymmetry of the magnetisation density cloud of the scatterer.
In our work, we have extended the theory of magnetic neutron diffraction to include these effects. Drawing analogies from the magneto-electric (ME) phenomena and standard magnetic neutron diffraction, we have developed a framework to calculate the associated ME form factor, size of the ME multipoles and the ME propagation vector.
Furthermore, we have identified several material systems, including CuO and $LiMnPO_4$, which can not only host these ions but also display an ordered arrangement of these scatterers. Alongside our density functional theory calculations of the corrections to the scattering amplitudes and form factor of these scatterers, we used spherical neutron polarimetry to provide evidence for the interactions between neutrons and these ions.

SESSION B3.08: Unconventional Superconductors and Related Materials
Session Chair: Shan Wu
Wednesday Afternoon, June 8, 2022
UMC East Ballroom 212

4:00 PM *B3.08.01
Carrier Tuning of Stoner Ferromagnetism in $Ca(Co_{1-x}Fe_x)_{2-y}As_2$ Benjamin G. Ueland[1,2], Santanu Pakhira[1,2], Bing Li[1,2], Aashish Sapkota[1,2], N. S. Sangeetha[1], Toby G. Perring[3], Yongbin Lee[1], Liqin Ke[1], D. C. Johnston[1,2] and R. J. McQueeney[1,2]; [1]Ames Laboratory, United States; [2]Iowa State University of Science and Technology, United States; [3]STFC Rutherford Appleton Laboratory, United Kingdom

$CaCo_{2-y}As_2$ is an unusual itinerant magnet with signatures of extreme magnetic frustration. The conditions for establishing magnetic order in such itinerant frustrated magnets, either by reducing frustration or increasing electronic correlations, is an open question. I will present results from inelastic neutron scattering and magnetic susceptibility measurements and density functional theory calculations that show hole doping in $Ca(Co_{1-x}Fe_x)_{2-y}As_2$ suppresses magnetic order by quenching the magnetic moment while maintaining the same level of magnetic frustration. The suppression is due to tuning the Fermi energy away from a peak in the electronic density of states originating from a flat conduction band. This results in the complete elimination of the magnetic moment by $x \approx 0.25$, providing a clear example of a Stoner-type transition.
This work is published in Phys. Rev. B **104**, L220410 (2021).

This research was supported by the U.S. Department of Energy, Office of Basic Energy Sciences, Division of Materials Sciences and Engineering under Contract No. DE-AC02-07CH11358. Experiments at the ISIS Neutron and Muon Source were supported by a beamtime allocation RB1810596 from the Science and Technology Facilities Council.

4:30 PM B3.08.02
Understanding Charge Density Wave superlattice structure and potential Quantum Spin Liquid behavior in 1T-TaS_2 and 1T-$TaSe_2$ Sharon S. Philip and Despina Louca; University of Virginia, United States

The transition metal dichalcogenide 1T-$TaS_{2-x}Se_x$ is an interesting system due to the multiple phase transitions both as a function of doping and temperature, that includes charge density wave (CDW) states, Mott insulator, superconductivity and more recently, quantum spin liquid (QSL) state. 1T-TaS_2, undergoes a series of phase transitions upon cooling, acquiring a commensurate CDW (CCDW) phase below 183 K. Moreover, the CCDW transition is accompanied by a periodic lattice distortion that forms a $\sqrt{13}\times\sqrt{13}$ supercell. In this, an in-plane trimerization of the Ta atoms with an inward displacement towards the center Ta creates the so-called Star-of-David motif. The Star-of-David formation alone cannot explain the electronic behavior of 1T-TaS_2 such as the QSL behavior, superconductivity, and the metal-insulator transition (MIT). On the other end of the phase diagram, 1T-$TaSe_2$, undergoes a single transition to a CCDW state at high-temperatures, in the absence of drastic changes in the electronic conductivity. Understanding the superlattice structure of 1T-TaS_2 and 1T-$TaSe_2$ in the CDW regime is very important to study the lattice, spin and electronic correlations in this system. From our recent work on this system, we observed clear evidence that the low-temperature crystal structure deviates from the long-range symmetry (P-3m1). Pair distribution function analysis suggests in-plane displacements of Ta atoms in 1T-TaS_2 in the CDW phase which shows significant differences from the model based on average structure. The Ta-Ta and S-S correlation distances modify in the CDW phase because of Ta in-plane and S atom movements in the superstructure. Distortions at the 12 Ta atoms making up the outer rim of the star are present in both 1T-TaS_2 and 1T-$TaSe_2$, which may very well lead to distortions in the spin structure.

4:45 PM B3.08.03
Structural Correlations in the Hole-Doped Cuprate $HgBa_2CuO_{4+\delta}$ Zachary W. Anderson[1], Damjan Pelc[2], Matthew Krogstad[3], Nikolaos Biniskos[4], Biqiong Yu[1], Yaohua Liu[5], Liam Thompson[1], Jack Zwettler[1], Richard Spieker[1], Nina G. Bielinski[1], Feng Ye[5], Stephan Rosenkranz[3], Raymond Osborn[3] and Martin Greven[1]; [1]University of Minnesota, United States; [2]University of Zagreb, Croatia; [3]Argonne National Laboratory, United

States; [4]Forschungszentrum Jülich GmbH, Germany; [5]Oak Ridge National Laboratory, United States

The cuprate superconductors form a large family of strongly-correlated materials that share several universal structural and electronic features. In addition to the universal bulk d-wave superconductivity at low temperature, these complex oxides share a common layered structure comprised of CuO_2 planes separated by perovskite-like buffer layers. We have used the CORELLI spectrometer at the Spallation Neutron Source to measure deviations from the average crystal structure in several cuprate materials, with a focus on the model system $HgBa_2CuO_{4+\delta}$ (Hg1201). Using structure modeling and the 3D-ΔPDF method, we are able to determine real-space correlations and their temperature- and doping-dependence. These structural correlations appear to be relevant to the emergence of superconductivity in the cuprates.

The work at the University of Minnesota was funded by the US Department of Energy through the University of Minnesota Center for Quantum Materials, under grant number DE-SC-0016371.

5:00 PM B3.08.04
Magnetic Fluctuations in Superconducting and Non-Superconducting 11 Iron Chalcogenides Igor Zaliznyak[1], Yangmu Li[2,1], Ovidiu Garlea[3], Andrei T. Savici[3], Zhijun Xu[4,1], Gu Genda[1] and John Tranquada[1]; [1]Brookhaven National Laboratory, United States; [2]Institute of Physics, China; [3]Oak Ridge National Laboratory, United States; [4]National Institute of Standards and Technology, United States

There are strong connections between magnetism and unconventional superconductivity in Fe-based superconductors. Recently, new aspects associated with their multi-band, multi-orbital nature and the band topology came to prominence in the iron chalcogenide family. Neutron scattering experiments revealed signatures of an orbital-selective electron localization (coherence-decoherence crossover) and ferro-orbital order in the FeTe parent material [1,2]. In the superconducting iron chalcogenide materials, the ARPES studies discovered significant temperature- and orbital-dependent correlation effects in the electronic band structure that were also indicative of orbital-selective Mott physics. More recently, superconductivity with the topological surface state was discovered in $Fe_{1+y}Te_{0.55}Se_{45}$, which was shown to be sensitive to compositional variations, such that both conventional and topological superconductivity, as well as non-superconducting charge-incoherent regions were shown to coexist on the same sample [4]. Here, we present polarized neutron scattering studies of magnetic dynamics in $Fe_{1+y}Te_{1-x}Se_x$ samples with and without superconductivity, which is tuned by Fe stoichiometry. We analyze the evolution of the dynamical magnetism with temperature and composition in order to understand the nature of magnetic and orbital electronic states and their relation with the superconductivity, such as the relation between Tc and the spin gap.
[1] I. A. Zaliznyak, et al. Phys. Rev. Lett. **107**, 216403 (2011).
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[3] M. Yi, et al. Nature Communications 6, 7777 (2015).
[4] Y. Li, et al. Nature Materials **20**, 1221–1227 (2021).

5:15 PM B3.08.05
Nematic Correlation Length in Iron-Based Superconductors Probed by Inelastic X-Ray Scattering Dmitry Reznik; University of Colorado-Boulder, United States

Nematicity is ubiquitous in electronic phases of high-T_c superconductors, particularly in the Fe-based systems. We used inelastic neutron and x-ray scattering to extract the temperature dependent nematic correlation length ξ from the anomalous softening of acoustic phonon modes in FeSe, underdoped $Ba(Fe_{0.97}Co_{0.03})_2As_2$, and optimally doped $Ba(Fe_{0.94}Co_{0.06})_2As_2$. In all cases, we find that ξ is well described by a power law $(T-T_0)^{-1/2}$ extending over a wide temperature range. Combined with the previously reported Curie-Weiss behavior of the nematic susceptibility, these results point to the mean-field character of the nematic transition, which we attribute to a sizable nematoelastic coupling that is likely detrimental to superconductivity.

SESSION BP3.09: Poster Session: Hard Condensed Matter
Wednesday Afternoon, June 8, 2022
5:30 PM - 7:30 PM
UMC Center Ballroom 210

BP3.09.01
Magnetic Excitations in the Highly Frustrated fcc Iridate K_2IrCl_6 Qiaochu Wang and Kemp Plumb; Brown University, United States

Magnetic materials with a high degree of geometrical frustration are prime candidates to realize exotic ground states governed by quantum fluctuations. K_2IrCl_6 is a model antiferromagnet with Ir^{4+} on a face-centered cubic lattice which is an exciting compound to explore Heisenberg-Kitaev exchange physics. The combination of crystal field and spin-orbit coupling renders a half-filled band of J_{eff}=1/2 character, which is localized by even moderate electronic correlations.[1] This J_{eff}=1/2 state and 90° Ir-O-Ir superexchange pathway favor both Heisenberg and Kitaev exchange interactions. In this talk, I will discuss inelastic neutron scattering measurements on K_2IrCl_6 and quantify the magnetic excitation spectra with a model including nearest and next-nearest neighbor Heisenberg interactions, Kitaev exchange and off-diagonal anisotropy. These measurements are essential to determine the Hamiltonian and thus where the material lies on the J-K-Γ phase diagram. The results of this study will provide experimental insights into the unconventional magnetic ground states and excitations in the frustrated Kitaev-type system and thus guide further searchers for quantum fluctuating phases and spin liquids.

[1] J.Rau et al, Condensed Matter Phys. 7, 195 (2016)

BP3.09.02
Fluctuating Pseudospin Dimers in J_{eff}-3/2 Cluster Mott Insulator Tsung-Han Yang[1], Tomoya Higo[2], Shinya Kawamoto[2], Joerg Neuefeind[3], Matthew Stone[3], SuYin Wang[4], Milinda Abeykoon[5], Yu-Sheng Chen[4], Satoru Nakatsuji[2] and Kemp Plumb[1]; [1]Brown University, United States; [2]The University of Tokyo, Japan; [3]Oak Ridge National Laboratory, United States; [4]Argonne National Laboratory, United States; [5]Brookhaven National Laboratory, United States

Spin, orbit, and lattice dynamics are coupled in strong spin-orbit coupling systems. The Lacunar spinel, $GaTa_4Se_8$, is a unique Mott insulator that hosts molecular spin-orbit J_{eff}-3/2 states as correlation units. However, little is known about the spin-orbital ground state and dynamics in this system. In this presentation, I will discuss a series of comprehensive x-ray and neutron pair total scattering phonon dynamics measurements that uncover a fluctuating pseudospin dimer phase in this cluster Mott insulator. $GaTa_4Se_8$ has concurrent structural and magnetic transitions at 50 K where a spin-orbital singlet

valence bond solid ground state forms. As a result of strong spin-orbit coupling and orbital degeneracy, the magnetic ground state couples with a lattice distortion. Our total scattering measurements show that the lattice is locally distorted up to at least 300 K, indicating the spin-orbital singlet transition is order-disorder type. Inelastic neutron measurements directly reveal the lattice fluctuations preceding the transition and provide direct evidence of a fluctuating spin-orbital liquid. This work provides insight into the interplay between spin, orbit, and lattice dynamics in the molecular J_{eff}-3/2 Mott insulators.

*This material is based upon work supported by the U.S. Department of Energy, Office of Basic Energy Sciences, under Award Number DE-SC0021223.

BP3.09.03
Temperature Dependence of Anharmonic Effects in NaBr by Inelastic Neutron Scattering and Machine Learning Interatomic Potentials Vladimir Ladygin[1], Claire N. Saunders[1], Camille Bernal[1], Michael E. Manley[2], Douglas L. Abernathy[2] and Brent Fultz[1]; [1]California Institute of Technology, United States; [2]Oak Ridge National Laboratory, United States

Harmonic and quasiharmonic theory of lattice vibrations have widely been used to explain the dynamics of solids. However, a large number of phenomena lie beyond their scope arising from higher-order phonon interactions. Recent inelastic neutron scattering experiments with ARCS at Oak Ridge National Laboratory have revealed new anharmonic effects. The rocksalt phase of NaBr has highly anharmonic phonons. Its thermal expansion is well explained by an anharmonic theory [1] DOI: 10.1103/PhysRevLett.125.085504. It also displays new anharmonic phenomenon: intrinsic localized modes and intermodulation phonon sidebands. These features occur in pairs resulting from anharmonic interactions of TA and TO phonons [2] DOI: 10.1103/PhysRevB.103.134302.

Here we describe another effect of high temperatures on the phonon dispersions of NaBr measured by INS. We find new spectral features in the optical modes, including patches of diffuse intensity and distinct changes in shapes of the dispersions. The acoustic modes are relatively unchanged. Prior observations of these features at 700 K were published previously in [1]. Recently we measured phonon dispersions in a single crystal of NaBr with ARCS at temperatures of 300, 450, and 650 K that confirm the original observations, and give information on the changes in optical modes with temperature. Data were post-processed using Mantid, reduced with an original multiphonon background correction, and folded back into a single irreducible wedge in the first Brillouin zone to obtain the 4D scattering function S(Q,E). The analyses of spectra allowed us to determine the temperature dependence of these new anharmonic features.

We support the experimental data with classical molecular dynamics simulations using machine learning interatomic potentials. The phonon spectrum along high symmetry directions is calculated via the mode-projected velocity autocorrelation function method to include infinite order anharmonic effects. Tentative interpretations of these high temperature phonon dynamics will be presented at the meeting.

This work is supported by DOE BES award No. DE-FG02-03ER46055.

BP3.09.05
High-Field Low-Energy Spin Dynamics in the Kitaev QSL Candidate α-RuCl3 Kiranmayi Dixit[1], Colin Sarkis[2], Barry Winn[2], Stephen Nagler[2], David Mandrus[2], Christian Balz[2] and Arnab Banerjee[1]; [1]Purdue University, United States; [2]Oak Ridge National Laboratory, United States

alpha-RuCl3 is one of the best candidates for materializing a Kitaev quantum spin liquid (QSL). The observation of the half-integer thermal quantum Hall effect in α-RuCl3 in its field-induced quantum spin liquid state, at a magnetic field of 8 T has led to the debate on the presence of a chiral spin gap in this phase. We investigate the magnetic excitations in this material using Neutron Scattering experiments to investigate if there is a spin gap and pay special attention to the overall nature of the low-energy excitations resolved in the 4D momentum, energy as a function of a magnetic field up to 13.5 T.

BP3.09.06
Giant Doping Response of Magnetic Anisotropy in MnTe Duncan H. Moseley, Keith Taddei, David Parker, Randy Fishman and Raphael P. Hermann; Oak Ridge National Laboratory, United States

Developing simple ways to control spin states in spintronic devices is a crucial step towards increasing their functionality. MnTe is a room-temperature antiferromagnet with promising spintronic properties, including for thermospintronics and magnon-based devices. Here, we show that, in MnTe, as little as 0.3% Li is sufficient to produce a dramatic spin reorientation as observed by neutron diffraction. The behavior of the 0001 magnetic Bragg peak reveals a significant reorientation of the Mn spins from planar in the pure material to almost completely axial with minimal Li doping. Temperature dependence of the magnetic peaks in Li-doped samples indicates that axial spins shift back to planar suddenly upon approaching the Néel temperature (T_N = 307 K). Density functional theory calculations support the idea that a shift in the Fermi level caused by doping is responsible for switching the material between two competing magnetic ground states. This takes an essential step towards further development of the easy switching of spin orientations in spintronic material and devices such as spin valves and promises to open new avenues of spintronics and transport research in antiferromagnetic semiconductors.

BP3.09.07
Unveiling the Magnetic Structure of the Topological Semimetal Co3Sn2S2 with Spherical Neutron Polarimetry Jian Rui Soh[1], ChangJiang Yi[2], Ivica Zivkovic[1], Navid Qureshi[3], Anne Stunault[3], Bachir Ouladdiaf[3], Jose Alberto Rodriguez-Velamazan[3], YouGuo Shi[4], Henrik M. Ronnow[1] and Andrew Boothroyd[5]; [1]EPFL, Switzerland; [2]Max Planck Institute for Chemical Physics of Solids, Germany; [3]Institut Laue-Langevin, 71 Avenue des Martyrs, France; [4]Institute of Physics, Chinese Academy of Sciences, China; [5]University of Oxford, Clarendon Laboratory, United Kingdom

Recently, $Co_3Sn_2S_2$ has been predicted to be the first clear example of a magnetic topological semi-metal in which the topological features in the electronic bands are generated spontaneously by its magnetic order. Since then, $Co_3Sn_2S_2$ has garnered a tremendous amount of interest, with more than 50 reports over the last two years.

In spite of the extensive literature on $Co_3Sn_2S_2$, up to now, there are no direct measurements of the magnetic structure, which is key to its behavior as a topological material. This is mainly due to the small size of the Co magnetic moment, which makes the experimental determination of the magnetic structure tremendously challenging.

In this work, we (1) unambiguously determine the ground state magnetic structure of $Co_3Sn_2S_2$, thereby solving a key issue about this material, and (2) experimentally discovered the nature of a transition that has been observed well below the magnetic ordering temperature. These achievements were made possible by the use of an advanced polarized neutron diffraction technique. The experimental evidence presented in our work, lends support to theoretical predictions that $Co_3Sn_2S_2$ is a magnetic Weyl semimetal.

BP3.09.10
Quantum Disordered State of Magnetic Charges in Nanoengineered Honeycomb Lattice George Yumnam[1], Yiyao Chen[2,1], Jiasen Guo[1], Jong Keum[3], Valeria Lauter[3], Pousali Ghosh[1] and Deepak K. Singh[1]; [1]University of Missouri, Columbia, United States; [2]Suzhou Institute of Nano-Tech and Nano-Bionics (SINANO), Chinese Academy of Sciences, China; [3]Oak Ridge National Laboratory, United States

A quantum magnetic state due to magnetic charges is never observed, even though they are treated as quantum mechanical variables in theoretical calculations. Here, the occurrence of a novel quantum disordered state of magnetic charges in a nanoengineered magnetic honeycomb lattice of ultra-small connecting elements is demonstrated. A magnetic honeycomb lattice with competing exchange interactions between Ising moments is theoretically predicted to exhibit disordered magnetic states with macroscopic degeneracy. The experimental research, performed using spin-resolved neutron scattering, reveals a robust massively degenerate ground state, comprised of low integer and energetically forbidden high integer magnetic charges, that manifests cooperative paramagnetism at low temperature. We explored the competing physics of energy vs. entropy in a thermally tuned magnetic phase transition. The system tends to preserve the degenerate configuration even under large magnetic field applications. It exemplifies the robustness of the disordered correlation of magnetic charges in a 2D honeycomb lattice. The realization of a quantum disordered ground state elucidates the dominance of exchange energy, which is enabled due to the nanoscopic magnetic element size in nanoengineered honeycomb. Consequently, an archetypal platform is envisaged to study quantum mechanical phenomena due to emergent magnetic charges.

BP3.09.11
***Q*-dependent Collective Relaxation Dynamics of Glass-Forming Liquid $Ca_{0.4}K_{0.6}(NO_3)_{1.4}$ Investigated by Wide-Angle Neutron Spin-Echo** Peng Luo[1], Yanqin Zhai[2], Peter Falus[3], Victoria García-Sakai[4], Monika Hartl[5], Maiko Kofu[6], Kenji Nakajima[6], Antonio Faraone[7] and Yang Zhang[2]; [1]University of Pennsylvania, United States; [2]University of Illinois at Urbana-Champaign, United States; [3]Institut Laue-Langevin, France; [4]ISIS Neutron and Muon Facility, United Kingdom; [5]European Spallation Source, Sweden; [6]J-PARC Center, Japan; [7]National Institute of Standards and Technology, United States

The relaxation behavior of glass formers exhibits spatial heterogeneity and dramatically changes upon cooling towards the glass transition. However, the underlying mechanisms of the dynamics at different microscopic length scales are not fully understood. Employing the recently developed wide-angle neutron spin-echo spectroscopy technique, we measured the *Q*-dependent coherent intermediate scattering function of a prototypical ionic glass former $Ca_{0.4}K_{0.6}(NO_3)_{1.4}$, in the highly viscous liquid states. In contrast to the structure modulated dynamics for $Q < 2.4$ Å^{-1}, i.e., at and below the structure factor main peak, for $Q > 2.4$ Å^{-1}, beyond the first minimum above the structure factor main peak, the stretching exponent exhibits no temperature dependence and concomitantly the relaxation time shows smaller deviations from Arrhenius behavior. This finding indicates a change in the dominant relaxation mechanisms around a characteristic length of $2\pi/(2.4$ Å$^{-1}) \approx 2.6$ Å, below which the relaxation process exhibits a temperature independent distribution and more Arrhenius-like behavior.

BP3.09.12
Anomalous Hall Effect due to Topological Magnetic Charge Correlation in Permalloy Honeycomb Lattice Jiasen Guo[1], Ashutosh Dahal[1], George Yumnam[1], Yiyao Chen[2], Pousali Ghosh[1], Valeria Lauter[3], Vitalii Dugaev[4], Arthur Ernst[5,6] and Deepak K. Singh[1]; [1]University of Missouri, Columbia, United States; [2]Suzhou Institute of Nano-Tech and Nano-Bionics,, China; [3]Oak Ridge National Laboratory, United States; [4]Rzeszow University of Technology, Poland; [5]Johannes Kepler Universitat, Austria; [6]Max-Planck-Institut fur Mikrostrukturphysik, Germany

Two-dimensional artificial honeycomb lattice of connected ferromagnetic elements undergoes temperature dependent evolution of magnetic phases and provides a facile platform for exploring novel electric and magnetic properties. Polarized neutron reflectometry and Hall probe measurements on a honeycomb lattice of connecting permalloy elements, with a typical length of 12 nm, suggest that as temperature decreases, the system undergoes a magnetic phase transition that renders the Hall resistance from the anomalous Hall effect (AHE) type to an unusual quasi-oscillatory type at low temperature. The oscillatory behavior in Hall resistance provides evidence to the emergence of topologically nontrivial chiral magnetic vortex loops in the novel spin solid state at low temperature. The experimental observation is supplemented by first-principle theoretical calculations based on a 2DEG model of the spin solid state. Finally, we will discuss the implication of the Hall probe results in the broader perspective of condensed matter physics research on 2D materials.

BP3.09.13
Structure, Dynamics of a Two-Dimensional Metal Halide Perovskite Haritha S. Rajeev; University of Virginia, United States

Hybrid organic-inorganic perovskites (HOIPs), such as CH3NH3PbI3 (MAPbI3) and CH(NH2)2PbI3 (FAPbI3), have shown a potential to become a commercially widely used solar cell due to their high- power conversion efficiency and low economic cost. The HOIPs based solar cell has hit the efficiency record above 20%[1], however, the biggest challenge of the high-efficiency perovskite solar cells is their device instability. Recent studies on two-dimensional (2D) HOIPs outlined their advantageous properties such as a photovoltaic efficiency of 12.52% without any obvious degradation over 2250 hours under standard light illumination, 650hrs at 65% relative humidity [2], and tunable bandgap and high light absorption [3,4]. Understanding these extraordinary properties requires thorough investigation of the crystal structures of 2D HOIPs.
Here we study temperature dependent structure evolution of a 2D perovskite, (BdA)PbI4 (BdA= NH3(CH2)4NH3). We investigate how the organic molecule and inorganic molecule configuration evolve with temperature. This question is important because our previous studies[4-6] on the 3D perovskite samples have demonstrated that the organic molecules in HOIPs play a central role in extending the lifetime of photoexcited charge carriers, leading to the high efficiency. In this purpose, quasi-inelastic neutron scattering (QENS) data measured at J-PARC will be reported, investigating the temperature-dependent rotational dynamics of (BdA)PbI4. We will also present how the inorganic molecule configuration evolve with temperature and how it effects the electronic structure and band gap of the system. We performed neutron diffraction measurements at Oak Ridge National Laboratory to obtain the crystal structure information and fitted the structure using GSAS Rietveld refinement. Ab-initio quantum mechanical molecular dynamic calculations to obtain the electronic band structure information were done using Oak Ridge Leadership Computing Facility.
References
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BP3.09.14
Influence of Synthesis Atmosphere on the Magnetic and Structural Properties of $TbMnO_3$ Multiferroic Polycrystals Gustavo S. Dias[1], Luiz F. Cotica[1], Ivair A. Santos[1], Adelino de Aguiar Coelho[2], Juraci Aparecido Sampaio[3], Luiz G. Danvanse da Silveira[4] and Fabiano Yokaichiya[4]; [1]State

University of Maringá, Brazil; [2]State University of Campinas, Brazil; [3]State University of Northern Fluminense Darcy Ribeiro, Brazil; [4]Federal University of Paraná, Brazil

There has been a revival in studying the magnetoelectric behavior/state of magnetic ferroelectrics since the discovery of magnetic field controllable ferroelectric polarization in perovskite manganite $TbMnO_3$ single crystals, and giant ferroelectric polarization in $BiFeO_3$ thin films. Specifically, the $TbMnO_3$ compound is a multiferroic magnetoelectric material at cryogenic temperatures due to a transition between two distinct magnetic states. In fact, $TbMnO_3$ single crystals exhibit a para-antiferromagnetic phase transition at 41 K, where the Mn^{3+} ions develop a sinusoidal ordering propagating along the b-direction of the unit cell. The wave vector of the magnetic modulation is incommensurate at the AF transition and becomes locked-in below 28 K, where $TbMnO_3$ becomes a ferroelectric. The incommensurate nature of the magnetic structures observed in $REMnO_3$ (RE = Rare Earths) comes from an increase of the frustration effects due to exchange competitions in Mn-O-Mn bonds. In this contribution, nanostructured $TbMnO_3$ polycrystals were synthesized by high-energy ball milling and annealed under different atmospheres (Argon or Oxygen). The obtained high-resolution X-ray powder diffraction (HR-XPD), X-ray absorption fine-structure (XAFS), high-resolution neutron powder diffraction, magnetic and dielectric measurements data allowed us to observe the influence of vacancies on structural and ferroic properties of nanostructured $TbMnO_3$ and will be discussed in detail.

SESSION B4.10: Spin Textures and Helimagnets
Session Chair: Andrew Christianson
Thursday Morning, June 9, 2022
UMC East Ballroom 212

10:15 AM *B4.10.01
Skyrmion Lattice Manipulation with Electric and Thermal Currents Morten R. Eskildsen; University of Notre Dame, United States

Magnetic skyrmions show promise for future data storage applications due to their topological protection and easy manipulation. An electric current exerts two forces on the skyrmions: a drag force parallel to the current and a Magnus force perpendicular. When these forces overcome pinning effects they drive a translation of the skyrmions.
Here we report on small-angle neutron scattering (SANS) studies of the SkL in MnSi under the influence of radial electric and thermal currents. Using a (semi)circular sample the electrical current has a $1/r$ dependence, resulting in a torque on the SkL and thus causing a rotation even in the absence of thermal gradients or other extraneous effects. Interestingly, the SkL rotation shows a nonmonotonic current dependence and changes sign at high currents. We understand the switching to be due to a competition between the electrical current and the accompanying thermal current in the sample. Experimentally, the two effects can be separated by having the two currents flowing in the same or opposite directions. Modelling of the data, including a finite element analysis of the sample current and temperature profiles, will be discussed.
Second, the magnetic self-field generated by the electrical current is perpendicular to both the current direction and the applied magnetic field. The skyrmions are weakly coupled to the crystalline directions in the host material and will closely follow the direction of the applied magnetic field. As a result, the self-field causes of a tilting of the the SkL which is observed directly in our SANS measurements.

10:45 AM B4.10.02
Neutron Diffraction Study of Complex Helical Magnetic Ordering in Ni-doped $EuCo_2As_2$ Single Crystals Tianxiong Han[1,2], Simon X. Riberolles[1], Benjamin G. Ueland[1], R. J. McQueeney[1,2], Yan Wu[3], Santanu Pakhira[1] and D. C. Johnston[1,2]; [1]Ames Laboratory, United States; [2]Iowa State University of Science and Technology, United States; [3]Oak Ridge National Laboratory, United States

$EuCo_{2-y}As_2$ crystallizes in the uncollapsed-tetragonal $ThCr_2Si_2$-type structure and exhibits *c*-axis helical antiferromagnetic (AF) ordering of the Eu below T_N=47 K with an AF propagation vector of t=(0,0,0.73(7)). $EuNi_{2-y}As_2$ is the electron-doped analogue which has the collapsed-tetragonal structure and shows *c*-axis helical-AF ordering below T_N=15 K with τ=(0,0,0.92). Here, we present results from neutron diffraction measurements on single-crystal samples that reveal changes to the magnetic ordering of $Eu(Co_{1-x}Ni_x)_{2-y}As_2$ across the doping series. We find good agreement with the magnetic phase diagram derived from previous thermodynamic and transport measurements[1]. For x=0.1, we find a *c*-axis helical structure with the Eu ferromagnetically aligned within the ab plane and the planes AF stacked along c consistent with τ~(0,0,0.67). With more Ni doping, upon cooling, ferromagnetic ordering of the Co/Ni sets in prior to AF ordering of the Eu. This creates a complex ground-state magnetic structure with a superposition of the in-plane helix structure and *c*-axis ferromagnetism. t changes with increasing x, reaching ~(0, 0, 0.85) at x=0.8.

This work is supported by the U.S. Department of Energy, Office of Basic Energy Sciences, Division of Materials Sciences and Engineering. Ames Laboratory is operated for the U.S. Department of Energy by Iowa State University under Contract No. DE-AC02-07CH11358. This research used resources at the High Flux Isotope Reactor, a DOE Office of Science User Facility operated by the Oak Ridge National Laboratory.

[1]. Magnetic phase transitions in $Eu(Co_{1-x}Ni_x)_{2-y}As_2$ single crystals. N. S. Sangeetha, Santanu Pakhira, D. H. Ryan, V. Smetana, A.V.Mudring, and D. C. Johnston, Phys. Rev. Materials **4**, 084407 (2020)

11:00 AM B4.10.03
Three-Dimensional Neutron Tomography of a Bulk Skyrmion Lattice Melissa E. Henderson[1], Benjamin Heacock[2], Markus Bleuel[2], Colin Heikes[2], Michael G. Huber[2], Jeff Krzywon[2], Olivier Nahman-Levesque[1], Mathew Pula[3], Dusan Sarenac[1], Kirill Zhernenkov[4], David Cory[1] and Dmitry Pushin[1]; [1]Institute for Quantum Computing, University of Waterloo, Canada; [2]NIST Center for Neutron Research, United States; [3]McMaster University, Canada; [4]Julich Centre for Neutron Science, Germany

Skyrmions are topologically protected structures with non-collinear spin configurations, thought to host a zoology of defects and unique spin textures. Hailed for their promise as information carriers in high density memory and spintronics applications, a detailed account of their lattice dynamics and stabilization in three-dimensions is crucial to their efficient manipulation in future devices. Skyrmion topological transitions are thought to be controlled by the proliferation and propagation of Bloch points, associated with emergent (anti)monopoles, in the bulk. However, three-dimensional visualizations of skyrmion lattices have thus far remained elusive. Here we report the observation of a room temperature skyrmion lattice in the bulk via the development and application of a novel small angle neutron scattering tomography algorithm. Reconstructions show a highly dynamic lattice, providing evidence of 3D skyrmion lattice topological transitions through two different emergent monopole-antimonopole defect pathways. We examine the equivalence of changes in local topology with defect mobility, tuning the contributions of internal

disorder, temperature, and external magnetic field. These observations provide access to skyrmions on bulk lengthscales, establishing experimental realizations and mechanisms of skyrmion formation/annihilation processes in the bulk. These results motivate future studies of true bulk systems unburdened by shape and thickness constraints, over a host of temperatures, fields, and compositions.

11:15 AM B4.10.05
Novel Magnetic Structures in M1/3TaS2 Junjie Yang and Yunpeng Gao; New Jersey Institute of Technology, United States

Exotic physical phenomena are bound to happen in chiral magnets, since broken space inversion, mirror symmetry and time reversal symmetry often accompany new physical phenomena. For example, the Dzyaloshinskii Moriya (*DM*) interactions, allowed by the lattice chirality, could bring about a twist between the magnetic moments and lead to various novel chiral magnetic structures (e. g., magnetic chiral solution and Skyrmion lattice). So far, very few chiral magnets have been discovered. Recently, we successfully synthesized single crystals of $M_{1/3}TaS_2$ (M = Cr, Co and Ni) which are new chiral magnet candidates. $M_{1/3}TaS_2$ crystallizes in a non-centrosymmetric chiral hexagonal space group $P6_322$ with M atoms intercalated between 2H-stacked TaS_2 layers. Strong *DM* interactions may exist in $M_{1/3}TaS_2$ and induce novel physicsl propeties. In this work, we will discuss the novel magnetic structures, bulk magnetic susceptibilties and magnetotransport properties of $M_{1/3}TaS_2$.

11:30 AM B4.10.06
Field-Tunable Toroidal Moment in a Chiral-Lattice Magnet Huibo Cao[1], Lei Ding[1], Xianghan Xu[2], Harald Jeschke[3], Xiaojian Bai[1], Erxi Feng[1], Admasu Alemayehu[2], Jaewook Kim[2], Feiting Huang[2], Qiang Zhang[1], Xiaxin Ding[4], Neil Harrison[4], Vivian Zapf[4], Daniel Khomskii[5], Igor Mazin[6] and Sang-Wook Cheong[2]; [1]Oak Ridge National Laboratory, United States; [2]Rutgers University, United States; [3]Okayama University, Japan; [4]Los Alamos National Laboratory, United States; [5]II. Physikalisches Institut, Universität zu Köln, Germany; [6]George Mason University, United States

A toroidal dipole moment appears independent of the electric and magnetic dipole moment in the multipole expansion of electrodynamics. It arises naturally from vortex-like arrangements of spins. A toroidal order breaks various combinations of global symmetries, such as time and space inversion. Observing and controlling spontaneous long-range orders of toroidal moments are highly promising for spintronics but remain challenging. Here we demonstrate that a vortex-like spin configuration with a staggered arrangement of toroidal moments, a ferritoroidal state, is realized in a chiral triangular-lattice magnet $BaCoSiO_4$. Upon applying a magnetic field, we observe multi-stair toroidal transitions correlating directly with metamagnetic transitions. We establish a first-principles microscopic Hamiltonian that explains both the formation of toroidal states and the metamagnetic toroidal transition as a combined effect of the magnetic frustration and the Dzyaloshinskii-Moriya interactions allowed by the crystallographic chirality in $BaCoSiO_4$.

Lei Ding, Xianghan Xu, Harald O. Jeschke, Xiaojian Bai, Erxi Feng, Admasu Solomon Alemayehu, Jaewook Kim, Feiting Huang, Qiang Zhang, Xiaxin Ding, Neil Harrison, Vivien Zapf, Daniel Khomskii, Igor I. Mazin, Sang-Wook Cheong, Huibo Cao, "Field-tunable toroidal moment in a chiral-lattice magnet", *Nature Communications*, **12**, 5339 (2021).

*The work is supported by the US DOE Early Career Research Program Award KC0402020 and used resources at HFIR and SNS, the DOE Office of Science User Facility operated by ORNL.

11:45 AM B4.10.07
Slow Relaxation with Signature of Glassiness in Non-Centroysmmetric Helimagnet ScFeGe Sunil K. Karna[1], John F. DiTusa[2], David Young[3], Wei Tian[4] and Adam Aczel[4]; [1]Norfolk State University, United States; [2]Indiana University-Purdue University Indianapolis, United States; [3]Louisiana State University, United States; [4]Oak Ridge National Laboratory, United States

Non-centrosymmetric ScFeGe hosts an incommensurate helimagnetic state along the *c*-axis below 36 K that matches well with a nesting condition found in its electronic structure. Here, we present the observation of low frequency dynamics in hexagonal helimagnet ScFeGe that are apparent in the *ac* susceptibility near the metamagnetic transition below 15 K, correlating well with the increased entropy and positive MR in this range of *H* and *T*. Single-crystal neutron diffraction refinement yields a magnetic moment $\approx 0.53\mu_B$ (confined to the *ab* plane), indicating highly itinerant magnetic system and displays a nesting vector along the *c*-axis and order in a helical state similar to rare Earth metal like Ho or Dy. Rare-Earth metals show multiple transitions such as fan or helifan state at higher temperatures and tend towards ferromagnetism at low temperature, whereas ScFeGe remains helical without a change in wave vector $k = (0\ 0\ 0.193)$ for T< 15 K and fan-type state at higher temperatures.

12:00 PM B4.10.09
Depth Profiles of Hybrid Magnetic Skyrmions Determined by Neutron Scattering WLNC Liyanage[1], Nan Tang[1], Lizabeth Quigley[1,2], Sergio Montoya[3], Julie A. Borchers[4], Alexander Grutter[4], Sunnil Sinha[3], Brian Maranville[4], Eric Fullerton[3], Lisa DeBeer-Schmitt[5] and Dustin A. Gilbert[1,1]; [1]The University of Tennessee, Knoxville, United States; [2]Purdue, United States; [3]University of California, San Diego, United States; [4]National Institute of Standards and Technology, United States; [5]Oak Ridge National Laboratory, United States

Magnetic skyrmions are topologically protected chiral structures which present opportunities for next generation magnetic data storage and logic information technologies. The topology of these structures originates in the geometric configuration of the magnetic spins – more generally described as its structure. Most often, the skyrmion structure is depicted as a 2D projection, however recent works have emphasized the role of the 3D structure in topology. In this work we use a combination of reflectometry, small-angle neutron scattering (SANS) and grazing-incidence small-angle neutron scattering (GISANS) to determine the depth profile of hybrid skyrmions. The structure of the hybrid skyrmions, which includes a combination of Néel-like and Bloch-like components along their length, is expected to strongly contribute to their stability. To interpret the data, micromagnetic simulations of the hybrid skyrmions were performed, then the corresponding diffraction patterns determined using a Born approximation transformation. The converged magnetic profile reveals the magnetic structure along the skyrmion depth, including the thickness of the Bloch and Néel segments, and diameter of the core.

12:15 PM B4.10.10
Revisiting Static and Dynamic Magnetic Correlations in the Chiral Helimagnet Cr1/3NbS2 Lisa DeBeer-Schmitt[1], Lazar Kish[2], Adam Aczel[1], Travis J. Williams[1], Huibo Cao[1], Timothy Charlton[1], Nirmal Ghimire[3], Jacob Ruff[4], Michael A. McGuire[5], Stephen J. Kuhn[6], Morten R. Eskildsen[7] and David Mandrus[8]; [1]ORNL, United States; [2]University of Illinois at Urbana-Champaign, United States; [3]George Mason University, United States; [4]Cornell University, United States; [5]Oak Ridge National Laboratory, United States; [6]Indiana University-Bloomington, United States; [7]University of Notre Dame, United States; [8]The University of Tennessee, Knoxville, United States

Transition metal dichalcogenides intercalated with magnetic ions can crystallize in a chiral, hexagonal space group. Competition between Heisenberg exchange, the Dzyaloshinskii-Moriya interaction, magnetocrystalline anisotropy and an applied magnetic field in the monoaxial chiral magnet Cr1/3NbS2 lead to interesting long-period magnetic structures such as a helimagnetic and chiral soliton lattice states. We recently performed a small-angle neutron

scattering experiment to characterize these states in detail. We observed higher-order peaks in our zero-field data, which are unexpected for an ideal helical state with perfect sinusoidal modulation. These signatures of anharmonic distortion could be due to undiscovered magnetocrystalline anisotropy, or metastable behavior due to disorder effects.

Funding Acknowledgments

This research used resources at the High Flux Isotope Reactor and Spallation Neutron Source, a DOE Office of Science User Facility operated by the Oak Ridge National Laboratory.

Soft Matter

* Invited Paper

SESSION C1.01: Field-Driven Structures in Soft Matter
Session Chair: Xiaodan Gu
Monday Morning, June 6, 2022
UMC Conference Room 235

10:15 AM *C1.01.01
Altering Block Copolymer (BCP) Self-Assembly and Phase Behavior via Magnetic Field Processing Grace V. Kresge, Karthika Suresh and Michelle A. Calabrese; University of Minnesota Twin Cities, United States

Block copolymers (BCPs) are attractive for developing novel materials due to their tunable properties and self-assembly via block chemistry, composition, and length. However, practical methods for processing BCPs into advanced materials with long-range order remain difficult, where techniques like magnetic field alignment are typically infeasible because of large required field strengths and limited range of responsive chemistries. We recently discovered anomalous magnetic field-induced ordering in weakly diamagnetic, aqueous block polymers (BCPs) exposed to low intensity magnetic fields ($0.05 \leq B \leq 0.5$ T). Prior work on magnetic field-directed assembly in BCPs has focused on alignment of a structure or phase with inherent anisotropy which leads to anisotropy in the magnetic susceptibility, $\Delta\chi$. Conversely, isotropic ferromagnetic particles can align into anisotropic, chain-like structures in applied fields due to dipole alignment; these structures relax upon field removal. However here, ordered phases are created by temporarily applying magnetic fields to low viscosity solutions of spherical BCP micelles with no inherent anisotropy, causing up to a six order of magnitude increase in the dynamic moduli. Additionally, induced phases are often stable for days upon field removal; thus dipole alignment and phase alignment are seemingly eliminated as mechanisms driving this behavior.

Using a combination of magnetorheology (MR) and small angle neutron and x-ray scattering (SANS/SAXS), we show that two types of ordered phases form under low intensity fields: those that remain composed of isotropic micelles (i.e. cubic phases), and those that transform into aligned phases. Interestingly, the induced elastic modulus, G'_B, is up to three orders of magnitude larger than the maximum modulus that results from thermal phase transitions at 0 T. This anomalous assembly is time-dependent, where the critical induction time and phase formed depend strongly on molecular weight and processing conditions. Evidence from SANS and vibrational spectroscopy suggests that low intensity magnetic fields facilitate phase transitions primarily by altering polymer-solvent interactions and hydrogen bonding. This new assembly strategy enables the discovery of structures and d-spacings inaccessible via traditional self-assembly, thus providing a platform for developing materials with long-range order using mild conditions and little energy input from external fields.

10:45 AM C1.01.02
Measuring and Modeling Interactions Between Orientable Nanoparticles in Flow Patrick T. Corona, Jiamin Zhang, L. Gary Leal and Matthew Helgeson; University of California Santa Barbara, United States

Flow processing of elongated particle dispersions produces a fluid with anisotropic material properties due to the coupling of particle alignment to the flow field. In non-dilute dispersions, the interplay between particle alignment, interparticle interactions and the flow field significantly changes the structure of such dispersions, and this flow-modification is critical for describing anisotropic material properties including rheology and dielectric properties. Previously developed theories make a number of different assumptions and treatments for interparticle interactions in flow, highlighting a need for new experiments and models to isolate and characterize the effects of flow on interparticle alignment and center-of-mass structure. To fill this gap, we report measurements of the structure of rod-like colloidal cellulose nanocrystal (CNC) dispersions from *in situ* flow small angle neutron scattering (flow-SANS) measurements across a range of flow types (including shear, extension and mixed flows) and deformation rates (characterized by the dimensionless rate, the Weissenberg number). By employing a recently developed method to independently infer particle orientation probability distribution functions (OPDFs) [1], we isolate the contributions to the OPDF from single-particle and pair-orientations and, in doing so, the anisotropic static structure factor $S(\mathbf{q})$ that captures interparticle interactions in flow. The resulting measurements are used to test assumptions about the flow-induced spatial structure that are typically made in theories for interacting particle dispersions. In general, we find that $S(\mathbf{q})$ for all flow types is accurately described by a generalized anisotropic form of the random phase approximation with a scalar, isotropic interaction parameter at sufficiently low Weissenberg numbers and concentrations, suggesting that, in this regime, flow-induced structuring of the fluid is dominated by thermodynamic interactions between suspended particles. At elevated shear rate and concentration, the apparent interaction parameter becomes rate-dependent, suggesting a transition to a regime where hydrodynamic interactions become important for determining interparticle structure. The results provide a simple modeling framework for describing the effects of flow on interparticle structure using the OPDF, and provide critical insights for testing and developing models for the dynamics of elongated particle dispersions that incorporate interparticle interactions.

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11:00 AM C1.01.03

Simultaneous Measurement of Structure and Rheology of Rod like Systems at High Shear Rates Katie M. Weigandt, Ryan P. Murphy, Steve Kuei, Paul Salipante and Steven D. Hudson; National Institute of Standards and Technology, United States

Our team is creating new tools to enable simultaneous rheology and neutron scattering measurements toward the development of structure-function relationships in complex fluids at high strain rates. Our current suite of tools include a prototype slit rheometer for relatively low viscosity or shear thinning fluids (infinite shear viscosity < 5 mPa s) at shear rates up to 100,000 s^{-1} and a capillary rheoSANS instrument capable of simultaneously measuring structure and rheology in excess of 10^6 s^{-1}. We will discuss recent work that uses a combination of Couette, capillary and slit rheometry to study structure and rheology in wormlike micelle solutions and fD virus suspensions. In both cases we are directly measuring fluids that continue to be strongly shear thinning well beyond the limits of traditional rheoSANS measurements. While fD virus provides a fixed "rigid" rod system for us to study, commercially available cationic and/or anionic surfactants can be formulated with sodium chloride and/or sodium salicylate in D_2O to form extended wormlike micelles of variable length. For instance, in the case of CTAB wormlike micelles, the salt/surfactant ratio is varied to tune the quiescent morphology of the micelles. The structure and rheology of these micelles systems are characterized at low shear rates (< 10^3 s^{-1}) with traditional rheoSANS and high shear rates (10^3-10^6 s^{-1}) with capillary rheoSANS to directly correlate rheology and structure. By controlling the quiescent length distribution we plan to examine the extent to which SANS can be used to characterize changes in the length distribution of aligned wormlike micelles with increasing shear rate which is critical to our unravelling the origin of the apparent reduction in alignment at high shear rates observed for some wormlike micelle solutions.

11:15 AM C1.01.04
Rheo-Small Angle Neutron Scattering Measurements of Shear-Thickening Colloidal Suspensions with Varying Interparticle Friction Yu-Fan J. Lee[1], Scott Brown[2] and Norman Wagner[1]; [1]University of Delaware, United States; [2]The Chemours Company, United States

Small angle neutron scattering under shear flow provides deep and unique insights into the micromechanics of flow of soft matter and biological systems. Of interest here is the underlying mechanism(s) of shear thickening in colloidal suspensions where there is substantial, current controversy about the nanoscale interparticle interactions that govern the macroscopic flow properties. As efforts in theoretical models and simulations seek to examine the fundamental physics for strong to discontinuous shear thickening, the current consensus reveals the key role of tangential constraint controlled by nanometric inter-particle interactions, such as enhanced lubrication hydrodynamics [1,2] or frictional contact forces <!--[endif]---->[3-5]. However, experimental tests of these latest theories are lacking especially in microstructure at particle level. In this work, we seek to understand the relationship between microstructure and rheology of colloidal suspensions with varying surface friction level. Microstructures under flow are measured via small angle neutron scattering in both velocity-gradient and velocity-vorticity plane. The results illustrate that, no matter the friction level of particles or the projection of plane, structures formations is always observed with a characteristic length scale of a few particle diameters. This is in agreement with prior rheo-USANS and direct rheo-microscopy measurements, but in conflict with all simulations, which predict similar, longer-range structure formation that evident in experiment. Meanwhile, it is found that an opposite trend in first normal stress differences for two model suspensions with different friction level can distinguish between these micromechanical mechanisms for shear thickening, and this difference is reflected in the shear-induced anisotropic microstructures measured in the velocity-gradient plane [6]. Statistical comparison of experiments with simulations indicates that better resolution of microstructures in simulation models are required to be validated by the experimental data presented. In general, investigation of rheology and microstructure provide quantitative information valuable to understand the mechanistic role of lubrication hydrodynamics and frictional contact in shear thickening as well as the overall microstructure in this intriguing rheological state. <!--[endif]---->
<!--[endif]---->[1] M. Wang, S. Jamali, and J. F. Brady, Journal of Rheology **64**, 379 (2020).
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[3] R. Mari, R. Seto, J. F. Morris, and M. M. Denn, Journal of Rheology **58**, 1693 (2014).
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11:30 AM C1.01.06
Extracting Meaning from Alignment Factor Peter Gilbert[1], Yun Liu[1,2] and Paul D. Butler[1]; [1]NIST Center for Neutron Research, United States; [2]University of Delaware, United States

A material's physical and rheological properties can be tuned by manipulating molecular and particle orientation. However, orientation distributions at the nanoscale can only be observed by a few techniques, including small-angle neutron scattering (SANS). Translation of 2D SANS patterns into 3D orientation distributions is challenging and computationally expensive. Alignment factors offer a simple method for estimating orientation, but there are no clear standards for computing alignment factors. Several methods exist to calculate alignment factor, where each method results in a different estimate of alignment. We explore each of these methods and produce recommended standards for their implementation. Using theoretical and experimental data for aligned and differently oriented systems, we examine how alignment factors relate to orientation and assess their utility as an analysis method. Our recommended process for producing alignment factor is the first step to expanding the usefulness of this simple 2D analysis technique.

11:45 AM C1.01.07
Probing Topological Transitions of Inverse Worm-like Micelles Subject to Transient Shear Flow using Dielectric RheoSANS Noah J. Cho[1] and Jeffrey J. Richards[2]; [1]Corning Korea, Korea (the Republic of); [2]Northwestern University, United States

Worm-like micelles (WLMs) are ubiquitous viscoelastic modifiers in consumer products, cosmetics, and pharmaceuticals. Though WLMs are often described as polymer-like fluids, they exhibit unique dynamics due to their self-assembled structure and ability to break and reform. These dynamics contribute to an abundant and not well-understood microstructural behavior in response to shear flow. Recent work has focused on the crucial role that chain worm topology plays in the non-linear rheology of WLMs including the onset of shear-banding in steady and transient flows. In particular, the rheological behavior of unbranched and branched WLMs exhibit distinct differences when subjected to transient flow. We have recently used dielectric rheology to study the quiescent properties of WLMs including the transition from unbranched to branched chains using an inverse microemulsion system formed from lecithin/water/decane. In this system, branching occurs as the micelles are swollen with water and is identified by a maximum in the zero-shear viscosity with water content. We have extended quiescent studies of the dielectric properties to steady flows and identified distinct dielectric signatures at low frequencies associated with micelle breakage that are unique in the branched samples versus unbranched. To interrogate this behavior under transient flow, we have developed a new technique that permits the acquisition of frequency dependent dielectric behavior during large amplitude oscillatory shear (LAOS) experiments. We combine these measurements with in-situ small-angle neutron scattering to probe the transient dielectric response, rheology, and microstructure simultaneously. Using this approach, we can distinguish the non-linear elastic response of branched micelles from that of unbranched micelles.

SESSION C1.02: Conjugated Polymers
Session Chair: Megan Robertson
Monday Afternoon, June 6, 2022
UMC Conference Room 235

2:00 PM *C1.02.01
Analysis of the Structure and Dynamics of Conjugated Polymers via Combined Neutron Scattering and Molecular Simulations Caitlyn M. Wolf[1] and Lilo D. Pozzo[2,2]; [1]NIST, United States; [2]University of Washington, United States

Conjugated polymer films, nanofibers, and networks can be ideal materials for the design of efficient photovoltaic devices, batteries, thermoelectric cells, light emitting diodes and many emerging energy technologies. It is also recognized that the structure and dynamics of organic semiconductor materials correlates strongly with large changes in optical, electronic and mechanical properties so that their control and manipulation is essential to advancing the field. This presentation outlines the use of neutron scattering techniques in the development of structure-property relationships for conjugated polymer nanomaterials. It also highlights recent results on the use of neutron and x-ray scattering techniques for improvements in molecular simulation force fields specifically produced for conjugated polymers. Quasi-elastic neutron scattering (QENS) experiments are used along with computationally efficient MD simulations to understand the nature of important nanoscale motions. X-ray and polarized neutron diffraction are also used to correlate experimental and model-generated polymer structures. QENS validation of MD force fields presents a unique opportunity to increase the accuracy of highly uncertain parameters used in simulation of conjugated polymers and other complex macromolecules. These parameters are currently estimated from quantum mechanical calculations such as density functional theory but, unlike many force fields for small molecules, they are not parameterized to available experimental data. Moreover, high variability is observed in parameters for the small number of force fields that have been proposed in the literature.

2:30 PM C1.02.02
Unraveling the Side Chain Effects on Solution Structure of Donor-Accepter Conjugated Polymers Zhiqiang Cao[1], Zhaofan Li[2], Miao Xiong[3], Guorong Ma[1], Luke Galuska[1], Song Zhang[1], Michael Ocheje[4], Gage Mason[4], Changwoo Do[5], Kunlun Hong[5], Ting Lei[3], Simon Rondeau-Gagné[4], Wenjie Xia[2] and Xiaodan Gu[1]; [1]University of Southern Mississippi, United States; [2]North Dakota State University, United States; [3]Peking University, China; [4]University of Windsor, Canada; [5]Oak Ridge National Laboratory, United States

The less studied chain conformation of donor-accepter conjugated polymers (D-A CPs) hinders the fundamental understanding of their optical and electronic properties. Here, we systematically studied the effect of side-chain length and branch point on the chain conformation of diketopyrrolopyrrole (DPP) and isoindigo (IID) based polymers. Using small-angle neutron scattering (SANS) measurements, we find that chain rigidity of DPP-based polymers increases with side-chain length, which directly correlates with increased charge carrier mobility until the insulating alkyl sidechains are bulky enough to hinder efficient intermolecular charge carrier transport. The chain rigidity of IID-based polymers increases when moving the branch position farther away to the backbone. The trend of the solution optical bandgap changes is in accordance with an odd-even effect of the branching point. Using atomistic molecular dynamics (MD), we examined how side-chain structure can be modified to control chain conformation which further validated the experimental results. This work fills the fundamental gaps in our knowledge of the most basic relationships between polymer structure and chain conformation.

2:45 PM C1.02.03
Effect of Polystyrene Additives and Solvent Quality on the Conformation and Self-Assembly of Conjugated Polymers in Mixed Solutions Sage Scheiwiller, Lilo D. Pozzo and Lorenzo Guio; University of Washington, United States

Blends of conjugated and commodity polymers provide a way to balance desirable electronic properties and physical properties of both components. This balance plays a critical role in the development and performance of organic electronic devices such as photovoltaics (OPVs), field-effect transistors (OFETs), bioelectronics, and wearable sensors. Understanding the fundamental interactions between the conjugated polymers and matrix polymers at all stages of processing is essential for designing and optimizing organic electronic devices. Our previous research on solid film blends of polystyrene and conjugated polymers has shown strong relationships between morphology and the resulting electronic properties. This work also shows differences in the self-assembly of conjugated polymers, with nanofibers emerging at low concentrations in compositionally-identical solid films cast from different solvents. In this work we determine the impact of the solvent identity and the addition of a co-dissolved matrix polymer (polystyrene) on the solution behavior of the conjugated polymers. We outline the effect that these interactions have on the morphology and on properties of the polymer blends.

Solution-state SANS with contrast matching is used to investigate the effects of matrix polymers on phase separation and assembly of the conjugated polymer. By selectively deuterating the matrix polymer and the solvent, we are able to isolate the assembly and morphology of the conjugated polymer while co-dissolved in solution. This deuteration allows us to observe changes to the conjugated polymer due to influences of the polystyrene, over a range of molecular weights, as well as the influence of the solvent quality. We observe dense P3HT nanofiber networks forming in the presence of polystyrene in solution phases, as well as variations in the extent of conjugated polymer assembly for various molecular weights of polystyrene. We also probe the effect of solvent quality on the interactions between the polystyrene and the conjugated polymer through the use of various solvents covering the range of “good” to “poor” solvents. The observed variation in conjugated polymer assembly is further supported by SANS measurements on solid-state samples cast to form identical blend compositions. With this work, we aim to expand our understanding of solution state interactions and behavior of conjugated polymer blends in order to refine the structure of solid state composite films.

3:00 PM C1.02.04
WITHDRAWN (C1.02) Chain Growth Kinetics of Conjugated Polymers on Ferromagnetic Nanoparticles Investigated by SANS Lilin He; Oak Ridge National Laboratory, United States

Ferromagnetic nanoparticles have shown promising applications in water treatment due to their large surface area, high reactivity, stability and reusability. However, their dissolution in aqueous environment greatly diminishes the usability as photocatalysts. Coating of conductive polymers on the surface of photocatalysts can reduce the dissolution without compromising the material’s photocatalytic properties. In this work, polypyrrole (PPy) and polyaniline (PANI) were coated on the surface of two magnetically separable MoO_3/Fe_3O_4 nanocomposites with different surface chemistries. We used small-angle neutron scattering (SANS) technique to *in situ* monitor the polymerization kinetics by taking advantage of neutron’s sensitivity on light elements. The scattering results reveal that PPy polymerization initiated faster yet had a slower rate than PANI. This study shows that the polymerization of PPy was more affected by the base material properties than PANI's. The ultimate size of polymer chains grafted on nanoparticles largely depended on the monomer

concentration and the surface properties of the nanoparticles. These observations provide valuable insights on the polymerization mechanisms of two conjugated polymers on the surface of nanocomposites.

3:15 PM C1.02.05
Combining Inelastic Neutron Scattering and Molecular Dynamics Simulation to Probe Conjugated Polymer Dynamics Xiaodan Gu[1], Zhiqiang Cao[1], Wenjie Xia[2] and Amirhadi Alesadi[2]; [1]University of Southern Mississippi, United States; [2]North Dakota State University, United States

Organic semiconducting polymers were widely studied due to their unique optoelectronic and mechanical property. They are the key component in various functional electronic devices, such as organic photovoltaic devices, flexible displays, wearable sensors, neuromorphic computing, and more recently bioelectronics. Despite tremendous progress were made in improving the charge carrier mobility and optimizing energy bandgap, the conjugated polymer's physical property was not widely studied such as chain rigidly, molecular entanglement behavior, and glass transition phenomenon. However, they are important for device stability, which is one of the major hurdles for polymer-based organic devices. Polymer dynamics could play an important role in dictating overall chain mobility and morphology stability in the bulk heterojunction.

In my talk, I will provide an overview of our effort in studying the conjugated polymer's dynamics using a wide range of unique characterization tools, from thin-film to bulk. I will discuss the challenge associated with accurately measuring the glass transition temperature for rigid conjugated polymers. I will discuss our approach to address this challenge using thin-film calorimetry and ellipsometry tools, as well as using molecular dynamic simulation and cheminformatic to accurately predict the glass transition temperature. Lastly, I will discuss how dynamics could impact thin film morphology and device performance at different operation temperatures and should be carefully considered when designing new polymers and devices.

SESSION CP1.03: Poster Session: Soft Matter
Session Chairs: John Riley and Javen Weston
Monday Afternoon, June 6, 2022
5:30 PM - 7:30 PM
UMC Center Ballroom 210

CP1.03.01
Understanding the Role of Topology on Deformation and Scission of Polymers in Dilute Solutions Under Extreme Shear Flows Using *In Situ* Neutron Scattering Anukta Datta[1], Xiaoyan Wang[2], Patrick Underhill[2] and Matthew Helgeson[1]; [1]University of California, Santa Barbara, United States; [2]Rensselaer Polytechnic Institute, United States

Applications of high molecular weight long-chain dilute polymer solutions as rheological modifiers involve extreme deformation rates that cause the polymer to mechanically degrade, thereby compromising performance. Previous attempts to characterize molecular-level deformation of polymers leading to mechanical scission have remained largely *ex situ* measurements due to a dearth of *in situ* characterization capabilities. Here, we propose *in situ* neutron scattering in a novel capillary device that operates at probe extreme shear rates ($>10^6$ s^{-1}) to directly develop quantitative, molecular-level insight for how topology affects polymer deformation, and its influence on mechanical scission of polymer backbone. We develop a modeling framework for comparing the results of scattering experiments to parameter-matched Brownian dynamics simulations that resolve single-molecule deformations of polymers at high Weissenberg number flows. Specifically, the model supports the estimation of molecular descriptors of deformation for chains under non-Gaussian deformations, a feature that becomes important in high-shear rate flows and for polymers with complex topology. This presentation will cover the development and validation of the model, including both forward and inverse scattering calculations from molecular configurations, as well as our initial attempts to apply the framework to capillary rheo-SANS measurements on a series of linear polymers at high Weissenberg number where chains experience considerable non-Gaussian deformations. The results allow us to formulate and test nonlinear rheological models for dilute polymer solutions. We anticipate that these newly confirmed relations along with experimental observations will facilitate the rational design of future rheological modifies, prolonging both their performance and lifetime in a wide range of applications.

CP1.03.02
Analysis of Engineered Nafion Surfaces via Neutron Reflectometry Natalie L. Schwab[1,2], Yuanchao Li[3], Trung van Nguyen[3], Robert M. Briber[1] and Joseph A. Dura[2]; [1]University of Maryland, United States; [2]National Institute of Standards and Technology, United States; [3]University of Kansas, United States

The thin film structures of Nafion[b], a model perfluorinated ionomer, impact practical advances in proton-exchange membrane fuel cells (PEMFCs). Engineered Nafion surfaces have been developed by *Dowd et al.* to alter the surface composition and wettability of Nafion[a]. Using neutron reflectometry (NR), we probe the through-plane structure of engineered Nafion thin films with hydrophobic and hydrophilic surfaces. Buried layers of phase-separated lamellae were previously observed at the interfaces of Nafion thin films on Au, Pt, and SiO_2 substrates by *Dura et al.*[b] and *DeCaluwe et al*[c]. We have used titanium dioxide (TiO_2) films on Si to minimize the neutron scattering contrast to Nafion interface structures, thus optimizing the sensitivity of NR to the presence and structure of the engineered Nafion surface layer. The neat and surface-engineered Nafion thin films are studied via neutron reflectometry in dry (0% RH) versus humidified (92% RH) environments in H_2O and D_2O vapor. Fitted depth profiles of the engineered Nafion thin films are compared to the neat samples to independently determine differences in water uptake as well as surface and interface structure for the hydrophobic and hydrophilic Nafion films.

References:
[a]10.1149/2.1081702jes [b]10.1021/ma802823j [c]10.1039/C4SM00850B
This work was supported by NIST cooperative agreement 70NANB17H301.

CP1.03.03
Crystal, Liquid, or Gel: A Thermodynamic Framework for Phase Behavior in Dilute Protein Solutions with Increasing Salt Concentration Brian Paul[1], Susana Teixeira[2], Norman Wagner[1], Eric Furst[1] and Abraham Lenhoff[3]; [1]University of Delaware, United States; [2]National Institute of Standards and Technology, United States

Soft matter and complex fluid behavior are of great interest to numerous industrial efforts, notably in pharmaceutical formulations and food processing.

Biopolymers are often extremely sensitive to temperature and pH variations, and the physical nature of the dense phase is strongly dependent on the solution conditions[1,2]. Despite the wide usage of biopolymers in industry, relatively little is known about the behavior of dense phases – such as gels, aggregates and precipitates - at either ambient or high-pressure conditions, namely the microstructures and mechanisms that lead to their formation. Greene et al. showed for the first time that macroscopically amorphous salted-out protein dense phases contain nanocrystalline regimes[3]. The work presented here pairs a completed structural investigation of salted-out ovalbumin under ambient conditions with an explanation of the underlying thermodynamics. Small- and Ultra-Small-Angle Neutron Scattering (SANS/USANS), Small- and Ultra-Small-Angle X-ray Scattering (SAXS/USAXS), and Static Light Scattering (SLS) are utilized to provide a full structural profile and place observed phase behavior in context of ovalbumin monomers as sticky hard ellipsoids. Attention is given to trends in microstructure development with varying salt concentration and how structural observations may serve as a predictor for macroscopic properties, such as gel mechanical moduli. Future work will optimize these studies with a novel instrument for simultaneous Small-Angle Neutron Scattering and Light Scattering Microrheology under high pressure[4,5]. The current results provide insights into competition between crystallization, gelation, and phase separation within dilute protein solutions of increasing attraction and build the groundwork for an in-depth study of pressure effects on biopolymer structure and rheology.

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3. Greene, D. G. M., S.; Wagner, N.J.; Sandler, S.I.; Lenhoff, A.M. (2015). Biophysical Journal 109, 1716-1723.
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5. Teixeira, S.C.M. (2019). Current Opinion in Colloid & Interface Science 42, 99-109.

CP1.03.04
Understanding the Role of Block Sequence on the Solution Aggregation of Polypeptoid Multi-Block Copolymers Meng Zhang[1], Yun Liu[2] and Donghui Zhang[1]; [1]Louisiana State University, United States; [2]NIST Center for Neutron Research, United States

Ionic block copolymers (BCPs) have a wide range of technical applications (e.g., nanolithography, drug delivery carrier, membrane materials, mobility control agent, etc.) due to their unique solution properties and various morphologies. Ionic block copolymers comprised of a hydrophobic segment and an ionic or ionizable hydrophilic segment can self-assemble into core-shell type structure with varying geometry including spheres, cylinders, bicontinuous structures, lamellas, vesicles, etc. in selective solvent. The aggregate morphology is influenced by several factors (e.g., electrostatic repulsion, counterion condensation, osmotic swelling, etc.). Electrostatic interactions are well-known to play an important role in the structure and function many naturally occurring biomacromolecules (e.g., polysaccharides, intrinsically disordered proteins, etc.). Polypeptoid polymers are an emerging class of peptidomimetic polymers featuring N-substituted polyglycine backbone. Due to N-substitutions, polypeptoids have reduced propensity to form secondary structure commonly observed for polypeptides and lack extensive hydrogen bonding interactions. These attributes make polypeptoids an ideal model system to study the effects of the sequence-encoded electrostatic interactions on the aggregate structures. The objective of this work is to investigate and understand the role of sequence-encoded electrostatic interactions on the micelle formation and micellar structures of ionic polypeptoid multi-block copolymers with well-defined block sequences. Earlier studies showed that the understanding of whether and how charge pattern of amphiphilic BCPs encoded in the block sequence influence their equilibrium aggregate structure in solution are limited. Recently, we investigated doubly charged ionic polypeptoid BCPs with an ionizable monomer positioned at varying location along the chains (i.e., at the hydrophilic chain terminus or at the hydrophilic-and-hydrophobic segment junction) and another ionizable monomer fixed at the hydrophobic chain ends. TEM and SANS analysis of these BCPs revealed ellipsoidal micelles in water. The SANS profiles can be best-fitted with core-shell ellipsoidal model and the radius of gyration and aggregation number of the micelles can be obtained from Guinier analysis at the Guinier region. The ionizable monomers position along the BCPs showed a significant effect on the micellar structures (i.e., core-shell dimension, hydration level).

CP1.03.06
Hofmeister Effect on Dynamics of Confined Water in Metal Ions Intercalated Graphene Oxide Gobin R. Acharya[1], Madhusudan Tyagi[2], Eugene Mamontov[3] and Peter M. Hoffmann[1]; [1]Wayne State University, United States; [2]NIST Center for Neutron Research, United States; [3]Oak Ridge National Laboratory, United States

When water is confined on a nanometer scale, it exhibits several unusual behaviors which include ordering, viscoelasticity, and long relaxation times. The presence of ions further changes the properties of water because ions influence ordering and interactions. Different ions, according to the Hofmeister series, have different effects on the ordering and dynamics of confined water. The combination of Quasi Elastic Neutron Scattering and Atomic Force Microscopy allows us to measure the dynamics and viscoelasticity of water in GO membranes at different time scales for direct comparison of dynamics and viscosity while matching size, composition, and temperature conditions. In the talk, we would like to present how the presence of three different cations (structure former, Na+, structure “destroyer”, Cs+ and an “intermediate” ion, K+) will affect the interlayer and intra-layer diffusion and viscosity of water in GO.

SESSION C2.04: Grafted Polymers and Nanocomposites
Session Chair: John Riley
Tuesday Morning, June 7, 2022
UMC Conference Room 235

10:15 AM *C2.04.01
Molecular Bottlebrushes: Scattering Measurements and Simulations Michael J. Hore and Raj Mukkamala; Case Western Reserve University, United States

Molecular bottlebrushes (BBs) are anisotropic macromolecules that consist of a central backbone which is densely grafted with sidechains. Recent developments in polymer chemistry have enabled the synthesis of BBs with a variety of chemical structures. BBs express a number of unique physical properties that have been harnessed for a several types of applications, such as for lithography, low modulus gels, and biomimicry. In this talk, I will highlight several small-angle neutron scattering (SANS) studies of BBs in solution which have provided insight into their conformation and how that influences the resulting physical properties of materials in which they are used. To interpret previous SANS measurements from the literature, we performed dissipative particle dynamics (DPD) simulations that investigate the role of backbone-solvent interactions on the BB conformation. In addition, we explore how these parameters influence the relaxation dynamics of BBs using proper orthogonal decomposition (POD). The simulation results are compared with experiments and simulations of both linear and star-branched polymers, and indicate that the BB structure and dynamics are governed

strongly by both the strength of the backbone-solvent interaction, as well as the spacing of sidechains along the backbone.

10:45 AM C2.04.02
Vanadium Ion Dynamics of Ionomer Nanocomposites Xueting Wang[1], Apoorva Balwani[1], Mayura S. Silva[1], Madhusudan Tyagi[2], Stephen Creager[1] and Eric M. Davis[1]; [1]Clemson University, United States; [2]National Institute of Standards and Technology (NIST) Center for Neutron Research (NCNR), United States

Ionomer nanocomposites have gained popularity as proton exchange membranes (PEMs) for redox flow batteries due to their chemical resistance and improved proton selectivity. To better understand the underlying mechanism of this improved ion selectivity, a myriad of research has been conducted, where it has been posited that the nanoparticles (NPs) reside within hydrophilic domains of the ionomer, physically blocking the crossover of bulky, hydrated vanadium ions. However, recent work suggests this reduction in crossover may be caused by the interaction between the NPs and the sulfonic acids groups in ionic channels, leading to inaccessibility of the sulfonic acid groups needed to facilitate vanadium ion transport. Herein, the dynamics of hydrated vanadium ions in Nafion nanocomposites were investigated via quasi-elastic neutron scattering. Specifically, high flux backscattering spectroscopy was utilized to measure vanadium ion dynamics in Nafion nanocomposite membranes prepared by both in situ sol-gel condensation and solution-casting processes. For both fabrication routes, the NP loading in these membranes was varied from 0 mass % to 10 mass %. The broadening of the energy spectra was attributed to the incoherent scattering from hydrogen atoms – both protons and water molecules in the vanadyl ion hydration sphere. The half width at half max (HWHM), as well as the elastic incoherent structure factors were extracted from analysis of the data using a jump diffusion within a sphere model. A delta function and a single Lorentzian function were applied in the fitting to extract information for both jump diffusion and local motion, providing direct insight into the vanadium ion dynamics in these ionomer nanocomposites. The radius of the sphere within which the dynamics occur was observed to increase with increasing temperature, while the residence time between jumps was seen to decrease. Additionally, the fraction of immobile hydrogen atoms was seen to decrease with increasing temperature. Notably, the loading of NPs had a significant effect on the fraction of immobile hydrogen atoms for solution-cast samples, where this fraction was observed to increase with increasing NP loading at all temperatures. Interestingly, the fraction of immobile hydrogen atoms for the sol-gel samples was not affected by the NP loading. This same correlation between NP loading and the residence time, as well as diffusion coefficient for both jump and local motion was observed. That is, the incorporation of NP was seen to increase the residence times and decrease the diffusion coefficients, though this effect was primarily observed for solution-cast membranes. To better understand the connection between water/vanadium dynamics and proton conductivity, the vanadium ion permeability and proton conductivity of these membranes were also characterized.

11:00 AM C2.04.04
The Microscopic Structure and Dynamics of PEO-Silica Nanocomposite: Effect of Nanoparticle Size on Short-Time Polymer Dynamics Jihyuk Kim[1], Antonio Faraone[2], Orsolya Czakkel[3], Michael Ohl[4], Stephan Forster[4] and Norman Wagner[1]; [1]University of Delaware, United States; [2]National Institute of Standards and Technology, United States; [3]Institut Laue-Langevin, France; [4]Research Center Juelich GmbH, Germany

Polymer nanocomposites (PNC) have enhanced mechanical, optical, and electrical properties relative to the host polymer and are already widely applied and studied because of their tunability, responsiveness, and functionality. However, an understanding of the structure-dynamics-property relationship of these materials, which is necessary for their rational formulation and processing, remains incomplete. Here, a well-studied model system comprised of silica nanoparticles (SiO_2) dispersed in poly(ethylene oxide) (PEO) is investigated in the melt state as model attractive PNC, using SANS, NSE, X-ray scattering and rheology to connect the molecular level structure and dynamics to the macroscopic rheological behavior in the melt. New results with nominal 10 nm nanoparticles are compared with previous literature reports on homologous systems with silica nanoparticles of larger size [1]. Small angle scattering techniques provide information on the dispersion of the nanoparticles as well as the conformation of the polymer chains. Neutron spin echo measurements of the single chain dynamics is achieved through contrast matching to determine the effect of nanoparticles on the Rouse dynamics and reptation tube diameter. Data are interpreted in terms of standard theories developed for bulk polymers providing a mean to connect the microscopic dynamics to the observed rheological behavior. The results point to the surface area to volume ratio as a key parameter determining the chain dynamics, providing microscopic insights on the effects of nanoparticle loading on the nanocomposite's viscoelastic properties and how the addition of nanoparticles induces mechanical reinforcement in well-dispersed PNCs.

[1] Senses, E. et al, *Macromolecules* **2020,** *53* (12), 4982-4989.

11:15 AM C2.04.05
The Impact of Graft Type on the Assembly of Nanoscale Organic Hybrid Materials in Solution using Small Angle Neutron Scattering Md Ashraful Haque[1], Tony G. Feric[2], Sara T. Hamilton[2], Ah-Hyung Park[2] and Mark Dadmun[1,3]; [1]University of Tennessee, Knoxville, United States; [2]Columbia University, United States; [3]Oak Ridge National Laboratory, United States

Functional polymer nanocomposites such as Nanoscale Organic Hybrid Materials (NOHMs) have recently gained attention due to their enhanced CO_2 capture and energy conversion capabilities. In this work, small-angle neutron scattering (SANS) studies investigate the assembly and structure of ionic and covalent NOHMs in aqueous solution (D_2O) with and without supporting electrolyte (0.1M $KHCO_3$). The synthesized ionic and covalent NOHMs consist of HPE polymer grafted to ~9nm silica nanoparticles. A combined model of core-shell sphere and polymer excluded volume is required to analyze the SANS patterns, providing insight into the assembly of ionic NOHMs (N-I-HPE) in solution. The combined model captures the presence of free polymer in the ionic NOHMs solutions that interacts with the grafted polymer layer and thereby alters the grafted layer structure. Moreover, the addition of supporting electrolyte in the solution indicates a decrease in the radius of gyration of the free polymer suggesting a decrease in the quality of the solvent. Careful analysis of the scattering profiles of the covalent NOHM (N-C-HPE) solutions indicates the existence of aggregated NOHM structures that co-exist with dispersed NOHMs in the solution. The clusters attain a size range of few hundred angstroms. Further investigation reveals that the addition of salt alters the conformation of grafted layer to a lesser extent than in the ionic NOHMs.

11:30 AM C2.04.06
Study of Interdiffusion of Bilayer Polymer Grafted Nanoparticle Films at Interfaces by Neutron Reflectivity Wenjie Wu[1], Kshitij Sharma[1], John F. Ankner[2], Jack Douglas[3] and Alamgir Karim[1]; [1]University of Houston, United States; [2]Oak Ridge National Laboratory, United States; [3]National Institute of Standards and Technology, United States

Polymer-grafted nanoparticles (PGNPs) have attracted a lot of attention in both academia and industries due to their extensive applications in areas ranging from electronics to medical diagnostics. The hybrid structure of the grafted polymer layer and the solid nanoparticle core in PGNPs offers additional complexity to the system. This work focuses on understanding the interdiffusion of bilayer PGNP films at interfaces. Particularly, we study the interdiffusion of bilayer dPMMA–SiO_2 (grafted deuterated poly(methyl methacrylate) on silica nanoparticles) on hPMMA–SiO_2 (protonated poly(methyl methacrylate)) films *via* neutron reflectivity (NR). NR gives us an excellent approach to analyze the dynamics at flat interfaces in thin films upon processing. This instrument is particularly suitable for the PGNP system as the great difference in scattering length density between hydrogen and

deuterium provides unique opportunities to tailor the system to selectively interrogate distinct structural parameters. The bilayer films were annealed using thermal annealing (TA). We investigated the interdiffusion as a function of deuteration rate and the inner core size. For the highly deuterated system, the enthalpic difference between the deuterated and protonated polymers ($\chi_{dh} > 0$) impeds the interdiffusion between the bilayer films. Furthermore, the interface width between the 3 nm core (in diameter) dPMMA-SiO_2 and hPMMA–SiO_2 particles increases from 3 nm to over 30 nm within 10 min, indicating a relatively fast interdiffusion. However, when the inner core size increases to 15 nm (in diameter), the interface width didn't evolve even with 24 hours of annealing at 100% higher temperature above T_g. This indicates that the surface energy variance due to the curvature of the inner particle core plays an important role in the interdiffusion of those bilayer films.

11:45 AM C2.04.07
A New Computational Method (CREASE) to Analyze and Interpret Small Angle Scattering Profiles from Assembled Structure in Polymer Solutions Zijie Wu and Arthi Jayaraman; University of Delaware, United States

In this talk we present a new method - *Computational Reverse Engineering of Scattering Experiments* (CREASE) that we have developed to analyze small angle scattering profiles and interpret assembled structure in macromolecular solutions. There are two steps within CREASE: the first step involves a genetic algorithm (GA) to determine the shape and dimensions of the domains in the assembled structure and the second step uses molecular simulations to reconstruct chain conformations and monomer level arrangements within the assembled structure. We validate the GA step within CREASE by taking input scattering intensity profiles from a variety of assembled shapes with known shapes and dimensions, and by producing outputs that match those known shapes and target dimensions. CREASE's power lies in its ability to interpret structural detail at a range of length scales for macromolecular solutions without relying on fitting with off-the-shelf analytical models that may be too approximate for novel polymers and/or unconventional assembled structures. In this talk we will show CREASE applied to analyze two structures of interest to researchers working with soft materials - vesicles and fibrillar networks.

12:00 PM C2.04.08
SESANS and SANS Studies to Understand the Presence of an Initial Opaque Phase in the Self-Assembly of Di-Block Copolymers Antonia Denkova; Delft University of Technology, Netherlands

Polymer micelles and vesicles (polymersomes) composed of block copolymers are often applied in drug delivery. Micelles are usually used to encapsulate hydrophobic substances while polymersomes can the also enclose hydrophilic compounds [1]. We have shown that both types of self-assemblies can be easily radiolabeled with imaging radioisotopes [2,3] allowing the combination of therapy and imaging. The preparation of self-assemblies differs depending on the building blocks but are often time consuming especially when involving block copolymers such as polybutadiene-polyethylene oxide (PB-PEO) which have slow self-assembly kinetics. To speed up the preparation process we have developed an inverse nanoprecipitation method to rapidly obtain polymersomes and micelles in a 'green' solvent, acetone. The block copolymers are initially dissolved in acetone, to which water is added dropwise. Upon the addition of the aqueous solution, an intermediate state appears in which the solution is opaque (5-10 vol% water), subsequently, it turns transparent again (>10% water), to finally be a whitish hue when the self-assemblies are formed (50 vol% water). To the best of our knowledge, we are the first to report the formation of the initial opaque phase. Understanding the mechanism governing the self-assembly of these self-assemblies at the various stagesis is essential to optimize the self-assembly morphology.

To understand the opaque phase, we have applied Spin Echo Small Angle Neutron Scattering (SESANS) which can be used to study structures between 50 nm to 20 microeter. The obtained scattering data has been fitted with a model corresponding to sphere resulting in a droplet size of 10 micrometers in diameter, suggesting the formation of large 'water and polymer' rich droplets in acetone. Subsequently, we have performed small angle neutron scattering (SANS) studies attempting to probe the internal structure of the large droplets. The SANS data reveals that in the absence of water, in the pure D-acetone phase, the block copolymer is dispersed in the solution as a unimer. Upon the addition of 6 vol% D_2O a distinct peak appears corresponding to 4.5 nm. By performing a contrast series we could clearly show that we are probing the internal structure of the droplets. Moreover, the data suggests that the droplets have small sub-domains rich on water and polymer. In addition, when heating the solution up to 40 °C the peak at 4.5 nm disappears, resulting in a scattering pattern associated with the unimers but it reappears with higher intensity once the sample is cooled down to room temperature.

Finally, when adding 50 vol% of water the SANS data changes again and can be fitted with a polymeric micelle model in accordance with Cryo-TEM results.

References:

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[3] A. C. Laan et al. EJNMMI research 6 (1):12, 2016.

SESSION C2.05: Surfactants and Emulsions
Session Chair: John Riley
Tuesday Afternoon, June 7, 2022
UMC Conference Room 235

1:45 PM *C2.05.01
Using Neutrons to Probe the Structure of PFAS Surfactant Micelles Marina Tsianou; University at Buffalo, The State University of New York, United States

Surface active per- and polyfluoroalkyl substances (PFAS) find niche applications because of their high chemical and thermal stability, their incompatibility with both water and hydrocarbons, and their unique ability to render surfaces non-stick. However, several widely used PFAS surfactants have been found extremely resistant to degradation, accumulate in the environment, and have long half-lives in humans, consequently causing great concern.
In the context of developing materials and processes for sequestering PFAS surfactants from aqueous media, we research how such surfactants associate

with (bind to) other molecules or particles/surfaces. To this end, we utilize complementary experiments (small-angle neutron scattering, SANS, with contrast variation) and modeling (molecular dynamics, MD), and present here examples on how the structure of micelles formed in water by the notorious PFAS surfactant perfluorooctanoate (PFOA) responds to the presence of various additives (salt [10.1016/j.colsurfa.2021.127313], urea [10.1021/acs.langmuir.1c00433], ethanol [10.1039/d1cp00049g]) and polymers (homopolymer poly(ethylene oxide) (PEO) [10.1021/jp5023168], PEO-based amphiphilic block copolymers [10.1016/j.jcis.2021.10.176]) across a wide range of compositions. A detailed picture emerges on how the additives or polymer segments distribute at the outer surface of the micelles and in their interior, which is used to rationalize the macroscopic behavior and various properties of the mixtures.
Fundamental knowledge of PFAS surfactant–polymer interactions supports the design of new materials to selectively capture and remove such surfactants from aqueous media. Self-assembly into micelles is a key feature of surfactants in aqueous solution and reveals how PFAS surfactants interact with themselves and with solvent (water) and other molecules present in solution. The capability established in our study to predict from first principles micelle formation and structure confirms that such multiple and often competing interactions have been properly accounted for. Micelles are relevant to environment and health in that PFAS surfactants, while typically found in very low bulk solution concentrations, they tend to concentrate in the vicinity of surfaces in the context of separations (activated carbon, ion exchange resins) and in the context of biointerfaces (proteins, lipid membranes).

2:15 PM C2.05.02
Controllable Nanostructures via a Bicellar Template – Characterized by Contrast-Variation SANS Chung-Hao Liu and Mu-Ping Nieh; University of Connecticut, United States

We report a facile shape-controllable polymerization utilizing lipid self-assembly, known as bicelles composed of 1,2-dipalmitoyl-sn-glycero-3-phosphocholine, 1,2-dihexanoyl-sn-glycero-3-phosphocholine and 1,2-dipalmitoyl-sn-glycero-3-phospho-(1'-rac-glycerol). This generalized template is capable of encapsulating hydrophobic monomers (e.g., styrene), producing size-controllable nano-rings and nano-meshes through tuning the monomer-to-lipid molar ratios. Contrast variation by very small angle neutron scattering (vSANS) plays a crucial role in structural identification. This platform allows for the manufacturing of functionalized polymers with a high interfacial area under well control in one-pot synthesis, providing excellent opportunities for protein separation, catalyst design, or nanomedicines.

2:30 PM C2.05.03
WITHDRAWN (C2.05) Chemical and Physical Control on the Nanostructures of Ionic Amphiphilic Oligomer Assemblies: Elucidated by Spectroscopy and Neutron Reflectivity Zening Liu, Hanyu Wang, Tianyu Li, Lu Lin, John Katsaras, Kunlun Hong, Jim Browning, Benjamin Doughty and Charles P. Collier; Oak Ridge National Laboratory, United States

Advances in building two-terminal memory elements from soft materials have resulted in new opportunities for the development of artificial neural networks. Assemblies of ionic amphiphilic oligomers (oligodimethylsiloxane-methylimidazolium cation, ODMS-MIM$^+$) (e.g., ODMS-MIM$^+$ droplet interface bilayers) showed short-term synaptic behaviors analogous to those consisting of phospholipid molecules. The advantages of using OMDS-MIM$^+$ oligomers rather than lipids in designing soft-matter memristive and memcapacitive devices include not only improved reliability and stability of the material, but also their salt-dependent and surface density-dependent electrochemical properties at surfaces and interfaces. According to our previous work, we have noticed these properties are tightly associated with the nanostructures of the ODMS-MIM$^+$ assemblies, which can be controlled through chemical and physical means. Here, we employed surface-active vibrational sum frequency generation (vSFG) spectroscopy to give insights into the orientation and (re)arrangement of the ODMS-MIM$^+$ assemblies formed at air-aqueous interface through the Langmuir trough technique under varied conditions. The Langmuir films were further transferred onto silicon wafers and characterized with neutron reflectometry (NR), which is a complementary characterization technique to vSFG spectroscopy, providing information about the thickness, roughness, and scattering length density of the ODMS-MIM$^+$ assemblies. These studies have shown that the nanostructures of charged, amphiphilic OMDS-MIM$^+$ assemblies are sensitive to both the overall ionic strength of the system and the surface density of the oligomers. This information will enable for the development of refined protocols for the construction of enhanced neuromorphic devices based on ionic, amphiphilic oligomers.

2:45 PM C2.05.04
Self-Association in Pluronic®-Cationic Surfactant Mixed System: A Scattering and Molecular Dynamics Approach Ketan C. Kuperkar[1], German Perez-Sanchez[2] and Pratap Bahadur[1]; [1]Veer Narmad South Gujarat University (VNSGU), Surat, India; [2]Campus Universitario de Santiago, Portugal

Nanoscale morphology and self-aggregation dynamics of the water-soluble non-ionic PEO–PPO–PEO [poly(ethylene oxide)–poly(-propylene oxide)–poly(ethylene oxide)]-*based* tri-block copolymer i.e., $EO_3PO_{43}EO_3$ (L81) in the presence of cationic surfactants with diverse amphiphilic characteristics [dodecyl trimethylammonium bromide (DTAB), didodecyldimethylammonium bromide (DDAB), andethanediyl-1,2-bis(dimethyldodecylammonium bromide) (12-2-12)]in aqueous solution are investigated using phase behaviour, and scattering approach. Cationic surfactants enhanced the aqueous solubility and exhibited *double* cloud point (CP) of copolymers in water. Progressive addition of cationic surfactants triggered the transformation of unstable vesicles into spherical mixed micelles. The small-angle neutron scattering (SANS) study probed the mixed-aggregate geometrical aspects of the Pluronics®-cationic surfactant assemblies from vesicles to spherical micelles. The SANS results are complemented by molecular dynamics (MD) simulations, using a simple and transferable coarse-grained model based on the MARTINI force field, to study the impact of cationic surfactants in Pluronics® aggregation in aqueous solutions. These complementary techniques yield further understanding of the nanoscale interactions directing the aggregation in these mixtures in terms of the alkyl chain length and the head group nature of the cationic surfactants.

3:00 PM C2.05.05
Variation of Bicontinuous Microemulsion Surface Structures on Hydrophilic and Amphiphilic Substrates Luke Heroux[1,2], Adam Imel[2], Brian Barth[2], Thomas Zawodzinski[2] and Mark Dadmun[2,1]; [1]Oak Ridge National Laboratory, United States; [2]The University of Tennessee, Knoxville, United States

The need for a large-scale renewable energy supply has prompted research into developing novel electrolytes in redox active batteries. Microemulsions (MEs) have recently been shown to be active in electrochemical applications which means they may play an important role in the development of such large-scale batteries. While understanding the structure of the bulk electrolyte is central to developing redox flow batteries, understanding the structure of electrolytes at a surface is also important. This is because the charge transfer processes of the system are affected by the surface structures formed at the electrode. In MEs, this includes both the electron transfer (at the electrode) and the ion transfer (across the surfactant). The near surface structure of microemulsions that consist of deuterated water (D_2O)/polysorbate-20/toluene with varying composition have been determined using neutron reflectivity on hydrophilic and amphiphilic surfaces. These MEs form mixed lamellar-type layers near a hard electrode surface, where the layers increase in number and purity with decreasing D_2O content. These mixed layers appear to be consistent with perforated lamellae and become purer with oil and water on amphiphilic surfaces. These changes in layer structure, thickness and purity impact the accessibility of the redox active species and availability of the charge transfer to satisfy the electron neutrality conditions.

3:15 PM C2.05.06
Measuring Co-surfactant Demixing Across Internal Nanodroplet Interfaces by SANS Tanvi Sheth, Nairiti Sinha and Matthew Helgeson; University of California, Santa Barbara, United States

The production of multiple (i.e., multi-phase) emulsion droplets has provided a versatile route for designing novel complex particle technologies and formulated chemical products. Generating *nano*scale multiple emulsions, however, has been limited by the capabilities of existing emulsification methods and characterization tools. We previously showed that co-surfactants of opposing spontaneous curvature produce ultra-low interfacial tensions that can be utilized to stabilize internal nanodroplet interfaces. Importantly, equilibrium free energy predictions across droplet interfaces for these systems suggest that internal interface formation coincides with co-surfactant demixing to promote multiple nanoemulsion formation. Crucially, SANS provides a powerful means of testing the thermodynamic model predictions of preferred nanodroplet morphology, as well as the potential for interfacially-driven demixing. Here, we utilize contrast variation SANS to probe the compositional differences between co-surfactant films across oil-water interfaces in a complex nanodroplet. To do so, we synthesize and employ selectively deuterated surfactants to quantify the surfactant composition at each interface from modeling of the droplet SLD profile. This enables us to critically test and further develop a new interfacial thermodynamic model that incorporates curvature-induced co-surfactant demixing to predict conditions for preferred multiple nanoemulsion formation.

SESSION C3.06: Bio-Inspired Soft Matter
Session Chair: Javen Weston
Wednesday Afternoon, June 8, 2022
UMC Conference Room 235

2:00 PM *C3.06.01
Understanding and Controlling the Solution Self-Assembly of Amphiphilic Polypeptoid Block Copolymers Donghui Zhang; Louisiana State University, United States

Polypeptoids, a class of biomimetic polymers featuring *N*-substituted polyglycine backbone, exhibit enhanced proteolytic stability, solution and thermal processability, and reduced propensity for secondary structure relative to polypeptides due to *N*-substitution. Polypeptoids have strong propensity to form ordered molecular packing due to the strong dipolar interaction among the backbones and structural coupling between the side chain and main chain. Polypeptoids upon crystallization adopt a universal cubic lattice where the domain spacing is determined by the *N*-substituent structure in a highly predictable manner. The combination of these attributes makes polypeptoids attractive building blocks for hierarchical assemblies. We have synthesized a series of amphiphilic polypeptoid block copolymers (BCPs) by controlled ring-opening polymerization method and investigated their solution aggregation by a combination of microscopic and scattering methods. We have found that the molecular shape of the solvophobic polypeptoids bearing *N*-alkyl sidechains in the solution micelles can be modulated by controlling the sidechain length and branching pattern. The crystalline and liquid crystalline packing of the solvophobic polypeptoid segments can drive the assembly of amphiphilic polypeptoid BCPs to form one, two and three-dimensional micelles with structural hierarchy and controlled sizes in solution. Furthermore, seeded epitaxial crystallization of the polypeptoid BCPs in solution can be made to occur in a controlled fashion, producing one-dimensional fibrillar micelles with well-defined size and narrow size distribution. The role of various *N*-substituent structure, polymer composition and block sequence in the formation of the solution micelles of varying geometry will be discussed.

2:30 PM C3.06.02
Phase Morphology of Amorphous Solid Dispersions using Small-Angle Neutron Scattering and Neutron Interferometry Caitlyn M. Wolf[1], Youngju Kim[2,1], Sarah M. Robinson[1], Michael Cyrus Daugherty[2,1], Ryan P. Murphy[1], Nikolai N. Klimov[1], Michael G. Huber[1], Peter N. Bajcsy[1], David Jacobson[1], Jacob M. LaManna[1], Paul A. Kienzle[1], Daniel S. Hussey[1] and Katie M. Weigandt[1]; [1]National Institute of Standards and Technology, United States; [2]University of Maryland, United States

A challenging property of most drug candidates is low water solubility, which limits their dissolution and uptake in vivo. One approach to improving their bioavailability is to administer them as an amorphous solid dispersion (ASD). ASDs are comprised of a polymer matrix that kinetically traps amorphous domains of the active pharmaceutical ingredient (API) to increase solubility, and therefore, efficacy in the body. These formulations can exist as tablets, pills, or implants, and production methods range from hot melt extrusion, spray drying, and milling to 3D printing. The spatial and size heterogeneity of API phase in ASDs directly impact dissolution behavior, and so understanding the influence of composition, molecular interactions, and production method on the final phase morphology is important for the development of effective formulations. In this work, small-angle neutron scattering (SANS) is used to characterize the bulk phase morphology in a model set of ASDs with a substitute “active” small molecule (palmitic acid) embedded in a biodegradable matrix (PLA, PLGA). Unfortunately, we are still limited in our understanding of the heterogeneity in these materials using this sample-averaged technique. However, a new type of far field interferometer (INFER) currently under development at the NIST Center for Neutron Research will enable us to probe spatially resolved structural information on the same length scales as SANS/USANS. This method could help us understand how spatial and phase heterogeneity in a range of ASDs influences the dissolution profile. In this talk, we will also discuss our first preliminary measurements collected with INFER on these ASD materials, as well as how this new technique could more broadly benefit polymer matrix blends and composites across a range of fields, such as aerospace, construction, medicine, and more.

2:45 PM C3.06.03
Microstructures of Starch Granules as Revealed by Scattering Techniques Yimin Mao[1,2] and Yong-Cheng Shi[3]; [1]National Institute of Standards and Technology, United States; [2]University of Maryland, United States; [3]Kansas State University, United States

Starch is a natural homoglucan biopolymer linked by α-1, 4 and α-1, 6 linkages. To better understand microstructures of starch granules, as-is (~ 10% moisture) and hydrated (50 % water by mass) granules of waxy potato (WP), waxy wheat (WW), waxy maize (WM), normal maize (NM), and high amylose maize (HAM) starches were investigated by using small-angle neutron and X-ray scattering (SANS and SAXS), wide-angle X-ray scattering (WAXS), and ultra-small-angle neutron scattering (USANS). SANS and SAXS data were fitted using the two-phase stacking model of alternating crystalline and amorphous layers. The partially crystalline lamellar structures inside the growth rings of granules were analyzed based on the inter-lamellar distances, thicknesses of the crystalline lamellae and amorphous layers, thickness polydispersities, and water content in each type of layer. The low-angle intensity upturn in SANS and SAXS was attributed to scattering from interfaces/surfaces of larger structures, such as growth rings and macroscopic granule surfaces. Data analysis based on model fitting and 1D correlation function (CF) were compared. It was emphasized that owing to inherent packing disorder inside starch granules, a comprehensive analysis of different parameters was essential in correlating the microstructures with starch properties.

3:00 PM C3.06.04
Glucose Induced Self-Assembly and Phase Separation in Hydrophilic Triblock Copolymer Solution and its Governing Mechanism Divya K. Patel[1], Ketan C. Kuperkar[1] and Pratap Bahadur[2]; [1]Sardar Vallabhbhai National Institute of Technology (SVNIT), India; [2]Veer Narmad South Gujarat University (VNSGU), India

Symmetrical poly(ethylene oxide)-poly(propylene oxide)– poly(ethylene oxide), EO–PO– EO, triblock copolymers with 80 % hydrophilicity typically stay molecularly dissolved as Gaussian chain at ambient temperature even at fairly high-concentrations (5 %w/v). The micellization is incited at lower concentration/ temperature in the presence of glucose. The outcomes on glucose prompted micellization from four such hydrophilic copolymers Pluronics®: F38, F68, F88, F98 and F108 obtained from phase behaviour and scattering techniques are described. At temperature near ambient, these block copolymers form micelles that consist central core of PO blocks, surrounded by corona of highly hydrated EO chains. Phase transitions of Pluronics® with Glucose demonstrated the dehydration of copolymer coil; a decrease in the micellization temperature with increase in [glucose] was configured with fluorescence spectroscopy conduct. Cloud point (CP) results showed a marked decrease with increase in PO molecular weight and also in the presence of glucose while the scattering investigation offered an insight in to the micellar dimension as a function of temperature and glucose concentration. The viscosity (η_{rel}) changes caused by glucose is attributed to the enhanced dehydration of the POE block of the polymeric amphiphile. Glucose is more effective apparently due to its higher hydration number. Dynamic Light Scattering (DLS) and small-angle neutron scattering (SANS) investigation showed that the size of hydrophobic core increases as a part of dehydration of EO-PO blocks with expanding temperature or on adding varying concentration of glucose. It has been observed that the impact of added glucose on the phase behaviour of block copolymer is analogous to that of temperature. In addition, the core-shell blended micelles framed by these copolymers are successfully upgraded for drug (Curcumin, Cur) solubilization as per the peak intensity observed from UV-Visible spectroscopy. Different kinetic models were employed to fit the release profile data that enabled this study to act as an ideal platform for drug delivery.

3:15 PM C3.06.05
Diffusion Coefficients of Anisotropic Particles Measured by NSE: A Case Study Using Monoclonal Antibody Yanqin Zhai[1], Nicos Martys[2], William L. George[2], Joseph E. Curtis[2], Jannatun Nayem[3], Yang Zhang[1] and Yun Liu[2,3]; [1]University of Illinois at Urbana-Champaign, United States; [2]National Institute of Standards and Technology, United States; [3]University of Delaware, United States

Diffusion coefficients can be measured by different techniques, such as DLS, NMR, and NSE. Different from other techniques, NSE is able to measure the diffusion coefficients at very large scattering wave vector, Q, and at the short-time limit of many nanoparticles, such as proteins. For the anisotropic particles, this diffusion coefficient measured by NSE changes as a function of Q and includes the contribution from both the rotational, translational, and internal motions of a particle. Interestingly, to study the internal domain motions of flexible proteins, such as monoclonal antibody (mAb), we need to first accurately evaluate the complex Q-dependent diffusion coefficient when mAb can be considered a rigid body anisotropic particle. Using the dissipative particle dynamics (DPD) simulation, the Brownian motions of a rigid mAb are investigated. Both the intermediate scattering functions and the Q-dependent diffusion coefficient are calculated for a rigid mAb. The diffusion coefficient is calculated with two different theoretical models and compared to the simulation results. The uncertainty introduced by the finite computer simulation time is also evaluated that could be useful for future computer simulations to understand NSE signals.

3:30 PM C3.06.06
Understanding the Microscopic Mechanism Behind the Dielectric Relaxation in Water using Inelastic Neutron Scattering Yadu Krishnan Sarathchandran[1], Yuya Shinohara[2], Wojciech Dmowski[1], Eugene Mamontov[2], Daniel M. Pajerowski[2] and Takeshi Egami[1,2]; [1]University of Tennessee, Knoxville, United States; [2]Oak Ridge National Laboratory, United States

Water exhibits an anomalously high dielectric constant, ε = 78, at ambient temperature and pressure. This is characterized by a Debye peak in the dielectric loss spectrum with a maximum at ~20 GHz. This strong dielectric loss of water at high frequencies is utilized in our daily life when we use a microwave oven to heat food. It is believed that this Debye peak reflects some hydrogen bond mediated, collective dipolar dynamics in water. Despite decades of research, the microscopic mechanism behind this unique dielectric response remains unclear. The traditional approach of using spectroscopic techniques to measure dynamic properties cannot fully describe this due to their lack of information on spatial extension. In this work, we utilize a method to study the real-space correlated dynamics with a time-delayed atomic pair-correlation function, the Van Hove function, $G(r, t)$, from inelastic neutron scattering measurements. The dynamic structure factor of water, $S(Q, E)$, is measured using inelastic neutron scattering over a wide energy and momentum transfer range by making use of the recent advances in the instrumentation of neutron scattering facilities. The dynamic structure factor, $S(Q, E)$, is double Fourier transformed to obtain $G(r, t)$ to investigate the temporal evolution of atomic/molecular correlations in water in real-space up to 10 picoseconds. We find that $G(r, t)$ at 5-6 Å shows a relaxation very similar to the dielectric relaxation in magnitude and temperature dependence. The results show that the dielectric relaxation behavior in water originates from collective proton dynamics in the hydrogen bond network involving several molecules, such as a resonant hydrogen bond ring dynamics with highly quantum-mechanical nature.

SESSION C4.07: Nanoparticles, Methods, and General Soft Matter
Session Chair: Javen Weston
Thursday Morning, June 9, 2022
UMC Conference Room 235

10:15 AM *C4.07.01
Frustrated Coulombic and Cation Size Effects on Nanoscale Boehmite Aggregation: A Tumbler Small- and Ultra-Small-Angle Neutron Scattering Study Lawrence M. Anovitz[1], Patricia Huestis[2], Nikhil Rampal[1], Andrew G. stack[1], Jay A. LaVerne[2], Xin Zhang[3], Geregory K. Schenter[3], Jaehun Chun[3], Benjamin A. Legg[3], Lili Liu[3], Markus Bleuel[4], Cedric Gagnon[4] and David F. Mildner[4]; [1]Oak Ridge National Laboratory, United States; [2]University of Notre Dame, United States; [3]Physical Sciences Division, United States; [4]National Institute of Standards and Technology, United States

Millions of gallons of DOE's legacy high-level waste (HLW), generated during plutonium production for nuclear weapons, are stored in tanks at the Hanford (WA) and Savannah River (SC) sites. To better understand the effects of solution chemistry on particle aggregation in the complex solutions in these tanks, we have performed a series of tumbler small and ultra-small angle neutron scattering experiments on 20 weight percent solid slurries of nanoparticulate aluminum oxyhydroxide (boehmite) in various concentrations of M^{+1} nitrates: H, Li, Na, K and Rb nitrates at 10^{-5}, 10^{-3}, 10^{-1}, 2 and 4 molal (m) concentrations, as well as in pure H_2O. Synthetic boehmite nanoparticles were used with a size range from ~20 to 30 nm. Although particles initially form individual rhombohedral platelets, once placed in solution they quickly form well-bonded stacks, primary aggregates, up to ~1500 Å long, and a

second level of aggregates whose concentration and structure varies as a function of cation type and concentration. Aggregation generally increases with increased solute concentration and with cation radius up to a concentration somewhat above10^{-1}m, at which point the trend reverses. Primary aggregates become more rod-like and larger. The Kirkwood-like reversal probably reflects a change from DLVO/Debye behavior controlled by surface chemistry to a frustrated Coulombic system controlled by the solution structure. In addition the effects of M^{+1} cation are not consistently monotonic, with Na falling out of the sequence of the other cations with greater aggregation than expected. In addition, gamma irradiation of the boehmite completely changes the order of cation effects. These data suggest that an understanding of the effects of salt concentration and chemistry on nanoparticle aggregate structures provides useful physical insights into the microscopic origin of slurry rheology in the Hanford and Savannah River legacy wastes.

10:45 AM C4.07.02
Investigating the Oxidation of Atmospheric Aerosols using Neutron Reflectometry Rebecca Welbourn[1] and Martin King[2]; [1]ISIS Neutron & Muon Source, United Kingdom; [2]Royal Holloway University of London, United Kingdom

The Earth's climate is a highly complex, kinetically controlled balance of components and conditions within the atmosphere, that we know has been adversely affected by human activities. In order to monitor changes and predict future effects, intricate models are used, which rely on accurate parameters to describe these components and their lifetimes. One important component is aerosols. These affect our climate through both direct scattering and absorption of solar radiation, and indirectly through acting as cloud condensation nuclei, which in turn effect cloud formation and precipitation.
Much of this aerosol contains an insoluble mineral dust core and has an organic film coating, from either natural or anthropogenic sources. This organic film influences the climatic effect of the aerosol particle. However, the atmosphere is highly oxidising and these organic films will react and become unstable. By comparing the rate of this process to the lifetime of aerosols in the atmosphere their importance in climate modelling can be determined. Using a flat surface as a proxy for a droplet, we have used neutron reflectometry to measure the kinetic changes in these surface-active films in the presence of various atmospherically important oxidative species. We have used both model species for these films as well as real aerosol, collected from the atmosphere from a range of locations. The results of these experiments have been combined with complementary laser tweezers experiments in a 1-D radiative transfer model to provide information about their effect on the climate.

11:00 AM C4.07.03
How Much Crosslinking Causes a Polymer Chain to Become a Nanoparticle? Jacob Fischer[1], Lu Han[2], Tomonori Saito[2] and Mark Dadmun[1,2]; [1]The University of Tennessee, United States; [2]Oak Ridge National Laboratory, United States

Soft nanoparticles have received great interest lately due to their potential use in a range of applications, including drug delivery, all polymer nanocomposites, and self-healing materials. These soft nanoparticles are generally formed via extensive branching and/or internal crosslinking of macromolecules to form nanoscale particle-like structures. Moreover, their unique properties often require particle like behavior, yet the correlation of crosslink density to particle like behavior is poorly defined. Will a single crosslink in a long polymer chain form a 'nanoparticle'? Is there a limiting percent of crosslinking that is needed to transition a polymer chain to a soft nanoparticle?
From a broader perspective, the definition of what makes a construct a 'nanoparticle' is also not clearly defined. In this presentation, we will discuss our small angle neutron scattering studies that seek to clearly define the transition from a polymer chain to a nanoparticle vis internal crosslinking. A combination of analyses of the SANS curves, including the dimensionless Kratky plot, the fractal dimension, and the ratio of R_g/R_H, provides insight into the particle-like properties of the examined polymer constructs. These results show that the primary characteristic that delineates a polymeric chain from a nanoparticle is not its size, but the presence of a surface, which can be detected via these analyses. Using the idea that a nanostructure requires the presence of a surface to be a nanoparticle leads to the surprising conclusion that crosslinking densities as low as 0.8%, or 1 crosslink for every 125 monomers, is sufficient to realize very strong particle-like characteristics.

11:15 AM C4.07.04
Molecular Deformation and Relaxation Dynamics of Ionomers Revealed by Complementary Small-Angle Scattering Techniques Christopher N. Lam, Wei-Ren Chen and Yangyang Wang; Oak Ridge National Laboratory, United States

The molecular relaxation dynamics of rubidium neutralized sulfonated polystyrene ionomers after a large step uniaxial extension are investigated by small-angle neutron (SANS) and x-ray (SAXS) scattering techniques. Due to the different scattering contrast for neutrons and x-rays, the nonequilibrium structures of polymers and ionic clusters are revealed by SANS and SAXS, respectively. It is found that the single-chain structure becomes highly anisotropic upon deformation, whereas the ionic structures hardly show any distortions. This observation stands in stark contrast to the classical transient network picture, where the physical crosslinks formed by ionic clusters drive the molecular deformation of the polymer chains. Further analysis of the SAXS spectra indicates that small ionic clusters form fractal aggregates, which provide structural rigidity to resist the applied deformation. Moreover, quantitative analysis of the SANS spectra using the spherical harmonic expansion technique shows that the presence of molecular association fundamentally alters the spatiotemporal dependence of the structural anisotropy relaxation compared to the neat polymers, with much stronger wavenumber dependence for the associating chains.

11:30 AM C4.07.05
Rapid Automated Morphology Identification and Parameter Determination from Small Angle Scattering Data via Machine Learning Graham W. Roberts, Mu-Ping Nieh, Anson Ma and Qian Yang; University of Connecticut, United States

We have developed a machine learning framework for automatically predicting morphologies and their corresponding structural parameters from small angle scattering data. Interpretation of scattering data is a challenging problem due to the large number of variations in morphology, structural dimensions, polydispersity, scattering contrasts, and other factors that may affect the observed spectra. In our method, we first leverage the forward models in the small angle scattering software package, SasView, to curate a database of computed spectra covering different morphologies and particle sizes of interest. Then, we utilize this database to train classification models for predicting morphologies from spectra, and subsequently regression models for predicting structural parameters of these spectra given their morphologies. In particular, we first utilize a hierarchical decision tree to distinguish between morphologies that are subsets of each other. Then, regression models are used to approximate the structural parameters. These parameters can be further refined if desired by using them to initialize fitting algorithms such as those found in SasView. Spectra from six morphologies: sphere, cylinder, disk, core-shell sphere, core-shell cylinder, and core-shell disk, were successfully classified with more than 90% accuracy. Our hierarchical classification method achieves better performance in comparison to direct multi-class classification using classical algorithms, and exhibits good extrapolation performance. Once a morphology has been correctly determined, our regression models are able to predict structural parameters such as radius, length, and core-shell thickness with a mean absolute percentage error of less than 10%. Our models require several hours to train and optimize, after which they are able to make classification and regression predictions on new spectra approximately instantaneously. This is a significant speedup in comparison to domain experts who may require hours to days to reliably determine morphologies and fit corresponding structural parameters with a similar accuracy. Our model was also validated with experimental spectra. Our methodology not only has the potential to lead to more efficient use of expert and facility time, but also prevents waste of large amounts of experimental data that are often left unanalyzed due to time constraints and/or interpretation difficulty.

11:45 AM C4.07.06
Automated SANS/SAXS Exploration of Soft Materials with the Autonomous Formulation Laboratory Peter A. Beaucage and Tyler B. Martin; National Institute of Standards and Technology, United States

While neutron scattering is a workhorse technique for characterizing model soft material formulations, its application to industrial and/or complex systems has been more elusive, largely because the large number of components (10-100) often precludes rational mapping between component fractions, structure, and function. Multimodal characterization and machine learning (ML) tools promise to greatly reduce the difficulty of exploring such highly multicomponent systems. Here we describe the development of the Autonomous Formulation Laboratory, a highly adaptable platform capable of autonomously synthesizing and characterizing liquid mixtures with varying composition using x-ray and neutron scattering in addition to a suite of secondary measurements such as optical imaging, UV-vis-NIR and capillary viscometry. We will demonstrate the application of the platform to systems ranging from model block copolymer formulations to industrial systems from personal care, biopharmaceutical, and alternative energy partner companies. Future directions in algorithms and instrumentation to study the far-from-equilibrium self-assembly processes that underlie many real products will also be discussed.

12:00 PM C4.07.07
Measurement of Time-Resolved Adsorption Profiles in PMMA-Methanol System with Neutron Imaging Martin Wissink[1], Michael Kass[1], Charles E. Finney[1], Jacob M. LaManna[2], David Jacobson[2] and Hassina Z. Bilheux[1]; [1]Oak Ridge National Laboratory, United States; [2]National Institute of Standards and Technology, United States

Adsorption of solvents into polymers is an important aspect in many polymer applications, but the transport mechanisms underlying various diffusion regimes are still a subject of debate. A key element to understanding the transport kinetics is the ability to quantitatively measure the evolution of solvent concentration profiles over time. However, accurate measurements have proven to be notoriously challenging in certain systems such as the classical polymethyl methacrylate (PMMA)-methanol, which exhibits Case II transport (linear with time). Colorimetric, optical, NMR, and Raman techniques have all been applied but face various limitations including indirect measurement, spatial resolution, temporal resolution, and introduction of unquantified uncertainty due to sample swelling. Here we present an approach with neutron imaging in which the isotopic sensitivity of neutrons is employed to create contrast by using deuterated methanol and protiated PMMA. A custom sample cell and alignment system were developed to enable time-resolved, through-plane imaging of a polymer disk suspended in a solvent bath which permeates only in the radial direction. White beam imaging with thermal neutrons demonstrated that radial concentration profiles can be extracted from the measured transmission images with adequate spatiotemporal resolution, but an unquantified uncertainty remains due to the unknown excess volume relationship of the PMMA-methanol system. To address this, a pathway to a fully quantitative measurement with dual-band (cold and thermal) or hyperspectral (time-of-flight) neutron imaging is proposed.

12:15 PM C4.07.08
Studying Morphology Transitions on Sequential Annealing of Lamellar Block Copolymer Thin Films via Neutron Reflectivity Kshitij Sharma[1], Maninderjeet Singh[1], Sushil K. Satija[2], John F. Ankner[3], Jack Douglas[2] and Alamgir Karim[1]; [1]University of Houston, United States; [2]National Institute of Standards and Technology, United States; [3]Oak Ridge National Laboratory, United States

Neutron reflectivity (NR) is an excellent technique for identifying and modeling the 1D structural characteristics of materials based on the components' differential scattering length density (SLD). With just NR, equilibrium parallel lamellar morphologies for symmetric block copolymer (BCP) assembly can be fully characterized. Here, we demonstrate the use of NR to identify transition mechanisms between two lamellar states with domain sizes of L_o and $L_o/2$ for thermally annealed (TA) and direct immersion annealed (DIA) thin films, respectively. This was done by trapping thin film morphologies at intermediate stages of development followed by NR. The acquired data were reduced with the reductus program by NIST and fitted to a 1D model of SLD variation with thickness using the NIST's reflpak software package. The fitted NR profiles show a fast-tracked transition (hours) from DIA to TA microstructure compared to TA alone (days). Fitting data for the reverse transitions reveal a transition morphology characterized by a highly rough surface. The modeled NR data provides insight into the internal structure of this rough surface and the cause of such surface instability on the DIA of a TA microstructure. The series of NR fits show how the surface instability progresses towards the regular DIA lamellar structure ($L_o/2$). Atomic force microscopy (AFM) conducted parallel to NR offers real space transition pictures between the two lamellar morphologies and corroborates the NR data. The sequential annealing technique discussed, DIA to TA, shows the expedited formation of the TA morphology compared to TA alone. The sequential TA to DIA experiments show the formation of a surface roughness instability due to rapid solvent incursion into an existing lamellar structure and reveal a significant asymmetry in the transition kinetics above (sluggish) and below (rapid) the entanglement molecular weight. The interesting structural crossovers between the two states for the same BCP as revealed by neutron reflectivity and AFM are explained with respect to interfacial energy, in-plane vs. through-plane chain diffusion, and in-plane chain junction density evolution.

12:30 PM C4.07.09
Thermodynamic Interactions in Polydiene/Polyolefin Blends Megan L. Robertson, Jialin Qiu and Ramanan Krishnamoorti; University of Houston, United States

Polymer blends exhibit properties that are highly dependent on interactions between components, typically quantified by the Flory-Huggins interaction parameter, χ. Prediction of blend phase behavior requires characterization of the temperature dependence of χ. Polyolefins and polydienes are important materials with commercial relevance in elastomer applications. The majority of previous studies on the thermodynamics in polyolefin and polydiene systems have focused on polymer pairs within the same class (i.e. polyolefin/polyolefin and polydiene/polydiene blends), which generally exhibit a small and weakly temperature dependent χ. There is little quantitative information on thermodynamic interactions in systems that contain both polydienes and polyolefins. We have characterized interactions in polyolefin and polydiene blends, composed of 1,2-polybutadiene, 1,4-polyisoprene, poly(ethyl ethylene), and poly(ethylene-*co*-ethyl ethylene), utilizing small angle neutron scattering (SANS). SANS data were analyzed through Random Phase Approximation and Zimm analyses in order to extract χ as a function of temperature. We observed an unusually large χ parameter in these polydiene/polyolefin blends. We also studied the impact of partial saturation of the polydiene on the thermodynamics of polydiene/polyolefin blends. The applicability of the random copolymer theory to predict $\chi(T)$ behavior in these blends was evaluated.

12:45 PM C4.07.10
Specific Salt Effects on Equilibrium and Flow Structure of Wormlike Micelles Javen S. Weston[1], Nour Alawami[1] and Katie M. Weigandt[2]; [1]The University of Tulsa, United States; [2]National Institute of Standards and Technology, United States

Adding a dissolved salt to aqueous solutions of surfactant molecules is a well-understood method of altering how the molecules self-assemble into micelles. In a certain range of salt and surfactant concentrations, the most energetically favorable micelle shape is a long, flexible cylindrical micelle referred to as a ‘wormlike’ micelle. In this work, we show that a variety of simple salts can be used to independently control the contour length and

persistence length (stiffness) of a model wormlike micelle system. Static neutron scattering was used to determine the micelle persistence length, and RheoSANS measurement are used to measure the segmental alignment of the micelles under shear flow conditions. We also develop a semi-empirical model to explain the relative importance of micelle stiffness, micelle length, and degree of entanglement on the onset of segmental alignment in these systems, and gain further insight into the fundamental physics of wormlike micelle solutions.

Neutron Physics

* Invited Paper

SESSION G1.01: Neutron Physics I
Session Chair: Dusan Sarenac
Monday Morning, June 6, 2022
UMC Aspen Room 285, 287, 289

10:15 AM *G1.01.01
Pendellösung Interferometry Measurement of the Neutron Charge Radius and Constraints on New Physics Benjamin Heacock[1], Fujiie Takuhiro[2], Robert W. Haun[3], Albert Henins[1], Katsuya Hirota[2], Takuya Hosobata[4], Michael G. Huber[1], Masaaki Kitaguchi[2], Dmitry Pushin[5], Hirohiko Shimizu[2], Masahiro Takeda[4], Robert Valdillez[6], Yutaka Yamagata[4] and Albert Young[6]; [1]National Institute of Standards and Technology, United States; [2]Nagoya University, Japan; [3]Tulane University, United States; [4]RIKEN, Japan; [5]University of Waterloo, Canada; [6]North Carolina State University, United States

Pendellösung interference occurs between internal neutron wave states in a Bragg-diffracting crystal. The absolute phase of the interference fringes is directly related to the neutron-crystalline potential. Recent measurements of silicon Bragg reflections with Miller indices (111), (220), and (400) have resulted in a determination of the neutron mean square charge radius of -0.1101 ± 0.0089 fm^2 and placed limits on a Yukawa modification to gravity on atomic length scales. Further measurements of other Bragg reflections and crystal species can improve the precision of the charge radius measurement, as well as further constrain Beyond the Standard Model forces.

10:45 AM G1.01.02
Measuring Higher Order Neutron-Silicon Structure Factors with Pendellösung Interferometry Using a Pulsed Beam Robert Valdillez[1], Leah Broussard[2], Matthew J. Frost[2], Robert W. Haun[3], Benjamin Heacock[4], Colin Heikes[5], Albert Henins[4], Katsuya Hirota[6], Shannon F. Hoogerheide[4], Takuya Hosobata[7], Michael G. Huber[4], Masaaki Kitaguchi[6], Dmitry Pushin[8], Hirohiko Shimizu[6], Masahiro Takeda[7], Fujiie Takuhiro[6], Yutaka Yamagata[7] and Albert Young[1]; [1]North Carolina State University, United States; [2]Oak Ridge National Laboratory, United States; [3]University of Colorado Boulder, United States; [4]National Institute of Standards and Technology, United States; [5]Northrop Grumman, United States; [6]Nagoya University, Japan; [7]RIKEN, Japan; [8]University of Waterloo, Canada

Dynamical diffraction gives rise to multiple waves inside perfect crystals when the incident wave nearly satisfies a Bragg scattering condition. The interference of these waves, called pendellösung, can be observed by "fringe-like" modulation of the intensity of the transmitted or diffracted beams exiting the crystal. Pendellösung interferometry can be used to precisely determine neutron-silicon structure factors, which may be used to investigate interactions Beyond the Standard Model, measure the internal structure of the neutron via the neutron charge radius, and provide information on thermal motion of the atoms in a lattice. While neutron-silicon structure factors have recently been measured for the (111), (220), and (400) reflections, quality data do not yet exist for the case of high-order reflections. Progress towards using the pulsed beam at the VULCAN beamline located at the Spallation Neutron Source run by Oak Ridge National Lab to measure the (333), (444), and (555) reflections simultaneously will be discussed. Leveraging the pulsed beam to measure multiple structure factors simultaneously will reduce some of the systematic uncertainties associated with the previous experiment. A successful measurement will allow for the study of anharmonic contributions, increase the precision of the determined neutron charge radius, and provide further constraints on an atomic length scale "fifth" force.

11:00 AM G1.01.03
Quantum Information Model for Neutron Diffraction Shows Promise for Neutron Optics Design Olivier Nahman-Lévesque[1], Dusan Sarenac[1], David Cory[1], Benjamin Heacock[2], Michael G. Huber[2] and Dmitry Pushin[1]; [1]University of Waterloo, Canada; [2]National Institute of Standards and Technology, United States

Designing precise optical components for neutrons would open the door to many high-performance measurement devices. However, it is a challenging task from a theoretical standpoint, due to the complexity of calculating the outcome of neutron diffraction by crystals. A new proposed model suggests that representing a crystal as a Galton board, where every peg is a quantum unitary gate acting on the neutron state, is an adequate theoretical description of the neutron diffraction problem, while being much less complex computationally. This proposition is supported by examples, where simulated profiles are a good match to theory in the Laue and Bragg geometries, and to data in mixed geometries where the wavefunction cannot be calculated. This is a promising tool for the field of neutron optics, which would allow simulation of complex diffraction systems containing imperfections, strains, and complex geometries. This could help improve the currently available optical technology and push the limit on the measurement of fundamental constants of nature.

11:15 AM G1.01.04
Neutron Interferometry and Current Advances Dmitry Pushin[1], Benjamin Heacock[2], Michael G. Huber[2], Dusan Sarenac[1], Chandra B. Shahi[3], Ivar Taminiau[1] and David Cory[1,1]; [1]University of Waterloo, United States; [2]National Institute of Standards and Technology, United States; [3]University of Maryland, United States

In this talk I will describe several current advances in our work with neutron interferometers (NIs). Neutron interferometry delivered numerous hallmark experiments exploring the nature of the neutrons and their interactions. In recent years we have developed new tools and approaches specifically for developing high quality neutron devices with sub-micron flatness, and sub-nanometer surface roughness. This will allow fabricating of large neutron

interferometers with extremely high contrast without the need for etching, which will provide better precision. Further integration of quantum information science techniques and quantum information models enables us to make a setup with a mode of operation which relaxes the stringent isolation requirements and lets us explore higher precision measurements of quantum phenomena and search for new physics.

11:30 AM G1.01.05
Generation and Detection of Structured Waves of Neutrons and Light Charles W. Clark[1], Dusan Sarenac[2], Melissa E. Henderson[2], Huseyin Ekinci[2], Chandra B. Shahi[1], David Cory[2], Lisa DeBeer-Schmitt[3], Michael G. Huber[1], Connor L. Kapahi[2] and Dmitry Pushin[2]; [1]National Institute of Standards and Technology, United States; [2]University of Waterloo, Canada; [3]Oak Ridge National Laboratory, United States

Work on structuring wavefronts of light and particles began in the early 1990s and has since blossomed into an active field of research and applications, [1] with vortex beams of neutral atoms and molecules being demonstrated just in the past year. [2]
We have investigated several approaches to applying such techniques to neutrons, [3-8]. These have also led to new techniques and applications of structured light. [9-13] Our methods include use of material phase plates in neutron interferometry and magnetic fields to induce neutron spin-orbit coupling.
A major challenge stems from the fact that neutron optical devices are limited to refractive indices on the order of $n \approx 1 \pm 10^{-5}$. By exploiting advances in precision manufacturing, we have designed and constructed a micro-meter period triangular grating with a high aspect ratio of 14.3. The manufacturing quality is demonstrated with white-light interferometric data and microscope imaging. We will present neutron scattering results in good agreement with refraction modelling. Capabilities of neutron Fresnel lenses based on this design are contrasted to existing neutron focusing techniques and the path separation of a prism-based neutron interferometer is estimated.
Our neutron experiments were performed at the NIST Center for Neutron Research, National Institute of Standards and Technology, and the High Flux Isotope Reactor, Oak Ridge National Laboratory.
1. "Roadmap on structured light," H. Rubinsztein-Dunlop, et al., *J. Optics* **19**, 013001 (2016)
2. "Vortex beams of atoms and molecules," A Luski, Y. Segev, R. David, O. Bitton, H. Nadler, A. Ronny Barnea, A. Gorlach, O. Cheshnovsky, I. Kaminer, E. Narevicius, *Science* **373**, 1105 (2021)
3. "Next-generation high transmission neutron optical devices utilizing micro-machined structures," C. Kapahi, D. Sarenac, M. Bleuel, D. G. Cory, B. Heacock, M. Henderson, M. G. Huber, I. Taminiau, D. A. Pushin, arXiv:2112.13176 (2021)
4. "Generation and detection of spin-orbit coupled neutron beams," D. Sarenac, C. Kapahi, W. C. Chen, C. W. Clark, D. G. Cory, M. G. Huber, I. Taminiau, K. Zhernenkov and D. A. Pushin, *Proc. Nat. Acad. Sci.* **161**, 20328 (2019)
5, "Methods for preparation and detection of neutron spin-orbit states," D. Sarenac, J. Nsofini, I. Hincks, M. Arif, C. W. Clark, D. Cory, M. Huber and D. Pushin, *New Journal of Physics* **20**, 103012 (2018)
6. "Holography with a neutron interferometer," D. Sarenac, M. G. Huber, B. Heacock, M. Arif, C. W. Clark, D. G. Cory, C. B. Shahi and D. A. Pushin, *Optics Express* **24**, 22528 (2016)
7. "Spin-orbit states of neutron wave packets," J. Nsofini, D. Sarenac, C. J. Wood, D. G. Cory, M. Arif, C. W. Clark, M. G. Huber, and D. A. Pushin, *Phys. Rev. A* **94**, 013605 (2016)
8. "Controlling Neutron Orbital Angular Momentum," C. W. Clark, R. Barankov, M. G. Huber, M. Arif, D. G. Cory and D. A. Pushin, *Nature* **525**, 504 (2015)
9. "Generation of a lattice of spin-orbit beams via coherent averaging," D. Sarenac, D. G. Cory, J. Nsofini, I. Hincks, P. Miguel, M. Arif, C. W. Clark, M. G. Huber and D. A. Pushin, *Phys. Rev. Lett.* **121**, 183602 (2018)
10. "Remote state preparation of single photon orbital angular momentum lattices," A. R. Cameron, S. W. L. Cheng, S. Schwarz, C. Kapahi, D. Sarenac, M. Grabowecky, D. G Cory, T. Jennewein, D. A. Pushin, K. J. Resch. arXiv:2106.08428 (2021)
11. "Psychophysical discrimination of structured light exhibiting spatially-dependent polarization," A. E. Silva, D. Sarenac, D. G. Cory, I. Taminiau, D. A. Pushin, B. Thompson, *J. Vision* **20**, 265 (2020)
12. "Direct discrimination of structured light by humans," D. Sarenac, C. Kapahi, A. E. Silva, D. G. Cory, I. Taminiau, B. Thompson, B. Thompson, D. A. Pushin, *Proc. Nat. Acad. Sci.* **117**, 14682 (2020)
13. "Talbot effect of orbital angular momentum lattices with single photons," S. Schwarz, C. Kapahi, R. Xu, A. R. Cameron, D. Sarenac, J. P. W. MacLean, K. B Kuntz, D. G Cory, T. Jennewein, K. J Resch, D. A Pushin, *Phys. Rev. A* **101**, 043815 (2020)

11:45 AM G1.01.06
Spin-Orbit Correlations in Neutron Beams Dusan Sarenac[1], Connor L. Kapahi[1], Wangchun Chen[2], Charles W. Clark[3], David Cory[1], Michael G. Huber[2], Ivar Taminiau[1], Kirill Zhernenkov[4] and Dmitry Pushin[1]; [1]University of Waterloo, Canada; [2]National Institute of Standards and Technology, United States; [3]Joint Quantum Institute, National Institute of Standards and Technology and University of Maryland, United States; [4]J\"ulich Centre for Neutron Science at Heinz Maier-Leibnitz Zentrum, Germany

In this talk I will cover our recent work on developing methods to induce spin dependant phase shifts that are directly correlated to neutron's transverse momentum as well as orbital angular momentum (OAM). These techniques create coherence between spin and momentum profiles and can be tailored to particular applications, such as neutron studies of topological materials as well as neutron studies with grating-based interferometers. Our practical demonstration made use of 3He neutron spin filters in conjunction with specifically oriented triangular coils to prepare neutron beams with lattices of spin-orbit correlations, as demonstrated by their spin-dependent intensity profiles.

12:00 PM G1.01.07
Measurements of the Neutron's Charge Distribution; a History Michael G. Huber[1], Benjamin Heacock[1], Robert Valdillez[2], Fujiie Takuhiro[3], Masaaki Kitaguchi[3], Hirohiko Shimizu[3], Katsuya Hirota[4], Masahiro Takeda[4], Yutaka Yamagata[4], Dmitry Pushin[5,6] and Albert Young[2]; [1]NIST, United States; [2]North Carolina State University, United States; [3]Nagoya University, Japan; [4]RIKEN, Japan; [5]University of Waterloo, Canada; [6]Institute for Quantum Computing, Canada

The neutron carries no known electrical charge, no known electric dipole moment, but does exhibit a known spherically symmetric charge distribution. This charge distribution created by the neutron's internal quark structure, is quantified by the mean-square charge radius ($\langle r_n^2 \rangle$). For the neutron $\langle r_n^2 \rangle < 0$ reflecting the fact that it has a slightly negative outer shell. The mean-square charge radius has been measured experimentally by its proportionality, through a series of known constants, to the neutron-electron scattering length (b_{ne}). The difficulty in measuring $\langle r_n^2 \rangle$ is that b_{ne} is ~1000x smaller than nucleus scattering. Prior measurements of $\langle r_n^2 \rangle$ utilized asymmetric neutron scattering from targets of noble gases or total cross-sectional scattering through dense liquids. The Particle Data Group (PDG) considers 5 such experiments and reports an average $\langle r_n^2 \rangle_{PDG} = -0.1161(22)$ fm^2 where the error has been inflated by 30% to account for minor discrepancies. Recently, there has been several new determinations of $\langle r_n^2 \rangle$. These include a χEFT approach using isotope shifts by Filin et al. (PRL 2020), an interpretation of electron scattering data by Atac et al. (Nat Com 2021), and a direct neutron measurement $\langle r_n^2 \rangle$ by Heacock et al. (Science 2021). For context on how these new determinations improve our understanding of $\langle r_n^2 \rangle$, the speaker

will discuss the current measurements making up the Particle Data Group's average.

12:15 PM G1.01.08
Precision Measurement of the Gravitational Constant via Three-Phase Grating Neutron Interferometry Connor L. Kapahi[1,2], Dusan Sarenac[2], Charles W. Clark[3], David Cory[1,2], Benjamin Heacock[3], Michael G. Huber[3], Youngju Kim[3] and Dmitry Pushin[1,2]; [1]University of Waterloo, Canada; [2]Institute for Quantum Computing, Canada; [3]National Institute of Standards and Technology, United States

The Gravitational Constant (G) is the least precisely known fundamental constant of nature. The source of this is discrepancies between the variety of measurement methods. Neutrons are ideal probes of nature due to their mass, and the fact that they are electromagnetically neutral. Neutron interferometers have long been proposed as gravitational measurement devices, but low neutron flux, small interferometer areas, and sensitivity to noise has prevented measurements of G. A new interferometer, called a Phase-Grating Moiré Interferometer (PGMI), has been shown to offer orders of magnitude increases in neutron flux and interferometer area. In this talk, the feasibility of this experiment is discussed using a model which calculates the effect of a test mass on a neutron and produces the expected interference pattern from the PGMI. This effect is compared to other gravitational effects which will be present during the experiment and potential sources of uncertainty are discussed.

SESSION G2.02: Neutron Physics II
Session Chair: Benjamin Heacock
Tuesday Afternoon, June 7, 2022
UMC Aspen Room 285, 287, 289

2:00 PM *G2.02.01
Fundamentals of Entangled Neutron Beams Gerardo Ortiz[1,2]; [1]Indiana University, United States; [2]Indiana University Quantum Science and Engineering Center, United States

Advancing the frontiers of science often requires the creation of new probes to uncover the underlying microscopic mechanisms giving rise to exotic macroscopic phenomena, such as high-temperature superconductivity. Can quantum entangled probes uncover the inherent entanglement of the target matter? We have recently [1,2] developed an entangled neutron beam where individual neutrons can be entangled in spin, trajectory and energy. To demonstrate entanglement in these beams we crafted neutron interferometric measurements of contextuality inequalities whose violation provided an indication of the breakdown of Einstein's local realism. In turn, the tunable entanglement (spin-echo) length of the neutron beam from nanometers to microns and energy differences from peV to neV opens a pathway to a future era of entangled neutron scattering in matter. What kind of information can be extracted with this novel entangled probe? A recent general quantum entangled-probe scattering theory [3] provides a framework to respond to this question. Interestingly, by carefully tuning the probe's entanglement and inherent coherence properties, one can directly access the intrinsic entanglement of the target material. This theoretical framework supports the view that our entangled beam can be used as a multipurpose scientific tool. We are currently pursuing several ideas and developing new spin-textured entangled beams with orbital angular momentum (OAM) for future experiments in candidate quantum spin liquids, unconventional superconductors, and chiral quantum materials.

[1] J. Shen et. al., Nature Commun. 11, 930 (2020).
[2] S. Lu et. al., Phys. Rev. A 101, 042318 (2020).
[3] A. A. Md. Irfan, P. Blackstone, R. Pynn, and G. Ortiz, New J. Phys. 23, 083022 (2021).

2:30 PM G2.02.02
How Entangled Neutron Beams Unveil Chiral Spin Orders Abu Ashik Md Irfan and Gerardo Ortiz; Indiana University, United States

We apply our entangled-probe scattering theory to an incident neutron beam entangled in spin and path degrees of freedom [1]. Previously, it has been shown that such entangled beam can distinguish entangled matter states from un-entangled ones when the neutrons scatter off spin dimer orders [2]. We extend this analysis to N spinors and study both short-and long-range chiral spin orders. We show that this entangled probe can successfully distinguish between different chiral spin orders in the plane wave limit when the neutrons scatter off a spin trimer. We expect this study sheds new light on the electronic correlations of materials with unconventional quantum orders.

[1] Nat. Commun. 11, 930 (2020)
[2] New J. of Phys. 23, 083022 (2021)

2:45 PM G2.02.03
Spin-textured Neutron Beams and Orbital Angular Momentum Sam McKay[1,1,1], Quan Le Thien[1,1], Fankang Li[2], Abu Irfan[1,1], Jiazhou Shen[1,1,1], Eric B. Dees[1,1,1], Stephen J. Kuhn[1,1,1], David V. Baxter[1,1,1], Gerardo Ortiz[1,1] and Roger Pynn[1,1,1]; [1]Indiana University, United States; [2]Oak Ridge National Laboratory, United States

Interest in structured beams of both polarized light and matter waves has grown over the last decade, specifically in beams with definite states of *orbital angular momentum* [1,2]. Orbital angular momentum (OAM) in this context means that the topology of the wavefronts of each particle directly links to a phase singularity $e^{il\varphi}$, where φ is the azimuthal angle about the direction of travel and l the OAM quantum number. We show that one can generate such OAM neutron beams using suitably focused *magnetic Wollaston prisms* (MWPs), birefringent neutron polarization devices based on superconducting technology [3].
When focused, the prisms produce a lattice of definite OAM states. Our analysis consists of both a quantum analytical calculation and a semiclassical simulation performed with the McStas ray-tracing program. For a few configurations of MWPs, we demonstrate the relationship between the generated OAM state and the corresponding spatial spin-texture of the beam, showing that our neutrons are in an *intraparticle* entangled state of spin and OAM (e.g., a spin-orbit state).

We conjecture that such spin-textured beams will be a direct probe of the intrinsic chirality and the topological properties of materials, and we present some expected beam-sample interactions. We propose future experiments that will test how such a spin-textured beam interacts with different chiral and topological systems, such as non-centrosymmetric crystals.

[1] D. Sarenac et al. *Proceedings of the National Academy of Sciences* (2019)
[2] Y. Shen et al. *Light: Science & Applications* (2019)
[3] F. Li et al. *Review of Scientific Instruments*, (2014)

Funding Acknowledgment: DOE STTR grant DE-SC0021482, US DOC NIST COOP Agreement No. 70NANB15H259

3:00 PM G2.02.04
New Determination of the ^{3}He Neutron Incoherent Scattering Length $\Delta b'_i$ Earl Babcock; Juelich Centre for Neutron Science, Germany

We report a new measurement of the ^{3}He neutron incoherent scattering length. The two previous measurements of this important quantity for nuclear physics theory were in disagreement by more than the standard deviations; the measurement presented here is in agreement with one of the prior values by M.G. Huber et .al, Phys. Rev. C 90, 064004 (2014). We measured the pseudomagnetic precession of neutrons through an in-situ polarized ^{3}He gas target which helped to eliminate sources of systematic error. The resulting measurement determines the difference $\Delta b'_i = b_+ - b_-$ in the two low-energy s-wave neutron-nucleus scattering amplitudes b_+ and b_-, respectively corresponding to the singlet J= 0 and triplet = 1 states of the neutron-3He interaction. The measurement was conducted at the FZ-Jülich neutron spin echo spectrometer at the Heinz Maier Leibnitz Zentrum (MLZ).

Biology, Biophysics and Biotechnology

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* Invited Paper

SESSION D1.01: Structure and Dynamics of Proteins and Peptide Assemblies
Session Chairs: Elizabeth Kelley and Haden Scott
Monday Afternoon, June 6, 2022
UMC Conference Room 235

4:00 PM *D1.01.01
Supramolecular Self-Assembly of Computationally Designed Coiled Coil Building Blocks Nairiti J. Sinha; University of California, Santa Barbara, United States

Computational design tools enable screening of peptide sequences that can assemble into functional nanomaterials and display target nanostructure. In my talk, I will discuss the assembly and characterization of computationally designed peptides that form coiled coil *bundlemers* having a hydrophobic core and side chains that are strategically chosen *in silico* to enable self-assembly under mild solution conditions. Via a feedback cycle between experiments and sequence-optimization algorithms, we have tested multiple designs for successful bundlemer formation. Taking advantage of their structural robustness, we modified bundlemers to incorporate thiol or maleimide groups that yielded tetra-functional building blocks. The bundlemers were reacted via Thiol-Michael click reaction which results in end-to-end polymerization of bundlemers. Short linkers between bundlemers yielded rigid rod-like polymers, whereas longer flexible linkers resulted in semi-rigid polymers. The differences in nanostructure and bending dynamics of rigid rod-like versus semi-rigid bundlemer polymers was investigated using a combination of Small-Angle Neutron Scattering, Neutron Spin Echo spectroscopy and Transmission Electron Microscopy techniques, these results will be discussed in this talk. The bundlemer-based polymers are not only useful for new material development but are also potential model systems to study sequence-driven structure-property relationships in natural 1D assemblies.

4:30 PM D1.01.02
Studying Internal Dynamics of the Monoclonal Antibody with SANS and NSE Roisin Donnelly[1,1,2], Yun Liu[2,1] and Norman Wagner[1,2]; [1]University of Delaware, United States; [2]NIST Center for Neutron Scattering, United States

Internal dynamics of a protein is key to many of its functions and covers a wide range of time and length scale. Here, we present our results from both SANS and NSE experiments which explore the internal dynamics of monoclonal antibody (mAb), an important class of proteins, of large interest to the pharmaceutical industry. Hydrogen deuterium exchange, (HDX) is of increasing interest for characterization of protein dynamics in solution, which can inform therapeutic stability and efficacy. Due to very large difference of the neutron scattering cross section between H and D, small angle neutron scattering (SANS), is a very sensitive technique to study the HDX in solution. SANS allows for the continual measurement of HDX over time and is a non-invasive technique to investigate the HDX in solution. By using SANS to study temperature, we probed the HDX of mAb, NISTmAb RM 8670, over the course of a couple of days. The HDX rate is observed to depend on the protein buffer conditions. We further use the neutron spin echo (NSE) to investigate the internal motions of the mAbs at similar buffer conditions and evaluate its potential correlation between internal domain motions and the HDX exchange rate. The detailed analysis of both SANS and NSE data will be discussed.

4:45 PM D1.01.03
Small-Angle Scattering to Understand Preservative-Induced Aggregation of Poloxamer 188 in Pharmaceutical Formulations Rachel R. Ford[1], Peter Gilbert[1], Ken Qian[2], Norman Wagner[3] and Yun Liu[1]; [1]NIST Center for Neutron Research, United States; [2]Eli Lilly & Company, United States; [3]University of Delaware, United States

Multi-injection pharmaceutical products such as insulin must meet certain requirements to ensure their safety and accurate dosage. As aggregation may lead to potential immunological responses and impact the efficacy of drugs, biotherapeutic formulations often require stabilizing excipients including

nonionic surfactants and antimicrobial agents. A commonly used nonionic surfactant is poloxamer 188 (P188), an ABA triblock copolymer of polyethylene oxide and polypropylene oxide. While P188 does not have a critical micelle concentration below 30°C, when in combination with the preservatives phenol and benzyl alcohol, P188 can aggregate and undergo large-scale phase separation over time. To understand what gives rise to this unexpected aggregation, we have conducted a systematic study of the P188/phenol/benzyl alcohol system using a variety of small-angle scattering techniques. We present here the differing impact of phenol and benzyl alcohol on P188 aggregate formation and the effect of temperature on aggregation and phase boundaries. Our results provide insight into P188/preservative solution stability that can help the pharmaceutical industry in the design of future formulations.

5:00 PM D1.01.04
Investigating Aggregation Surfaces In Thawed Bispecific Antibody Fragments Julia Greenfield; National Institute of Standards and Technology, United States

The field of therapeutic antibodies has rapidly grown since the FDA approved the first anybody drug Orthoclone OKT3 in 1986. Since then, more antibody-based therapeutics have been approved for treating a wide range of ailments. The precise binding nature of antibodies allows them to function in a variety of applications. However, antibody-based drugs' production, purification, and formulation are not trivial. Storage and stability concerns are incredibly important to maintain an antibody drugs' functionality and structural integrity. The manufacturers of antibodies-based therapeutics often use freezing as a long or intermediate-term storage method. Freezing preserves the protein/ biologics while inhibiting degradation and microbial contamination. However, freezing is not without its shortcomings on the biologic and can introduce some structural changes and diminished function in some antibody-based biologics.

Freezing can cause proteins to experience localized unfolding due to interacting with the ice-water interface. Also, freezing causes freeze concentration, where a protein sample is concentrated as free water and turned into ice. Both protein unfolding and protein freeze concentration can impact the post-frozen protein sample. It can facilitate aggregation/ self-association in the post-frozen sample that is not seen before the freezing process. Excipients are added to promote biological stability and functionality in liquid or the frozen state, but they may not eliminate the freezing's negative effects. The tendency of antibodies to form higher-order aggregates is of particular concern because of their potential to produce an immune response. Investigating antibody crowding in both the frozen state and antibody self-association in the post frozen state could help understand the antibody behavior during the phase transition.

Size exclusion chromatography coupled with multi-angle light scattering and small-angle x-ray scattering is a powerful technique to separate oligomeric species, determine size and shape information. Two standard bispecific monoclonal antibody fragments of different sizes were analyzed after thawing. The antibody samples contained high molecular weight species not present before freezing. Structural information from the SAX data and molecular dynamics is used to model the surface of interaction in the dimers.

5:15 PM D1.01.05
Characterize Conformational Flexibility of Monoclonal Antibodies using Small-Angle Scattering Amy Xu; Louisiana State University, United States

Understanding the behavior of protein molecules in crowded environments paves the ground for both fundamental and applied research. In particular, the ability to characterize the conformation and intermolecular interactions of monoclonal antibodies (mAbs) in concentrated formulations is of fundamental importance towards a better understanding of their physical stability and viscosity behavior. The high concentrations of mAb and excipients pose significant challenges for conventional biophysical characterization methods. Among others, small-angle scattering represents an ideal characterization approach for concentrated protein samples. Recent study from our group demonstrates that the preferred configurations of mAbs in concentrated formulations can be determined directly from the scattering data by integration of molecular simulations. In particular, we were able to identify the change of a model mAb structure with increasing protein and NaCl concentration where the most likely configurations were extended. The demonstrated methodology and results extend the use of analyzing S(q)eff from smallangle scattering data, thus allowing comprehensive characterization of the conformational flexibility and intermolecular interactions of mAbs in the relevant physicochemical conditions.

SESSION D3.02: Insights into Lipid Membrane Properties and Protein-Lipid Interactions
Session Chairs: Roisin Donnelly and Nairiti Sinha
Wednesday Morning, June 8, 2022
UMC Conference Room 235

10:15 AM *D3.02.01
The Orientation of KRAS at the Plasma Membrane Helps Recruit RAF Kinase Andrew Stephen; Frederick National Laboratory for Cancer Research, United States

KRAS is a small GTPase that is frequently mutated in human cancers, particularly pancreatic, colorectal and lung cancer. Engagement of mitogens with transmembrane receptor tyrosine kinases result in the activation of KRAS from an inactive GDP to active GTP bound state. Localized on the inner leaflet of the plasma membrane, active KRAS-GTP serves to recruit RAF kinase to the membrane where it undergoes activation. Oncogenic mutations lock KRAS in the active GTP state resulting in dysregulated cellular growth and proliferation. We have used a combination of biophysical methods and computational simulations to investigate the molecular details of these first steps in signal transduction. Specifically, NMR and neutron reflectivity of KRAS bound to lipid mimetics coupled with coarse grained molecular dynamics simulations reveal that KRAS is predominantly extended from the membrane making only transiently interacts with the membrane. This orientation at the membrane allows KRAS to efficiently recruit RAF from the cytoplasm for activation at the plasma membrane.

10:45 AM D3.02.02
Characterization of Structurally Disordered Peripheral Membrane Proteins with Neutron Reflectometry Frank Heinrich[1,2]; [1]Carnegie Mellon University, United States; [2]National Institute of Standards and Technology, United States

A significant fraction of biomedically relevant peripheral membrane proteins has structurally disordered regions of amino acids that provide configurational flexibility, often with functional implications. Neutron reflectometry is the method of choice to structurally characterize such membrane-bound proteins in a biomimetic environment, albeit at low spatial resolution. Thermodynamic studies, complementary structural techniques, and molecular simulations are part of an integrative approach that supplements the limited information from NR. The goal is to derive a high-resolution, dynamic structural model that provides relevant information for a biomedical application. I will demonstrate this approach using the cancer-related KRAS [1] and Arf [2] proteins and the HIV-I accessory protein Nef. I will highlight recent work on a quantitative comparison of scattering data to other techniques and integrative modeling using MD and Monte Carlo simulations.

[1] Heinrich F. et al. *Membrane-Bound KRAS Approximates an Entropic Ensemble of Configurations.* Biophysical Journal 120:4055 (2021)
[2] Soubias O. et al. *Membrane surface recognition by the ASAP1 PH domain and consequences for interactions with the small GTPase Arf1.* Science Advances 6:eabd1882 (2020)

11:00 AM D3.02.04
The Transmembrane Helix of pHLIP Slows Down Membrane Thickness Fluctuations and Translational Diffusion Haden L. Scott[1], Violeta Burns-Casamayor[2], Andrew Dixon[3], Robert Standaert[4], Christopher B. Stanley[1], Laura Stingaciu[1], Jan Michael Carrillo[1], Bobby G. Sumpter[1], John Katsaras[1], Wei Qiang[5], Frederick Heberle[3], Blake Mertz[2], Rana Ashkar[6] and Francisco Barrera[3]; [1]Oak Ridge National Laboratory, United States; [2]West Virginia University, United States; [3]The University of Tennessee, Knoxville, United States; [4]East Tennessee State University, United States; [5]Binghamton University, The State University of New York, United States; [6]Virginia Tech, United States

Cell membranes arrange diverse biological processes through coordinated interactions with membrane proteins. While the effects of lipid membranes on membrane proteins are becoming better understood, knowledge on how membrane protein conformational changes influence membranes remains quite limited. Here, we combined experiment and simulation to selectively study the dynamic response of a lipid membrane to changes in the conformation of pH-low insertion peptide (pHLIP). pHLIP is highly sensitive to changes in pH, transitioning from surface-associated (SA) state at neutral pH, to a transmembrane (TM) α-helix under acidic conditions. Neutron spin echo (NSE) measurements on DOPC-cholesterol membranes revealed that membrane thickness fluctuations are not noticeably impacted by SA pHLIP but experience strong dampening in the presence of TM pHLIP. Small-angle neutron / X-ray scattering (SANS / SAXS) data showed no changes in membrane thickness in either state of pHLIP, suggesting that the suppression of thickness fluctuations by TM pHLIP is driven by pHLIP-induced changes in the membrane viscosity. These conclusions are consistent with MD simulations and solid-state NMR data that show that pHLIP promotes distortion in the conformation of the lipid acyl chains in the vicinity of pHLIP, resulting in increased membrane viscosity and reduced lateral lipid diffusion. These findings suggest a possible regulatory mechanism, where TM proteins cause the redistribution of lipid tail conformations, consequently altering membrane fluctuations and viscosity – properties that cells can sense and calibrate.

11:15 AM D3.02.05
Relationship between Viscosity and Acyl Tail Dynamics in Lipid Bilayers Michihiro Nagao[1,2,3], Elizabeth Kelley[1], Antonio Faraone[1], Makina Saito[4], Yoshitaka Yoda[5], Masayuki Kurokuzu[6], Shinichi Takata[7], Makoto Seto[6] and Paul D. Butler[1]; [1]National Institute of Standards and Technology, United States; [2]Unviersity of Maryland, United States; [3]Unviersity of Delaware, United States; [4]Tohoku University, Japan; [5]Japan Synchrotron Radiation Research Institute, Japan; [6]Kyoto University, Japan; [7]J-PARC, Japan

Biological membranes are unique borders that define cell boundaries and separate the cell interior from its surrounding environment. These membranes are rigid enough to maintain the cell shape and protect the cell from collapsing and leaking its components, and at the same time, are soft enough to deform and exchange molecules across the border. Although the bending and compression elasticity describe the membrane rigidity, these properties do not explain how fast embedded components can move inside the membrane. Instead, the viscous properties of the membranes determine these response times. As molecular diffusion and membrane viscosity are related to one another, various techniques have been developed to measure molecular diffusion in membranes and extract the corresponding membrane viscosity using hydrodynamic models. More recently, several techniques that directly measure membrane viscosity have been developed, including computational simulations. However, the measured values vary by several orders of magnitude, and a more thorough understanding of the origin of membrane viscosity is necessary. Here we employed quasi-elastic x-ray and neutron scattering techniques to access coherent lipid acyl tail correlation dynamics. [1] Two relaxation processes were observed in fluid phase dimyristoyl phosphocholine bilayers with relaxation times of ≈ 30 ps and ≈ 500 ps. The fast relaxation was attributed to density fluctuations of the lipid acyl tails, while the slow mode was assigned to the lipid molecular rearrangements. Based on the relationships for the density fluctuation motions and viscosity in molecular liquids, we estimated the membrane viscosity from the slower relaxation time scales. Furthermore, these two modes become a heterogeneous single mode with relaxation times on the order of 10s to 100s of ns in the gel phase. The calculated membrane viscosity from the relaxation times is about an order of magnitude larger in the vicinity of the melting transition temperature compared to the fluid phase and increased significantly with decreasing temperature.

Reference
[1] M. Nagao, E.G. Kelley, A. Faraone, M. Saito, Y. Yoda, M. Kurokuzu, S. Takata, M. Seto and P.D. Butler, Phys. Rev. Lett. 127, 078102 (2021).

11:30 AM D3.02.06
The Structural Origins of Lipid Bilayer Dynamic Response James E. Fitzgerald[1], Elizabeth Kelley[2], Norman Wagner[1], Michihiro Nagao[2] and Edward Lyman[1,1]; [1]University of Delaware, United States; [2]National Institute of Standards and Technology, United States

Recent theoretical developments suggest that membrane viscosity may be critical to the interpretation of Neutron Spin Echo measurements of unilamellar vesicles, but experimental measurements of membrane viscosity are challenging. In order to understand the factors which determine the viscosity of a membrane, we ran equilibrium all-atom simulations of single component lipid bilayers and calculated their viscosities. The viscosity was calculated via a Green-Kubo relation, with the stress tensor autocorrelation function fit to a stretched exponential by a maximum-likelihood Markov chain Monte Carlo method. By simulating a series of lipids at different temperatures, we establish the dependence of viscosity on several aspects of lipid chemistry, including hydrocarbon chain length, unsaturation, and backbone structure.

11:45 AM D3.02.07
Determination of Sphingomyelin Molecular Structure using SANS, SAXS, NMR, and Molecular Dynamics Jacob J. Kinnun[1,1], Milka Doktorova[2], Norbert Kučerka[3,4], Jianjun Pan[5], Drew Marquardt[6], Haden L. Scott[1,1], Richard Venable[7], Richard Pastor[7], Stephen Wassall[8], Frederick Heberle[9] and John Katsaras[1,9,1]; [1]Oak Ridge National Laboratory, United States; [2]University of Texas Health Science Center, United States; [3]Joint Institute for Nuclear Research, Russian Federation; [4]Comenius University, Slovakia; [5]University of South Florida, United States; [6]University of Windsor, Canada; [7]National

Institutes of Health, United States; [8]Indiana University-Purdue University Indianapolis, United States; [9]University of Tennessee, United States

Sphingomyelin is a major component in the formation of lipid domains. Here, we present the molecular structure for fluid phases of palmitoyl sphingomyelin (PSM) and stearoyl sphingomyelin (SSM) as determined using the scattering density profile (SDP) method, where small-angle neutron and X-ray scattering data are co-refined using a bilayer parsing scheme derived from molecular dynamics (MD) simulations. To expand upon structural detail, unconstrained all-atom simulations of PSM bilayers at 55 °C were carried out using the C36 CHARMM force field. The simulations resulted in an area per lipid of 56 $Å^2$, a value that is 10% lower than determined by SDP analysis (61.9 $Å^2$). Moreover, scattering form factors calculated from the unconstrained simulations were in poor agreement with experimental form factors, even though order parameter (S_{CD}) profiles calculated from the simulations were in relatively good agreement nuclear magnetic resonance (NMR) experiments. Conversely, constrained area simulations (i.e., 61.9 $Å^2$ area per lipid) resulted in good agreement between the simulation and experimental scattering form factors, but not with S_{CD} profiles from NMR. Possible reasons for the discrepancies between the different data used to validate MD force fields will be presented.

12:00 PM D3.02.08
Structural Studies of mRNA Vaccines Using Combined SANS/SAXS and CryoEM Thomas E. Cleveland[1,2], Manuel Carrasco[3], Lacey Wright[3], Michael Buschmann[3] and Alexander Grishaev[1]; [1]National Institute of Standards and Technology, United States; [2]University of Maryland, United States; [3]George Mason University, United States

Lipid nanoparticles (LNPs) can be formulated to deliver nucleic acids to cells, e.g. mRNA for vaccines, siRNA to silence the expression of genes involved in disease processes, or DNA for gene therapy applications. Retention of the nucleic acid cargo in LNPs under conditions of neutral pH, and targeted release upon acidification once internalized into cells, is facilitated through the use of ionizable lipids with carefully chosen pKa values. Typically, LNP formulations also include additional lipids such as DSPC, cholesterol, and a small proportion of lipid with PEGylated head groups. The identity and proportions of these additional lipids can be adjusted in order to optimize LNP properties such as their diameter and stability. The utility of LNPs for mRNA delivery has been dramatically illustrated by the success of the SARS-CoV-2 mRNA vaccines. However, many fundamental questions remain about LNP morphology and internal lipid/RNA distribution, and particularly how these relate to the stability and efficacy of drug formulations. We have performed parallel Small-Angle Neutron and X-ray Scattering (SANS/SAXS) and cryogenic electron microscopy (CryoEM) studies on an LNP system consisting of a reporter mRNA formulated with the ionizable lipid DLin-KC2-DMA along with lipids DSPC, cholesterol, and DMG-PEG2k. Using SANS/SAXS measurements on formulations prepared (1) in the presence and absence of mRNA; (2) with and without deuteration of the lipid DSPC; and (3) at several solvent D2O concentrations, we have obtained experimental information on the distribution of lipids and mRNA within the LNPs. For all conditions, CryoEM was used as a complementary technique to observe the morphology of LNPs, and to inform and constrain the models used in analyzing the scattering data.

SESSION D3.03: New Tools and Methods for Biological Scattering Experiments
Session Chairs: Rachel Ford and Jacob Kinnun
Wednesday Afternoon, June 8, 2022
UMC Conference Room 235

3:45 PM D3.03.01
Time-Resolved In-Situ Reaction SANS Study Details Structural Changes to Noncellulosic Biopolymer in Switchgrass Plant Cell Wall Sai Venkatesh Pingali[1], Zhi Yang[2], Marcus Foston[3], Hugh O'Neill[1], Volker S. Urban[1], Arthur Ragauskas[4], Barbara Evans[1] and Brian Davison[1]; [1]Oak Ridge National Laboratory, United States; [2]Massey University, New Zealand; [3]Washington University in St. Louis, United States; [4]The University of Tennessee, Knoxville, United States

Plant cell wall structure consisting of several carbohydrate polymers encased in hydrophobic lignin polymer to protect against degradation, is an intricate and complex structure. This complex structure of the plants was designed to naturally restrict access to the carbohydrate polymers. Consequently, production of second-generation bioethanol from such a system in a commercially viable manner is not simple and thermochemical approaches are required to open this complex plant cell wall structure to improve enzyme access. However, the recalcitrant nature of lignin negatively affects efficient enzymatic access. Several different thermochemical pretreatments have been extensively developed and employed, but the exact nature of plant cell wall recalcitrance and the most efficient and economical approach to alter plant cell wall structure via pretreatment remains elusive. To understand the role of noncellulosic switchgrass polymers on the overall efficiency of pretreatment, the structural evolution of the noncellulosic polymers of the plant cell wall were investigated during dilute acid pretreatment (DAP) by employing in-situ small-angle neutron scattering (in-situ SANS). In this study, we observed real-time structural changes not possible to observe by any other technique. To deconvolute the structural evolution of lignin and hemicellulose polymers during DAP, native switchgrass (NATV), and isolated holocellulose (HOLO) and cellulose (CELL) fractions from NATV were studied. Our results show that aggregate particles first appear around 80 °C for NATV and HOLO samples. Due to the low temperature and pretreatment severity condition, these particles are likely derived from hemicellulose. The formations of much larger aggregate particles, only observed in the NATV sample, were attributed to lignin. For the HOLO sample, as the temperature and pretreatment severity condition increased, hemicellulose-derived aggregate particle sizes increased, suggesting this process was the nucleation and early stage formation of pseudolignin particles. Consistent with our interpretation of structural evolutions in NATV and HOLO samples, no formation of aggregate particles was observed in CELL samples for the entire duration of the pretreatment. These results suggest that not only lignin but also hemicellulose can form aggregate particles within plant cell walls during pretreatment.

4:00 PM D3.03.02
Developing DENSS for Neutron Contrast Variation Data -- DENSS Multiple Shuo Qian; Oak Ridge National Laboratory, United States

DENsity from Solution Scattering (DENSS), developed by Thomas Grant in 2018, is an iterative algorithm for ab initio structure reconstruction of small angle scattering data. DENSS can reconstruct the scattering density directly from biological small angle x-ray and neutron scattering data without the use of modeling. Here we further developed the method to work simultaneously on multiple datasets from small angle neutron scattering (SANS) contrast variation data. DENSS Multiple, the new method, provides a single structural result that include all the information represented by different contrast. The resolution of results is usually improved compared to that of a single dataset. And more subtle features can be enhanced in the density map. We will discuss the test results on various examples including simulated and experimental data, its applications, and limitations.

4:15 PM D3.03.03
Structure-Based Calculation of Biomolecular Neutron Scattering Contrast Match Points with Explicit Deuteration Alan Hicks[1], Paul Abraham[1],

Qiu Zhang[1], Jeremy Smith[2,1], Hugh O'Neill[1] and Loukas Petridis[1]; [1]Oak Ridge National Laboratory, United States; [2]The University of Tennessee, Knoxville, United States

The contrast variation method is a fundamental neutron scattering technique for determining structural correlations and shape distributions of macromolecular complexes and their components. Calculation of contrast match points of these complexes is critical for the preparation of SANS experiments. We present a 3D structure-based model with explicit deuteration and solvent for predicting contrast match points of protein, RNA and DNA monomers and complexes. Our method combines a structural model for hydrogen deuterium exchange and an empirical model for deuterium incorporation into non-exchangeable hydrogen sites from mass spectrometry. We demonstrate that this method accurately predicts contrast match points over a wide range of contrast variation conditions. We believe our method can be easily expanded to other solvents, carbohydrates, lipids and eventually soft-matter systems.

4:30 PM D3.03.04
Applying New Models to Describe Biomembrane Structure and Solvent Partitioning in Living Cell Membranes and Membrane Mimics Luoxi Tan[1], Micholas Smith[2,3], Haden L. Scott[3], John Katsaras[3], Sai Venkatesh Pingali[3], Jeremy Smith[2,3], Brian Davison[3], James Elkins[3] and Jonathan Nickels[1]; [1]University of Cincinnati, United States; [2]The University of Tennessee, Knoxville, United States; [3]Oak Ridge National Laboratory, United States

Amphiphilic co-solvents have a significant impact on the structure, organization, and physical properties of the cell membrane. The cell membrane is an approximately five nanometer thick selectively semi-permeable lipid (and protein) membrane. Compositionally complex, dynamic, and organized in both the transverse and lateral dimensions, understanding the cell membrane structure – and the role that structure plays in cellular function, communication, and environmental sensing is an active scientific effort. Describing the mutual impact of partitioning and induced structure changes is therefore a crucial consideration in bioenergy research for microbial solvent tolerance in the production of biofuels and other fermentation products.
Small angle neutron scattering (SANS) is a key method for studying lipid and polymer bilayer structures, with many models for extracting bilayer structure (thickness, area per lipid, etc.) from scattering data in use. However, the molecular details of co-solvent partitioning are conflated with induced changes to bilayer structure, making interpretation and modeling of the scattering curves a challenge. To address this issue, we present a model of bilayer structure which includes a two-term partition constant accounting for the localization of the co-solvent within the bilayer.
We validate this model using a series of SANS measurements of lipid vesicles in the presence of the co-solvent THF; showing several strategies of how to deploy the two-parameter partition coefficient model to describe scattering data and extract both structure and partitioning information from the data. Molecular dynamics (MD) simulations have been used to both evaluate underlying assumptions of the new data fitting model and illustrate its complementary approach to the data fitting procedure for our model membrane.
The new structure/partitioning model has been applied to solvent partitioning in the cell membrane of *Bacillus subtilis* and a *B. subtilis* cell membrane extract. Previously, a novel isotopic labelling approach was developed to enable direct *in vivo* structural study of the cell membrane of the gram-positive organism, *B. subtilis*. This was accomplished through a genetic inhibition of fatty acid degradation (*DyusL*) and a chemical inhibition of fatty acid biosynthesis. Here, we utilize an updated and improved method of isotopic labelling by introducing a dCas9/sgRNA-*fabF* complex that blocks transcription of the essential *fabF* gene when under xylose induction. This leads to greater sensitivity to cerulenin, more robust cell growth when supplementary fatty acids are introduced and a subtle change in fatty acid uptake. This enables investigation of cellular uptake and utilization of fatty acids, cell membrane structure and organization as a phenotypic response to metabolic and environmental changes.

4:45 PM D3.03.05
Low-Background Neutron Reflectometry Measurement Strategies for Solid/Liquid Interfaces David P. Hoogerheide[1], Joe Dura[1], Frank Heinrich[1,2], Brian Maranville[1], Paul A. Kienzle[1] and Charles F. Majkrzak[1]; [1]National Institute of Standards and Technology, United States; [2]Carnegie Mellon University, United States

Liquid cells are a common sample environment for neutron reflectometry experiments and are critical for measuring the properties of materials at solid/liquid interfaces. Background scattering determines the maximum useful scattering vector, and hence the spatial resolution, of the neutron reflectometry measurement. The primary sources of background are the liquid in the cell reservoir and the materials forming the liquid cell itself. Thus, characterization and mitigation of these background sources are necessary for improvements in the signal to background ratio and resolution of neutron reflectometry measurements employing liquid cells. Here we demonstrate two recent advances in reduction of background from liquid cells. First, we measure and calculate the background field generated by scattering from a thin liquid reservoir. We extensively characterize the scattering arising from each of the cell materials and show that fully encasing the liquid reservoir in single crystal silicon and minimizing the liquid reservoir thickness yields the lowest background scattering. We also show that background subtraction utilizing the entire calculated background field improves data modeling and reduces experimental uncertainties associated with localized background subtraction. Second, we show that inelastic scattering from the liquid reservoir and single crystal silicon at high scattering vectors represents a large fraction of the background signal and adds spurious scattering to the NR measurement. A significant improvement in the measurement quality is thus achieved by using energy-analyzed detection. Combining thin liquid reservoirs with energy analyzed detection and the high flux of the CANDOR polychromatic reflectometer at the NIST Center for Neutron Research, a background-subtracted neutron reflectivity smaller than 10^{-8} from a liquid cell sample is reported in a reasonable measurement time.

SESSION DP3.04: Poster Session: Biology, Biophysics and Biotechnology
Session Chairs: Elizabeth Kelley and Amy Xu
Wednesday Afternoon, June 8, 2022
5:30 PM - 7:30 PM
UMC Center Ballroom 210

DP3.04.01
Effect of Cholesterol on the Elastic and Viscous Properties of Saturated Lipid Bilayers – A Neutron Spin Echo Study Kuo-Chih Shih[1], Elizabeth Kelley[2], Paul D. Butler[2], Norman Wagner[1] and Michihiro Nagao[2,3,1]; [1]University of Delaware, United States; [2]National Institute of Standards and Technology, United States; [3]University of Maryland, United States

Cholesterol (Chol) is an important molecule in cell membranes and is found at concentrations ranging from <5% (mole %) in mitochondrial membranes to upwards of 40% in the plasma membrane of eukaryotic cells. In addition, Chol is a primary regulator of membrane properties and affects the lipid membrane phase behavior, structure, elasticity and viscosity. A detailed phase diagram of 1,2-dimyristoyl-sn-glycerol-3-phosphocholine (DMPC)/Chol mixture based on electron spin resonance (ESR) experiments was proposed in a previous study by Almeida et al. [1] We know from the phase diagram that

at temperatures higher than the melting transition temperature (T_m) of DMPC, with increasing Chol mole concentration, DMPC undergoes phase transitions from a liquid disordered phase (L_d) to a liquid ordered phase (L_o) with a phase coexistence region (L_d+L_o) in between. The aim of the present study is to understand the physical properties of model DMPC/Chol mixtures in the L_d and L_d+L_o phases.
In this study, we utilize small-angle neutron scattering (SANS) as well as neutron spin echo (NSE) to access the static and dynamic properties of DMPC membranes containing up to 20% Chol over a temperature range from 25 °C to 55 °C. The studied temperature and Chol concentration ranges cross the phase boundary between the L_d and L_d+L_o phases. First, densitometry measurements were carried out to determine the molecular volume of the DMPC/Chol mixtures as well as the phase boundaries as we used tail-deuterated DMPC (d_{54}-DMPC) molecules for our neutron experiments. SANS data were fitted by a smeared 6-slab model by implementing the molecular volume obtained from densitometry to extract the structural parameters. By adopting the structural information from the SANS measurements to the NSE data analysis, the NSE results also show that the effective bending modulus as well as membrane viscosity in the L_d phase scale with the area per lipid as proposed in our previous studies of mixed lipid membranes. [2]

Reference
[1] Almeida, P. F.; Vaz, W. L.; Thompson, T., Lateral diffusion in the liquid phases of dimyristoylphosphatidylcholine/cholesterol lipid bilayers: a free volume analysis. *Biochemistry* **1992,** *31* (29), 6739-6747.
[2] Kelley, E. G.; Butler, P. D.; Ashkar, R.; Bradbury, R.; Nagao, M., Scaling relationships for the elastic moduli and viscosity of mixed lipid membranes. *Proceedings of the National Academy of Sciences* **2020,** *117* (38), 23365-23373.

DP3.04.02
Effects of 3-DeEhydroshikimate Dehydratase Expression Levels in the Organization of Cellulose Microfibrils in Poplar Mutants Manjula P. Senanayake Mudiyanselage[1], Chien-Yuan Lin[2], Aymerick Eudes[2], Hugh O'Neill[1] and Sai Venkatesh Pingali[1]; [1]Oak Ridge National Laboratory, United States; [2]Joint Bioenergy Institute, United States

Lignocellulosic biomass is of particular interest as a sustainable, carbon dioxide neutral, and non-competitive source of sugars and platform chemicals for conversion into renewable fuels and advanced byproducts. Using traditional methods, converting biomass to bioenergy has proved costly and energy demanding. Different biomass deconstruction approaches have been employed, and the strong binding between lignin and carbohydrate polymers resulted in lignin recalcitrance issues that severely hindered improvement in the bioconversion process. In order to reduce lignin recalcitrance, transgenic poplar trees with lower lignin content have been developed by overexpressing the bacterial 3-dehydroshikimate dehydratase (QsuB) enzyme. Three different transgenic tree lines with reduced lignin content were compared to the wildtype (WT), and their level of QsuB expression varied as QsuB1>QsuB15>QsuB5>WT. Conversion of biomass into sugars follows the same order, which agrees with increasing the biomass conversion efficiency. A biochemical study determined 15% less lignin and 13% higher hemicellulose content in the Q1 and Q15 lines than Q5 and WT. Small-angle neutron scattering (SANS) and wide-angle X-ray scattering (WAXS) were used to study the effects of QsuB expression on the structure and morphology of the cellulose microfibril ordering. SANS data were modeled by cylindrical form factor coupled with unified fit to calculate the dimension of the cellulose microfibrils. The fitting results illustrated the cellulose microfibril cross-sectional sizes are not significantly different in mutants and the WT. However, the distance between neighboring cellulose microfibrils progressively increased with increasing QsuB expression levels, and the highest expressing line QsuB1 showed no regular order. WAXS data agrees with decreasing crystallinity for increasing QsuB overexpression and showed the presence of a mixture of cellulose I and II in the mutants. Together, comprehensive knowledge in the relation between QsuB overexpression levels and cellulose microfibril size and order in poplar stems was achieved, paving the way to develop a science-driven approach to tune poplar stems' characteristics from overcoming recalcitrance and enhancing biomass conversion efficiency.

DP3.04.03
Calcium Carbonate Polymorphism and Microstructure in Fish Otoliths Bryan Chakoumakos[1], Brenda M. Pracheil[1], R. Seth Wood[2], Alison Loeppky[3], Kassandra M. Merks[4] and W. G. Anderson[4]; [1]Oak Ridge National Laboratory, United States; [2]Washington University in St. Louis, United States; [3]North/South Consultants Inc, Canada; [4]University of Manitoba, Canada

Fish otoliths, or “ear bones”, are comprised of one or more of the $CaCO_3$ polymorphs (aragonite, calcite and vaterite). Otoliths are part of a fish's auditory system and act as accelarometers to determine and control the fish's orientation and motion. Otoliths are frequently used in environmental studies to infer temporally-explicit environmental conditions or fish life history events such as migrations based on concentrations of trace elements within their daily-to-annual growth rings. Proper characterization of the mineralogical composition and microstructure of fish otoliths feeds into the interpretation of trace element chemistry. The polymorph phase abundance in an otolith depends on, as yet, unexplained genetic and environmental factors, particularly when multiple polymorphs are present. Most fish otoliths are comprised of the densest $CaCO_3$ polymorph, aragonite, and concentric growth rings are the expected microstructure, but these notions are potentially outdated with many recent reports showing otherwise. We have been employing a variety of materials science methods (Polarized Light Microscopy, X-ray Diffraction, Neutron Diffraction, X-ray Tomography, Raman Spectroscopy, Neutron Vibrational Spectroscopy, Synchrotron XRF, Gas Pycnometry, and Crystal Growth Screens) to characterize the $CaCO_3$ polymorph distributions and microstructures of fish otoliths to develop a basis for understanding their ontogenetic, environmental, phylogenetic controls. Our work suggests examining the $CaCO_3$ polymorph composition of otoliths should become more common, especially in studies where results may inform fisheries management decisions. Future research should work to attribute controls on otolith $CaCO_3$ polymorph expression using a combination of omics and material science approaches to improve life history and environmental inferences obtained from otoliths. Research conducted at ORNL's High Flux Isotope Reactor and Spallation Neutron Source were sponsored by the Scientific User Facilities Division, Office of Basic Energy Sciences, U.S. Department of Energy.

Structural Materials and Engineering

* Invited Paper

2:00 PM *F1.01.01
Determining Residual Stress in Weldments and Additively Manufactured Parts by Neutron Diffraction Hamid Eisazadeh; Old Dominion University, United States

Due to rapid localized heating and subsequent cooling during heating and cooling, highly non-uniform temperature distribution occurs across the weldment and metal additive manufactured parts, resulting in the formation of significant residual stress (RS). Difficult to measure or to estimate theoretically, RS remains one of the largest unknowns in industrial damage situations. The deep penetration capability of neutrons into most metallic materials makes neutron diffraction a unique and powerful tool in understanding the material structures and properties. We review the challenges and opportunities that we are facing in measuring residual strain in welding and additive manufactured parts.

2:30 PM F1.01.02
Application of Neutron Grating Interferometry in Metal Additive Manufacturing Youngju Kim[1], Caitlyn M. Wolf[2], Sarah M. Robinson[2], Michael Cyrus Daugherty[2], Jacob M. LaManna[2], David Jacobson[2], Eli Baltic[2], Paul A. Kienzle[2], Katie M. Weigandt[2], Nikolai N. Klimov[2], Michael G. Huber[2], Peter N. Bajcsy[2], Ryan P. Murphy[2], Jongyul Kim[3], Wook Jin Lee[4], Seung Wook Lee[4] and Daniel S. Hussey[2]; [1]University of Maryland, United States; [2]National Institute of Standards and Technology, United States; [3]Korea Atomic Energy Research Institute, Korea (the Republic of); [4]Pusan National University, Korea (the Republic of)

Neutron grating interferometry (nGI) is a unique, nondestructive analysis technique for materials research, providing three forms of image contrast, namely transmission, differential phase, and dark-field images. Most notably, the dark-field image contrast is generated by the small-angle neutron scattering (SANS) of material structure on a length scale, represented by the autocorrelation length (ξ) of the nGI [1]. ξ is an nGI system parameter, which is proportional to the product of the neutron wavelength and the sample-detector distance divided by the interference fringe period. Acquiring dark-field images over a broad range of ξ allows one to map out the pair correlation function and quantitatively assess the structural parameters of the microstructure such as size, concentration, etc. In this study, we report on the use of nGI to ascertain the microstructural properties of steel samples produced by metal additive manufacturing (MAM). The dark-field images generated by nGI on our MAM samples were shown to be sensitive to the microstructure (defects, void, etc). Specifically, the selective laser melted (SLM) SS316 tensile specimens were prepared, and tensile strain was applied perpendicular or parallel to the build direction at three different tensile levels of 0 %, 75 %, and 100 % for fracture. The SS316 compression rods manufactured by SLM and directed energy deposition (DED) methods were prepared with four compression levels of 10 %, 20 %, 30 %, and 40 % of the initial length of the rod. Two types of nGI setups were used, Far-field interferometer (FFI) and Talbot-Lau interferometer (TLI). The microstructure behavior according to the mechanical deformation of tension and compression that was observed by nGIs can represent the mechanical properties and further predict features such as deformation, fracture, etc. We will present the nGI analysis of these MAM samples to demonstrate how the method can be used to explore hierarchical structures.

2:45 PM F1.01.03
Stresses Due to Friction Stir Weld Repair of Simulated Cracks in 304L Stainless Steel Plates Thomas Gnaupel-Herold[1], Madhumanti Bhattacharyya[2], Indrajit Charit[2], Krishnan Raja[2], Jens Darsell[3] and Saumyadeep Jana[3]; [1]NIST, United States; [2]University of Idaho, United States; [3]PNNL, United States

In order to investigate the stress effects of crack repair by friction stir welding in 304L stainless steel narrow notches were produced by electrical discharge machining to a depth of 0.5x thickness (5 mm /10 mm). The notches were subsequently 'healed' by friction stir welding with different weld temperatures. Residual stresses were mapped using both X-ray diffraction (direct surface and layer removal) and neutron diffraction for detailed depth profiles in both base metal and weld metal. Due to the sample being partially joined prior to welding the stresses exhibit differences to what has been previously shown in the literature, including a pronounced depth dependence as well as dependence on the weld temperature [1].

References
[1] M. Bhattacharyya et al., Materials Science & Engineering A 826 (2021) 141982

3:00 PM F1.01.04
Implications of Machining on Residual Stresses and Mechanical Properties of 316L Walls fabricated via Hybrid Additive Manufacturing Christopher Fancher, Rangasayee Kannan, Kyle Saleeby, Thomas Feldhausen and Peeyush Nandwana; Oak Ridge National Laboratory, United States

Additive Manufacturing (AM), often called 3D printing, offers the unique opportunity to transform how materials are consolidated into bulk 3D structures, with powder bed fusion and directed energy deposition (DED) becoming two prominent techniques for metal AM. The localized melt strategies used in metal AM introduce non-equilibrium thermal conditions that can enable the fabrication of bulk components from difficult-to-process materials. AM parts are often heat-treated to relieve residual stresses and then machined to achieve the required tolerances. Hybrid AM was developed to streamline the process for producing bulk parts by combining DED AM with 5-axis computer numerical controlled machining (CNC). Optimizing the build/machining schedule can reduce the total build time from 20 hrs (4 hr AM and 16 hr CNC) to 6.5 (4 hr AM and 2.5 hr CNC). The reduction in build time is achieved by shifting the machining from occurring after the AM process to an interrupted schedule where machining occurs after predefined intervals. Neutron residual stress mapping was performed at the High-Intensity Diffractometer for Residual Stress Analysis at the High Flux Isotope Reactor to determine the implication of the machining schedule on the bulk residual stresses in as processed 316L wall test coupons. Residual stress data evidence that the machining schedule dramatically affects the underlying stresses that evolve during the hybrid processing. For example, performing the CNC setup without coolant shifts the strain near the base plate from compressive to tensile. This presentation will discuss the implication of the hybrid machining schedule on the residual stress evolution and mechanical properties of hybrid AM fabricated 316L components.

FP1.01.01
Minimizing Helium Pressure Inhomogeneities Across Large Samples at Low Temperatures Juscelino B. Leao; National Institute of Standard and Technology, United States

In-situ inert gas high pressure vessels for neutron scattering for pressures of up to 1.0 GPa and temperatures as low as 1.5 K pose a particular problem due to the P x T phase diagram of the pressure media. Passing thru the P x T of helium to achieve low temperatures (1.5 K ≤ T < 30 K), when using pressure media to achieve hydrostatic pressure under constant pressure and volume, will cause an overall pressure reduction of at most 25 percent of the pressure read at the hydrostatic conditions temperature. Here it is discussed a methodology for pressurization to ensure minimal pressure loss as temperature falls below the pressure media phase change. The reduction in pressure inhomogeneity throughout the length of the sample; as well as calibration data are presented within this work.

FP1.01.02
Development of Quantitative Texture Analysis Routines at the WAND² and HIDRA Nate Peterson[1], Christopher Fancher[2], Matthias D. Frontzek[2], Jeffrey Bunn[2], Edward A. Payzant[2], Ke An[2] and S. Agnew[1]; [1]University of Virginia, United States; [2]Oak Ridge National Laboratory, United States

Data collection and analysis strategies have been developed for efficient and reliable crystallographic texture measurements at two recently upgraded neutron diffractometers: the Wide Angle Neutron Diffractometer Squared (WAND²) and the High Intensity Diffractometer for Residual Stress Analysis (HIDRA) at the High Flux Isotope Reactor (HFIR) located at Oak Ridge National Laboratory (ORNL). These methods are demonstrated using measurements on a variety of textured samples, including multi-phase steel composites and polycrystalline calcite ($CaCO_3$). Reference measurements were also made at Vulcan, the engineering diffractometer located at the Spallation Neutron Source (SNS). The texture data obtained on the different instruments agree with each other, and WAND² is more *time*-efficient than HIDRA. Two analysis methods were investigated, single peak fitting to obtain individual pole figures for inversion and Rietveld texture analysis using MAUD. The impact of the variances between the various textures obtained was evaluated through the calculation of diffraction elastic constants, which is one application of the collected texture data. Both instruments were found to provide texture data which is suitable for complimenting other analyses, such as residual stress mapping. These measurements also provide the ORNL contribution to an ongoing neutron diffraction texture measurement round robin study, which has participants from the other major neutron sources around the world (LANSCE, NCNR, ISIS, J-PARC, FRM-II, JINR).

SESSION F3.02: Using Neutrons for Large Scale Engineering Applications
Session Chair: Jeffrey Bunn
Wednesday Afternoon, June 8, 2022
UMC Aspen Room 285, 287, 289

2:00 PM *F1.02.01
Application of Neutron Diffraction for Industrial Materials & Manufacturing Development Shenyan Huang; GE Research, United States

Neutron scattering techniques have been increasingly adopted by industries to solve engineering problems or understand structure-property relationships. This talk will introduce how neutron diffraction has provided unique values in the industrial research and development of advanced structural materials and manufacturing at GE Research. A few cases of ex-situ or in-situ neutron diffraction studies coupled with heating or loading will be presented, including high temperature behavior of single crystal or directionally solidified superalloys, deformation of thin-wall FeCrAl accident tolerant fuel cladding tubes, residual stress mapping of functionally graded material manufactured by direct energy deposition. Industrial perspective of future research interests and new capability needs at neutron facilities will be discussed.

2:30 PM F1.02.02
Effects of Mechanical Deformation on Dislocation Density, Phase Separation and Hydrogen Diffusion in Pipeline Steel Zachary Buck[1], Matthew Connolly[1], May Martin[1], Damian Lauria[1], Peter Bradley[1], Andrew Slifka[1], Ke An[2], Yan Chen[2] and Naresh C. Osti[2]; [1]National Institute of Standards and Technology, United States; [2]Oak Ridge National Laboratory, United States

The effects of hydrogen embrittlement in metals, particularly ferritic steels, has remained a persistent obstacle in designing and manufacturing reliable structural materials for use in energy storage and transportation applications. Pipeline steels, such as those used to transport natural gas, are particularly susceptible to embrittlement. Therefore, understanding mechanisms promoting the embrittlement process, including how mechanical forces induce changes to the microstructure and hydrogen diffusion within these materials, is of great importance. Our investigation into this phenomenon begins with interrupted uniaxial tensile tests performed at the National Institute of Standards and Technology – Boulder on AISI 4130 steel under ambient conditions and high-purity hydrogen environments ranging in pressures from 260 psi to 2600 psi. Results from neutron diffraction measurements collected using beamline #7 (VULCAN) at ORNL on samples mechanically deformed in air reveal a partitioning of ferrite and martensite phases as a function of applied strain. Dislocation densities of the two individual phases were extracted by analyzing the broadening of their Bragg peaks using a Williamson-Hall approach. In addition, a phase transformation from predominantly martensitic steel (at little to no applied strain/stress) to a ferritic steel (beyond the ultimate tensile strength) was observed for the in-air samples. Measurements are currently in progress on samples strained under various hydrogen pressures to determine its effects on the formation of dislocations and phase partitioning. These results demonstrate our ability to fabricate samples with the desired microstructure for upcoming quasielastic neutron scattering (QENS) measurements aimed at quantifying hydrogen diffusion as a function of dislocation density.

2:45 PM F1.02.03
Damage Modes in Hydrogen-Assisted Fatigue Probed by Neutron and X-Ray Scattering Matthew Connolly[1], Zack Buck[1], May Martin[1], Robert Amaro[2], Peter Bradley[1], Damian Lauria[1], Jun-Sang Park[3] and Andrew Slifka[1]; [1]National Institute of Standards and Technology, United States; [2]AMTT, United States; [3]Argonne National Laboratory, United States

Elucidating the mechanisms of hydrogen embrittlement of steels is complicated by the fact that multiple mechanisms may be activated at once or may even require a synergistic co-existence for activation. Some leading preposed mechanisms of hydrogen embrittlement include hydrogen-enhanced decohesion (HEDE), the hydrogen-enhanced localized plasticity (HELP) mechanism, and the Nano-Void Coalescence Mechanism (NVC). At their core, these mechanisms differ either in the way in which damage manifests when a material is strained, or in the maximum amount of damage the material can withstand prior to fracture. Specifically, in HEDE, accumulation of hydrogen at locations of high triaxial stresses lead to the weakening of Fe-Fe bonds, reducing the amount of plastic deformation by intra-granular separation a material can withstand; in HELP, the introduction of hydrogen gas creates areas

of extended dislocations in the Fe lattice and enhances plastic deformation for a given applied load; in NVC, hydrogen is predicted to lead to the stabilization and promotion of vacancy ("nano-scale void") agglomeration, leading to plastic deformation by void coalescence and growth. It is critical to probe all three to assess which mechanisms are dominant under typical use conditions. Here we present neutron Transmission Bragg Edge, High Energy X-ray Diffraction (HEXRD), and Small-Angle X-ray Scattering (SAXS) measurements during fatiguing of a steel crack. Neutron Transmission Bragg Edge probe HEDE through a determination of crack-tip strain fields; HEXRD measurements probe HELP through measurement of dislocation density; SAXS measurements probe NVC through a determination of nano-pore size distribution. We will present strain, dislocation density, and pore size distributions maps ahead of crack tips grown in air and in hydrogen. We will discuss the differences in each between the crack tips grown in air and in hydrogen in the context of the HELP, HEDE, and NVC mechanisms.

3:00 PM F1.02.04
Elevated Temperature Dislocation Density Reductions in Cold-Worked ASTM A586 High-Strength Steel Wire Jumari A. Robinson, Adrian Brugger and Raimondo Betti; Columbia University, United States

As much of the nation's infrastructure surpasses its originally intended service life, the importance of quantifying robustness towards natural and anthropogenic hazards grows. For suspension bridges, the two to four irreplaceable main cables (which carry the entire weight of the bridge deck) are particularly susceptible to damage from vehicular fires on the roadway at mid-span and anchorage. These cables are composed of thousands of high-strength, cold drawn ASTM A586 steel wires, which have been shown to experience much higher strength reductions at and after exposure to elevated temperatures than the mild steels typically used in construction. This study seeks qualitative and quantitative understanding of one of the many metallurgical factors attributing to these permanent changes in stress-strain behavior at elevated temperatures. Evolving dislocation density, as evidenced by changing peak-widths at elevated temperature, was examined for wires heated ex-situ (SMARTS diffractometer at Los Alamos National Laboratory) and in-situ (VULCAN engineering materials diffractometer at Oak Ridge National Laboratory). Results from both experiments show significant peak-narrowing in the diffraction spectra of both in-situ and ex-situ heated wires, qualitative indication of permanent decreases in dislocation density. Dislocation line profile analysis of the data confirms quantitatively that significant reductions in dislocation density occur within the wire at temperatures ranging from 400°C to 700°C – shedding angstrom-scale light on the observed macroscale reductions in yield stress at and after exposure to elevated temperatures.

Emerging Applications of Neutron Scattering in Engineering, Arts and Sciences

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* Invited Paper

SESSION H2.01: Emerging ML Applications—Neutrons and Beyond
Session Chairs: Tyler Martin and Alan Tennant
Tuesday Morning, June 7, 2022
UMC Aspen Room 285, 287, 289

10:15 AM *H2.01.01
Machine Learning: A Data-Driven Spectrometer for Neutron Scattering Mingda Li; Massachusetts Institute of Technology, United States

Neutron scattering has made remarkable experimental progress in the past decades, but understanding the microscopic interaction mechanisms of quantum material from neutron data remains challenging. With ever-increasing neutron scattering data, machine learning brings new hope and can serve as a new probe to perform neutron scattering data analysis. In this colloquium, I will introduce how machine learning can be used to reveal the hidden information in neutron scattering data and elucidate the quantum materials. We first introduce an improved identification of the proximity effect from neutron reflectometry, an effect that can lead to dissipationless spintronics or topological quantum computing. We then introduce the symmetry-preserved neural network and the capability to predict phonon density-of-states for vibrational spectroscopies. Finally, we will introduce our recent work aiming to predict ground-state magnetic orderings from neutron diffraction. We highlight the importance of the representations and envision a few scattering problems that can benefit from machine learning.

10:45 AM *H2.01.02
Accelerate Discovery of New Chemical Synthesis Pathways using Autonomous Experiments Combined with AI Agents Kristin Schmidt, Dmitry Zubarev, Sarath swaminathan, Renato Fontoura de Gusmao Cerqueira, Nathaniel Park, Tim Erdmann, Daniel Sanders and Jed Pitera; IBM Research, United States

It can take over 10 years to develop new materials. At IBM Research, we're looking to accelerate the discovery process by developing cutting-edge technologies, demonstrating scalable processes, and deploying new models of collaborative innovation. Our goal is to unlock new properties, materials and processes to address global challenges in computing, health and sustainability. We will show our end-to-end AI-powered workflow on the discovery of more sustainable, viable photoacid generators which are important materials in the semiconductor industry that became the subject of regulatory scrutiny. We will specifically highlight how AI agents and autonomous experiments speed up the design of chemical synthetic pathways.

11:15 AM *H2.01.03
Autonomous Control at X-Ray Sources from Accelerator to Detector Daniel Ratner; SLAC, United States

As the dimensionality and resolution of photon science experiments increases, it is no longer tenable for users to simply scan parameters during a beam time. Similarly on the machine side, the large number of control parameters needed to optimize advanced operating modes precludes an exhaustive search

during setup. Instead “smart” scans are needed for both, adaptively probing the sample to focus on the highest value measurements or searching through parameter space to find the most likely regions to optimize performance. I will show examples from both the accelerator and user perspectives, using a variety of methods including Bayesian optimization and reinforcement learning.

SESSION H2.02: Emerging ML Applications—Diffraction to Dynamics
Session Chair: Alan Tennant
Tuesday Afternoon, June 7, 2022
UMC West Ballroom 208

2:00 PM H2.02.01
Using AI to Determine Space Group from Neutron Powder Diffraction Data William Ratcliff[1,2], Haotong Liang[2], Aaron G. Kusne[1,2] and Ichiro Takeuchi[2]; [1]NIST, United States; [2]University of Maryland, United States

We use deep learning to determine the space group of materials based on neutron powder diffraction measurements. Our approach utilizes sem-supervised learning to take advantage of the large volumes of unlabled data available at facilities. Our method outperforms previous deep learning approaches, using less data, covering 144 space groups (limited by the representation of structures within the ICSD database).

[1] "A Semi-Supervised Approach for Automatic Crystal Structure Classification", Satvik Lolla, Haotong Liang, A> Gilad Kusne, Ichiro Takeuchi, William Ratcliff, arXiv:2111.01287 (2021)

2:15 PM H2.02.02
Super-resolution Dispersions Measured by Direct Geometry Spectrometers Jiao Lin[1], Gabriele Sala[1], Matthew Stone[2] and Andrew D. Christianson[2]; [1]Oak Ridge National Lab, United States; [2]Oak Ridge National Laboratory, United States

Direct-geometry time-of-flight chopper neutron spectroscopy is instrumental in studying excitations in single crystal systems. Neutron scattering from these excitations are characterized by the 4-dimensional (4D) scattering function in reciprocal space, S(Q, E), where Q is the momentum transfer vector and E the energy transfer. In recent years the use of highly pixelated detector arrays in conjunction with direct-geometry chopper spectrometer (DGS) instruments has allowed for efficient measurements of 4D S(Q, E) functions over large ranges of Q and E. Typically 2D slices are extracted from these 4D datasets, with energy transfer as the vertical axis and a high symmetry direction in momentum transfer as the horizontal axis. Dispersion curves are then obtained from these slices. We report here that real-space techniques in optical imagery can be adapted to obtain super resolution dispersion curves for phonon or magnetic excitations from single-crystal neutron spectroscopy measurements. The procedure to reconstruct super-resolution energy dispersion of excitations relies on accurate determination of the momentum and energy-dependent point spread function, and a dispersion correction technique inspired by an image disparity calculation technique commonly used in stereo imaging. Applying these methods to spinwave dispersion data demonstrates ~5-fold improvement over nominal energy resolution.

2:30 PM H2.02.03
A Method for Determining Mid-Range Order in Semi-Crystalline Materials using Inelastic Neutron Scattering and Density Functional Theory Makena Dettmann[1], Lucas S. Cavalcante[1], John Anthony[2] and Adam Moule[1]; [1]University of California, Davis, United States; [2]University of Kentucky, United States

Atomic vibrations, or phonon modes, impact several material properties ranging from charge mobility in organic semiconductors to correlated defects in metal-organic frameworks. One of the best tools to measure these phonon modes is inelastic neutron scattering because it gives a weighted vibrational density of states for a massive energy range (1-1000 meV). However, this also means that spectra can be difficult to interpret for complex materials. For example, semi-crystalline molecular solids will have short-range order but often contain multiple molecular configurations within a single sample. In this study, we demonstrate a method to accurately model complex, semi-crystalline materials using multiple DFT simulations. We then apply this method to semi-crystalline BDT trimers. We further demonstrate the ability to use this method to describe the types of mid-range disorder present in the sample and even tie regions of the spectrum to areas of the molecule. This method offers a powerful tool to explore more complex systems than previously available with traditional modeling techniques.

2:45 PM H2.02.04
Machine Learning for Sample Alignment in Neutron Scattering Experiments Abdourahmane Diaw[1], Kevin Bruhwiler[1], Chris Hall[1], Jonathan Edelen[1], Stuart Calder[2] and Christina Hoffmann[2]; [1]RadiaSoft LLC, United States; [2]Oak Ridge National Laboratory,, United States

Access to neutron scattering facilities, such as Oak Ridge National Laboratory (ORNL) and the NIST Center for Neutron Research (NCNR), provides researchers with instruments to investigate various scientific and technologically important problems in physics, chemistry, biology, and engineering. In these experiments, the quality of collected data is susceptible to sample positioning, requiring sample alignment in the beam and stabilization if sample environment conditions change in the experiment. This process has relied on human intervention to tune the sample position and beam characteristics, which is often tedious, imprecise, and time-consuming. Due to the irregular sample shapes, varying mount geometries, and a high amount of noise in the case of neutron camera data, robust identification of the sample center is a fundamental challenge for the automation of sample alignment. Here, we bring experiment and convolutional neural to a nexus for solving this sample alignment problem, improving the accuracy and speed of sample alignment. We introduce an image segmentation approach based on the U-Net algorithm to automatically identify samples from neutron beamlines images. We demonstrate the adaptability of the approach on optical images from the TOPAZ beamline and neutron camera images from the HB-2A beamline, both at ORNL.

SESSION H3.03: Emerging ML Applications—Soft Matter and Chemistry
Session Chair: Tyler Martin
Wednesday Afternoon, June 8, 2022
UMC West Ballroom 208

4:00 PM H3.03.01
Machine Learning for Neutron Reflectometry Mathieu Doucet, William Heller and Richard Archibald; Oak Ridge National Laboratory, United States

Machine learning has the potential to help identify plausible models for neutron scattering data, greatly helping users. Such a tool would be of great benefit for neutron reflectometry, where finding an initial thin film structure can be a time-consuming endeavor for novice users. We report on the use of a neural network to predict a two-layer thin film structure from measured reflectometry data. The predictions were found to be stable against statistical fluctuations of the data and an excellent starting point for final refinement. The approach was applied to data measured at the Magnetism Reflectometer at SNS and found to be very close to results obtained with traditional methods. We will also discuss where such approaches may realistically be applied at reflectometers.

4:15 PM H3.03.02
Davis Computational Spectroscopy workflow - from structure to spectra Lucas Samir Ramalho Cavalcante, Makena Dettmann, Ambarish Kulkarni and Adam Moule; UC Davis, Brazil

Metal-organic frameworks (MOF) hold great promise in applications on gas adsorption, catalysis and supercapacitors because of their porous structure and modularity. Despite having a symmetrical and apparently well-organized structure formed by metal clusters connected via organic linkers, these materials have low thermal, mechanical and chemical stability, yielding a high defect density and disorder. To overcome the stability problem, a zirconium-based MOF called UiO-66 was presented as a solution due to its high connectivity with 12 connected clusters in the face-centered-cubic topology. However, a better understanding of the nature of defects and disorder in UiO-66 MOFs is still required.

Inelastic Neutron Scattering (INS) has been proven to be a good ally in the investigation of structural and dynamic disorder but it requires a detailed modeling of the system in order to characterize peak positions and intensities, and subsequently materials properties. The high computational cost of the electronic modeling has limited investigations with INS to crystalline materials, dampening the study of disordered large systems such as MOFs.

To address the trade-off between simulation cost and accuracy, we developed an automated workflow that connects various atomic simulation tools in order to investigate the relationship between material properties, lattice dynamics, and INS spectra. This workflow allows an accurate and efficient method of calculating phonon modes and the INS spectrum with the use of a broad range of quantum mechanical approximations, including density functional theory (DFT) and density functional tight-binding (DFTB). We have also implemented a machine-learned force field based on Chebyshev polynomials (Chebyshev Interaction Model for Efficient Simulation - ChIMES) to improve the accuracy of the DFTB simulations with ~100x reduction in computational expense while retaining most of the accuracy of DFT. Besides the benefits of a tool that automates the simulation and consequent analysis of the INS spectrum, our efforts expand the possibilities of investigating more complex structures that would be unfeasible with ab initio methods.

4:30 PM H3.03.03
Machine Learning-Enabled Inverse Analysis of Small Angle Scattering Data Graham W. Roberts, Mu-Ping Nieh, Anson Ma and Qian Yang; University of Connecticut, United States

We propose a framework for utilizing machine learning-based models to derive information from scattering data that human users cannot currently determine quickly without the help of computationally expensive data analysis. In addition to identifying the morphology, the models also inform the quantitative value of structural parameters without requiring time-consuming optimization procedures. Leveraging physical knowledge from the known forward models, we have developed novel algorithms that are designed to accurately extrapolate to different structural parameter ranges, unlike traditional machine learning models which are designed to be predictive over the same distribution as the training data. In this presentation, we will discuss how to curate a simulated dataset for training our machine learning models to enable high quality predictions on experimental data. The machine learning software we developed can be readily integrated with existing scattering analysis software such as SasView and deployed at scattering facilities. Our machine learning framework will help enable real-time data analysis for experiments, significantly increase the efficiency of expert and facility time, and democratize scattering research, making it more accessible to the broader scientific community.

4:45 PM H3.03.04
Machine Learning Augmented Computational Reverse-Engineering Analysis for Scattering Experiments of Assembled Mixtures of Nanoparticles Christian Heil and Arthi Jayaraman; University of Delaware, United States

Nanoparticle assembly is a common route to generate materials with specific properties. The assembled material must undergo characterization at multiple length scales to link the structural features with the macroscopic properties. Small angle scattering (SAS) is a useful method to characterize nanoparticles' assembled structure. The output of SAS experiments is the averaged intensity at various wave vectors, $I(q)$ vs. q, whose interpretation often relies on fitting with analytical models. This model selection can be a limitation when appropriate analytical models do not exist. We present an open-source computational method, CREASE, to analyze the scattering results from spherical nanoparticle mixtures assembled in various confinement geometries. We test the strengths and limitations of our method by using a variety of *in silico* $I(q)$ obtained from simulations of binary nanoparticle assemblies and nanoparticle solutions with varying mixture composition/concentration, nanoparticle size distribution, and degree of mixing/aggregation. Furthermore, the developed machine learning model, linking features of the nanoparticle solutions (e.g., concentration, nanoparticles' tendency to aggregate) to computed scattering profile, is generic and applicable to different nanoparticle sizes without the need for additional data to retrain the model to be specific to the size of interest. We will also present extensions of this method for systems (e.g., concentrated solution of micelles) where the form factor of the particle (i.e., micelle) is unknown, and CREASE analyzes both structure and form factor in the assembled mixture.

5:00 PM H3.03.05
Designing an Active Learning Agent for Autonomous Small-Angle Scattering Tyler B. Martin, Aaron G. Kusne, Austin McDannald and Peter A. Beaucage; National Institute of Standards and Technology, United States

Most neutron scattering experiments consist of simple grid searches through composition, temperature, or applied field space. The use of these simple approaches is driven by a scarcity of neutron measurement time, a lack of well-tagged data sets, and the challenge of adapting intelligent search methods to neutron scattering. Driven by the needs of the Autonomous Formulation Lab (AFL), we have developed a virtual instrument for generating synthetic data and a challenge course of phase mapping problems for benchmarking active learning agents. The virtual instrument combines the ranges, resolutions, and uncertainties from real SANS data with models from the SasView package. The challenge course is designed to test many common factors in neutron scattering data including low signal to noise, high incoherent background, second-order phase boundaries, and kinetically trapped interphases. We also benchmark our agent against other intelligent and non-intelligent search approaches such as random grid search or multi-dimensional integration. Here, we

will discuss how this virtual approach has shaped the development of our active learning agent and highlight design rules for tailoring agents to bespoke measurement challenges.

SESSION HP3.04: Poster Session: Emerging Applications in Neutron Scattering—Machine Learning and Data Science
Session Chairs: Tyler Martin and Alan Tennant
Wednesday Afternoon, June 8, 2022
5:30 PM - 7:30 PM
UMC Center Ballroom 210

HP3.04.01
Measuring Coefficients of Spin-Spin Correlation Functions on Quantum Hardware Norhan M. Eassa[1], Zoe Holmes[2], Jeffrey Cohn[3], Andrew T. Sornborger[2], Joe Gibbs[2], Gavin L. Hester[1], Paul Kairys[4], Bilal Khalid[1] and Arnab Banerjee[1]; [1]Purdue University, United States; [2]Los Alamos National Laboratory, United States; [3]IBM Almaden Research Center, United States; [4]Oak Ridge National Laboratory, United States

Quantum spin systems can demonstrate a variety of interesting quantum phenomena, ranging from soliton lattices to quantum spin liquids, with a potential to constitute key elements in various quantum applications. Inelastic scattering experiments, such as using inelastic neutron scattering (INS) and NMR, provide key insights into the dynamics of spins inside quantum magnets. To understand such spin dynamics, we compute the magnetic neutron cross-section in a qubit-based hardware extending the prescription laid in Nature Phys. 15, 455 (2019). We have utilized the IBM quantum devices to simulate the time evolution of different Hamiltonians acting on an initial state for spin-½ systems, starting from 2-spin systems and extending above, and have been able to measure the coefficients of the correlation functions. In order to mitigate the backend noise and to capture longer time dynamics, we use a fast-forwarding ansatz, and compare the results to that of trotterization, as well as compare between direct (ancilla qubit-free circuits) and indirect (circuits including ancilla qubit) measurements. These results will be discussed in the context of the existing literature on spin dimer models and their neutron scattering studies. We end with ongoing ideas to expand to larger spin networks to emulate scattering results from INS measurements.

Materials Chemistry and Energy

* Invited Paper

SESSION E1.01: Materials Chemistry and Energy I
Session Chair: Graeme Luke
Monday Afternoon, June 6, 2022
UMC Aspen Room 285, 287, 289

3:45 PM *E4.04.01
Excess Vibrational Entropy in Metallic and Molecular Glasses Hillary Smith[1], Claire N. Saunders[2], Camille Bernal[2], Stefan H. Lohaus[2], Douglas L. Abernathy[3], Jiao Lin[3], Marios Demetriou[4] and Brent Fultz[2]; [1]Swarthmore College, United States; [2]Caltech, United States; [3]Oak Ridge National Laboratory, United States; [4]Glassimetal, United States

Liquid fragility characterizes how quickly viscosity decreases above the glass transition in glass-forming liquids. This property is fundamental to glass physics and of critical importance for engineering applications. Competing theories have suggested that vibrational properties may play a role in describing fragility by contributing vibrational entropy to the large excess entropy of the undercooled liquid over the crystal above the glass transition [1]. We performed the first in situ inelastic neutron scattering measurements of metallic glasses in the undercooled liquid phase and reported that at most 5% of the entropy of the glass transition originates from vibrational entropy in $Cu_{50}Zr_{50}$ and $Cu_{46}Al_{46}Zr_8$ [2]. We have extended this work to include two extremely fragile Pt-based metalloid glasses and the canonical fragile glass ortho-terphenyl (1,2-diphenylbenzene, $C_{18}H_{14}$). In situ measurements of the phonon densities of states were performed on ARCS during constant heating through the glass transition and above the crystallization temperature. The phonon density of states is extracted for 2-25K changes in temperature to quantitatively evaluate the vibrational entropy in the glass, liquid, and crystalline phases. No measurable change is observed in the phonon spectra across the glass transition in the Pt-based glasses. This talk will discuss the correlation between excess vibrational entropy in the undercooled liquid and liquid fragility. These new results will be considered in the context of a universal understanding of vibrational contributions to the thermodynamics of the glass transition.
1. Martinez, L.-M. & Angell, C. A. Nature **410**, 663–667 (2001).
2. Smith, H. L. et al., Nature Physics **13**, 900–905 (2017).

4:15 PM E4.04.02
The Two-Dimensional Nature of Dynamic Disorder in Hybrid Metal Halide Perovskite Semiconductors Nicholas Weadock[1], Tyler C. Sterling[1], Matthew Krogstad[2], Feng Ye[3], David Voneshen[4], Julian Vigil[5], Ballal Ahammed[6], Peter Gehring[7], Hans-Georg Steinrueck[8], Elif Ertekin[6], Hemamala Karunadasa[5], Dmitry Reznik[1] and Michael Toney[1]; [1]University of Colorado Boulder, United States; [2]Argonne National Laboratory, United States; [3]Oak Ridge National Laboratory, United States; [4]Rutherford Appleton Laboratory, United Kingdom; [5]Stanford University, United States; [6]University of Illinois at Urbana-Champaign, United States; [7]National Institute of Standards and Technology, United States; [8]Universität Paderborn, Germany

Hybrid metal halide perovskites are a novel class of semiconductor that require anharmonic structural and electronic calculations to explain the contradiction of a soft, defective lattice and remarkable optoelectronic performance of fabricated devices. [1,2] One consequence of the anharmonicity is octahedral tilting instabilities and soft phonon modes driving structural phase transitions. [3,4] These instabilities have led to predictions of dynamic domains of the tetragonal phase persisting within the high temperature cubic phase. [5] In this work, we utilize neutron and X-ray single crystal diffuse

scattering to probe structural correlations hidden within the cubic phases of $CH_3NH_3PbI_3$ and $CH_3NH_3PbBr_3$. Energy discrimination afforded by the CORELLI spectrometer allows us to distinguish between inelastic scattering from phonons and static scattering from defects. We find rods of diffuse intensity extending along the Brillouin zone edge, intersecting at the R-points, which imply the cubic phase contains two dimensional regions of tilted PbX_6 octahedra. Furthermore, we calculate the theoretical S(Q,E) from large supercell molecular dynamics (MD) simulations [6] and find a remarkable agreement with our experimental results. The MD simulations allow us to visualize the octahedral correlations and further reveal that similar correlations occur on the organic cation sublattice. The lifetime of these domains is at least 3ps, as determined by inelastic neutron scattering. Finally, we discuss the impact of this two-dimensional network on optoelectronic properties.
This work was supported by CHOISE, an Energy Frontier Research Center funded by the Department of Energy Office of Basic Energy Sciences.
[1] M. Z. Mayers, L. Z. Tan, D. A. Egger, A. M. Rappe, and D. R. Reichman, *How Lattice and Charge Fluctuations Control Carrier Dynamics in Halide Perovskites*, Nano Lett. **18**, 8041 (2018).
[2] D. A. Egger, A. Bera, D. Cahen, G. Hodes, T. Kirchartz, L. Kronik, R. Lovrincic, A. M. Rappe, D. R. Reichman, and O. Yaffe, *What Remains Unexplained about the Properties of Halide Perovskites?*, Advanced Materials **30**, 1800691 (2018).
[3] J. Klarbring, *Low-Energy Paths for Octahedral Tilting in Inorganic Halide Perovskites*, Phys. Rev. B **99**, 104105 (2019).
[4] J. S. Bechtel and A. Van der Ven, *Octahedral Tilting Instabilities in Inorganic Halide Perovskites*, Phys. Rev. Materials **2**, 025401 (2018).
[5] A. N. Beecher, O. E. Semonin, J. M. Skelton, J. M. Frost, M. W. Terban, H. Zhai, A. Alatas, J. S. Owen, A. Walsh, and S. J. L. Billinge, *Direct Observation of Dynamic Symmetry Breaking above Room Temperature in Methylammonium Lead Iodide Perovskite*, ACS Energy Lett. **1**, 880 (2016).
[6] T. Zhu and E. Ertekin, *Mixed Phononic and Non-Phononic Transport in Hybrid Lead Halide Perovskites: Glass-Crystal Duality, Dynamical Disorder, and Anharmonicity*, Energy Environ. Sci. **12**, 216 (2019).

4:30 PM E4.04.03
Neutron Scattering to Characterize Adsorbents and Their Hosts Craig M. Brown[1], Ryan Klein[2], Benjamin A. Trump[1] and Hayden A. Evans[1]; [1]NIST Center for Neutron Research, United States; [2]National Renewable Energy Laboratory, United States

Adsorption of molecules in functionalized and high surface area metal-organic frameworks (MOFs) is of emergent technological importance in a multitude of areas ranging from chemical separations to energy storage. We have been studying the properties of MOFs and other porous materials for storage and separations of industrially important small molecules such as hydrogen, oxygen, carbon dioxide, noble gases, and short chain organics. Besides the geometrical and porosity control available in MOF chemistry, the properties of the frameworks can be tweaked to elevate electrostatic interactions by exposing open metal cation sites or through enhanced van der Waals contacts via functionalizing ligands and introducing flexibility. We will highlight some of the most recent results on optimized interactions of hydrogen with open metal sites to obtain room temperature adsorption and a range of dynamic frameworks pore binding in MOF-like materials that optimize hydrogen adsorption using close-contacts. These results illustrate the governing characteristics of these material properties and the interactions with the guest molecules.

4:45 PM E4.04.05
Structure Modulation of $LnMnFeO_4$ upon Oxidation into $LnMnFeO_{4.5}$ (Ln=Y, Yb, Lu) Tianyu Li and Efrain E. Rodriguez; University of Maryland, United States

Hexagonal layered $LnFe_2O_4$ type material can uptake oxygen into its lattice and undergoes a structure transition from R-3m symmetry to P-3 symmery. Mn substituted version $LnMnFeO_4$ maintian oxygen uptake behavior and can be readily oxidized into $LnMnFeO_{4.5}$. We found the $LnMnFeO_{4.5}$ actually actopts a incommensurate modulated structure with an averaged structure of P-3. We first examined the structure and valence difference between the $LnFe_2O_4$ and $LnMnFeO_{4.5}$ with diffraction techniques, Raman spectroscopy and EELS to understand how does the structure evolve under oxidation. Then we focus on undersanding the modulated incommensurate structure of the oxidized phase $LnMnFeO_{4.5}$ with the help of HRTEM, electron diffraction, neutron diffraction and PDF analysis. Finally, the magetic properties and magnetic structure of $LnFe_2O_4$ and $LnMnFeO_{4.5}$ are with Neutron diffraction.

5:00 PM E4.04.06
Diffusion Dynamics of FLiNaK Molten Salt Characterized with Quasi-Elastic Neutron Scattering Brent J. Heuser, Golam Rakib and Yang Zhang; University of Illinois, United States

The Cold Neutron Chopper Spectrometer at the Spallation Neutron Source was used to measure the incoherent quasi-elastic neutron scattering response of molten FLiNaK, a potential coolant and fuel for molten salt reactors. Two incident beam energies, 3 meV and 12 meV, were used. The 12 meV data yields the structure factor at the elastic lines and includes accoustic vibrational modes. The element weighted self diffusion coefficient was measured by fitting the 3 meV QENS broadening as a function of temperature from 500 C to 800 C. The corresponding dynamic range was approximately 1 ps to 30 ps, appropriate for the fast dynamics associated with molten FLiNaK. Enriched Li-7 was used to reduce the strong absorption from Li-6. We determine the activation energy associated with self diffusion from the temperature dependence of the diffusivity. Discussion will focus on the comparison of our measured diffusivities and advanced computations of the same system.

SESSION E3.02: Materials Chemistry and Energy II
Session Chair: Allyson Fry-Petit
Wednesday Morning, June 8, 2022
UMC Aspen Room 285, 287, 289

10:15 AM *E3.01.01
Search for Broken Symmetries in Kagome Lattice Superconductor CsV_3Sb_5 Graeme Luke[1,2], Jonah Gautreau[1], Sudarshan Sharma[1], Mathew Pula[1], Yasutomo Uemura[3], Stephen D. Wilson[4], Brenden Ortiz[4] and Yuzki Oey[4]; [1]McMaster University, Canada; [2]TRIUMF, Canada; [3]Columbia University, United States; [4]University of California, Santa Barbara, United States

The kagome lattice, consisting of geometrically frustrated corner-sharing triangle units has a rich history of novel electronic and magnetic phenomena including spin liquid and topological phases. Kagome metals have recently received considerable attention, especially since the discovery of superconductivity in AV_3Sb_5 (where A=K, Cs, Rb). Theoretical calculations indicate that this family of materials has a non-trivial topological band structure, while experiments have demonstrated a variety of phase temperatures such as charge density wave (CDW) order, in addition to superconductivity. I will discuss our muon spin rotation/relaxation studies of pure CsV_3Sb_5, as well as charge-doped $CsV_3Sb_{5-x}Sn_x$ where we observed multi-gap superconductivity with preserved time-reversal symmetry, but with weak broken time-reversal symmetry in the normal state, distinct from the

CDW ordering.

10:45 AM E3.01.02
Phonon Dynamics and Thermal Transport in Tl_3VSe_4 Yoel F. Lencina Wendt[1], Qingan Cai[1], Brian Sales[2], Ayman Said[3] and Chen Li[2,1,1]; [1]University of California, Riverside, United States; [2]Oak Ridge National Laboratory, United States; [3]Argonne National Laboratory, United States

Solids with ultralow thermal conductivity have attracted interest in applications of thermoelectrics. At room temperature, bulk crystalline Tl3VSe4 has been reported to possess one of the lowest lattice thermal conductivity values among the bulk thermoelectrics. It is known that the conventional phonon picture provided by Peierls-Boltzmann equation is insufficient in describing the experimentally observed ultralow thermal conductivity of this thermoelectric material, and supplemental theories have been proposed in literature, including the existence of an additional hopping channel based on uncorrelated localized vibrations. We investigated the phonon dynamics in Tl3VSe4 as a function of temperature with inelastic x-ray scattering. The temperature dependence of acoustic phonons has been observed to be inconsistent with literature. This may provide the groundwork to challenge previous work on this material.

11:00 AM E3.01.03
High Throughput Operando Neutron Diffraction at the Nanoscale Ordered Materials Diffractometer (NOMAD) Jue Liu, Zhijia Du, Xianyang Wu and Michelle Everett; Oak Ridge National Laboratory, United States

Recently, we have successfully commissioned the first high throughput *operando* neutron diffraction studies of batteries at the Spallation Neutron Source (SNS)'s Nano-Scaled Materials Diffractometer (NOMAD). The very high neutron flux together with large detector coverage of NOMAD allows for fast data collection of structural refinable neutron diffraction data using our newly designed *in situ* electrochemical cell. In this talk, I will showcase some of our recent efforts and progress in this direction. I will highlight our recent discovery of a universal four-stage structural evolution mechanism for Ni-rich high energy density battery cathodes. Particulary, we discovered an anomalous increase of average TM-O bond lengths when more than ~ 0.75 Li^+ are removed during the initial charge, and this transition is fully reversible during discharge. This anomalous structural transition is strongly correlated with the oxygen gas release and the universal decline of capacity retention in Ni-rich cathodes, suggesting this overlooked bulk structural transition may play an indispensable role in the degradation process.

11:15 AM E3.01.05
Lithium-Polymer Batteries—A Microscopic View! Michael Ohl[1,2], Juergen Allgaier[1], Marcella Cabrera-Berg[1], Changwoo Do[3], Yuya Doi[4], Rene Halver[1], Eugene Mamontov[3], Ridhima Nain[5], Naresh C. Osti[3], Godehard Sutmann[1,6], Hui Wang[7], Stephan Forster[1] and Takeshi Egami[3,2]; [1]Forschungszentrum Jülich GmbH, Germany; [2]Univ. Tennessee, United States; [3]Oak Ridge National Laboratory, United States; [4]Nagoya University, Japan; [5]Indian Institute of Technology Delhi, India; [6]Ruhr-University Bochum, Germany; [7]Stanford Linear Accelerator Center, United States

Batteries play an important role in our daily life with a variety of applications. To this day a lot of money resources had have been put in researchdirected to develop high-performance and especially safe batteries, for which solid polymer electrolytes are excellent candidates. To find the right electrolyte withoutand to avoid prolongedany trial and error effort, innovative studies are necessaryintend to develop a microscopic picture of the Li – ion conductivity process, which is difficult to obtain from standard in-house laboratory techniques. However, Neutron scattering in combination with MD simulation form a suitable basis for such studiesthis. Also, Poly-ethylene oxide (PEO) is the most commonly used polymer electrolyte due to its high conductivity, easily up up to to 10^{-2} S/cm even in its pure occurrence. In PEO based Lithium batteries, it is generally believaccepted that the charge transportation mechanism of the Li+ ion is directly dependsing on the segmental and backbone motions of the polymer. If optimized, these processes can make the conductivity process of Li+ faster, which directly results in faster charging and discharging of the battery. With MD simulation we can access and developenlighten the microscopic picture and once confirm ited with QENS. Here we report on the results of a study of a comb PEO polymer which leads to the development of 4-arm star PEO polymers. We describe hile all of them offer their own specific conductivity mechanism, the technique of tailoring polymers towards better and higher performanceing polymers, throughespecially studying the conductivity mechanism of various components is reported. A variety of Li-salt/ polymer candidates will be simulated alongside with the study via QENS measurements.

SESSION E3.03: Materials Chemistry and Energy III
Session Chair: Hillary Smith
Wednesday Afternoon, June 8, 2022
UMC Aspen Room 285, 287, 289

3:45 PM E3.02.01
The Symmetry Relationship Between Magnetic Order and Toroidal Moments in $LiM_xMn_{1-x}PO_4$ (M = Co, Fe) Stephanie Gnewuch and Efrain E. Rodriguez; University of Maryland, United States

While the $LiM_xMn_{1-x}PO_4$ (M = Co, Fe) series of materials are best known as cathode materials for Li-ion batteries, the end members $LiFePO_4$, $LiCoPO_4$, and $LiMnPO_4$ have been known for decades to exhibit a linear magneto-electric effect.[1] In recent years, the magneto-electric effect within these materials has been linked to a fourth type of ferroic ordering called ferrotoroidicity, in which there are a net alignment of toroidal moments in a material.[2] This is analogous to ferromagnetism, in which there are a net alignment of magnetic moments within the material. While the magnetic structure *Pn'm'a'* of $LiMnPO_4$ does not support net toroidization, it is allowed in the magnetic structure *Pnma'* of $LiFePO_4$ and $LiCoPO_4$.[3] The candidate $LiCoPO_4$ in particular is reported to display the expected domain structure and hysteretic behavior expected of a ferroic material.[4]

Given this promising structure type, we set out to prepare and study the magnetic ordering of solid solutions of this material. The objective was to manipulate the magnitude and distance between the magnetically active ions, while maintaining the magnetic structure which permits ferrotoroidal ordering. Previously at this conference I presented my analysis of powder neutron diffraction of various compositions of $LiFe_xMn_{1-x}PO_4$ (x = 0.2 - 0.6). By varying the Fe to Mn ratio on the 4*c* site within the *Pnma* structure type, we were able to observe a clear evolution of the antiferromagnetic transition temperature between the samples (about 40 - 47 K), but the same magnetic structure *Pnma'.* Since then we continued this work and prepared additional samples of the structurally analogous system $LiCo_xMn_{1-x}PO_4$ (x = 0.3 - 0.7). We observed a similar evolution in the transition temperature (about 34 - 30 K), but with the same magnetic structure *Pnma*' revealed by powder neutron diffraction.

In this presentation, we discuss the the structural similarities between the compositions, leading to the similar magnetic structures. We then present our

most recent understanding of how the structural symmetry can support the formation of ordered toroidal moments within this series of materials. We also discuss how the changes in the observed magnitudes and distances between the magnetic moments determined from the neutron diffraction data can rationalize how to tune the magnitude of toroidal moments in this structure type.

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4:00 PM E3.02.03
Raising the Transition Temperature of Olivines $Li_{1-x}Fe_xMn_{1-x}PO_4$ Through Selective Li-Deintercalation Timothy J. Diethrich and Efrain E. Rodriguez; University of Maryland, College Park, United States

The Li*M*PO_4 (*M* = Mn, Fe, Co, Ni) olivines have been utilized and applied in countless fields of study including batteries, quantum materials, and magnetic materials. This work is specifically focuses on broadening our current understanding of magnetoelectrics and ferrotoroidic materials. Ferrotoroidics undergo a spontaneous alignment of toroidal moments at a critical temperature. For a toroidal moment to exist, a specific orientation of magnetic moments is required, because of this, only certain magnetic point groups are allowed. $LiFePO_4$, for example, has an 'allowed' magnetic point group of *m'mm*; $LiMnPO_4$ and $FePO_4$ have 'forbidden' point groups of *m'm'm'* and 222 respectively. Our work focuses on lithiated and delithiated mixed iron-manganese phosphates. The $Li_{1-x}Fe_xMn_{1-x}PO_4$ solid solution series was chosen with the specific aim to simultaneously increase the critical temperature at which long-range order is introduced while maintaining an 'allowed' magnetic point group.

We successfully synthesized the entire phase-pure lithiated $LiFe_xMn_{1-x}PO_4$ powder and single crystal series as well as the entire delithiated $Li_{1-x}Fe_xMn_{1-x}PO_4$ powder series. The purity of the sample as well as the expected structural properties were verified with powder x-ray diffraction. A SQUID MPMS was used on both the single crystals and powders to show that the antiferromagnetic transition temperature increases significantly upon removal of lithium and increases in iron concentration. The transition temperature increases from 32 K to 152 K depending on the iron content and amount of lithium deintercalated.

Following preliminary structural and magnetic analysis, we collected neutron powder diffraction data on lithiated and delithiated samples. Representational analysis was performed on the neutron data at 5 K. The observed commensurate antiferromagnetic reflections were fit to a propagation vector k = (0,0,0) which was then used to solve the magnetic structures. It became clear that even when removing only 20% of the lithium from these solid solutions, the resultant magnetic contributions are no longer colinear and demonstrate a second magnetic contribution. While this differs from the lithiated materials, it is still an 'allowed' magnetic point group for ferrotoroidicity. We believe that this change in magnetic structure is due to the unit cell contracting and the super exchange pathway between metal centers getting smaller. We continued to analyze samples with more iron and less lithium until we found the concentration where the magnetic symmetry was equivalent to the $FePO_4$ 'forbidden' 222 point group. To the best of our knowledge, this is the first time that the magnetic structures of any partially delithiated transition metal phosphate has been determined. The resultant magnetic points groups from neutron refinements were used to determine the material with the largest transition temperature while maintaining an allowed magnetic point group for ferrotoroidicity.

4:15 PM E3.02.04
Van Hove Correlation Function of Magnesium Chloride Molten Salt Yuya Shinohara[1], Alexander S. Ivanov[1], Garrett E. Granroth[1], Douglas L. Abernathy[1] and Takeshi Egami[2,1]; [1]Oak Ridge National Laboratory, United States; [2]The University of Tennessee, Knoxville, United States

Molten salt has gained renewed attention as coolants and reaction media of molten salt reactors as well as electrolytes and heat transfer media. Despite extensive research, little is known about the local atomic motion of molten salt because of experimental and computational challenges. We have recently developed an approach to study local dynamics in liquid using Van Hove correlation function—spatial and temporal correlation function in real space. We measured inelastic neutron scattering spectra of magnesium chloride molten salt using ARCS of the Spallation Neutron Source at Oak Ridge National Laboratory and converted them into the Van Hove correlation function by carrying out Fourier transformation over both momentum and energy transfer. This real-space approach will elucidate the local dynamics that is hardly discernible in reciprocal space.

This work was supported by the U.S. Department of Energy (DOE), Office of Science, Office of Basic Energy Science (BES), Materials Sciences and Engineering Division.

4:30 PM E3.02.05
Mapping the Light Elements in Complex Oxides for High-rate Lithium-ion Batteries Kent Griffith and Kenneth R. Poeppelmeier; Northwestern University, United States

Complex early transition metal oxides have emerged as leading candidates for fast charging lithium-ion battery anode materials, particularly those based on multielectron niobium redox. Framework crystal structures with frustrated topologies are good electrode candidates because they may intercalate large quantities of guest ions with minimal structural response. Starting from the empty perovskite (ReO_3) framework, shear planes and filled pentagonal columns are examples of motifs that decrease the structural degrees of freedom. As a consequence, many early transition metal oxide shear and bronze structures do not readily undergo the tilts and distortions that lead to phase transitions and/or the clamping of lithium diffusion pathways that occur in a purely corner-shared polyhedral network.

In this work, we explore the relationship between composition, crystal structure, and electrochemical performance in a variety of mixed alkali, transition metal, and main group oxides. By expanding beyond binary niobium-based shear and bronze structural archetypes, we explore the structural modifications and performance implications upon incorporation of alkali and/or heavy main group metals. High-resolution neutron diffraction (with joint synchrotron X-ray diffraction for simultaneous Rietveld refinement) is used to resolve structural questions in the host materials related to sodium disorder, space group subtleties, lithium positions, and distortions of the second-order Jahn–Teller d^0 Nb^{5+}/W^{6+} and lone-pair $6s^2Bi^{3+}$ redox centers. The structure models from diffraction are informed by the local structure perspective from solid-state NMR spectroscopy. Prospects for tunability and implications for charge rate and structural stability will be discussed.

4:45 PM *E3.02.06
Exploring Oxygen Motion Through Perovskites $La_{0.9}Sr_{0.1}Co_{1-y}Fe_yO_{3-\delta}$ with In Situ Diffraction Allyson M. Fry-Petit[1], Mara Milhander[1,2] and Jose Gonzalez Jimenez[1,2]; [1]California State University, Fullerton, United States; [2]Rutgers, The State University of New Jersey, United States

$La_{0.9}Sr_{0.1}Co_{1-y}Fe_yO_{3-\delta}$ (y=0, 0.25, 0.75, 1) is one example of a broad family of perovskite materials for which oxygen selectively moves through the structure, known as oxygen transport membranes. These materials have potential uses in solid oxide fuel cells, oxygen storage materials, and chemical looping reactions, but despite their many applications and current use in industry the mechanism of their operation at the structural level is poorly understood. We will present the work performed to understand the complex structural changes that $La_{0.9}Sr_{0.1}Co_{1-y}Fe_yO_{3-\delta}$ (y=0, 0.25, 0.75, 1) undergoes under operating conditions. Through the use of *in situ* neutron and synchrotron diffraction studies we will show that the operating temperature, which was known to depend upon changes in the Co to Fe ratio, is intimately tied to small structural changes in the material. Moreover, we will show quantitative analysis of the multiple phases that are present throughout the cycling of these materials. Lastly, we will discuss further instrumental improvements that need to be made to enhance the quality of this type of analysis.

SESSION EP3.03: Poster Session: Materials Chemistry and Energy
Wednesday Afternoon, June 8, 2022
5:30 PM - 7:30 PM
UMC Center Ballroom 210

EP3.03.01
Time-Resolved Neutron Reflectometry Study of Li-Mediated Electrochemical Nitrogen Reduction Mathieu Doucet[1], Sarah J. Blair[2,3], Jim Browning[1], Hanyu Wang[1], Candice Halbert[1], Adam Nielander[3], Alessandro Gallo[3] and Thomas F. Jaramillo[2,3]; [1]Oak Ridge National Laboratory, United States; [2]Stanford University, United States; [3]SLAC National Accelerator Laboratory, United States

Li-mediated electrochemical reduction of nitrogen to NH_3 has been identified as an alternative to the resource-demanding Haber-Bosch ammonia process. Through this Li-mediated process, an initial electrodeposition of Li is followed by a reaction to form Li_3N and subsequent ethanolysis of Li_3N to form NH_3. We report on the study of the initial phase of this process with time-resolved neutron reflectometry at the Liquids Reflectometer at SNS. We studied Li electrodeposition on a molybdenum thin film in contact with deuterated THF with 0.2 M $LiClO_4$ and 1 v% EtOH at a current density of -0.1 mA/cm^2. The neutron reflectometry data acquired during the first five minutes of Li plating was sliced in 60-second intervals in a wide enough Q range ($0.017 < Q < 0.05$ 1/Å) to allow time-resolved modeling of the plating process. The observed Li layer and the absence of a clear solid-electrolyte interphase layer in this time frame is of particular interest since cycling on the time scale of minutes has been identified as a way to enhance selectivity towards NH_3.

EP3.03.02
A Room temperature Polar and Weak-ferromagnetic Oxide with Low Dielectric Loss Nagamalleswari Katragadda[1], Pranab Mandal[1], Premakumar Yanda[2], A Sundaresan[2], S. D. Kaushikc[3], Weiguo Zhang[4], P. Shiv Halasyamani[4] and Alicia M. Manjon Sanz[5]; [1]SRM University, India; [2]Jawaharlal Nehru Centre for Advanced Scientific Research, India; [3]UGC-DAE Consortium for Scientific Research Mumbai Centre, R-5 Shed, BARC, India; [4]University of Houston, United States; [5]Oak Ridge National Laboratory, United States

Single phase materials that are simultaneously ferroelectric and ferromagnetic at room temperature are promising for non-volatile random access memory devices. Perovskite $BiFeO_3$ which crystallizes in rhombohedral structure (*R*3*c*), is both ferroelectric and antiferromagnetic at room temperature. The synthesis of $BiFeO_3$ is challenging in ceramic and in thin film forms. Here, we report a family of perovskite oxides in the $BiFeO_3$ – $Bi_{2/3}TiO_3$ – $ATiO_3$ (where A = Ca, Sr, Ba) that are polar as well as weak ferromagnetic at room temperature. We achieved nearly pure A-site perovskite phase $Bi_{0.9167}Ca_{0.075}Fe_{0.9}Ti_{0.1}O_3$ by using solid state reaction method. Powder X-ray and neutron diffraction experiments suggest that the room temperature structure is rhombohedral (space group *R*3*c*) as corroborated by second harmonic generation (SHG) experiments. Room-temperature powder neutron diffraction confirms G-type antiferromagnetic ordering consistent with weak ferromagnetism that onsets at T_N = 557 K. These perovskites show a low dielectric loss, and impedance spectroscopy reveal that electrical response is dominated by grain contribution below 723 K.

EP3.03.03
Phonon Lifetimes and Mode Softening in Cubic $Cs_2AgBiBr_6$ Zihan Zhang[1], Nicholas Weadock[2], Peter Gehring[3], Julian Vigil[4], Tao Hong[5], Johan Klarbring[6], Adam Slavney[7] and Michael Toney[2]; [1]University of Colorado, Boulder, United States; [2]University of Colorado Boulder, United States; [3]National Institute of Standards and Technology, United States; [4]Stanford University, United States; [5]Oak Ridge National Laboratory, United States; [6]Linköping University, Sweden; [7]Harvard University, United States

Hybrid organic-inorganic metal halide perovskite (HOIP) materials have attracted major research interest due to their ease of fabrication and the rapid rise in efficiency as absorbers in solar cells. The best performing HOIP devices, however, contain toxic lead, resulting in a search for lead-free perovskite formulations. One promising lead-free archetype is the double perovskite $A_2B^IB^{III}X_6$ formulation, with $Cs_2AgBiBr_6$ reported to have a device relevant 1.95 eV bandgap. [1] In addition to the favorable bandgap, $Cs_2AgBiBr_6$ is stable in ambient conditions, and has promise as a direct x-ray and gamma-ray detector. [1,2] $Cs_2AgBiBr_6$ crystallizes in the cubic Fm-3m phase and undergoes a cubic-tetragonal phase transition at 122 K. [3] Recent anharmonic lattice dynamics calculations have suggested that the phase transition proceeds by the condensation of a soft, zone-center optical phonon. [4] Raman scattering studies have not identified this phonon in the cubic phase, therefore no experimental confirmation of the phenomenon exists. [5] In addition to the soft mode prediction, the calculations predict zone-boundary anticrossing behavior and phonon lifetimes on the order of 10 ps. This anticrossing behavior (phonon repulsion) demonstrates that there is a strong anharmonic coupling which may contribute to low thermal conductivity. To date there has been no experimental quantification of the phonon lifetimes, limiting our understanding of thermal properties and electron-phonon interactions in Cs2AgBiBr6. In this work, we use inelastic neutron scattering to characterize the acoustic and low energy optical phonons modes with the SPINS and BT4 triple-axis spectrometers at NIST. The acoustic phonon energies identified from constant Q-scans match well with room temperature phonon dispersions calculated from fully anharmonic first-principles lattice dynamics. [4] We also calculate the acoustic phonon lifetimes after careful removal of the instrument resolution. Compared to the prototypical HOIP, MAPI, $Cs_2AgBiBr_6$ acoustic modes exhibit a larger group velocity and longer lifetime, resulting in a longer phonon mean free path (MFP). As a result, we propose that the acoustic modes are the dominant contribution to the thermal conductivity in $Cs_2AgBiBr_6$. This result contrasts with MAPI, in which the low-energy optical Pb-I cage modes are the main contributors to the thermal conductivity. [6] We confirm the presence of a zone-center optical soft mode and track the temperature dependence using the CTAX triple-axis spectrometer at the High Flux Isotope

Reactor (HFIR) at ORNL. The soft mode frequency is measured from 150K to 450K and shows a linear decay approaching the phase transition temperature of 122K. This confirms a first-order cubic-to-tetragonal transition in $Cs_2AgBiBr_6$. Ultimately, this work helps to understand the free charge carrier properties in double-perovskite $Cs_2AgBiBr_6$ which are largely governed by electron-phonon interactions.

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EP3.03.05
Ratiometric Thermometry Using Thermochromic Tb^{3+}:Mn^{4+}:$Na_4Mg(WO_4)_3$ Phosphors Dinesh K. Amarasinghe and Federico A. Rabuffetti; Wayne State University, United States

The utility of Tb^{3+}:Mn^{4+}:$Na_4Mg(WO_4)_3$ phosphors as ratiometric luminescent thermometers in the 200–450 K temperature range is demonstrated. Targeted substitution of Tb^{3+} for Na^+ and Mn^{4+} for Mg^{2+} yields phosphors of formula $Na_{4-12x}Tb_{4x}Mg_{1-2y}Mn_y(WO_4)_3$. The average crystal structure of the phosphors is probed using neutron diffraction and the site of substitution of Tb^{3+} activators is determined via X-ray absorption spectroscopy. UV excitation of the $Na_4Mg(WO_4)_3$ host results in green emission from terbium (544 nm) and red emission from manganese (682 nm). Differential thermal quenching of these emissions renders $Na_{3.976}Tb_{0.008}Mg_{0.990}Mn_{0.005}(WO_4)_3$ thermochromic between 78 and 450 K, with an orange-to-green color change that is particularly noticeable above 200 K. As a result, this phosphor serves as a ratiometric and colorimetric thermometer between 200 and 450 K. Quantitative assessment of its thermometric performance yields a maximum relative sensitivity of 2.5×10^{-2} K^{-1} at 375 K, along with a resolution of 0.5 K and a repeatability of 98%. These findings highlight the versatility of $Na_4Mg(WO_4)_3$ as a platform to realize luminescent thermometers via targeted aliovalent substitutions.

EP3.03.06
Structural Resolution and Mechanistic Insight into Hydrogen Adsorption in Flexible ZIF-7 Ryan Klein[1,2], Sarah Shulda[2], Craig M. Brown[1] and Michael McGuirk[3]; [1]NIST Center for Neutron Research, United States; [2]National Renewable Energy Laboratory, United States; [3]Colorado School of Mines, United States

Flexible metal–organic frameworks offer a route towards high useable hydrogen storage capacities with minimal swings in pressure and temperature via step-shaped adsorption and desorption profiles. Yet, the understanding of hydrogen-induced flexibility in candidate storage materials remains incomplete. Here, we investigate the hydrogen storage properties of a quintessential flexible metal–organic framework, ZIF-7. We use high-pressure isothermal hydrogen adsorption measurements to identify the pressure–temperature conditions of the hydrogen-induced structural transition in ZIF-7. The material displays narrow hysteresis and has a shallow adsorption slope between 100 K and 125 K. To gain mechanistic insight into the cause of the phase transition correlating with stepped adsorption and desorption, we conduct powder neutron diffraction measurements of the D_2 gas-dosed structures at conditions across the phase change. Rietveld refinements of the powder neutron diffraction patterns yield the structures of activated ZIF-7 and of the gas-dosed material in the dense and open phases. The structure of the activated phase of ZIF-7 is corroborated by the structure of the activated phase of the Cd congener, CdIF-13, which we report here for the first time based on single crystal X-ray diffraction measurements. Subsequent Rietveld refinements of the powder patterns for the gas-dosed structure reveal that the primary D_2 adsorption sites in the dense phase form D_2–arene interactions between adjacent ligands in a sandwich-like adsorption motif. These sites are prevalent in both the dense and the open structure for ZIF-7, and we hypothesize that they play an important role in templating the structure of the open phase. We discuss the implications of our findings for future approaches to rationally tune step-shaped adsorption in ZIF-7, its congeners, and flexible porous adsorbents in general. Lastly, important to the application of flexible frameworks, we show that pelletization of ZIF-7 produces minimal variation in performance.

EP3.03.07
Structure Investigation of the Transition Between LnMnFeO4 and LnMnFeO4.5 (Ln=Y, Yb, Lu) Tianyu Li and Efrain E. Rodriguez; University of Maryland, United States

Hexagonal layered $LnFe_2O_4$ type material can uptake oxygen into its lattice and undergoes a structure transition from R-3m symmetry to P-3 symmetry. Mn substituted version $LnMnFeO_4$ maintains oxygen uptake behavior and can be readily oxidized into $LnMnFeO_{4.5}$. We found that the $LnMnFeO_{4.5}$ actually adopts an incommensurately modulated structure with an averaged structure of P-3. We examined the structure and valence difference between the $LnFe_2O_4$ and $LnMnFeO_{4.5}$ with diffraction techniques, Raman spectroscopy, and EELS to understand how does the structure evolve under oxidation. Then we focus on understanding the modulated incommensurate structure of the oxidized phase $LnMnFeO_{4.5}$ with the help of HRTEM, electron diffraction, neutron diffraction, and PDF analysis. Finally, the magnetic properties and magnetic structure of $LnFe_2O_4$ and $LnMnFeO_{4.5}$ are also investigated.

EP3.03.08
Local Ordering of Cations in Thermoelectric Alloys Vanessa Meschke, Andrew Novick and Eric Toberer; Colorado School of Mines, United States

Alloys of thermoelectric materials have been studied for over 100 years as they often increase a compound's thermoelectric figure of merit (zT) by lowering the lattice thermal conductivity and tuning the electronic structure. In this work, we focus on bulk samples of alloys we have synthesized between GeTe and MnTe (i.e., $Ge_{1-x}Mn_xTe$), which show an increased zT accompanied by a change in symmetry driven by alloying. The structure observed in our experiments versus those we modeled using the special quasirandom structures approach, which models alloys as totally random structures, do not agree, suggesting the presence of local ordering that is undescribed by both theory and experiment. Motivated by the discrepancy, we have developed a novel computational method that acknowledges the energetic importance of local structural motifs in alloys. With this framework, we predict the local structure at varying temperatures, which can be paired with neutron diffraction data to develop a deeper understanding of possible local ordering in the $Ge_{1-x}Mn_xTe$ alloy space.

EP3.03.09

Study of Magnetic Properties and Structural Distortions in $Bi_{1-x}Nd_xFeOc_3$ Compositions Luiz F. Cotica, Igor B. Catellani, Odair G. Oliveira, Gustavo S. Dias and Ivair A. Santos; State University of Maringá, Brazil

In this work, to study the Nd doping on $Bi_{1-x}Nd_xFeO_3$ multiferroics (with x ranging from 0.025 to 0.20), ceramic powders were synthesized by a high energy milling method followed by sintering. Neutron diffraction measurements were performed to obtain data on crystalline and magnetic structures of the studied compositions. Rietveld refinements verified that the gradual substitution of bismuth to neodymium induces a transition from the rhombohedral phase (*R*3*c*) to the orthorhombic phase (*Pbam*). In addition, a third phase, whose assists in the transition from the rhombohedral to orthorhombic phase, was identified. From the refinements of the neutron diffraction patterns, we verify that the structural parameters s, t, d, z, and *w* of the FeO_6 octahedron directly influence the evolution of the Fe^{3+} magnetic moment intensity. The increase in distortions signified an increase in the magnetic moment value of the Fe^{3+} ion in a weak ferromagnetic (WFM) phase. The resulting magnetic moment of the WFM phase was observed, and its behavior appears to be linked to *w* in the FO_6 octahedron. It was observed, at room temperature, a continuous smooth process of spin reorientation up to x = 0.125. Out of x = 0.15, a reorientation from a state slightly different from Gx to a GxGyGz state was revealed. Concerning the cycloid structure of the magnetic moments, an expansion in the period until x = 0.125 was detected (reaching 1064 Å). Observing the correlation among the electrical polarization, the unit cell volume, and the ionic displacement along the c axis, we can state that it increases for valudes with a higher concentration of Nd. In the same way, an increase in the WFM magnetic moment was observed. These results suggest that the magnetoelectric coupling increases with increasing Nd concentration in $Bi_{1-x}Nd_xFeO_3$ compositions.

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Neutrons Diffraction Studies Under Static Electric Fields Ivair A. Santos[1], Gustavo S. Dias[1], Luiz F. Cotica[1], Valdirlei F. Freitas[2] and Fabiano Yokaichiya[3]; [1]State University of Maringá, Brazil; [2]Midwestern Parana State University, Brazil; [3]Federal University of Paraná, Brazil

Recent advances in the harvesting of energy using the perovskite solar cells have reinvigorated the efforts to develop materials with perovskite structure. For such an application, an interesting alternative emerges with the multifunctional multiferroic materials, especially magnetoelectrics, like the $(0.6)BiFeO_3$-$(0.4)PbTiO_3$ compound. This compound, in fact, potentially gives these devices more degrees of freedom to be explored to improve their photovoltaic conversion efficiency. However, for practical applications, it is still necessary to understand and solve fundamental questions about the multiferroism in displacive ferroics as, for example, the origin of magnetoelectric effect/coupling. This is because the prior knowledge of these mechanisms would allow the control and improvement of related properties, allowing the engineering of new materials for different applications in this area. In this way, to investigate the origin of magnetoelectric effect/coupling in displacive multiferroics and mainly to test, understand and solve the intrinsic relation between ferroelectricity, magnetism and structural properties in these perovskite-structured compounds, La modified $(0.6)BiFeO_3$-$(0.4)PbTiO_3$ (BFPT-L) ceramics were processed and investigated (structural-magnetic-related properties) by applying high-resolution neutron powder diffraction under external static electric fields. Thus, we conducted high-resolution neutron diffraction experiments (at 300 K and 2 K) at the E2 equipment of the Helmholtz Zentrum Berlin in BFPT-L ceramics under static electric fields (0, 2 and 4 kV/mm), turning the sample around its position, and consequently, changing the sample's polarization vector (q = 0, 45 and 90 degrees) in relation to the propagation direction of the neutron beam. The results were investigated by Rietveld refinement using the Fullprof software, and structural and magnetic properties were obtained on the three distinct angular orientations. Substantial changes in the intensities of neutron diffraction pattern were identified, as a function of the orientation angle, indicating different direction and intensities of magnetic dipoles despite any significant change in the structural parameters. These results are clear evidence of the intrinsic and intimate link between ferroelectric and magnetic orders in these perovskite-structured materials.