WORKSHOP AGENDAS AND ABSTRACTS

Tuesday, May 7

WK1 Joint APS/CNM: Driving Scientific Discovery with Artificial Intelligence, Advanced Data Analysis, and Data Management in the APS-U Era

Location: Building 402, Room E1100/1200

Time: 8:45 – 5:00

Organizers: Nicholas Schwarz (APS/ANL), Chengjun Sun (APS/ANL), and Mathew Cherukara (CNM/ANL)

The APS and CNM are in the position to help solve some of the most challenging and novel scientific questions facing the nation. The design of new materials to manipulate classical and quantum information with high fidelity and ultralow power consumption, enabling systems for efficient energy storage, transportation, and conversion that will drive the emerging economy based on renewable energy, and understanding the structure and motion of protein molecules to enable individualized medicine are just a few examples. Addressing these scientific opportunities will be aided by the intrinsic capabilities of APS-U era facilities along with new measurement techniques and technological advances in detectors.

These advances are expected to drastically increase the amount and complexity of data generated by instruments at the APS, especially in the areas of coherence driven, imaging, high-energy scattering, advanced x-ray spectroscopy, and multi-modal techniques enabled by APS-U era facilities. However, methods for understanding data have not always kept pace. Manual analysis and management of data is too time consuming and cumbersome for large, complex datasets. State-of-the-art mathematics and computer science tools will help automate the understanding process. The scientific mission of the APS-U era facility will only be achieved by coupling the capabilities of the facility with advanced data analysis and data management resources.

For example, machine learning methods including deep learning methods, have been employed to accelerate data analysis for advanced x-ray spectroscopy and time-resolved x-ray and electron imaging, to design new molecular structures, to automate manufacturing and to accelerate the materials design loop. This workshop is organized to discuss the state-of-the art and potential of artificial intelligence, and data analysis and management for the APS and CNM. It is expected to provide an opportunity for academics, R&D scientists in industry, and students to exchange ideas and think creatively about new avenues for collaborations.

Topics include, but are not limited to:

- Data analysis and machine learning algorithms and software
- Experiment design and real-time experiment steering
- Deep learning approaches for inverse imaging problems, and feature detection and labeling
- Machine learning to accelerate high-throughput computational x-ray absorption spectroscopy
- Data driven material selection and synthesis, property prediction and inverse design of materials
- On-demand utilization of high-performance computing resources
- Data management and data collaboration workflow tools
- Deep learning at scale
- Introduction to machine learning for x-ray diffraction, spectroscopy, and imaging

The goal is to elaborate on how users can benefit from advanced data analysis and management techniques, and to identify needs to enable new scientific discovery in the APS-U era.

8:45 – 9:00	Opening Remarks
9:00 – 9:30	TBD *coming soon*
9:30 – 10:00	Elise Jennings (Argonne National Laboratory) *Scientific Machine Learning at Argonne Leadership Computing Facility*
10:00 – 10:30	Break
10:30 – 11:00	Shuai Liu (University of California-Berkeley, Lawrence Berkeley National Laboratory) *Driving Scientific Discovery with Artificial Intelligence, Advanced Data Analysis and Data Management in the APS-U Era*
11:00 – 11:30	TBD *coming soon*
11:30 – 12:00	Nouamane Laanait (Oak Ridge National Laboratory) *Decoding Inverse Imaging Problems in Materials with Deep Learning and Supercomputing*
12:00 – 1:00	Lunch
1:30 – 2:00	Shyam Dwaraknath (Lawrence Berkeley National Laboratory) *The Materials Project: Conception to Confirmation in a Virtual Lab*
2:00 – 2:30	John Rehr (University of Washington) *High-throughput Computational X-ray Absorption Spectroscopy*
2:30 – 3:00	Maria Chan (Argonne National Laboratory) *Combining Characterization and Modeling Data for the Determination of Nanoscale Structures*
3:00 – 3:30	Break
3:30 – 4:00	Saugat Kandel (Northwestern University and Argonne National Laboratory) *Learning Phase Retrieval with Backpropagation*
4:00 – 4:30	Daniela Ushizima (Lawrence Berkeley National Laboratory) *Thin Film Structure Identification through Convolutional Neural Networks Applied to Scattering Patterns*
4:30	Discussion and Close Out

WK1

Scientific Machine Learning at Argonne Leadership Computing Facility

Elise Jennings
Argonne National Laboratory, Lemont, IL 60439

Scientific machine learning has the potential to transform science and energy research and has found a broad use across DOE user facilities. Scientific progress and discovery is being driven by massive data together with optimized software for predictive models and algorithms running on heterogeneous high-performance computing platforms. The ALCF data science program (ADSP) supports projects which push the state-of-the-art in data-centric and data-intensive computing; as well as in machine learning, deep learning, and other AI methods at scale. I will discuss the data science research at ALCF and give an overview of current ADSP projects relevant to the APS.

WK1

Driving Scientific Discovery with Artificial Intelligence, Advanced Data Analysis and Data Management in the APS-U Era

Shuai Liu
University of California-Berkeley, Berkeley, CA 94720
Lawrence Berkeley National Laboratory, Berkeley, CA 94720

X-ray scattering has a variety of applications in science to investigate the structure of different materials systems. However, it remains challenging to analyze large-scale x-ray scattering data in a high-throughput and effective fashion. We propose to approach this problem using machine learning to categorize the data and to determine the underlying structure of the materials. Here we propose a hierarchical categorization method to cluster the features in the x-ray scattering data and to manage large-scale experimental data. Moreover, this method is applied to several *in situ* experimental systems in order to detect the structure transitions automatically. Second, we illustrate a machine learning methodology to classify the underlying materials structure from x-ray scattering data. The machine learning model is trained using data generated from the high performance GISAXS (HipGISAXS) simulation package. This methodology is applied to binary superlattices in thin films to characterize the crystal structure. Finally, we introduce a new neural network architecture, which proved to be more effective than conventional convolutional neural networks looking at x-ray scattering and XRD datasets.

WK1

Decoding Inverse Imaging Problems in Materials with Deep Learning and Supercomputing

Nouamane Laanait
Computational Sciences and Engineering Division,
Oak Ridge National Laboratory, Oak Ridge, TN 37830

Modern Scanning Transmission Electron Microscopes (STEM) provide sub-Angstrom beam sizes, high spatial coherence and fast frame rates. In principle, STEM imaging could enable characterization of local materials properties at unit-cell spatial resolutions. The fundamental challenge to a quantitative analysis of scattering data in the STEM, however, has remained the same for 80 years, namely an intractable inverse problem where dynamical scattering is prevalent. In this talk, we present a few examples where a deep learning approach to this inverse problem in the STEM was successfully applied and discuss its advantages and current limitations. First, we show that deep convolutional neural networks (DCNN), trained on multislice simulations, learn to accurately predict the 3-D oxygen octahedral rotations in complex oxides from annular bright-field (ABF) images, with sub-degree accuracy and unit-cell spatial resolutions. Second, we show that distributed deep learning, implemented on Oak Ridge National Lab's Summit supercomputer (and scaled to 10,000 GPUs) can successfully "invert" coherent convergent-beam electron diffraction (CBED) data to uncover the projected atomic potential for a limited set of material classes, with sub-Angstrom spatial resolutions.

WK1

The Materials Project: Conception to Confirmation in a Virtual Lab

Shyam Dwaraknath
Lawrence Berkeley National Laboratory, Berkeley, CA 94720

The materials project (MP) is an open searchable database of computed materials properties to enable a transition to materials by design. MP currently documents over 133,000 materials and a range of associated properties from the electronic structure, elastic tensor, dielectric tensors, piezoelectric tensors, phonons, substrates recommendations, and more. This rich data set enables scientists to search for materials with appropriate functionality and identify those that are the most likely to be thermodynamically stable both chemically and dynamically. Building on this, the local spectroscopy data initiative (LSDI) is an effort in MP to build a database of computed x-ray absorption spectroscopy (XAS) spectra and tools to enable researchers to investigate the characterizability of a material. Only a small percentage of the materials community can take full advantage of emergent capabilities at light sources.

Enabling broad access to simulated data for materials discovery will greatly increase the user-base at APS by allowing researchers to "experiment" *in silico*; greatly reducing the necessary expertise and resource risks. While this data-set provides a powerful first step in democratizing XAS, it also creates new challenges. For instance, the over 600,000 site-specific XAS spectra at LSDI makes spectral matching in a traditional human-in-the-loop fashion rather difficult. To solve these big-data challenges, the LSDI is also developing tools to enable integration into materials discovery pipelines and eventually enable automatic online identification of local environments.

WK1

High-throughput Computational X-ray Absorption Spectroscopy

John J. Rehr[1,2], Joshua J. Kas[1], Fernando D. Vila[1], Kiran Mathew[3], Chen Zheng[4], Donald Winston[5], Chi Chen[4], Yiming Chen[4], Alan Dozier[6], Shyam Dwaraknath[3], Shyue Ping Ong[4], and Kristin A. Persson[3]

[1] Department of Physics, University of Washington, Seattle, WA, 98195

[2] Department of Photon Science, SLAC National Accelerator Laboratory, Menlo Park, CA, 94025

[3] Department of Materials Science, University of California Berkeley, Berkeley, CA 94720

[4] Department of Nanoengineering, University of California San Diego, La Jolla, CA 92093

[5] Energy Technologies Area, Lawrence Berkeley National Laboratory, Berkeley, CA 94720

[6] Division of Applied Research and Technology, National Institute for Occupational Safety and Health, Centers for Disease Control, Cincinnati, OH 45226

Modern electronic structure theory and computational methods now permit efficient calculations of many ground state materials properties, as exemplified by results from the Materials Project [1]. Complementary advances in the theory of excited states have led to efficient methods for calculations of x-ray and electron spectroscopies (e.g., the real-space Green's multiple scattering theory in the FEFF9 code [2]). Here we discuss these developments and their application to high throughput calculations of x-ray absorption spectra [3]. In particular, a database of computed spectra has been developed which currently contains over 800,000 K-edge spectra for over 40,000 materials, which has also been used in ensemble-learned matching of near edge spectra [4]. The database is freely available from the Materials Project [1], and extensions to other spectra are in progress.

This work was a product of the Data Infrastructure Building Blocks (DIBBS) Local Spectroscopy Data Infrastructure project funded by NSF under Award 1640899. The FEFF code development is supported primarily by the DOE Office of Science, BES Grant DE-FG02-45623 (JJR,JJK,FDV). Computational resources were provided by the NSF DIBB funding and the Triton Shared Computing Cluster (TSCC) at the University of California, San Diego. The online data and search capabilities were funded by the DOE Office of Science, BES, MSE Division under Contract No. DE-AC02-05-CH11231: Materials Project program KC23MP.

[1] A. Jain et al. (2013). "The Materials Project: A materials genome approach to accelerating materials innovation," *APL Materials* **1**: 011002; www.materialsproject.org.

[2] John J. Rehr et al. (2009). "*Ab initio* theory and calculations of X-ray spectra," *Comptes Rendus Physique* **10**: 548; John J. Rehr et al. (2010). "Parameter-free calculations of X-ray spectra with FEFF9," *Physical Chemistry Chemical Physics* **12**: 5503.

[3] K. Matthew, et al. (2018). "High-throughput computational X-ray absorption spectroscopy," *Scientific Data* **5**: 180151.

[4] Chen Zheng et al. (2018). "Automated generation and ensemble-learned matching of X-ray absorption spectra," *npj Computational Materials* **4**: 12.

WK1

Combining Characterization and Modeling Data for the Determination of Nanoscale Structures

Maria Chan

Center for Nanoscale Materials, Argonne National Laboratory, Lemont, IL 60439

Determining the atomistic structures in the nanoscale is a challenging problem. Although there are both experimental and computational methods to determine these nanoscale structures, they both possess limitations. We developed the fully automated nanoscale to atomistic structure from theory and eXperiment (FANTASTX) code to overcome the limitations of either by combining both experimental and computational data, with the help of machine learning. The approach will be illustrated using x-ray and electron-microscopy data, combined with first principles density functional theory (DFT), or empirical force field (FF) guidance, and machine learning tools.

WK1

Learning Phase Retrieval with Backpropagation

Youssef S.G. Nashed[1], Saugat Kandel[2], Ming Du[3], and Chris Jacobsen[4,5]

[1] Mathematics and Computer Science Division, Argonne National Laboratory, Lemont, IL 60439

[2] Applied Physics, Northwestern University, Evanston, IL 60208

[3] Department of Materials Science and Engineering, Northwestern University, Evanston, IL 60208

[4] Department of Physics and Astronomy, Northwestern University, Evanston, IL 60208

[5] Advanced Photon Source, Argonne National Laboratory, Lemont, IL 60439

Synchrotron radiation light source facilities are leading the way to ultrahigh resolution x-ray imaging. High resolution imaging is essential to understanding the fundamental structure and interaction of materials at the smallest length scale possible. Coherent diffraction imaging (CDI) achieves nanoscale imaging by replacing traditional objective lenses by pixelated area detectors and computational image reconstruction. We present our work for solving CDI reconstruction problems through fitting a physics based model to measured data. The model parameters are learned in a similar manner to deep neural networks, utilizing the backpropagation method as implemented in Google TensorFlow package. This approach has advantages in terms of speed and accuracy compared to current state of the art algorithms, and demonstrates re-purposing the deep learning backpropagation algorithm to solve general phase retrieval problems that are prevalent in lensless microscopy research.

WK1

Thin Film Structure Identification through Convolutional Neural Networks Applied to Scattering Patterns

Shuai Liu[1,2], Charles N. Melton[1], Singanallur Venkatakrishnam[3], Ronald J. Pandolfi[1], Guillaume Freychet[1], Dinesh Kumar[1], Haoran Tang[2], Alexander Hexemer[1], and Daniela M. Ushizima[1,2]

[1] University of California, Berkeley, Berkeley, CA 94720

[2] Lawrence Berkeley National Laboratory, Berkeley, CA 94720

[3] Oak Ridge National Laboratory, Oakridge, TN 37830

Nano-structured thin films have a variety of applications from waveguides, gaseous sensors to piezoelectric devices. Among the methods to investigate such materials, grazing incidence small angle x-ray scattering (GISAXS) consists of a surface-sensitive technique that enables probing complex morphologies ranging from the fields of polymer and soft matter science, to hard-condensed matter. One challenge is to determine structure information from these scattering patterns alone. Our recent work showcase how GISAXS images work as signatures, which depend on the size, shape and arrangement of nanostructured components, ultimately enabling the classification of thin films.

The CNN-based classification scheme categorizes seven combinations of 3D nanoparticle lattice orientations from simulated x-ray scattering data based on observable features in scattering patterns. Training data are obtained using HipGISAXS [1] scattering simulator. Several million scattering patterns were generated for seven different nanoparticle crystal lattices at various orientations. We verify the robustness of the CNN by subjecting the training data to various noise sources that simulate typical data artifacts. This talk presents details about work published at the Materials Research Society Communications, Cambridge Press [2]. It describes the design of multiple Convolutional Neural Networks to classify nanoparticle orientation in a thin film by learning scattering patterns. The network was trained on several thin films with a success rate of 94%. We demonstrate CNN robustness under different artifacts as well as show the potential of our proposed approach as a strategy to decrease scattering pattern analysis time.

This work was supported by the Center of Advanced Mathematics for Energy Research Applications (CAMERA) through the Office of Science, of the U.S. Department of Energy under Contract No. DE-AC02-05CH11231 and the Early Career Program. This research is funded in part by the Gordon and Betty Moore Foundation through Grant GBMF3834 and by the Alfred P. Sloan Foundation through Grant 2013-10-27 to the University of California, Berkeley.

[1] S.T. Chourou, A. Sarje, X.S. Li, E.R.Chan, and A. Hexemer (2013). "HipGISAXS: a high performance computing code for simulating grazing-incidence x-ray scattering data," *J. Appl. Crystallogr.* **46**: 1781–1795. https://hipgisaxs.github.io/.

[2] S. Liu, C. Melton, S. Venkatakrishnam, R. Pandolfi, G. Freychet, D. Kumar, H. Tang, A. Hexemer, and D. Ushizima (2019). "Convolutional Neural Networks for Grazing Incidence X-ray Scattering Patterns: Thin Film Structure Identification," Special Issue on Artificial Intelligence, Materials Research Society, Cambridge Press (accepted).

Wednesday, May 8

WK2 Joint APS/CNM: Topological Quantum Information Science

Location: Building 401, Room A1100

Time: 9:00 – 5:30

Organizers: Yue Cao (MSD), Pierre Darancet (CNM), Nathan Guisinger (CNM), Jessica McChesney (APS), Subramanian Sankaranarayanan (CNM), Hua Zhou (APS)

The conceptual design and experimental observation of topological phases of matter have opened up new avenues for Quantum Information Science. In particular, topologically protected states can feature longer electron coherence time, and offer control over dissipation channels via the time and space group symmetry of materials.

Despite the tremendous success of the past decade in predicting, synthesizing, and characterizing topological materials, many topological phases of matter central to quantum information science, such as Majorana fermions and topological bosons have yet to be observed. Moreover, realistic application of topological materials will require protocols that precisely control the material properties responsible for the propagation of information (e.g., the flow of spin current, or the braiding of Majorana fermions). This workshop will address the main progress and challenges in the research of topological materials, and cover the entire life cycle of these studies, from computational design, experimental realization, to control methods and device schemes towards topological devices. Speakers will feature major players in all the aforementioned sub-areas of research.

9:00 – 9:15 Yue Cao (Argonne National Laboratory)
Opening Remarks

9:15 – 9:30 John Mitchell (Argonne National Laboratory)
Welcome

Session 1: Topological Materials: Where Do We Find Them? (Pierre Darancet)

9:30 – 10:05 Ivar Martin (Argonne National Laboratory)
Hamiltonian Engineering with Majorana Modes

10:05 – 10:40 Aash Clerk (University of Chicago)
Photonic and Bosonic Analogues of Topological Superconductors

10:40 – 11:00 Break

Session 2: Topological States in the Bulk (Nathan Guisinger)

11:00 – 11:35 John Mitchell (Argonne National Laboratory)
Putative Topological Features in $Co_{1/3}NbS_2$ and Pd_3Pb

11:35 – 12:10 Ni Ni (University of California, Los Angeles)
Experimental of Topological Materials

12:10 – 1:30 Lunch

Session 3: Artificial Topological States on the Surface and Interface (Hua Zhou)

1:30 – 2:05 Seongshik Oh (Rutgers, the State University of New Jersey)
Tunable Proximity-coupled Topological Superconductor Heterostructures for Quantum Computation

2:05 – 2:40 Tai Chiang (University of Illinois, Urbana-Champaign)
Playing with Topological Insulators: Superconductivity and Strain Effects

2:40 – 3:00 Break

Session 4: Identifying Fermionic and Bosonic Topological States (Jessica McChesney)

3:00 – 3:35 Liuyan Zhao (University of Michigan)
Magnetic Excitations in a Honeycomb Ferromagnet

3:35 – 3:35 Mark Dean (Brookhaven National Laboratory)
Observation of Double Weyl Phonons in Parity-breaking FeSi

4:10 – 4:45 Yong P. Chen (Purdue University)
Topological Insulator-based Quantum Devices: From Spin Batteries to Josephson Junctions

4:45 – 5:30 Subramanian Sankaranarayanan (Argonne National Laboratory)
General Discussion

5:30 Adjourn

WK2

Hamiltonian Engineering with Majorana Modes

Ivar Martin[1] and Kartiek Agarwal[2]

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

[2] Department of Physics, McGill University, Montrèal, Quèbec H3A 2T8, Canada

We discuss a new possibility of using braiding operations on Majoranas to engineer novel Hamiltonians. Our proposal is based on the topological robustness of the π phase acquired by Majoranas upon 2π-braiding operations. Repeated application of such braids can be stable to heating if the braiding is performed at high frequencies, by way of pre-thermalization, or by introducing disorder, by way of many-body localization. Repeated braiding can then be used to selectively eliminate certain terms in the Hamiltonian, or enhance others, can be used to engineer additional symmetries and thus realize novel symmetry protected topological phases. The protocol can also be used to stabilize qubits in quantum computers.

Work at Argonne National Laboratory was supported by the U.S. Department of Energy, Office of Science, Materials Science and Engineering Division.

WK2

Photonic and Bosonic Analogues of Topological Superconductors

Aashish Clerk

Institute of Molecular Engineering, University of Chicago, Chicago, IL 60637

Interest continues to grow in bosonic analogues of topological electronic phases realized using photonic or phononic degrees of freedom. Such devices have a range of potential applications, ranging from new kinds of non-reciprocal devices to sources of entangled photons. Such systems are however typically non-interacting, and have the same band structure and edge state structure as their fermionic counterparts. In this talk, I will discuss recent theory work in my group on a class of bosonic systems where this correspondence fails. They involve using parametric “two-photon” driving, and have Hamiltonians that superficially resemble those of topological superconductors. Among the surprising effects that emerge are the presence of topologically-protected instabilities that can be harnessed for non-reciprocal quantum amplification [1] and squeezing light generation, and effective non-Hermitian dynamics in a bosonic analogue of the Kitaev-Majorana chain [2]. I will discuss

how these ideas could be realized in a variety of different experimental platforms.

[1] V. Peano, M. Houde, F. Marquardt, and A.A. Clerk (2016). *Phys. Rev. X* **6**: 041026.

[2] A. McDonald, T. Pereg-Barnea, and A.A. Clerk (2018). *Phys. Rev. X* **8**: 041031.

WK2

Putative Topological Features in $Co_{1/3}NbS_2$ and Pd_3Pb

J.F. Mitchell[1], Nirmal Ghimire[1], Mojammel Khan[1], Antia Botana[2], and Sam Jiang[1]

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

[2] Department of Physics, Arizona State University, Tempe, AZ 85281

The recent realization of topological electronic states such as Dirac and Weyl fermions in real materials and their potential for future energy and electronics applications has motivated interest in the study of new forms of topological behavior embodied through new materials. We report here our recent experimental and theoretical work on two such materials, $Co_{1/3}NbS_2$ and Pd_3Pb.

Recently it has been shown that intrinsic part of the anomalous Hall effect (AHE) can be non-zero in non-collinear antiferromagnets as well as in topological materials hosting Weyl nodes near the Fermi energy. We have found a large anomalous Hall effect with Hall conductivity of 30 Ω^{-1} cm^{-1} in a collinear antiferromagnet $Co_{1/3}NbS_2$ with a non-centrosymmetric crystal structure. It orders below 27.5 K with the moments lying in the ab-plane. Magnetization measurement shows the presence of a small ferromagnetic component (0.0013 Bohr magnetons) along c-axis. The large AHE, observed only for H || c, is difficult to reconcile in light of the small ferromagnetic component. Here we will discuss potential alternative explanations of the AHE in $CoNb_3S_6$ framed in terms of topological band structure features or spin texture [1].

Pd_3Pb has been shown theoretically [2] to host unique topological features, including a dispersionless band near the Fermi level and triple nodal points hosting Dirac fermions and open Fermi arcs. Here, we report the crystal growth and electric transport properties of Pd_3Pb. Our low field magnetoresistance measurements indicate an anisotropic Fermi surface. We found that Pd_3Pb manifests a large transverse magnetoresistance, which reaches 650% at 1.8 K and 14 T, and pronounced Shubnikov-de Haas (SdH) oscillations. Preliminary analysis of the field dependence of the SdH oscillations points to the likelihood of nontrivial Berry phase in Pd_3Pb. Preliminary analysis of studies in high field limit will be presented to explore unique features of this unusual fermi surface [3].

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

[1] N.J. Ghimire et al. (2018). *Nature Communications* **9**: 3280.

[2] K-H Ahn et al. (2018). *Phys. Rev. B* **98**: 035130.

[3] N.J. Ghimire et al. *Phys. Rev. Materials* **2**: 081201.

WK2

Experimental Exploration of Topological Materials

Ni Ni

Physics and Astronomy, University of California, Los Angeles, Los Angeles, CA 90095

New materials are the driving force for technology innovations and our progressive understanding of condensed matter physics. In the last decade, breakthroughs have been made on 3D topological materials. The discovery of bulk materials with non-trivial topology has led to rich new emergent phenomena, including Fermi arc surface state, chiral pumping effect, colossal photovoltaic effect, quantum anomalous Hall effect, etc. In this talk, I will present the exciting progress made in our lab, ranging from the “hydrogen type” topological nodal line semimetal to magnetic topological insulators.

WK2

Tunable Proximity-coupled Topological Superconductor Heterostructures for Quantum Computation

Seongshik Oh

Department of Physics and Astronomy, Rutgers, the State University of New Jersey, Piscataway, NJ 08854

Since the seminal work by Fu and Kane in 2008, heterostructures composed of superconductors (SC) and topological insulators (TI) have been considered a promising platform for producing topological superconductivity (TSC) that can host Majorana particles, the key component for topological quantum computation (TQC). However, despite the relatively well-established theoretical ideas, there has been little progress experimentally and the current prospects to utilize TSC heterostructures for TQC are unclear. The main experimental problems have been the lack of reliable SC/TI materials platforms that have the proper control knobs. Even theoretically, it is only within a small parameter window where the SC/TI heterostructures can lead to the desired TSC phase with Majoranas via a proximity effect in the TI surface states. Therefore, in order to experimentally implement such ideas, it is essential to construct a materials platform where the key materials parameters such as the coupling strength between SC and TI and the Fermi level of TI are under full control. However,

such a materials platform has been so far lacking. Here, I will show that with a series of materials innovations [1–5], we have now come very close to implementing such tunable topological superconductor platforms. With this talk, I would like to open discussion on the prospects and limitations of such tunable topological heterostructures for topological quantum computation.

[1] L. Wu, ..., S. Oh, and N. P. Armitage (2016). "Quantized Faraday and Kerr rotation and axion electrodynamics of the surface states of three-dimensional topological insulators," *Science* **354**: 1124.

[2] M. Salehi, ..., S. Oh (2016). "Finite-Size and Composition-Driven Topological Phase Transition in (Bi1–xInx)2Se3 Thin Films," *Nano Lett.* **16**: 5528.

[3] J. Moon, ..., S. Oh (2018). "Solution to the hole-doping problem and tunable quantum Hall effect in Bi2Se3 thin films," *Nano Lett.* **18**: 820.

[4] P. P. Shibayev, ..., S. Oh (2019). "Engineering topological superlattices and phase diagrams," *Nano Lett.* **19**: 716.

[5] M. Salehi, ..., S. Oh (2019). "Quantum-Hall to insulator transition in ultra-low-carrier-density topological insulator films and a hidden phase of the zeroth Landau level," arXiv:1903.00489.

WK2

Playing with Topological Insulators: Superconductivity and Strain Effects

T.-C. Chiang

University of Illinois at Urbana-Champaign, Urbana, IL 61801

Topological insulators are semiconductors characterized by bulk band gap inversion often caused by strong spin-orbit coupling. By analytic continuation, this gap must close at the surface and reopen outside in vacuum where the gap is noninverted (and infinite). The gap closing at the surface results in metallic surface states, or topological states, which are spin-polarized and span the bulk gap. They carry a surface spin current, largely independent of the details of the surface, which is a feature of strong interest for spintronic applications. This talk will focus on thin films of a prototypical topological insulator Bi_2Se_3 that are (1) made superconducting by proximity coupling to a simple superconducting substrate or (2) strained by stretching a flexible plastic film substrate. Angle-resolved photoemission and x-ray diffraction were employed to determine the electronic structure and the lattice structure. A novel "flip-chip" technique was employed to prepare epitaxial films of Bi_2Se_3 on the superconducting or flexible plastic substrates. We show how the topological surface states are modified under these conditions. Effects of superconducting pairing, coherence, bulk-surface coupling, and electron-lattice coupling will be discussed.

WK2

Magnetic Excitations in a Honeycomb Ferromagnet

Liuyan Zhao

Department of Physics, University of Michigan, Ann Arbor, MI 48109

Two-dimensional (2D) honeycomb ferromagnets are predicted to host massless Dirac magnons because of the two equivalent magnetic sites per unit cell of the honeycomb lattice, mimicking Dirac electrons in graphene. More interestingly, the introduction of the next-nearest-neighbor Dzyaloshinskii-Moriya interaction breaks the sublattice equivalency and suggests the emergence of topological magnons in these honeycomb ferromagnets. In this talk, I will present polarization-resolved Raman spectroscopy studies on magnetic excitations in a honeycomb ferromagnet CrI_3, as a function of temperature, layer number, and magnetic field.

WK2

Observation of Double Weyl Phonons in Parity-breaking FeSi

H. Miao[1], T.T. Zhang[2,3], L. Wang[2,3], D. Meyers[1], A.H. Said[4], Y.L. Wang[1], Y.G. Shi[2], H.M. Weng[2,5], Z. Fang[2,5], and M.P.M. Dean[1]

[1] Condensed Matter Physics and Materials Science Department, Brookhaven National Laboratory, Upton, NY 11973

[2] Beijing National Laboratory for Condensed Matter Physics, Institute of Physics, Chinese Academy of Sciences, Beijing 100190, China

[3] University of Chinese Academy of Sciences, Beijing 100049, China

[4] Advanced Photon Source, Argonne National Laboratory, Lemont, IL 60439

[5] Collaborative Innovation Center of Quantum Matter, Beijing 100190, China

Condensed matter systems have now become a fertile ground to discover emerging topological quasiparticles with symmetry protected modes. While many studies have focused on fermionic excitations, the same conceptual framework can also be applied to bosons yielding new types of topological states. Motivated by Zhang et al.'s recent theoretical prediction of double Weyl phonons in transition metal monosilicides [*Phys. Rev. Lett.* **120**: 016401 (2018)], we directly measure the phonon dispersion in parity-breaking FeSi using inelastic x-ray scattering. By comparing the experimental data with theoretical calculations, we make the first observation of double Weyl points in FeSi, which will be an ideal material to explore emerging bosonic excitations and its topologically nontrivial properties [1].

H.M. and M.P.M.D. acknowledge A. Alexandradinata, C. Fang, L. Lu, and D. Mazzone for insightful discussions. This material is based upon work supported by the U.S. Department of Energy, Office of Basic Energy Sciences, Early Career Award Program under Award No. 1047478. Work at Brookhaven National Laboratory was supported by the U.S. Department of Energy, Office of Science, Office of Basic Energy Sciences under Contract No. DE-SC0012704.

The IXS experiments were performed at 30ID in the Advanced Photon Source, a U.S. Department of Energy (DOE) Office of Science User Facility operated for the DOE Office of Science by Argonne National Laboratory under Contract No. DE-AC02-06CH11357. We acknowledge the support from the National Key Research and Development Program of China (Grant No. 2016YFA0300600), the National Natural Science Foundation of China (Grants No. 11674369 and No. 11774399), and the Chinese Academy of Sciences (XDB07020100 and QYZDB-SSW-SLH043).

[1] H. Miao et al. (2018). *Phys. Rev. Lett.* **121**: 035302.

WK2
Topological Insulator Based Quantum Devices: From Spin Batteries to Josephson Junctions

Yong P. Chen
Purdue University, West Lafayette, IN 47907

Topological insulators (TI) feature insulating bulk and topologically conducting surfaces with “spin-helical” electrons that may enable a variety of topological quantum information devices. We have demonstrated spin-helical electronic transport [1,2] on the surface of TIs, and further discovered a “topological spin battery effect” [3], opening the possibility to electrically induce and readout a nuclear and electronic spin polarization with exceptionally long lifetime—which we present as a remarkable demonstration of the “topological protection” unique to TI. We further observe unusual behaviors in superconducting Josephson junctions and SQUIDs made out of our Tis [4,5], paving the way for using such spin-helical electrons to realize “topological superconductor” proposed to harbor “majorana fermions” that could enable scalable, topologically-protected quantum computing.

[1] J. Tian et al. (2015). “Electrical injection and detection of spin-polarized currents in topological insulator Bi2Te2Se,” *Sci. Rep.* **5**: 14293.

[2] J. Tian et al. (2019). “On the understanding of current-induced spin polarization of three-dimensional topological insulators,” *Nature Comm.* **10**: 1461.

[3] J. Tian et al. (2017). “Observation of current-induced, long-lived persistent spin polarization in a topological insulator: a rechargeable spin battery,” *Science Advances* **3**: e1602531.

[4] M. Kayyalha et al. (2019). “Anomalous low-temperature enhancement of supercurrent in topological-insulator nanoribbon Josephson junctions: evidence for low-energy Andreev bound states,” *PRL* **122**: 047003.

[5] M. Kayyalha et al. (2019). “Highly skewed current-phase relation in superconductor-topological insulator-superconductor Josephson junctions,” arXiv:1812.00499.

Tuesday, May 7

WK3 APS:
Workshop on Chemical Separations

Location: Building 401, A1100

Time: 8:45 – 5:00

Organizers: Lynne Soderholm (ANL) and Aurora Clark (Washington State)

Chemical-separations researchers represent an untapped user group for synchrotron facilities. X-ray photons offer a variety of opportunities for the chemical separations community, who are struggling with materials performance issues as they look to develop better, more efficient ways to refine natural resources such as petroleum, purify specialty chemicals from rare-earth ores, or deal with effective radioactive-waste disposal. To date researchers in this field have concentrated largely on in-house resources, despite the fact that their problems are on the forefront of some of the most interesting, cutting-edge challenges under study today. For example, most of their materials can be classified as soft matter, including structured solutions, membranes, polymers, and bio-inspired organics. Many systems are amorphous, with hierarchical structuring playing a largely unexplored role in processes of interest.

The opportunity for a mutually beneficial collaboration between chemical-separations researchers and synchrotron-facility personnel lays in answering fundamental questions of structure-function relationships in complex, evolving soft-matter systems. From a separations perspective, synchrotrons can provide information about structuring from local to mesoscopic length scales, providing here-to-fore unavailable insights into the free-energy drivers for these processes. Spectroscopic, scattering, and imaging studies such as those available at the APS would provide new windows into how these separations systems work. The impact of this work is further enhanced when combined with advanced molecular simulations methods that provide a bridge between the molecular level structure and ensemble average measured data. For example, very recent XPCS studies at the APS on the liquid-liquid phase transitions known to impede separations efficacy are resolving questions in favor of a micellar model over a simple biphasic model, thus revealing promising new avenues to resolve this problem. From the APS perspective, SAXS experiments can be used to quantify the structural changes occurring as the phase transition is approached by varying temperature. Dynamics studies reveal aggregate fluctuations on the order of 20 microseconds, just at the limit of current APS capabilities. These studies are informing APS efforts to prepare for the upgrade by providing opportunities to quantify issues ranging from detector-speeds to big-data analysis scenarios.

This workshop will bring together leaders in the fields of separation sciences, modeling and simulation, and machine learning with synchrotron experts in spectroscopy, scattering, and imaging to discuss mutual interests and set the groundwork for collaborative efforts that would benefit all communities involved. Expected outcomes include a roadmap for new ways to integrate these techniques to increase the breadth, applicability, and impact of APS experiments to a diverse and growing chemical-separations user base. Separations is acknowledged as an important impact area within the national research landscape, as demonstrated by the ongoing National Academy of Sciences committee study on A Research Agenda for a New Era of Separations Science.

8:45 – 9:00	Opening Remarks
9:00 – 9:30	Joan Brennecke (The University of Texas at Austin) *Ionic Liquids with Aprotic Heterocyclic Anions for Post-combustion CO_2 Capture*
9:30 – 10:00	Ahmet Uysal (Argonne National Laboratory) *A Necessary Engagement: Surface Science and Chemical Separations*
10:00 – 10:30	David A. Dixon (The University of Alabama) *Predicting the Properties of Actinide Complexes in the Gas Phase and in Solution*
10:30 – 10:45	Break
10:45 – 11:15	Eric Schelter (University of Pennsylvania) *Rare Earth Metals: Challenging Separations and Opportunities for Innovation*
11:15 – 11:45	Uta Ruett (Argonne National Laboratory) *How High-energy X-rays Can Provide Insight into Structure of Liquids*
11:45 – 12:15	Ilan Benjamin (University of California at Santa Cruz) *Surface-active Molecules Influence on Charge Transfer Reactions at Liquid Interfaces*
12:15 – 1:30	Lunch
1:30 – 2:00	Jenifer Shafer (Colorado School of Mines) *Extractant Aggregation in f-element Science: Current Understanding and New Opportunities*
2:00 – 2:30	Mark Antonio (Argonne National Laboratory) *A Telescoping View of Chemical Separations with Synchrotron Radiation*
2:30 – 3:00	David Wu (Colorado School of Mines) *Hierarchical Organization in Associated Structured Phases*
3:00 – 3:15	Break
3:15 – 3:45	Seth Darling (Argonne National Laboratory) *Interface Engineering in Separation Technologies for Water Treatment*
3:45 – 4:15	Michael Servis (Washington State University) *Surfactant-enhanced Heterogeneity of the Aqueous Interface Drives Water Extraction into Organic Solvents*
4:15 – 4:45	Discussion
4:45	Closing Remarks

WK3

Ionic Liquids with Aprotic *Heterocyclic* Anions for Post-combustion CO_2 Capture

Seungmin Oh[1], Tangqiumei Song[2], Gabriela M. Avelar Bonilla[1], Oscar Morales-Collazo[1], and Joan F. Brennecke[1]

[1] McKetta Department of Chemical Engineering, University of Texas at Austin, Austin, TX 78712

[2] Department of Chemical and Biomolecular Engineering, University of Notre Dame, Notre Dame, IN 46556

Ionic liquids (ILs) present intriguing possibilities for removal of carbon dioxide from a wide variety of different gas mixtures, including post-combustion flue gas, pre-combustion gases, air, and raw natural gas streams. Even by physical absorption, many ILs provide sufficient selectivity over N_2, O_2, CH_4 and other gases. However, when CO_2 partial pressures are low, the incorporation of functional groups to chemically react with the CO_2 can dramatically increase capacity, while maintaining or even enhancing selectivity. Towards this end, we have developed a series of ionic liquids that include reactive aprotic heterocyclic anions. By incorporating the amine on the anion, they react with CO_2 stoichiometrically (one mole of CO_2 per mole of IL), thus doubling the molar capacity compared to conventional aqueous amine solutions. Moreover, these ILs do not exhibit any viscosity increase upon complexation of the IL with CO_2, because the aprotic heterocyclic anions (AHA ILs) eliminate the pervasive hydrogen bonding and salt bridge formation that is the origin of the viscosity increase.

We have used x-ray scattering to study the structure of these ILs both neat and after reaction with CO_2. Interestingly, the liquid structure is insensitive to the specific anion and whether or not they are reacted with CO_2. We will also describe the discovery of AHA ILs whose melting points when reacted with CO_2 are more than 100°C below the melting point of the unreacted material. These materials allow one to dramatically reduce the energy required for CO_2 release and regeneration of the absorption material because a significant amount of the energy needed for the regeneration comes from the heat of fusion as the material releases CO_2 and turns from liquid to solid. Most recently, we have investigated the effect of encapsulating the AHA ILs in polymeric shells to combat the mass transfer challenges associated with the relatively high viscosity of ILs and show that they maintain their high capacity and can be cycled even in the presence of water.

WK3

A Necessary Engagement: Surface Science and Chemical Separations

Ahmet Uysal

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

Heavy elements, such as lanthanides and actinides, are very important to modern strategic technologies, including electronics, magnets, and nuclear energy. Therefore, refining and reprocessing of heavy elements in an efficient way is an important challenge, from obtaining the raw materials to handling the waste, during the industrial processes. Solvent extraction, the surfactant-aided preferential transfer of a species from an aqueous to an organic phase, is the most common technique used in heavy and precious metal separations. However, fundamental physical and chemical phenomena behind this process is still not well-understood.

Interfaces play a crucial role in solvent extraction, as the selective adsorption and transfer of ions happen at phase boundaries. Although electrostatics is the major factor determining the interfacial energetics landscape, non-Coulombic weak interactions as well as the interfacial solute and solvent structures are key to selective transfer of similarly charged and sized ions. Therefore, a predictive understanding of interfacial phenomena in heavy element separations require a molecular scale description of all these factors under process conditions, which presents experimental and theoretical challenges. For instance, most empirical and theoretical models of interfacial ion distributions are developed for lighter/smaller ions in low ionic strength solutions. Also, liquid surfaces/interfaces are difficult to access with most structural probes.

We address these challenges by combining state-of-the-art interfacial x-ray scattering and vibrational spectroscopy tools, integrated with molecular dynamics simulations. Interface specific x-ray reflectivity, grazing incidence diffraction, and x-ray fluorescence techniques provide structural information about interfacial ion distributions and ion-extractant structures in the most direct way possible [1]. Sum frequency generation spectroscopy measurements complement the interfacial picture presenting crucial information about interfacial water structures [2]. The molecular dynamics simulations provide a tangible understanding of experimental results [1,2]. With a combination of these methods we acquire a detailed understanding of energetics of ion adsorption at soft interfaces, and structural details of organization and reorganization of metal-amphiphile complexes that drive ion transfer. These studies are naturally integrated with our efforts on chemistry and speciation of heavy ions in aqueous solutions and metal-extractant structures formed in organic solutions.

This work is supported by the U.S. Department of Energy, BES-CSGB, Contract No. DE-AC02-06CH11357. The use of the Advanced Photon Source is also supported by U.S. DOE, BES. ChemMatCARS Sector 15 is supported by the National Science Foundation under grant number NSF/CHE-1346572.

[1] A. Uysal et al. (2017). *J. Phys. Chem. C* **121**(45): 25377–25383.

[2] W. Rock et al. (2018). *J. Phys. Chem. C* **122**(51): 29228–29236.

WK3

Predicting the Properties of Actinide Complexes in the Gas Phase and in Solution

David A. Dixon

Department of Chemistry and Biochemistry, University of Alabama, Tuscaloosa, AL 35487

There is a need to develop new approaches for separations for the actinides for the development of new nuclear reactors for efficient energy generation and for cleaning up the environment at nuclear weapons production sites. In addition, the actinides present a relatively less well-understood domain of chemistry and the presence of *5f* electrons can make the chemistry significantly different from that in other parts of the Periodic Table. Thus, there is substantial interest in understanding the properties and reactivity of molecules containing actinides, including their reactions in aqueous solution. We will describe how computational electronic structure theory can play a role in helping us to understand the actinides and the difficulties in employing such methods for the reliable prediction of the properties of the actinides, for example, the role of relativistic effects. Hydrolysis reactions are important in transformations of actinide ions and oxides into hydroxides. These processes impact environmental transport, nuclear fuel processing, and waste treatment and disposition.

We will describe applications to the hydrolysis chemistry of actinyl dioxide cations AnO_2^+ for An = Pa to Lr, including how their behavior changes with the number of *5f* electrons. The impact of anionic ligands on AnO_2^{2+} will also be discussed. The acidities of aqueous An = Th - Cf in different oxidation states have been calculated with multiple solvation shells and exhibit some interesting trends. These are the first available estimates of the pK_a's for some of these species. Issues in the design of new organic separation agents such as frustrated Lewis basic complexants will be discussed including the potential for separating actinides from lanthanides and issues in the design and synthesis of new complexants, for example solubility.

This work is supported by the U.S. Department of Energy Office of Science (BES) under the Heavy Element and Separations Programs.

WK3

Rare Earth Metals: Challenging Separations and Opportunities for Innovation

Eric J. Schelter

University of Pennsylvania, Philadelphia, PA 19104

The rare earth metals (La-Lu, Sc, and Y) comprise some of the most challenging inter-element separations. Countercurrent solvent extraction is the industry standard for achieving pure materials used applications ranging from permanent magnets to lighting phosphors. These conditions typically operate at or near equilibrium for a single pass extraction or stripping stem, and result in low selectivities. Work in our group has examined different modes of separation. Recently, we initiated a program to examine separations of rare earth metals by kinetic means. We have demonstrated that rate differences for chemical a chemical process tied to rare earth cations can provide means for separation. For example, we have used variable rates of oxidation reactions to partition late rare earth metals. Simple rate equation models also allow for prediction of separations factors. The presentation will highlight recent efforts in this area for the simplified separations of pairs of rare earth species, especially for applications in recycling rare earths from anthropogenic matrices.

WK3

How High-energy X-rays Can Provide Insight into Structure of Liquids

Uta Ruett and Olaf Borkiewicz

Advanced Photon Source, X-ray Science Division, Argonne National Laboratory, Lemont, IL 60439

High-energy x-rays above 50 keV photon energy offer unique possibilities for structural analysis of matter from only locally ordered liquids to long-range ordered single crystals. Systems in complex environments under realistic conditions can be studied because of the high penetration into matter, which is comparable to neutrons.

The Structural Science group at the APS is operating four beamlines with high-energy x-rays. The beamline 11-ID-B is dedicated to the study of nanocrystalline, amorphous and liquid matter and enables collection of total scattering data suitable for pair distribution function (PDF) analysis of wide range of materials. The beamline offers various *in situ* and *operando* capabilities, and can accommodate special sample environments provided by users.

We are continuously developing our capabilities towards multi-modal studies of materials. The station offers a DRIFT spectrometer for certain applications, and allows moderate SAXS measurements providing insights into the shape of particles, information complementary to the PDF. The

future options enabled by the upgrade of the APS will be outlined here.

This presentation will also discuss how the high-energy x-rays can contribute to the understanding of the mechanism of phase separation through structural analysis.

WK3

Surface-active Molecules Influence on Charge Transfer Reactions at Liquid Interfaces

Ilan Benjamin

Department of Chemistry and Biochemistry, University of California, Santa Cruz, CA 95064

Charge transfer reactions such as electron and ion transfer as well as reactions with significant difference in charge distribution between the reactants and products (such as S_N2 and ionization reactions) are very sensitive to the solvent polarity. The dynamics and thermodynamics of these reactions are strongly influenced by the heterogeneous character of the liquid/liquid and liquid/vapor interfaces where the polarity of the media changes rapidly over a sub-nanometer length scale and by surface density fluctuations. Surface-active molecules that are adsorbed at the interface and interacts with the reactants can provide a local reaction environment whose polarity is further distinguished from that of the neat interface. We examine how these surface-active molecules affect reactivity and how their effect is influenced by the local structure and dynamics of the interface.

WK3

Extractant Aggregation in F-element Science: Current Understanding and New Opportunities

M.J. Servis[1], A.G. Baldwin[1], D. Wu[1], A. Clark[2], and J.C. Shafer[1]

[1] Department of Chemistry, Colorado School of Mines, Golden, CO 80401

[2] Department Chemistry, Washington State University, Pullman, WA 99163

Supramolecular assembly has been integral to the management of used nuclear fuel since the initial, ether-based solvent extraction process used to purify uranium and attempted during the Manhattan project. Solvating extractants, which rely on supramolecular assembly, are an ideal partner for purifying the *f*-elements away from the rest of the periodic table. The general mechanism for metal recovery by a solvating reagent includes the interaction of a charge neutral, strong dipole amphiphile and charge balancing counter ions with a metal center to solubilize this metal center into the organic phase. This general mechanism is well depicted by a coordination chemistry model, where the metal sits in the center of the coordination complex and the amphiphile and counter-ions populate the metal coordination sphere, however this mechanism probably underestimates the importance of hydrogen bonding and other ion-dipole interactions that control intermediate (second and third coordination shell) and long (bulk material) range interactions. These interactions can control other important phenomena, such as third phase formation, and the combination of x-ray scattering techniques and molecular dynamics simulation is proving to be a powerful partnership in providing molecular level resolution with experimentally determined measurables. This presentation will be an overview of recent findings from this research group, as well as a review of the broader literature pertaining to solvating extractants and extractant aggregation, to highlight the opportunities for using beamline techniques to examine extraction mechanisms most relevant to *f*-element science.

WK3

A Telescoping View of Chemical Separations with Synchrotron Radiation

Mark R. Antonio

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

The advent of intense, tunable- and scanable-wavelength x-rays has revolutionized many disciplines in physical sciences and engineering. Although the separation science communities have been slow to embrace the myriad characterization capabilities available at synchrotron user facilities, it is not hyperbole to state that synchrotron x-radiation is advancing the field of chemical separations in ways that would be difficult, if not impossible, to realize with conventional tube sources. For example, in applications related to equilibrium separations based on differences in solute transfer across liquid-liquid and liquid-gas phase boundaries, the techniques of x-ray absorption spectroscopy and small-angle x-ray scattering are opening new vistas of understanding with unprecedented sensitivity and resolution. Examples from the literature of liquid-liquid extraction will be discussed to benchmark the role that synchrotron research plays in unraveling wickedly complex, fundamental issues in the profusion of separation methods used in industrial- and bench-scale process chemistry. The way forward includes the use of the full armamentarium of synchrotron instrumentation and techniques—scattering, spectroscopy, and imaging alike—to provide a transformative, telescoping view of separation science on interconnected length-, time-, and energy-scales. A multimodal, multiscale approach towards characterization of separations processes will provide a unified view of targeted solutes in equilibrium phases and at phase boundaries,

thereby enabling deliberate and rational design of high-performance systems by tailoring mass transfer processes in a hierarchical fashion in both space and time.

This work is supported by the U.S. Department of Energy, Office of Science, Office of Basic Energy Sciences, Division of Chemical Sciences, Biosciences and Geosciences, under Contract No. DE-AC02-06CH11357.

WK3
Hierarchical Organization in Associated Structured Phases

David T. Wu
Department of Chemistry and Department of Chemical and Biological Engineering, Colorado School of Mines, Golden, CO 80401

Chemical separations involve the partitioning of molecules according to differences in interactions, such as hydrophobic, hydrogen bonding, charge, polar, steric and ligand complexation interactions. From a condensed matter perspective, balance and frustration between such competing interactions, such as those that exist in amphiphilic surfactant systems or analogous frustrated magnetic systems, can lead to rich structured phases. Moreover, this competition at the molecular level often leads to structure at greater length scales, for instance, the formation of micelles or other extended mesophases that emerge from the association of surfactant molecules. The characterization and understanding of this higher-order structure can be relevant for chemical separation processes.

In this talk, I will present an overview of systems we have studied exhibiting such hierarchical organization:

- Cooperative association of methane and water molecules in controlling the nucleation of clathrate hydrate lattices
- Electric-field directed assembly of anisotropic colloids into complex phases
- Virus capsid protein assembly: autocatalysis and error-correction
- Association and aggregation of extractant/water/acid molecules into hydrogen-bonded networks in solvent extraction systems

I will highlight our use of simulation and theory, in close connection with experiment when possible, to understand both equilibrium and kinetic properties of these systems.

[1] M.R. Walsh, C.A. Koh, E.D. Sloan, A.K. Sum, and D.T. Wu (2009). "Microsecond Simulations of Spontaneous Methane Hydrate Nucleation and Growth," *Science* **326**: 1095–1098.

[2] F. Ma, D.T. Wu, and N. Wu (2013). "Formation of Colloidal Molecules Induced by Alternating-Current Electric Fields," *J. Am. Chem. Soc.* **135**: 7839–7842.

[3] F.D. Ma, S.J. Wang, D.T. Wu, and N. Wu (2015). "Electric-field-induced assembly and propulsion of chiral colloidal clusters," *P. Natl. Acad. Sci. USA* **112**: 6307–6312.

[4] G.L. Casini, D. Graham, D. Heine, R.L. Garcea, and D.T. Wu (2004). "*In vitro* papillomavirus capsid assembly analyzed by light scattering," *Virology* **325**: 320–327.

[5] M.J. Servis, D.T. Wu, and J.C. Braley (2017). "Network analysis and percolation transition in hydrogen bonded clusters: nitric acid and water extracted by tributyl phosphate," *Phys. Chem. Chem. Phys.* **19**: 11326–11339.

[6] M.J. Servis, D.T. Wu, J.C. Shafer, and A.E. Clark (2018). "Square supramolecular assemblies of uranyl complexes in organic solvents," *Chem. Commun. (Camb)* **54**: 10064–10067.

WK3
Interface Engineering in Separation Technologies for Water Treatment

Seth B. Darling
Institute for Molecular Engineering, Argonne National Laboratory, Lemont, IL 60439

Advanced Materials for Energy-Water Systems EFRC, Argonne National Laboratory, Lemont, IL 60439

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

Institute for Molecular Engineering, University of Chicago, Chicago, IL 60615

Driven by climate change, population growth, development, urbanization, and other factors, water crises represent the greatest global risk in the coming decades. Advances in materials represent a powerful tool to address many of these challenges. Understanding—and ultimately controlling—interfaces between materials and water are pivotal [1]. In this presentation, Dr. Darling will lay out the challenges and present several examples of work in his group based on materials science strategies for addressing applications in water. In each instance, manipulation of interfacial properties provides novel functionality, ranging from selective transport to energy transduction to pollution mitigation.

This work was supported as part of the Advanced Materials for Energy-Water Systems (AMEWS) Center, an Energy Frontier Research Center funded by the U.S. Department of Energy, Office of Science, Basic Energy Sciences. Use of the Center for Nanoscale Materials, an Office of Science user facility, was supported by the U.S. Department

of Energy, Office of Science, Office of Basic Energy Sciences, under Contract No. DE-AC02-06CH11357.

[1] *J. Appl. Phys.* **124**: 030901 (2018).

WK3

Surfactant-enhanced Heterogeneity of the Aqueous Interface Drives Water Extraction into Organic Solvents

Michael J. Servis[1] and Aurora E. Clark[1,2]

[1] Department of Chemistry, Washington State University, Pullman, WA 99163

[2]Pacific Northwest National Laboratory, Richland, WA 99352

Liquid/liquid interfaces are essential to certain chemical separations processes, such as solvent extraction. In solvent extraction, an amphiphilic extractant molecule forms complexes with aqueous-soluble molecules at the liquid/liquid interface to extract those aqueous molecules into an organic solvent. Despite the importance of those complexation reactions on the underlying kinetics of solvent extraction, understanding of the molecular structure at the interface is lacking in large part due to the challenges associated with probing the interfacial environment with experimental approaches such as x-ray reflectivity. Many of those challenges stem from molecular-scale thermal fluctuations of the interface, which impart structural heterogeneity across the aqueous surface. That heterogeneity is further enhanced by the presence of the surface-active extractant molecules.

Here, molecular dynamics simulations are applied to investigate the structural characteristics of the water/extractant hydrogen bonded network at the interface in response to extractant adsorption. Additionally, a mechanism by which the extractant transports water from the aqueous interface into the organic phase is identified. A graph theory topological analysis method is developed to quantify the role of hydrogen bonded clusters of water and extractant protruding from the aqueous surface in the water transport process. The chemical insight and analysis methodology presented here may be broadly applied to study molecular structure and reactions at surfactant-laden liquid/liquid interfaces and assist in interpretation of experimental data.

Tuesday, May 7

WK4 CNM:
Photon Qubit Entanglement and Transduction

Location: Building 401, Room A5000

Time: 8:30 – 5:00

Organizers: Xuedan Ma, Xufeng Zhang, and Stephen Gray (all CNM)

Various quantum systems that are suitable for specific applications have been developed in the past few years based on a broad range of physical platforms. To further exploit the advantages of quantum technology for complicated tasks in computation, communication, and sensing, it has becoming increasingly pressing to develop distributed quantum networks that combine the advantages of different quantum systems. Such distributed quantum networks require the development of individual quantum systems that operate in both the microwave and optical domains, as well as high fidelity transductions among different systems to interconnect quantum information of different formats. Despite of the fast development in microwave qubits based on superconducting technologies, deterministic optical single-photon sources and the related optical photonic components are still underdeveloped, which, as a result, hinders the development of long-distance quantum links. On the other hand, high-efficiency quantum interconnections between superconducting and optical systems are also missing because of the intrinsic incompatibility of the superconducting technique with optical photons and their huge frequency difference, which pose a significant challenge in encoding/decoding the microwave quantum information to the optical quantum link.

In this workshop, we aim to bring together researchers working on various aspects of photon qubits and quantum transduction systems to discuss pathways towards addressing these grand challenges faced by the quantum network community. This is a highly interdisciplinary research topic that involves efforts from a broad community including quantum optics, superconducting qubits, optomechanics. Effective communication among researchers of different backgrounds will greatly help to explore novel opportunities for designing and implementing photon qubit entanglement and transduction. It will also help create synergies between Argonne/CNM and the worldwide scientific community. With CNM's efforts towards developing a high standard user facility to support the increasing need for quantum infrastructure, this workshop will be a great opportunity for outreaching to potential users and obtaining valuable feedbacks.

Main topics in this workshop include:

- Quantum photonic materials and devices
- Novel quantum transduction approaches and material systems
- Photonic quantum networks and links
- Quantum repeaters and memories

Session 1: Quantum Optics and Nanophotonics (Session Chair: Xuedan Ma)

8:30 – 9:15 Qiang Lin (University of Rochester)
Manipulating Quantum States of Photons on Integrated Photonic Chips

9:15 – 10:00 Virginia Lorenz (University of Illinois, Urbana-Champaign)
Engineering Photonic Quantum States for Quantum Applications

10:00 – 10:30 Break

10:30 – 11:15 Michael Reimer (University of Waterloo)
On-demand Generation of Bright Entangled Photon Pairs

11:15 – 12:00 German Kolmakov (NYC College of Technology, CUNY)
Optical Detection and Storage of Entanglement in Plasmonically Coupled Quantum-dot Qubits

12:00 – 1:30 Lunch

1:30 – 2:15 Nick Vamivakas (University of Rochester)
Quantum Optics with Atomically Thin Materials

Session 2: Quantum Transduction Systems (Session Chair: Xufeng Zhang)

2:15 – 3:00 Hailin Wang (University of Oregon)
Mechanically Mediated Quantum Networks of Spins in Diamond

3:00 – 3:30 Break

3:30 – 4:15 Hoi-Kwan Lau (The University of Chicago)
High-fidelity Bosonic Quantum State Transfer Using Imperfect Transducers and Interference

4:15 – 5:00 Miguel Levy (Michigan Technological University)
Faraday Effect Enhancement in Nanoscale Iron Garnet Films

5:00 Adjourn

WK4

Manipulating Quantum States of Photons on Integrated Photonic Chips

Qiang Lin
Department of Electrical and Computer Engineering, University of Rochester, Rochester, NY 14627

Recent advances in quantum photonics have resulted in broad applications ranging from secure communication, metrology, sensing, to advanced computing. Chip-scale implementation would not only enhance the complexity and capacity of information processing, but also enable novel functionalities, which are otherwise inaccessible in room-wide/table-top experiments.

Lying in the heart of these applications is the capability of generating and manipulating versatile high-purity entangled photonic quantum states. In this talk, we will discuss our recent effort in engineering micro/nanophotonic device structures for producing and manipulating photonic quantum states on various chip-scale device platforms, by taking advantage of enhanced four-wave mixing and parametric down conversion processes, via the second-order and third-order optical nonlinearities.

WK4

Engineering Photonic Quantum States for Quantum Applications

Virginia O. Lorenz
Department of Physics, University of Illinois at Urbana-Champaign, Urbana, IL 61801

Photonic quantum states are good carriers of quantum information because they are robust to environmental fluctuations, but generating photons with just the right properties for specific applications is still a challenge. I will present our work on generating, engineering and characterizing photonic quantum states for quantum applications, including our demonstrations of novel photon-pair sources, bi- and tri-partite entangled states, and efficient characterization techniques.

WK4

On-demand Generation of Bright Entangled Photon Pairs

M.E. Reimer

Institute for Quantum Computing, and Department of Electrical and Computer Engineering, University of Waterloo, Waterloo, ON N2L 3G1, Canada

The on-demand generation of bright entangled photon pairs is an essential resource in quantum optics, quantum communication and quantum sensing. However, a bright entangled photon source with near-unity fidelity and efficiency is currently lacking. In this talk, I will present the generation of dephasing-free entangled photon pairs from a nanowire quantum dot with high brightness and collection efficiency [1]. We prove through our research that it is possible to reach perfect entanglement fidelity with current technology by also considering the detection process *in addition to* the generation process, even in an indium rich quantum dot with a large nuclear spin.

Finally, I will present two approaches to reach the perfect entangled photon source with near-unity fidelity and efficiency. These two approaches rely on the need to remove the fine structure splitting of the intermediate exciton states in the biexciton-exciton cascade as a finite fine structure splitting has been shown to degrade the entanglement fidelity. First, I will discuss our novel gating strategy with a quadrupole electrostatic potential, which shows that the fine structure splitting can be erased for any quantum dot dipole orientation without comprising the quantum dot brightness [2]. Second, I will present our all-optical approach to compensate the fine structure splitting by employing a fast rotating waveplate emulated by a high frequency shifter [3]. This latter approach has the unique advantage that the fine structure splitting of quantum dots in photonic nanostructures such as nanowires and micropillars can be directly compensated for without the need for further sample processing.

In this presentation, I will discuss these two important points: dephasing free entangled photons and no fine structure splitting, both of which lead us further towards the perfect source of entangled photons. With this work, we make great strides forward to transition out of the lab into practical and powerful real-world applications, such as information security for the day-to-day user as well as quantum radar which positively influences strategies of national defence.

[1] A. Fognini et al. (2017). "Path to perfect photon entanglement with a quantum dot," arXiv:1710.10815.

[2] M. Zeeshan et al. (2018). "Quadrupole electric field for erasing the fine structure splitting in a single quantum dot," arXiv:1809.02538.

[3] A. Fognini et al. (2018). "Universal fine-structure eraser for quantum dots," *Opt. Express* **26**: 24487–24496.

WK4

Optical Detection and Storage of Entanglement in Plasmonically Coupled Quantum-dot Qubits

Matthew Otten[1], Stephen K. Gray[1], and German V. Kolmakov[2]

[1] Center for Nanoscale Materials, Argonne National Laboratory, Lemont, IL 60439

[2] Physics Department, New York City College of Technology, City University of New York, Brooklyn, NY 11201

Recent proposals and advances in quantum simulations, quantum cryptography, and quantum communications substantially rely on quantum entanglement formation. Contrary to the conventional wisdom that dissipation destroys quantum coherence, coupling with a dissipative environment can also generate entanglement. We consider a system composed of two quantum-dot qubits coupled with a common, damped surface plasmon mode; each quantum dot is also coupled to a separate photonic cavity mode. Cavity quantum electrodynamics calculations [1] show that upon optical excitation by a femtosecond laser pulse, entanglement of the quantum-dot excitons occurs, and the time evolution of the $g^{(2)}$ pair correlation function of the cavity photons is an indicator of the entanglement. We also show that the degree of entanglement is conserved during the time evolution of the system. Furthermore, if coupling of the photonic cavity and quantum-dot modes is large enough, the quantum-dot entanglement can be transferred to the cavity modes to increase the overall entanglement lifetime. This latter phenomenon can be viewed as a signature of entangled, long-lived quantum-dot exciton-polariton formation. The preservation of total entanglement in the strong-coupling limit of the cavity–quantum-dot interactions suggests a novel means of entanglement storage and manipulation in high-quality optical cavities

The authors thank Xuedan Ma for helpful discussions. G.V.K. gratefully acknowledges support from the U.S. Department of Energy, Office of Science, Office of Workforce Development for Teachers and Scientists (WDTS) under the Visiting Faculty Program (VFP). G.V.K. is also grateful to the U.S. Department of Defense for partial support under Contract No. W911NF1810433. This work was performed, in part, at the Center for Nanoscale Materials, a U.S. Department of Energy Office of Science User Facility, and supported by the U.S. Department of Energy, Office of Science, under Contract No. DE-AC02-06CH11357.

[1] M. Otten, S.K. Gray, and G.V. Kolmakov (2019). "Optical detection and storage of entanglement in plasmonically coupled quantum-dot qubits," *Physical Review A* **99**(3): 032339.

WK4

Quantum Optics with Atomically Thin Materials

A. Nick Vamivakas

The Institute of Optics, University of Rochester, Rochester, NY 14627

Department of Physics, University of Rochester, Rochester, NY 14627

Center for Coherence and Quantum Optics, University of Rochester, Rochester, NY 14627

Two-dimensional, atomically-thin, materials have received enormous interest as a result of their unique mechanical, electrical and optical properties. Particularly exciting are the transition metal dichalcogenides—atomically-thin semiconductors that possess an electronic band gap in the visible. Although these materials have been investigated for applications in opto-electronics, not much work has focused on these systems as a platform for quantum photonics and quantum optics.

In this talk, I will describe two approaches that leverage atomically thin semiconductors, and other two-dimensional materials, assembled in layered van der Waals heterostructures for applications in these areas. In the first part of the talk, I will describe the unique photophysical properties of quantum emitters hosted by single layer transition metal dichalcogenides. I will describe our recent efforts to control the confined excitons and subsequent single photon generation. Finally, I will report on the observation of the coherent evolution of quantum emitters in the insulator hexagonal boron nitride.

WK4

Mechanically-mediated Quantum Networks of Spins in Diamond

Hailin Wang

Department of Physics, University of Oregon, Eugene, OR 97403

Phonons, which are the quanta of mechanical waves, are immune to scattering loss into vacuum and can couple to a variety of quantum systems. These distinct properties can enable new experimental platforms for quantum computing and quantum communication. In this talk, I will discuss the design and experimental implementation of a phononic quantum network of spins, in which individual spin qubits in diamond couple to a network of nanomechanical oscillators. I will also consider a spin-photon interface that explores quantum transduction between spins and photons through their coupling to a common mechanical oscillator. These mechanically-mediated networks can overcome or circumvent many of the inherent difficulties of direct optical networks of spins qubits.

WK4

High-fidelity Bosonic Quantum State Transfer Using Imperfect Transducers and Interference

Hoi-Kwan Lau and Aashish A. Clerk

Institute for Molecular Engineering, University of Chicago, Chicago, IL 60637

We consider imperfect two-mode bosonic quantum transducers that cannot completely transfer an initial source-system quantum state due to insufficient coupling strength or other Hamiltonian non-idealities. We show that such transducers can generically be made perfect by using interference and phase-sensitive amplification. Our approach is based on the realization that a particular kind of imperfect transducer (one which implements a swapped quantum non-demolition (QND) gate) can be made into a perfect one-way transducer using feed-forward and/or injected squeezing. We show that a generic imperfect transducer can be reduced to this case by repeating the imperfect transduction operation twice, interspersed with amplification. Crucially, our scheme only requires the ability to implement squeezing operations and/or homodyne measurement on one of the two modes involved. It is thus ideally suited to schemes where there is an asymmetry in the ability to control the two coupled systems (e.g., -to-optics quantum state transfer). We also discuss a correction protocol that requires no injected squeezing and/or feed-forward operation.

Hoi-Kwan Lau and Aashish A. Clerk (2019).
npj Quantum Information **5**: 31.

WK4

Faraday Effect Enhancement in Nanoscale Iron Garnet Films

Miguel Levy[1], Olga Borovkova[2], and Vladimir Belotelov[2]

[1] Michigan Technological University, Physics Department, Houghton, MI 49931

[2] Russian Quantum Center, Moscow, Russia

This presentation reports on a newly discovered enhancement mechanism of the magneto-optic Faraday Effect in nanoscale bismuth-substituted iron garnet films. Iron garnets are widely used in the fabrication of optical isolators, circulators and modulators because of the nonreciprocal nature of this effect.

In the last quarter of the 20th century, elemental substitutions were found to enhance the nonreciprocal response of these systems, and advances in isolator device on-chip integration made extensive used of this technology, especially for telecom applications. However, no further elemental substitutions have been found to yield comparable improvement in recent decades. Other

approaches such as photonic-crystal and ring resonator techniques were developed since, but at the expense of optical bandwidth. Present day industrial need for nanoscale nonreciprocal photonic on-chip and broad band devices in the telecom regime thus call for further advances in magneto-optical functional strength.

The work we present in this talk reports on an as yet untapped approach to nonreciprocal functional magnification. The methodology does not rely on resonant effects or elemental substitution mechanisms, but rather on symmetry-breaking at the surface. A seven-fold magnification of the magneto-optic gyrotropy parameter responsible for the Faraday Effect is observed, occurring within a few monolayers of the iron garnet crystal surface. Rigorous electromagnetic calculations show that classical theory cannot account for the observe phenomena, pointing to fundamental departures from the classical magneto-optic response.

Wednesday, May 8

WK5 APS:
RIXS after the APS Upgrade: Science Opportunities

Location: Building 402, Room E1100/1200

Time: 8:30 – 5:00 followed by a one-hour beamline tour

Organizers: Diego Casa and Jungho Kim (Argonne National Laboratory)

For resonant inelastic x-ray scattering (RIXS) at beamline 27-ID, the APS upgrade offers unprecedented opportunities for enhancements of the technique and the beamline, which will greatly benefit users of the facility. The vastly increased brightness of the x-ray source and a higher overall flux will enable ultra-high-resolution measurements (<10meV) that are not feasible today. These experiments will require both smaller focal spot sizes (<5um) and tighter incident energy bandpasses (<10meV).

A novel prototype RIXS flat-crystal spectrometer was recently implemented at beamline 27-ID at the Advanced Photon Source in anticipation of the upgraded source. It established a new record resolution for RIXS below 10 meV, together with a promise to do even better soon. To fully harness the benefits of such a vastly improved source and optics, parallel improvements to beamline instrumentation will include a more precise (<10um sphere-of-confusion) and efficient spectrometer, a more flexible high-resolution tunable monochromator with selectable bandpass down to ~5meV, and crucially practical implementations of meaningful in situ sample environments, including high-pressure and strong magnetic fields.

The aim of this workshop is to bring together the RIXS community to chart the roadmap ahead, considering how the technical upgrades can enable advances on the many exciting scientific opportunities from materials science from both practical and fundamental perspectives. In addition to presentations from our user community on forefront RIXS science, a review of RIXS experimental considerations and design, along with RIXS data simulation, numerical methods and analysis will be offered, culminating in an open session and a beamline tour.

8:30 – 8:40 Thomas Gog (Argonne National Laboratory)
Opening Remarks

8:40 – 9:20 Jeroen van den Brink (Institute for Theoretical Solid State Physics, IFW Dresden)
Resonant Inelastic X-ray Scattering on High-Tc Cuprates and Magnetic Iridates

9:20 – 10:00 Michel van Veenendaal (Northern Illinois University)
Non-equilbrium X-ray Spectroscopy in Fe-Co Prussian Blue Analogue Spin-crossover Complex

10:00 – 10:30 Break

10:30 – 11:00 Gábor B. Halász (Oak Ridge National Laboratory)
Probing Quantum Spin Liquids with Resonant Inelastic X-ray scattering

11:00 – 11:30 Yilin Wang (Brookhaven National Laboratory)
EDRIXS: An Open Source Toolkit for Simulating Spectra of Resonant Inelastic X-ray Scattering

11:30 – 12:00 Diego Casa and Ayman Said (Argonne National Laboratory)
Proposed RIXS Instrumentation

12:00 – 1:30 Lunch

1:30 – 2:00 Jason Hancock (University of Connecticut)
Local and Itinerant Electronic Structure in Rare-earth Intermetallics

2:00 – 2:30 Alex Frano (University of California, San Diego)
RIXS with In Situ *Magnetic Field: Towards Understanding the Complex Magnetism of Li_2IrO_3*

2:30 – 3:00 Gilberto Fernandes Lopes Fabbris (Argonne National Laboratory)
RIXS at High Pressure: Challenges and Opportunities

3:00 – 3:30 Break

3:30 – 4:00 Wentao Jin (University of Toronto)
RIXS Study of Sr_2IrO_4 Single Crystal under Uniaxial Strain

4:00 – 4:30 Ryan Hadt (California Institute of Technology)
Applications of Resonant X-ray Emission and Inelastic X-ray Scattering in Chemistry and Biology

4:30 – 4:55 Jungho Kim (Argonne National Laboratory)
Open session

4:55 – 5:00 Thomas Gog (Argonne National Laboratory)
Wrap-up

5:00 Mary Upton (Argonne National Laboratory)
27-ID RIXS Beamline tour

WK5

Resonant Inelastic X-ray Scattering on High-Tc Cuprates and Magnetic Iridates

Jeroen van den Brink
Institute for Theoretical Solid State Physics, Leibniz Institute for Solid State and Materials Research Dresden Helmholtzstraße 20, 01069 Dresden, Germany

Department of Physics, Washington University, St. Louis, MO 63130

Resonant inelastic x-ray scattering (RIXS) provides direct access to elementary charge, spin and orbital excitations in complex oxides. As a technique, it has made tremendous progress with the advent high-brilliance synchrotron x-ray sources. From the theoretical perspective, a fundamental question is to precisely which low-energy correlation functions RIXS is sensitive. Depending on the experimental RIXS setup, the measured charge dynamics can include charge-transfer, phonon, d-d, and orbital excitations. The focus of this talk will be on RIXS as a probe of spin dynamics and the electron-phonon coupling in high-Tc cuprates and the combined magnetic and orbital modes in strongly spin-orbit coupled iridium-oxides.

WK5

Nonequilbrium X-ray Spectroscopy in Fe-Co Prussian Blue Analogue Spin-crossover Complex

M. van Veenendaal
Northern Illinois University, DeKalb, IL 60115

Nonequilibrium x-ray spectroscopy is studied for a model spin crossover compound. The ultrafast spincrossover is calculated for Fe-Co Prussian blue analogues using a dissipative quantum-mechanical model of a cobalt ion coupled to a breathing mode [1,2]. All electronic interactions are treated on an equal footing. The divalent cobalt ion reaches 90% of the $S = 3/2$ value within 20 fs after photoexciting a low-spin Co^{3+} ion by an iron-to-cobalt charge transfer. The doublet-to-quartet spin crossover is significantly faster than the oscillation period of the breathing mode. The system relaxes to the lowest manifold of divalent cobalt ($^4T_1$) in 150–200 fs. Resonant inelastic x-ray scattering and x-ray absorption are calculated from the time-dependent nonequilibrium wavefunction. The resulting spectra cannot be decomposed into incoherent superpositions of transient states. Oscillations in the spin-orbit coupling gives rise to strong variations in the isotropic branching ratio. The RIXS shows crystal field oscillations and strong anti-Stokes effects.

[1] M. van Veenendaal (2017). *Scientific Reports* **7**: 6672.

[2] M. van Veenendaal (2018). *Phys. Rev. B* **97**: 125108.

WK5

Probing Quantum Spin Liquids with Resonant Inelastic X-ray Scattering

Gábor B. Halász

Oak Ridge National Laboratory, Oak Ridge, TN 37830

It is difficult to identify quantum spin liquids in experimental candidate materials due to the lack of direct "smoking gun" signatures. We propose that resonant inelastic x-ray scattering (RIXS) is an effective probe to detect spin-liquid physics in potential material incarnations of the Kitaev spin liquid (such as the honeycomb iridates and α-$RuCl_3$). Different RIXS channels pick up the fractionalized excitations of the Kitaev spin liquid separately and, in particular, the non-spin-flip channel can probe the characteristic momentum dispersion of the gapless Majorana excitations. As a signature of symmetry fractionalization, the corresponding RIXS response is suppressed around the Γ point of the Brillouin zone.

WK5

EDRIXS: An Open Source Toolkit for Simulating Spectra of Resonant Inelastic X-ray Scattering

Y.L. Wang[1], G. Fabbris[1], M.P.M. Dean[1], and G. Kotliar[1,2]

[1] Department of Condensed Matter Physics and Materials Science, Brookhaven National Laboratory, Upton, NY 11973

[2] Department of Physics and Astronomy, Rutgers University, Piscataway, NJ 08856

In this talk, we present an open source toolkit (dubbed EDRIXS) [1] to facilitate the simulations of RIXS spectra of strongly correlated materials based on exact diagonalization (ED) of certain model Hamiltonians. The model Hamiltonian can be from a single atom, small cluster or Anderson impurity model, with model parameters from density functional theory plus Wannier90 or dynamical mean-field theory calculations. The spectra of x-ray absorption spectroscopy (XAS) and RIXS are then calculated using Krylov subspace techniques. This toolkit contains highly efficient ED, XAS and RIXS solvers written in modern Fortran 90 language and a convenient Python library used to prepare inputs and set up calculations. We first give a short introduction to RIXS spectroscopy, and then we discuss the implementation details of this toolkit. Finally, we show several examples [2] to demonstrate its usage.

[1] Y.L. Wang, G. Fabbris, M.P.M. Dean, and G. Kotliar, arXiv:1812.05735.

[2] Y.L. Wang, Ruitang Wang, Jungho Kim, M.H. Upton, D. Casa, T. Gog, G. Cao, G. Kotliar, M.P.M. Dean, and X. Liu, arXiv:1810.05243. Accepted by PRL.

WK5

Local and Itinerant Electronic Structure in Rare-earth Intermetallics

Jason N. Hancock[1], Donal Sheets[1], Maxim Dzero[2], Daniel Mazzone[3], and Ignace Jarrige[3]

[1] Department of Physics and Institute for Material Science, University of Connecticut, Storrs, CT 06269

[2] NSLS-II, Brookhaven National Laboratory, Upton, NY 11973

[3] Department of Physics, Kent State University, Kent, OH 44242

Rare earth materials represent the ultimate limit of electronic correlations and present a rich landscape for exploring phenomena in electron-correlated and topological quantum matter. Central to the physics of these materials is the admixture of strongly correlated and localized magnetic degrees of freedom with itinerant states across the electronic bandwidth. Resonant spectroscopies like resonant inelastic x-ray scattering present unique opportunities to control and direct interrogation of quantum wavefunctions between the local and itinerant limits. In this talk, we will show how progress in this direction has been evolving over the past decade with strong support from synchrotron endstation improvements at the APS and around the world. In particular, we will show results at the rare-earth *L* edges crossing the valence transitions in $YbInCu_4$ and $Sm_{1-x}Y_xS$, with complementary work at the soft x-ray *M* edges of these and related materials. Together, these science-driven benchmarks in discovery make a strong and compelling case to support upgrades such as the APS-U project and show the synergy between the user community and pioneers of instrumentation at the APS can strengthen old and usher new activity areas in science.

[1] Donal Sheets et al., submitted 2019.

[2] J.N. Hancock, M. Dzero, M. Guarise, M. Grioni, J. Sarrao, and T. Schmitt (2018). "Kondo lattice excitation observed using resonant inelastic x-ray scattering at the Yb M5 edge," *Phys. Rev. B* **98**: 075158.

[3] J.N. Hancock and I. Jarrige (2016). "The promise of resonant inelastic x-ray scattering for f-electron materials," *Journal of Magnetism and Magnetic Materials* **400**: 41.

[4] I. Jarrige, A. Kotani, H. Yamaoka, N. Tsujii, K. Ishii, M. Upton, D. Casa, J. Kim, T. Gog, and J.N. Hancock (2015). "Kondo interactions from band reconstruction in $YbInCu_4$," *Physical Review Letters* **114**: 126401.

WK5

RIXS with *in situ* Magnetic Field: Towards Understanding the Complex Magnetism of Li_2IrO_3

Alejandro Ruiz[1], Anthony Allen[1], Vikram Nagarajan[2], Isaac Zinda[3], Jake Koralek[4], Mary Upton[5], Jungho Kim[5], Nicholas Breznay[3], James Analytis[2], Diego Casa[5], and Alex Frañó[1]

[1] Department of Physics, University of California, San Diego, La Jolla, CA 92093

[2] Department of Physics, University of California, Berkeley, Berkeley, CA 94720

[3] Department of Physics, Harvey Mudd College, Claremont, CA 91711

[4] SLAC National Accelerator Laboratory, Menlo Park, CA 94025

[5] Advanced Photon Source, Argonne National Laboratory, Lemont, IL 60439

Unconventional magnetism appears in correlated electron systems with strong spin-orbit coupling. 5d iridate compounds coordinated in a honeycomb lattice like Li_2IrO_3 are a promising avenue to realize the peculiar anisotropic, frustrated magnetism scenario of Kitaev's Hamiltonian [1,2] which could host a quantum spin liquid ground state. Experimentally, the material displays a degeneracy of magnetic ground states upon the application of a magnetic field [3]: one incommensurate spiral and a commensurate 'zigzag' phase. The mechanism by which the magnetic order and the associated low-energy excitations vary with magnetic field is unknown. Measuring the excitation spectrum associated with each of the two magnetic phases is challenging for two reasons: the energy scale of said excitations is low and thus at the limit of experimental resolution of RIXS spectrometers, and the application of in-situ magnetic fields within RIXS chambers is highly challenging.

Using the state-of-the-art RIXS spectrometer at Sector 27 of the Advanced Photon Source, we have overcome both challenges to measure the spin excitations of both spiral and commensurate phases by tracking low-energy excitations near their respective q-vectors in an applied magnetic field of 2 Tesla (as well as 0 Tesla). We have identified magnon branches near each q-vector.

Surprisingly, the magnon velocity near the spiral q-vector is much higher than the expected *sin(qa/2)* behavior, possibly suggesting a larger magnetic coupling strength than previously thought. This data may provide insight into the interactions between pseudo-spins in this fascinating material. Finally, we will discuss how the application of modest magnetic fields within modern high-resolution RIXS setups is now possible and will open up interesting new avenues of research in exotic 5d-magnetism.

This research used resources of the Advanced Photon Source, a U.S. Department of Energy (DOE) Office of Science User Facility operated for the DOE Office of Science by Argonne National Laboratory under Contract No. DE-AC02-06CH11357.

[1] G. Jackeli and G. Khalliulin (2009). *Phys. Rev. Lett.* **102**: 017205.

[2] J. Chaloupka et al. (2010). *Phys. Rev. Lett.* **105**: 027204.

[3] A. Ruiz, A. Frano, and N. Breznay et al. (2017). *Nature Communications* **8**: 961.

WK5

RIXS at High Pressure: Challenges and Opportunities

Gilberto Fabbris

X-ray Science Division, Argonne National Laboratory, Lemont, IL 60439

Pressure is a key thermodynamic variable that can be used not only to understand material properties, but also to drive novel phenomena. However, high-pressure techniques often impose strict experimental constraints, which drastically limits the pressure range available for certain techniques. This is particularly problematic for magnetic and electronic properties, for which, despite recent advances, probes such as inelastic neutron scattering and magnetometry are often restricted to pressures below 10 GPa. Over the last decade, RIXS has emerged as a key material science technique due to its ability to probe various types of magnetic, electronic and lattice excitations, making its implementation in the high-pressure regime highly desirable [1]. In this talk, I will focus on the technical challenges of probing RIXS at high-pressure. This will be done with the aid of two scientific examples: the nature of Sr_2IrO_4 high-pressure quantum paramagnetism [2], and the interplay between Cu 3d and Ir 5d orbitals in Cu_2IrO_3 [3]. I will also discuss potential instrument developments and the impact of APS-U on high-pressure RIXS experiments.

Work at Argonne is supported by the U.S. Department of Energy, Office of Science, Office of Basic Energy Sciences, under Contract No. DE-AC-02-06CH11357.

[1] J. Kim (2016). *High Pressure Research* **36**: 391–403.

[2] D. Haskel et al. (2012). *Physical Review Letters* **109**: 027204.

[3] M. Abramchuk et al. (2017). *Journal of the American Chemical Society* **139**: 15371–15376.

WK5

RIXS Study of Sr_2IrO_4 Single Crystal under Uniaxial Strain

Wentao Jin[1], Blair Lebert[1], Jungho Kim[2], Diego Casa[2], Xiao Wang[3], Yixi Su[3], and Young-June Kim[1]

[1] Department of Physics, University of Toronto, Toronto, ON M5S 1A7, Canada

[2] Advanced Photon Source, Argonne National Laboratory, Lemont, IL 60439

[3] Jülich Centre for Neutron Science JCNS at Heinz Maier-Leibnitz Zentrum (MLZ), Forschungszentrum Jülich GmbH, D-85747 Garching, Germany

It has been shown that the structural details, such as the Ir-O-Ir angle and the Ir-Ir distance, play an important role in influencing the magnetic properties of spin-orbital entangled iridates. But it is not well understood how the electronic structure and magnetic interaction in the iridates respond to structural distortion. The application of uniaxial strain within the *ab* plane can break tetragonal symmetry and induce the lattice distortion. In addition, the distortion can be continuously tuned, allowing one to examine the pseudospin-lattice coupling. Using our homemade piezoelectric-based strain apparatus, we have performed Ir L_3-edge RIXS measurements on single-crystal Sr_2IrO_4 under uniaxial tensile/compressive strain applied along the in-plane (1 0 0) and (1 1 0) directions. In both cases, we are able to drive the lattice distortion to the magnitude of 10^{-3} and tune the magnetic domain structure in Sr_2IrO_4 accordingly, while the change in the magnetic excitation is found to be quite marginal within this level of distortion. This indicates that the in-plane magnon gap in Sr_2IrO_4 is relatively robust with respect to the uniaxial perturbation, and a larger magnitude of orthorhombic distortion than 10^{-3} is needed to induce the gap change.

WK5

Applications of Resonant X-ray Emission and Inelastic X-ray Scattering in Chemistry and Biology

Ryan G. Hadt

Division of Chemistry and Chemical Engineering, California Institute of Technology, Pasadena, CA 91125

Transition metal electronic structure plays important roles across chemistry, biology, and physics. This presentation will discuss recent applications of resonant x-ray emission spectroscopy (RXES) and resonant inelastic x-ray scattering (RIXS) in defining the roles of transition metals in bioinorganic chemistry, catalysis, and molecular inorganic photophysics. Within biology, RXES/RIXS has provided detailed insights into how cytochrome *c* can function as both an electron transfer protein in oxidative phosphorylation and a peroxidase enzyme in programmed cell death (i.e., apoptosis). In catalysis, RXES/RIXS is a powerful, site-selective probe of the electronic structures of heterogeneous catalysts and their molecular analogs and has provided insights into the mechanism of O–O bond formation in the electrocatalytic oxygen evolution reactivity of cobalt oxides. Lastly, we demonstrate the unique power of high-resolution L-edge RIXS to define the ligand field excited state manifolds of inorganic complexes, with an emphasis on the spin- forbidden, photochemically active electronic states of metal-metal quadruply-bonded Re_2 dimers. Generally, we further showcase the high-level insights that can be gained by coupling density functional theory and multiplet ligand field calculations to the unique information content of RXES and RIXS.

Wednesday, May 8

WK6 CNM:
Optoelectronic Devices and Mechanical Systems Based on Ultra-thin 2D Materials

Location: APCF Conference Room, Building 446

Time: 8:30 – 5:30

Organizers: Irma Kuljanishvili (St. Louis University) and Saptarshi Das (Penn State)

Workshop aims to facilitate and strengthen the synergetic approach to experimental and theoretical works of experts from academia, national labs and industries to discuss the latest developments in the synthesis, characterization, and application of ultra-thin 2D materials and device architectures with the emphasis to explore their unique potential as smart/adaptive devices, and substrate or other stimulus-responsive systems.

Properties of these layered 2D materials can be tuned by lateral confinements and vertical multi-layer z-stacking. This offers opportunity for development of next generation of multi-functional devices whose properties can be engineered/designed to specific performance. Particular emphasis would be on optoelectronic and mechanical systems.

The materials of interest include but not limited to van der Waals materials (graphene, hBN, and TMDC or other mono-elemental layered 2D systems and heterostructures). Recent advances in the development of 2D materials and understanding of their electronic, optical, mechanical or thermal, properties have demonstrated vast potentials in these systems.

Center for Nanoscale Materials at Argonne has expertise and state-of-the-art facilities in the synthesis, functionalization and characterization of 2D materials and has a strong research program focused around the fundamental studies on physical properties of these materials. Workshop will include topics covering fundamental physical aspects of 2D materials and heterostructures, their growth, characterization and applications. Computational modeling offers opportunity for computational experimentation and testing of physical, chemical and mechanical properties with emphases on manipulating interfaces of these layered materials by geometric al designs and/or surface interaction for targeted outcome and/or applications. The workshop will facilitate discussions that explore routes for scalable and transformative technologies.

This workshop is expected to provide an opportunity for academics, R&D scientists in industry, and students to exchange ideas, think creatively about new avenues for collaborations with user facilities at Argonne and to work on expanding knowledge of nanoscience into development and implementation of new frontiers in nanotechnology.

Workshop topics include:

- Advances in fabrication and characterization, of 2D materials and heterostructures with emphasis on fundamental science, and their potential for scalability and/or commercialization.
- Novel device architectures with emphasis on optoelectronic and mechanical systems made with 2D materials and heterostructures, their fundamental properties, manufacturing, and testing.
- Optoelectronic devices made with 2D heterostructures on flexible substrates, functional and adaptable optoelectronic and optomechanical devices.
- Computational studies interface designs, mechanical: Computational approach for understanding interfaces interactions and mechanical properties with emphases on interface engineering.

8:30 – 8:35	Opening Remarks
8:35 – 9:25	Jiwoong Park (University of Chicago) *Atomically Thin Materials for Technology*
9:25 – 10:15	Ali Javey (University of California at Berkeley) *Making Semiconductor Monolayers Perfectly Bright*
10:15 – 10:30	Break
10:30 – 11:20	Zhihong Chen (Purdue University) *2D Cu Diffusion Barriers for Ultra-scaled Interconnect Technologies*
11:20 – 12:10	Mark Hersam (Northwestern University) *Mixed-dimensional van der Waals Heterostructures for Electronic and Energy Applications*
12:10 – 1:40	Lunch
1:40 – 2:30	Philip Feng (Case Western Reserve University) *Atomic Layer Semiconductor and Heterostructure Nanoelectromechanical Systems for Information Processing*
2:30 – 3:20	Ani Sumant (Argonne National Laboratory) *Advances in Superlubricity Using Hybrid 2D Materials-based Lubricants*
3:20 – 3:40	Break
3:40 – 4:30	Xiaodong Xu (The University of Washington) *Moiré-excitons in $MoSe_2$/WSe_2 Heterobilayers*
4:30 – 5:20	Vikas Berry (University of Illinois-Chicago) *Chemical and Structural Manipulation of Graphene and Other 2D Nanomaterials for Electronics and Optoelectronics*
5:20 – 5:30	Closing Remarks
5:30	Adjourn

WK6

Atomically Thin Materials for Technology

Jiwoong Park

University of Chicago, Chicago, IL 60637

Manufacturing of paper, which started two thousand years ago, simplified all aspects of information technology: generation, processing, communication, delivery and storage. Similarly powerful changes have been seen in the past century through the development of integrated circuits based on silicon. In this talk, I will discuss how we can realize these integrated circuits thin and free-standing, just like paper, using two-dimensional inorganic and organic materials and how they can impact future technology. In order to build these atomically thin circuits, we developed a series of chemistry-based approaches that are scalable and precise. They include wafer-scale synthesis of three atom thick semiconductors and heterojunctions (*Nature*, 2015; *Science*, 2018), a wafer-scale patterning method for one-atom-thick lateral heterojunctions (*Nature*, 2012), and atomically thin films and devices that are vertically stacked to form more complicated circuitry (*Nature*, 2017). Once realized, these atomically thin circuits will be foldable and actuatable, which will further increase the device density and functionality.

WK6

Making Semiconductor Monolayers Perfectly Bright

Ali Javey

Electrical Engineering and Computer Sciences, University of California, Berkeley, CA 94720

Two-dimensional semiconductors exhibit excellent device characteristics, as well as novel optical, electrical, and optoelectronic characteristics. In this talk, I will present our recent advancements in surface passivation, contact engineering, surface charge transfer doping, and heterostructure devices of layered chalcogenides. We have developed a passivation technique that allows for observation of near-unity photoluminescence quantum yield in monolayer semiconductors. I will discuss the mechanism by which non-radiative recombination can be fully removed in monolayers. The work presents the first demonstration of an opto-electronically perfect monolayer, and highlights one of their unique properties. Finally, I will discuss an AC carrier injection mechanism to enable bright light emitting devices using monolayers, overcoming the problem of Schottky contacts.

WK6

2D Cu Diffusion Barriers for Ultra-scaled Interconnect Technologies

Zhihong Chen

School of Electrical and Computer Engineering and Birck Nanotechnology Center, Purdue University, West Lafayette, IN 47907

Cu is used as the interconnect material in IC chips because of its superior conductivity. However, the diffusion of Cu into the dielectric between two interconnects can cause shorting and create chip failures, while the diffusion to transistors can introduce deep-level traps to Si and affect the transistor performance. Ta/TaN bilayer stacks have been used as diffusion barrier/liner materials to isolate Cu from surrounding intra/inter-layer dielectrics. Along with the scaling of VLSI, the dimensions of interconnects and vias need to be scaled as well. To maximize Cu volume for lower line resistance and reduce thevia resistance, the thickness of the barrier/liner bilayer should be scaled accordingly. However, these materials lose their capability of blocking Cu diffusion as their thickness is extremely reduced. In this talk, I will show that the diffusion barrier and liner properties of several 2D materials are investigated by various characterization methods. We observe that the lifetime of the dielectrics surrounding Cu electrodes can be significantly extended with the presence of the tested 2D barriers, providing strong evidence for promising alternative barrier/liner solutions.

WK6

Mixed-dimensional van der Waals Heterostructures for Electronic and Energy Applications

Mark C. Hersam

Department of Materials Science and Engineering, Northwestern University, Evanston, IL 60208

Layered two-dimensional (2D) nanomaterials interact primarily via van der Waals bonding, which has created new opportunities for heterostructures that are not constrained by epitaxial growth [1]. However, it is important to acknowledge that van der Waals interactions are not limited to interplanar interactions in 2D materials. In principle, any passivated, dangling bond-free surface interacts with another via non-covalent forces. Consequently, the emerging layered 2D nanomaterials can be integrated with a diverse range of other materials, including those of different dimensionality, to form mixed-dimensional van der Waals heterostructures [2]. In order to efficiently explore the vast phase space for mixed-dimensional heterostructures, our laboratory employs solution-based additive assembly [3]. In particular, constituent nanomaterials (e.g., carbon nanotubes, graphene, transition metal dichalcogenides, black phosphorus, boron nitride, and indium selenide) are isolated in solution, and then deposited into thin films with scalable additive manufacturing methods (e.g., inkjet, gravure, and screen printing) [4]. By achieving high levels of nanomaterial monodispersity and printing fidelity [5], a variety of electronic and energy applications can be enhanced including digital logic circuits [6], photodetectors [7], and lithium-ion batteries [8]. Furthermore, by integrating multiple nanomaterial inks into heterostructures, unprecedented device function is realized including anti-ambipolar transistors [9], gate-tunable photovoltaics [10], and neuromorphic memtransistors [11]. In addition to technological implications for electronic and energy technologies, this talk will explore several fundamental issues including band alignment, doping, trap states, and charge/energy transfer across previously unexplored mixed-dimensional heterointerfaces [12].

[1] X. Liu et al. (2018). *Advanced Materials* **30**: 1801586.

[2] D. Jariwala et al. (2017). *Nature Materials* **16**: 170.

[3] J. Zhu et al. (2017). *Advanced Materials* **29**: 1603895.

[4] G. Hu et al. (2018). *Chemical Society Reviews* **47**: 3265.

[5] J. Kang et al. (2017). *Accounts of Chemical Research* **50**: 943.

[6] M. Geier et al. (2015). *Nature Nanotechnology* **10**: 944.

[7] J. Kang et al. (2018). *Advanced Materials* **30**: 1802990.

[8] K.-S. Chen et al. (2017). *Nano Letters* **17**: 2539.

[9] V.K. Sangwan et al. (2018). *Nano Letters* **18**: 1421.

[10] D. Jariwala et al. (2016). *Nano Letters* **16**: 497.

[11] V.K. Sangwan et al. (2018). *Nature* **554**: 500.

[12] S.B. Homan et al. (2017). *Nano Letters* **17**: 164.

WK6

Atomic Layer Semiconductor and Heterostructure Nanoelectromechanical Systems for Information Processing

Philip Feng

Electrical Engineering, Case School of Engineering, Case Western Reserve University, Cleveland, OH 44106

Atomically thin crystals (including semiconducting, metallic and insulating layers) and their heterostructures offer compelling new platforms for 2D electronic, photonic devices and nanoelectromechanical systems (NEMS), where their unconventional and unique properties can be harnessed for engineering both classical signal processing and quantum transduction schemes. In this presentation, I will describe some of my research group's latest effort on advancing device physics and engineering of 2D heterostructures and NEMS. I will first show atomically thin radio-frequency (RF) NEMS resonators with excellent electrical tunability and remarkably broad dynamic range. I will then demonstrate how the unusually strong and efficient coupling effects have led to ultra-broad resonance tuning of van der Waals heterostructure resonators, as well as stable, robust graphene NEMS operating at glowing temperatures with simultaneous light emission. I will also describe h-BN nanomechanical resonators and phononic waveguides, as well as other 2D building blocks, and device engineering effort on new, energy-efficient signal transduction and information processing, toward realizing chip-scale integrated platforms.

[1] Islam, van den Akker, and Feng (2018). "Anisotropic Thermal Conductivity of Suspended...," *Nano Letters* **18**: 7683–7691.

[2] Ye, Lee, and Feng (2018). "Glowing Graphene Nanoelectromechanical Resonators at Ultra-High Temperature...," *IEDM 2018* **4.4**: 87–90.

[3] Islam, Lee, and Feng (2018). "Atomic Layer GaSe/MoS_2 van der Waals Heterostructure...," *ACS Photonics* **5**: 2693–2700.

[4] Lee, Wang, and Feng et al. (2018). "Electrically Tunable Single & Few-Layer MoS_2...," *Science Advances* **4**: eaao6653.

[5] Ye, Lee, and Feng (2018). "Electrothermally Tunable Graphene Resonators Operating...," *Nano Letters* **18**: 1678–1685.

[6] Yang, Lee, Feng, et al. (2017). "Tuning Optical Signature of Single- and Few-Layer...," *Nano Letters* **17**: 4568–4575.

[7] Ye, Lee, and Feng (2017). "Atomic Layer MoS_2-Graphene van der Waals Heterostructure...," *Nanoscale* **9**: 18208–18215.

[8] Wang, Jia, and Feng et al. (2016). "Resolving and Tuning Mechanical Anisotropy...," *Nano Letters* **16**, 5394-5400.

[9] Wang, Lee, and Feng (2014). "Spatial Mapping of Multimode Brownian Motions...," *Nature Communications* **5**: 5158.

[10] Lee, Wang, and Feng et al. (2013). "High Frequency MoS_2 Nanomechanical Resonators," *ACS Nano* **7**: 6086–6091.

WK6

Advances in Superlubricity Using Hybrid 2D Materials-based Lubricants

Anirudha V. Sumant

Center for Nanoscale Materials, Argonne National Laboratory, Lemont, IL 60439

Recent advances in achieving near zero friction at macroscale in sliding interfaces using solid lubricants based on 2D materials-nanoparticles hybrid combination opened new possibilities for reducing wear and friction related losses in various mechanical systems involving rotational/sliding motion [1]. The energy loss due to wear and friction in moving mechanical systems amounts to quarter of total energy loss worldwide. Oil-based lubricants used in majority of moving mechanical system have to be either spent (burned) or dumped into the environment after their use, creating major problems to the environment. Various alternatives including bio-degradable oil are being developed; however, more effort and research is needed to develop green lubricants.

Our recent work on utilizing combination of 2D materials (such as graphene and MoS_2) with nanodiamond as an oil-free lubricant have shown that we can achieve superlubricity (near zero friction) as well as exceptional wear resistance of the sliding components, at macroscale [2,3]. This discovery presents a paradigm shift from conventional oil-based lubrication and allows us to explore untapped potential of nanomaterials as a dry solid lubricant in many rotational/sliding mechanical systems. We show that near zero friction is possible due to structural reorganization in case of graphene-nanodiamond forming nanoscale ball-bearings and formation of onion-like-carbon (OLC) in case of MoS_2 and nanodiamond through tribochemical reaction at the sliding interface. The formation of OLCs is self-generating providing unlimited supply of lubricant and thus helping to increase the wear-life significantly.

I'll discuss the detailed experimental studies along with theorical investigations that suggest the overall mechanism of superlubricity. I'll also discuss our efforts towards commercializing this technology by working collaboratively with industry.

[1] Berman and Sumant et al. (2018). *ACS Nano* **12**(3): 2122.

[2] Berman and Sumant et al. (2015). *Science* **348**(6239): 1118.

[3] Berman and Sumant et al. (2018). *Nature Communications* **9**: 1164.

WK6

Moiré-excitons in $MoSe_2/WSe_2$ Heterobilayers

Xiaodong Xu

Department of Physics, Department of Materials Science and Engieering, University of Washington, Seattle, WA 98105

The creation of moiré patterns in crystalline solids is a powerful approach to manipulate their electronic properties, which are fundamentally influenced by periodic potential landscapes. In two-dimensional (2D) materials, a moiré pattern with a superlattice potential can form by vertically stacking two layered materials with a twist and/or finite lattice constant difference. This unique approach has led to emergent electronic phenomena, including the fractal quantum Hall effect [1–3], tunable Mott insulators [4,5], and unconventional superconductivity [6]. Furthermore, theory predicts intriguing effects on optical excitations by a moiré potential in 2D valley semiconductors [7–9], but these signatures have yet to be experimentally detected. In this talk, I will report our experimental evidence of interlayer valley excitons trapped in a moiré potential in $MoSe_2/WSe_2$ heterobilayers. We observe quantum-dot-like photoluminescence of interlayer excitons with the inheritance of unique valley-contrasting properties from the heterobilayer bulk. Twist angle dependent studies further support our observation of moiré excitons. Our results open opportunities for 2D moiré optics with twist angle as a unique control knob.

[1] Dean, C.R. et al. (2013). *Nature* **497**: 598–602.

[2] Hunt, B. et al. (2013). *Science* **340**: 1427-1430.

[3] Ponomarenko, L.A. (2013). *Nature* **497**: 594–597.

[4] Cao, Y. et al. (2018). *Nature* **556**: 80.

[5] Chen, G. et al. (2018). *Nature Physics*, Preprint: arxiv.org/abs/1803.01985.

[6] Cao, Y. et al. (2018). *Nature* **556**: 43.

[7] Yu, H et al. (2015). *Phys. Rev. Lett.* **115**: 187002.

[8] Yu, H. et al. (2017). *Sci. Adv.* **3**: e1701696.

[9] Wu, F. et al. (2018). *Phys. Rev. B* **97**: 35306.

WK6

Chemical and Structural Manipulation of Graphene and Other 2D Nanomaterials for Electronics and Optoelectronics

Vikas Berry

Department of Chemical Engineering, University of Illinois at Chicago, Chicago, IL 60607

The presentation will outline several chemical and structural manipulation techniques developed in our laboratory to control the properties of 2D nanomaterials. (a) Structural manipulation includes formation of wrinkles, nanoribbons and quantum dots, and (b) Chemical manipulation includes non-destructive functionalization, nanoparticle incorporation and biomolecular interfacing. The following topics will be discussed: (i) selective desiccation of bacterium under impermeable and flexible graphene *via* a flap-valve operation to produce axially-aligned graphene-wrinkles with anisotropic electrical properties; (ii) nanotomy process to cleave graphite nanoblocks and their exfoliation to produce graphene nanostructures (nanoribbons and quantum dots) with controlled electrical properties; (iii) wrinkling of MoS_2 to control photoluminescence; (iv) unique eta-6 organometallic approach to functionalize graphene in a vapor-phase process, while retaining its structural and electrical properties and offering chemical sites for nano-interfacing of plasmonic centers for enhanced photovoltaics; (v) biointerfacing of graphene and MoS_2 with cancer cells and bacteria for bionanotechnology and biomedical applications; and (vi) Photovoltaics via electronic coupling of graphene atop a bulk semiconductor to induce interfacial energy-band reorganization for light-sensitive junctions only one atom below the front surface.

Tuesday, May 7

WK7 APS: *In Situ* and Multimodal Microscopy for APS-U

Location: Building 446, Conference Room

Time: 8:30 – 5:00

Organizers: Ross Harder (APS XSD), Jörg Maser (APS XSD), Wonsuk Cha (APS XSD), and Michael Stuckelberger (DESY)

The Upgrade of the Advanced Photon Source with a multi-bend achromat magnetic lattice will revolutionize coherent imaging and microscopy techniques with significantly improved spatio-temporal resolution, greater sensitivity to a variety of contrast mechanisms, and multimodal imaging capabilities.

With x-ray fluorescence beginning to be sensitive to just a handful of atoms in an illuminated volume and coherent imaging reaching down to length scales approaching single atomic crystallographic defects, there is tremendous opportunity for cross-platform and in situ studies of materials across many decades of length scales in the suite of planned instruments at the APS. Examples are the study of individual catalytically active particles, study of photovoltaic and battery materials and devices, and study of evolution and kinetics during materials synthesis, operation, and degradation.

Accomplishing truly cross-platform investigations of individual samples will require the development of *in situ* instruments and methodology compatible with the many imaging, microscopy and spectroscopy instruments planned for the APS Upgrade. This workshop will bring together both domain experts and instrument scientists working at the forefront of *in situ* and *operando* x-ray characterization to identify high-impact opportunities for cross-platform and multimodal investigations.

8:30 – 8:45 Introduction

8:45 – 9:30 Michael Stuckelberger (Deutsches Elektronen Synchrotron DESY)
In Situ *Meets* Operando *Meets Multi-modal: Opportunities and Challenges of X-ray Microscopy for Solar Cells*

9:30 – 10:15 Andrej Singer (Cornell University)
Nucleation of Dislocations and Their Dynamics in Layered Oxides Cathode Materials during Battery Charging

10:15 – 10:45 Break

10:45 – 11:30 Amelie Rochet (Brazilian Synchrotron Light Laboratory, LNLS)
Coherent X-ray Diffraction Imaging Unravelling Structure-activity Relationships of Gold Catalysts

11:30 – 12:30 Sebastian Kalbfleisch (MAX IV Laboratory, Lund University)
The NanoMAX Beamline at MAX IV

12:30 – 1:30 Lunch

1:30 – 2:15	Paul Evans (University of Wisconsin, Madison) *Expanding the Scope of Nanobeam Diffraction: Dynamical Diffraction in Epitaxial Electronic Materials*
2:15 – 2:35	Junjing Deng (Argonne National Laboratory) *Correlative X-ray Ptychographic and Fluorescence Imaging at the Advanced Photon Source*
2:35 – 3:00	Christian Roehrig (Argonne National Laboratory) In Situ *and Multimodal Microscopy for APS-U Workshop*
3:00 – 3:30	Break
3:30 – 4:15	Richard Sandberg (Los Alamos National Laboratory) *Studying Materials Deformation and Failure with Bragg Coherent Diffraction Imaging*
4:15	Changyong Park (Argonne National Laboratory) *Challenges in* In Situ *Observation of Texture and Microstructure Evolution of Polycrystals under Pressure with Diffraction-based Imaging and Microscopy Techniques*

WK7

In Situ Meets *Operando* Meets Multi-modal: Opportunities and Challenges of X-ray Microscopy for Solar Cells

Michael E. Stuckelberger[1,2], Christina Ossig[1,3], T. Nietzold[2], Mariana. I. Bertoni[2], and Christian G. Schroer[1,3]

[1] Deutsches Elektronen-Synchrotron, 22607 Hamburg, Germany

[2] Arizona State University, Tempe, AZ 85287

[3] Department Physik, Universität Hamburg, 20355 Hamburg, Germany

Today, we are utilizing scanning probe x-ray microscopy endstations at 3rd generation synchrotrons to answer questions about photovoltaic materials such as: How do interfaces look like in solar cell stacks? How can thin-film devices be manufactured without sacrificing the efficiency due to lateral inhomogeneity? Which defects limit the device performance, and how can they be engineered to be less detrimental? How can solar cells be cost-effectively fabricated using abundant and non-toxic materials?

Based on experiments at DESY, APS, ESRF, and NSLS II, we will showcase examples of multimodal solar cell measurements during *in situ* growth and operation of solar cells, involving x-ray beam induced current (XBIC) and voltage (XBIV), x-ray fluorescence (XRF), x-ray diffraction (XRD), ptychography, and x-ray excited optical luminescence (XEOL). We have found correlations between the optical / electrical performance, composition, and strain, and will highlight the significance of correlations between performance and material properties for the solar cell development.

Tomorrow, the questions will be different, and we will be able to use x-ray microscopes at 4th generation synchrotrons to answer them. Where the frontiers of scanning microscopy will be pushed to? Which challenges arise with the new opportunities, and how can we tackle them? We will present ideas of 'dream experiments' and discuss the challenges related to multimodal scanning x-ray microscopy.

WK7

Nucleation of Dislocations and Their Dynamics in Layered Oxides Cathode Materials during Battery Charging

Andrej Singer

Department of Materials Science and Engineering, Cornell University, Ithaca, NY 14853

Lithium-rich layered oxides (LRLO) are among the leading candidates for the next generation cathode material for energy storage, delivering 50% excess capacity over commercially used compounds. Despite excellent prospects, voltage fade has prevented effective use of the excess capacity, and a major challenge has been the lack of understanding of the mechanisms underpinning the voltage fade. I will present our recent results, where using *operando* three-dimensional Bragg coherent diffractive imaging, we directly observe the nucleation of a mobile dislocation network in LRLO nanoparticles. The dislocations form more readily in LRLO as compared with a classical layered oxide, suggesting a link between the defects and voltage fade. We show microscopically how the formation of partial dislocations contributes to the voltage fade. The insights allow us to design and demonstrate an effective method to recover the original high voltage functionality.

Our findings reveal that the voltage fade in LRLO is reversible and call for new paradigms for improved design of oxygen-redox active materials.

WK7

Coherent X-ray Diffraction Imaging Unravelling Structure-activity Relationships of Gold Catalysts

Amélie Rochet

Brazilian Synchrotron Light Laboratory (LNLS), Brazilian Center for Research in Energy and Materials (CNPEM), 13083-970, Campinas, SP, Brazil

Chemical properties of catalytic materials are dependent on dynamic changes of the three-dimensional structure of the catalysts as well as the reactive environment. The active sites formed under reaction conditions are short-lived species and their identification remains a great challenge. Besides, defects and lattice strain dynamics of nanocrystals directly tuned their catalytic properties. To understand the nature of the active sites and the relationship between catalysts structure and activity, the study of catalysts under *operando* conditions is crucial. Bragg coherent x-ray diffraction imaging (BCDI) provides a unique opportunity to follow in 3D under realistic reaction conditions the strain and defects dynamics in the surface and inner core of the nanomaterials.

In this talk, I will present our *in situ/operando* BCDI investigations of gold catalysts during CO oxidation. Under the catalytic reaction conditions, we investigate the defects dynamics of gold nanocrystals with twin domains and show the *in situ* formation of a nanotwin network that correlates with the catalytic properties. Besides, we follow at the single nanoparticle-level, the highly dynamic 3D strain distribution during the hysteresis phenomenon occurring through CO oxidation reaction cycles.

WK7

The NanoMAX Beamline at MAX IV

Sebastian Kalbfleisch, Ulf Johansson, Gerardina Carbone, and Alexander Björling

MAX IV Laboratory, Lund University, Lund 22592, Sweden

The MAX IV synchrotron has started operation during 2016 and its 3 GeV ring is considered pioneering in the world of diffraction limited sources. One of the beamlines which stands to profit most is the NanoMAX beamline [1], a hard x-ray nanoprobe, set for best performance between 8 and 15 keV. It exploits especially the low vertical emittance of the ring by employing a horizontal Bragg diffracting crystal Si(111) monochromator, used to select the working energy in the range 6–20 keV, and a 1:1 focusing scheme of the source size on the secondary source aperture (SSA) placed at 51m from the undulator, allowing full control of flux versus coherence. A superpolished KB-mirror pair is used to focus the light onto the sample at 98m, with an energy dependent spot size: from 200 nm at 5 keV, down to 50 nm at 20 keV. With SSA settings optimized for coherence, the beamline delivers a maximum flux of around 1E10 on the sample.

In this talk, we will highlight some of the characteristics of the beamline, as well as present results from various types of experiments (e.g., forward ptychography and CDI, scanning diffraction and coherent imaging in Bragg geometry, x-ray fluorescence) performed at the first of two planned end-stations taken during the beamline's first two years of operation.

[1] U. Johansson, D. Carbone, S. Kalbfleisch, A. Björling, A. Rodriguez-Fernandez, T. Stankevic, M. Liebi, B. Bring, A. Mikkelsen, and U. Vogt (2018). *Microsc. Microanal.* **24**(2): 250.

WK7

Expanding the Scope of Nanobeam Diffraction: Dynamical Diffraction in Epitaxial Electronic Materials

Paul G. Evans

Materials Science and Engineering, University of Wisconsin-Madison, Madison, WI 53706

Epitaxial electronic materials include a thin film or nanostructure that shares key aspects of its crystallographic structure with a thick underlying substrate. In key applications of these epitaxial materials in the development of semiconductor quantum computing, the control of this structure at the scale of tens to hundreds of nanometers has been an important challenge. Semiconductor quantum computing devices based on heterostructures formed in the AlGaAs/GaAs system involve thin epitaxial layers with tiny lattice mismatches on the order of 10^{-4} or smaller and rely on the formation of qubits defined at the lateral scale of tens of nanometers. The further mechanical distortion of these layers during the formation of devices causes significant electronic perturbations due to the piezoelectric and deformation potential effects. Ultimately, the mechanical distortion and the accompanying electronic perturbation affects the operation of devices. It has been challenging, however, to characterize the mechanical distortion of these epitaxial layers, in part because the close lattice match between the substrate and the thin film has posed a difficult problem for x-ray nanobeam diffraction techniques. We have found that this problem can be addressed through the development of new nanobeam diffraction methods in which dynamical diffraction simulations are employed to interpret the complex diffraction patterns arising from latticed-matched heterostructures [1].

Nanobeam diffraction studies of lattice-matched GaAs/AlGaAs quantum computing devices reveal mechanical

distortions due to device fabrication, including from the residual stress and thermal expansion coefficient mismatch associated with metallic electrodes [2,3]. This new understanding the magnitude and role of these effects has the potential to allow the design of improved device geometries and potentially to lead to devices in which strain, in addition to electrostatic effects, is used to define quantum devices.

Beyond quantum computing structures, similar dynamical diffraction effects appear in problems associated with ferroelectric domains and domain boundaries in epitaxial $BaTiO_3$ thin films and in strain sharing effects in nanoscale complex oxides [4].

[1] A. Pateras, J. Park, Y. Ahn, J. Tilka, M.V. Holt, H. Kim, L.J. Mawst, and P.G. Evans (2018). "Dynamical scattering in coherent hard x-ray nanobeam Bragg diffraction," *Phys. Rev. B* **97**: 235414.

[2] A. Pateras, J. Park, Y. Ahn, J.A. Tilka, M.V. Holt, C. Reichl, W. Wegscheider, T. Baart, J. P. Dehollain, U. Mukhopadhyay, L.M.K. Vandersypen, and P.G. Evans (2018). "Mesoscopic Elastic Distortions in GaAs Quantum Dot Heterostructures," *Nano Lett.* **18**: 2780.

[3] A. Pateras, J. Carnis, U. Mukhopadhyay, M.-I. Richard, S.J. Leake, T.U. Schulli, C. Reichl, W. Wegscheider, J.P. Dehollain, L.M.K. Vandersypen, and P.G. Evans (2019). "Electrode-induced lattice distortions in GaAs multi-quantum-dot arrays," *J. Mater. Res.* https://doi.org/10.1557/jmr.2019.61.

[4] J.A. Tilka, J. Park, Y. Ahn, A. Pateras, Z. Cai, and P.G. Evans (2018). "Stressor-layer-induced elastic strain sharing in SrTiO3 Complex Oxide Sheets," *Appl. Phys. Lett.* **112**: 091905.

WK7
Correlative X-ray Ptychographic and Fluorescence Imaging at the Advanced Photon Source

Junjing Deng[1], Si Chen[1], Yudong Yao[1], Yi Jiang[1], Qiaoling Jin[2], Chris Jacobsen[1,2,3], Zhonghou Cai[1], Barry Lai[1], and Stefan Vogt[1]

[1] Advanced Photon Source, Argonne National Laboratory, Lemont, IL 60439

[2] Department of Physics and Astronomy, Northwestern University, Evanston, IL 60208

[3] Chemistry of Life Processes Institute, Northwestern University, Evanston, IL 60208

Accurate knowledge of elemental distributions of specimens within their structural context is very important for understanding their roles and the resulting sample behaviors. At the Advanced Photon Source (APS), we have developed a correlative imaging method via a combination of x-ray fluorescence microscopy (XFM) and ptychography [1,2]. XFM offers high sensitivity for quantitative mapping of elements in samples with a spatial resolution that limited by beam size, while ptychography provides a complementary approach to visualize ultrastructure beyond focusing-optic resolution limit. We have applied this correlative method on the study of materials samples including solar cell film [3] and electrocatalysts [4], as well as 3D frozen-hydrated cells [5]. To speed up the imaging speed, we have developed photon-efficient fly-scan techniques [6] together with novel instrumentations [7]. By using continuous scanning, novel data analysis, and a new generation of synchrotron light source provided by APS-U, it will enable high-throughput high-resolution imaging which provides opportunities to implement this correlative tool for *in situ* and *operando* measurements.

This research used resources of the Advanced Photon Source, a U.S. Department of Energy (DOE) Office of Science User Facility operated for the DOE Office of Science by Argonne National Laboratory under Contract No. DE-AC02-06CH11357.

[1] J Deng et al. (2015). *Proc. Natl. Acad. Sci.* **112**: 2314.

[2] J Deng et al. (2017). *Sci. Rep.* **7**: 445.

[3] M. Stuckelberger et al. (2018). *Microscopy and Microanalysis* **24**: 434.

[4] M. Wang, et al. (2019). *ACS Applied Materials & Interface* **11**: 5682.

[5] J Deng et al. (2018). *Sci. Adv.* **4**: eaau4548.

[6] J. Deng, et al. (2015). *Optics Express* **23**: 5348.

[7] C. Preissner et al. (2016). *Proceedings of MEDSI2016*, TUAA02.

WK7
In Situ and Multimodal Microscopy for APS-U Workshop

Christian Roehrig

X-ray Science Division, Advanced Photon Source, Argonne National Laboratory, Lemont, IL 60439

The increased coherent flux that will become available with the APS Upgrade will provide exciting opportunities for new types of experiments. Critical to the realization of these experiments are instruments that are able to handle a variety of scan trajectories, have sufficient precision and bandwidth, and provide for the high-speed collection of both position and x-ray data. These goals necessitate control systems that are flexible and dynamic and built upon robust mechanics and position metrology. In this talk, I will discuss the current development efforts at the APS on scanning microscopes. I will then talk about performance targets for some of the APS Upgrade beamline control systems and the challenges that must be met to get there.

WK7

Studying Materials Deformation and Failure with Bragg Coherent Diffraction Imaging

Richard L. Sandberg[1], Mathew J. Cherukara[2], Reeju Pokharel[3], Timothy O'Leary[1], Jonathan Gigax[1], Eric E. Hahn[3], Wonsuk Cha[2], Ross J. Harder[2], and Saryu J. Fensin[3]

[1] Laboratory for Ultrafast Materials and Optical Science, Center for Integrated Nanotechnologies, Los Alamos National Laboratory, Los Alamos, NM 87545

[2] Advanced Photon Source, Argonne National Laboratory, Lemont, IL 60439

[3] Materials Science and Technology Division, Los Alamos National Laboratory, Los Alamos, NM 87545

We are developing Bragg coherent diffraction imaging (BCDI) to study materials damage and failure in structural metals. BCDI can provide nanometer scale images of strain in crystalline materials. Our team performed the first proof of principle demonstration of combined *in situ* BCDI experiment guided by MD diffraction simulations on a thin film specimen of deformed copper [1]. We are developing compatible tensile and compressive loading cells to stress materials at the nanometer scale. We are also developing a tuneable monochromator for the coherent x-ray scattering beamline at APS (34ID-C) as part of a partner user proposal to conduct in-situ orientation mapping of polycrystalline thin films, thus enabling more rapid identification of desired nanocrystals or grains. Finally, we are also studying materials implanted with helium ions as a way to understand how materials in nuclear reactors lose strength under high radiation environments. Techniques such as these may enable us to understand materials damage in reactors.

This work was supported by the U.S. Department of Energy (DOE) at Los Alamos National Laboratory (LANL) under Contract No. DE-AC52-06NA25396 through the LANL LDRD Program 20180683ER and the Institute for Materials Science 2016 Rapid Response program. We also acknowledge funding support from the LANL Dynamic Materials Campaign (C2). This work was also supported by Argonne National Laboratory LDRD 2015-149-R1 (Integrated Imaging, Modeling, and Analysis of Ultrafast Energy Transport in Nanomaterials) and Argonne National Laboratory LDRD 2018-019-N0 (A.I. C.D.I: Atomistically Informed Coherent Diffraction Imaging).

The experiment was performed at the 34-IDC beamline of the Advanced Photon Source (APS). This work was supported, in part, by the Center for Nanoscale Materials. The Advanced Photon Source, the Center for Nanoscale Materials and the Argonne Leadership Computing Facility are supported by the U.S Department of Energy, Office of Science, Office of Basic Energy Sciences (BES), under Contract No. DE-AC02-06CH11357. The work was in part supported by the Center for Integrated Nanotechnologies, a U.S. DOE BES user facility.

[1] M.J. Cherukara, R. Pokharel, T.S. O'Leary, J.K. Baldwin, E. Maxey, W. Cha, J. Maser, R.J. Harder, S.J. Fensin, and R.L. Sandberg (2018). "Three-dimensional x-ray diffraction imaging of dislocations in polycrystalline metals under tensile loading," *Nat. Commun.* **9**: 3776.

WK7

Challenges in *In Situ* Observation of Texture and Microstructure Evolution of Polycrystals under Pressure with Diffraction-based Imaging and Microscopy Techniques

Changyong Park

HPCAT, X-ray Science Division, Argonne National Laboratory, Lemont, IL 60439

Textures and microstructures of polycrystals evolve with compression, resulting in nonuniformity in the strain distributions and phase equilibria. While the anisotropy is readily expected under non-hydrostatic conditions, recent observations of the high-pressure micro-XRD using diamond anvil cells show more and more evidences for inherently anisotropic responses of polycrystals to the external pressure even under hydrostatic conditions. The nonuniformity of strain distribution under hydrostatic compression raises a fundamental question to determining isothermal equation of state of crystalline solids and to describing the macroscopic behaviors as to be thermodynamically equilibrated phases.

The intergranular interactions that are responsible to dynamically changing textures with external pressure seem to be intimately correlated with the intragranular response to the local forces forming anisotropic strain fields inside single grains. Shear-induced phase transition can be triggered by this localized deformation processes, which in turn further complicates the microstructure evolution. The microscopic (nucleation and growth) to mesoscopic (intragranular development) to macroscopic (texture and microstructure evolution and phase equilibria) link is extremely difficult to observe *in situ* due to the orders of magnitude changes in its scale but has yet to be fully described to better model the predictable phase behaviors under compression.

The spatially resolved x-ray diffraction imaging and microscopy would be required to address these highly localized phenomena. Some x-ray diffraction-based imaging techniques are promising for *in situ* observations of the pressure-induced local variations, but there are several challenges to be overcome for the successful applications. In this presentation, a case of pure Zr polycrystal is presented with the results of 2D x-ray diffraction imaging (XDI) and grain Bragg coherent diffraction imaging (gBCDI). The challenges involved in the technical development and the future perspectives are discussed.

Portions of this work were performed at HPCAT (Sector 16), 1-ID-E, and 34-ID-C beamlines, Advanced Photon Source (APS), Argonne National Laboratory. HPCAT operations are supported by DOE-NNSA's Office of Experimental Sciences. The Advanced Photon Source is a U.S. Department of Energy (DOE) Office of Science User Facility operated for the DOE Office of Science by Argonne National Laboratory under Contract No. DE-AC02-06CH11357.