2018 APS/CNM USERS MEETING

WORKSHOP AGENDAS AND ABSTRACTS

Wednesday, May 9

APS Workshop 1
Resolution@speed: Advanced X-ray Spectroscopies with Upgraded APS (APS_U)

Location: Building 402, Room E1100/1200

Organizers: Chengjun Sun (APS) and Anne Marie March (CSE)

Advanced x-ray emission and absorption spectroscopies are important routes to explore the structural and electronic properties of complex materials and devices in the fields of physical science and engineering. With the increases in brightness, flux and sub-micron focusing provided by APS_U, it will become easier to apply high-energy-resolution detection methods at high speed to a wider variety of experiments, such as x-ray emission for ultra-diluted samples, high pressure x-ray emission spectroscopy, time–resolved x-ray emission/XANES, high-resolution x-ray absorption spectroscopy, and spin-resolved x-ray absorption spectroscopy.

This symposium will consider prospects of instrumentation and applications of high-energy-resolution and high speed (e.g., *in operando* and time-resolved) x-ray spectroscopies that will take advantage of APS_U. The symposium includes various related topics in the field, such as the overall picture of advanced x-ray spectroscopies at APS_U, the proposal/design of a next generation miniature x-ray emission spectrometers (miniXES), high-resolution x-ray emission and absorption spectroscopy for high-pressure research, the perspective of advanced x-ray spectroscopies from a theory viewpoint, time-resolved XAS of photochemical reactions, and time-resolved x-ray emission spectroscopy with MHz pink beam at 7-ID.

Session 1 Chair: Chengjun Sun

8:25 – 8:30 Assemble

8:30 – 9:00 Steve Heald (Advanced Photon Source, Argonne National Laboratory)
Opportunities for Advanced Spectroscopies at the APS-U and Sector 25

9:00 – 9:30 Gilles Doumy (CSE, Argonne National Laboratory)
Advanced Hard X-ray Spectroscopies for Tracking Optically Driven Molecular Dynamics

9:30 – 10:00 Yang Ding (HPSTAR)
High-pressure X-ray Spectroscopic Research and Its Prospect for APS-U

10:00 – 10:30 Break

Session 2 Chair: Anne Marie March

10:30 – 11:00 Robert Gordon (Simon Fraser University)
Upgrading S20 XES Capabilities for Improved Efficiency and Compatibility for High Pressure, Catalysis and Imaging Applications

11:00 – 11:30 Yves Joly (Institut NEEL)
Interpretation of Advanced X-ray Spectroscopies Using ab initio *Simulations*

11:30 – 12:00 Amy Cordones-Hahn (SLAC National Accelerator Laboratory)
Photochemistry of Transition Metal Complexes Probed by Time-resolved XAS

WK1

Advanced Hard X-ray Spectroscopies for Tracking Optically Driven Molecular Dynamics

Gilles Doumy
Chemical Sciences and Engineering Division,
Argonne National Laboratory, Argonne, IL 60439

The ability to follow and possibly control the dynamics of light-induced reactions in solution is a longstanding goal in chemistry. After light absorption, a series of mechanistic steps take place, which can involve charge transfer, geometric and electronic rearrangement, bond breaking and making, ligand elimination or substitution, and electron solvation. Time-resolved hard x-ray spectroscopies have become invaluable tools to study those dynamics. Use of high repetition rate laser excitation, fast refreshing targets and flexible data gating capabilities allows for high fidelity measurements of transient properties on various timescales ranging from ~100ps to microseconds.

Complementary techniques such as XAS, XES, RXES can be successfully combined to get a more comprehensive picture, and it is even possible to access some of the valence electronic properties directly responsible for chemical reactivity. I will show how using MHz pink beam for non-resonant emission spectroscopy represents another game changer for those studies, before contemplating the challenges and opportunities brought by the APS-U characteristics.

Work was supported by the U.S. Department of Energy, Office of Science, Chemical Sciences, Geosciences, and Biosciences Division.

WK1

Upgrading S20 XES Capabilities for Improved Efficiency and Compatibility for High Pressure, Catalysis and Imaging Applications

Robert Gordon
Department of Physics, Simon Fraser University,
Burnaby, BC V5A 1S6, Canada

The miniXES style of x-ray emission spectrometer was designed to use flat crystals, in a pseudo-Johann or pseudo-von Hamos configuration, to get a spectrum in a single shot without having to scan an analyzer crystal. *In situ* calibration and readily swappable crystals to change energy range are hallmarks of the design, but the design targeted individual samples of small size, and user demand for higher efficiency and larger sample clearance has increased since inception. In particular, for these who are studying *in situ* catalysis, high pressure and imaging, there is a need to upgrade to a system with improved clearance and improved collection efficiency. This can be achieved by an expanded crystal array in conjunction with a larger position-sensitive detector. Within the upgrade (S20 moving to S25), an improvement in efficiency of ~500 is anticipated.

WK1

Photochemistry of Transition Metal Complexes Probed by Time-resolved XAS

Amy Cordones-Hahn
PULSE Institute, SLAC National Accelerator Laboratory,
Menlo Park, CA 94025

Time-resolved x-ray absorption spectroscopy (XAS) is a powerful tool for following changes in oxidation state and bonding that occur during a chemical reaction in real-time. Several examples that combine an optical laser pump and x-ray absorption probe will be presented, highlighting the effectiveness of this method to study photochemical reactions of transition metal complexes. As an example, the photoinduced linkage isomerization of a Ru-sulfoxide complex will be discussed. In this study, time-resolved XAS at the metal and ligand atomic edges were combined to achieve a localized view of the electronic excited states that drive the chemical reaction [1]. The properties of APS and APS-U enable similar time-resolved experiments of photochemical processes that occur on timescales of hundreds of picoseconds or longer, making it ideal for studies of photo-redox reactions or other diffusional chemistry.

[1] A.A. Cordones, J.H. Lee, K. Hong, H. Cho, K. Garg, M. Boggio-Pasqua, J.J. Rack, N. Huse, R.W. Schoenlein, and T.K. Kim (2018). "Transient Metal-Centered States Mediate Isomerization of a Photochromic Ruthenium-Sulfoxide Complex." *Nature Comm.* (in press).

Tuesday, May 8

APS Workshop 2
Workshop on Past, Present, and Future of Insertion Devices at the APS: A Tribute to Efim Gluskin, Emil Trakhtenberg, and Isaac Vasserman

Location: Building 401, Room A1100

Organizers: Dennis M. Mills, Yury Ivanyushenkov, and E. Ercan Alp (APS)


Efin Gluskin


Emil Trakhtenberg


Isaac Vasserman

Undulators were the key components of the third-generation synchrotron radiation sources built in early 1990s. Their successful development and implementation led to the installation of fully energy-tunable devices and later polarization-switchable devices that revolutionized x-ray scattering, imaging, and spectroscopy methods. The exceptional ability in the building of and precise magnetic tuning of the undulators led the way towards the free electron lasers of today. Building novel vacuum chambers and developing critical technologies for manufacturing, machining, magnetic tuning, and installation were all parts of the successful integration of undulators into APS' modern storage ring.

The current innovations in superconducting undulators, revolving devices with different periods, and fast polarization switching abilities will determine the success of the APS Upgrade Project, and many similar projects around the world.

This workshop will celebrate past successes, take a critical look at the future developments that are in progress all around the globe, and recognize the unique and invaluable contributions of three key individuals who have worked at the APS: Efim Gluskin, Emil Trakhtenberg, and Isaac Vasserman.

Session Chair: David Moncton

8:30 – 8:50	Stephen Streiffer (Advanced Photon Source) *Welcome Remarks*
8:50 – 9:00	David Moncton (Massachusetts Institute of Technology)
9:00 – 9:30	Herman Winick, (SLAC National Accelerator Laboratory, Emeritus) *Experience with Wigglers and Undulators at Harvard in the 1960s and Stanford in the 1970s*
9:30 – 10:00	Joachim Pflüger (European XFEL) *Undulator Technology for X-ray Free Electron Laser*
10:00 – 10:30	Kwang-je Kim (Argonne National Laboratory, University of Chicago) *Not to be Taken Lightly*
10:30 – 10:45	Break

Session Chair: Ercan Alp

10:45 – 11:15 (video)	John Galayda (SLAC National Accelerator Laboratory) *FEL Undulators at BNL, ANL and SLAC*
11:15 – 11:45 (video)	Toshiya Tanabe (NSLS-II, Brookhaven National Laboratory) *Evolution of Insertion Devices at NSLS and NSLS-II*
11:45 – 12:15 (video)	Nikolay Vinokurov (Budker Institute of Nuclear Physics) *Undulator Development in Budker INP*
12:15 – 1:30	Lunch

Session Chair: Liz Moog

1:30 – 2:00	Tetsuya Ishikawa (SPring-8) *Insertion Devices at SPring-8*
2:00 – 2:30	Johannes Bahrdt (BESSY-II) *New Developments in Cryogenic Permanent Magnet Undulator Technology*
2:30 – 3:00	Shigemi Sasaki (Hiroshima University, Emeritus; Shanghai Tech University) *APPLE U, Quasi-Periodic U, LCLS U, Knot-APPLE U, What's Next?*
3:00 – 3:15	Break

Session Chair: Dennis Mills

3:15 – 3:45	Yury Ivanyushenkov (Advanced Photon Source) *Development of Superconducting Undulators at the APS*
3:45 – 4:15	Emil Trakhtenberg (Advanced Photon Source) *25+ Years with APS/ANL*
4:15 – 4:45	Efim Gluskin (Advanced Photon Source)
4:45 – 5:00	Dennis Mills, Closing Remarks (Advanced Photon Source) *Closing Remarks*

WK2

Undulator Technology for X-ray Free Electron Lasers

Joachim Pflueger
European XFEL, 22869 Schenefeld, Germany

Today FELs in the hard x-ray regime make use of undulator technology on a large scale. For example the European XFEL, which recently became operational, provides system lengths of 220m to produce radiation wavelengths as low as 0.5 Angstroem.

This talk gives an outline over the technological developments and challenges of undulator technology over the past 20 years. It begins with the first developments for systems in visible/VUV/soft x-ray regime, LEUTL, TTF, FLASH realized at APS and DESY. The present state of the art is described by the LCLS, SACLA, PAL-XFEL, European XFEL and SwissFEL. Some challenges of future developments are described and speculated about.

WK2

Not to be Taken Lightly

Kwang-je Kim
Argonne National Laboratory, Argonne, IL 60439
University of Chicago, Chicago, IL 60637

Based on personal and scientific contacts, we will prove that these colleagues are not to be taken lightly.

WK2

Evolution of Insertion Devices at NSLS and NSLS-II

Toshiya Tanabe
NSLS-II, Brookhaven National Laboratory, Upton, NY 11973

National Synchrotron Light Source (NSLS) project was started in 1977 and started operation in 1982 as the first 2nd generation light source. In the same period, Synchrotron Radiation Source (SRS) at the Daresbury Laboratory and Photon Factory at KEK started operation. The 2nd generation light sources were designed to use bending magnet radiation as a primary source. However, towards the end of their life, various insertion devices (IDs) were installed to take advantage of their superior source characteristics.

In this paper, evolution of various IDs employed towards the end of NSLS operation and those of its successor, NSLS-II, is delineated.

WK2

Undulator Development in Budker INP

Nikolay Vinokurov
Budker Institute of Nuclear Physics SB RAS,
Novosibirsk 630090, Russia

Budker INP develops different undulators (wigglers) since 1979, when the first superconducting wiggler (20 poles, 9 cm period, 3.3 T) was commissioned at VEPP-3 storage ring. Few tens of insertion devices were

designed, manufactured and tested during next 39 years. Among them were variable-gap permanent magnet undulators (1979), hybrid permanent magnet undulators (1983), superconducting wigglers, non-superconducting electromagnetic undulators (including variable-polarization ones) and variable-period permanent magnet undulators. Original experiments with undulator radiation were performed. Four free electron lasers were built. Three of them are in operation now at the user facility "Novosibirsk free electron laser".

WK2

Insertion Devices at SPring-8

Tetsuya Ishikawa
RIEN SPring-8 Center, Sayo-gun, Hyogo 679-5148 Japan

Insertion devices at SPring-8 and SACLA, including new development for SPring-8-II and Laser-Plasma Accelerator Light Source, will be discussed.

WK2

New Developments in Cryogenic Permanent Magnet Undulator Technology

Johannes Bahrdt
Helmholtz-Zentrum Berlin, 14109 Berlin, Germany

In the last two decades, short period in-vacuum undulators became indispensable in many synchrotron radiation facilities worldwide. These devices extend the photon energy range to short wavelengths at existing 3rd and next generation accelerators. In the last ten years, cryogenically cooled planar undulators (CPMUs) were developed, aiming for even shorter period lengths. After a prototyping phase, today full-scale CPMUs are built and operated at several synchrotron radiation facilities. Different technical approaches have proven feasibility and reliability. New measurement techniques were developed and optimized. As an example, the results from the HZB CPMU17 will be discussed in more detail. Future options for a mild field enhancement will be discussed.

WK2

APPLE U, Quasi-Periodic U, LCLS U, Knot-APPLE U, What's Next?

Shigemi Sasaki
ShanghaiTech University and Hiroshima University

Along with the development of light source storage rings and FELs, progresses of insertion device are significant in recent 30 years. Typical example is a development of variably polarizing insertion devices that started from Onuki-type undulator followed by the elliptical wiggler, Helios undulator, APPLE undulator, and some other variations. It could be achieved in the circumstances of international competition and collaboration in the third generation light source constructions.

In my talk, "time-line" and "how it came" of various undulators development that I contributed is presented. Also, some prospects in further development of insertion device are presented.

WK2

Development of Superconducting Undulators at the APS

Yury Ivanyushenkov on behalf of the APS SCU Team
Argonne National Laboratory, Argonne, IL 60439

Development of superconducting undulator (SCU) technology at the Advanced Photon Source (APS) has started more than a decade ago. The first superconducting undulator SCU0 was installed on the APS storage ring in December 2012, and after commissioning, became a user device in January 2013. Currently, there are three SCUs in user operation at the APS. A short review of development of SCUs at the APS is given in this talk.

WK2

25+ Years with APS/ANL

Emil Trakhtenberg
APS, Argonne National Laboratory, Argonne, IL 60439

I came to the USA on 08/19/1991 in the framework of an international collaboration between Budker Institute of Nuclear Physics and SSC laboratory. At BINP, I was the Head of the Engineering Department responsible for the colliding beam storage rings and synchrotron radiation sources. I was a chief engineer for the storage ring VEPP-2M and two synchrotron radiation sources we build for Kurchatov Institute: "Siberia-I" and "Siberia-II". In June 1992, I started to work for the APS.

In this presentation are briefly described my major works done during this period: extruded ID vacuum chambers, four-motor drive system for APS planar undulators, LCLS-I project, HGVPU undulator, and different types of superconducting undulators.

Help and collaboration from many of my colleagues made this work successful.

Tuesday, May 8

CNM Workshop 3
Next-generation Quantum Systems Based on Topological Phases and Integrated Quantum Photonics

Location: Building 401, Room A5000

Organizers: Xuedan Ma and Dafei Jin (CNM)

Quantum information science is an emerging field that can potentially revolutionize multiple technologies extending from sensing and measurement precision to quantum computation and communication. The field leverages explicit quantum properties of matter such as entanglement and superposition to enable phenomena that break classical limits. The quest to understand and control such intriguing phenomena, in turn, stimulates progress in the development of quantum materials.

The goal of this workshop is to bring together researchers working on various aspects of topological materials and quantum photonic systems, and create synergies between Argonne/CNM and the worldwide scientific community. Main topics in this workshop include:

1. Exotic topological materials
2. Photonic quantum networks exploiting quantum entanglement effects
3. Novel photonic metamaterials and cavities

Quantum materials transforming electronics and photonics from the classical to the quantum regime may enable fundamentally new approaches to information technologies. One vibrant research area in quantum materials is topological materials, including topological insulators and superconductors, semi-metals, and more. These systems feature remarkable properties such as gapless boundary modes and fractionalized quasi-particle excitations. These topological properties remain stable even if the system is subject to external perturbation. Qubits encoded in such states are topologically protected and permit creation of topological quantum computers that are robust against noise and disorder.

Aside from the exotic topological phases, rich interactions between light and quantum materials offer promising opportunities for secured quantum communication, vast increase of data-storage capacity, and ultrasensitive quantum metrology. While photons are excellent quantum-state carriers for long-distance transmission, their entanglement with matter qubits in quantum materials implement deterministic quantum interfaces. On-chip solid-state qubit architecture has emerged as one of the most promising implementations of quantum photonic circuits. Integration of functional photonic cavities and metamaterials may lead to groundbreaking technologies such as quantum repeaters that enable globally entangled quantum networks.

Session 1: Quantum and Nano Photonics (Session Chair: Xuedan Ma)

8:30 – 9:15 Arka Majumdar (University of Washington, Seattle)
Towards Single Photon Nonlinear Nanophotonics

9:15 – 10:00 Edo Waks (University of Maryland)
Quantum Nanophotonics: Controlling Photons with Spin on a Semiconductor Chip

10:00 – 10:30 Break

Session 2: Quantum and Topological Materials (Session Chair: Dafei Jin)

10:30 – 11:15 Michael Manfra (Purdue University)
Hybrid Superconductor/Semiconductor Systems for Topological Quantum Computing with Majorana Zero Modes

11:15 – 12:00 Lu Li (University of Michigan)
Quantum Oscillations in Kondo Insulators

12:00 – 1:30 Lunch

Session 3: Quantum and Nano Photonics (Session Chair: Xuedan Ma)

1:30 – 2:15 Koray Aydin (Northwestern University)
Nanophotonics in the Flatland: Engineering Novel Photonic Platforms from 2D Materials to Metadevices and Metamaterials

2:15 – 3:00 Han Htoon (Los Alamos National Laboratory)
Electronic Structure and Quantum Optics of Carbon Nanotube Defects

3:00 – 3:30 Break

Session 4: Quantum and Topological Materials (Session Chair: Dafei Jin)

3:30 – 4:15 Weida Wu (Rutgers University)
Direct Evidence of Ferromagnetism in a Quantum Anomalous Hall System

4:15 – 5:00 Ulrich Welp (Argonne National Laboratory)
Bulk Topological Superconductors, Gap Structure, and Effect of Electron Scattering

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WK3

Towards Single Photon Nonlinear Nanophotonics

Arka Majumdar
Electrical Engineering and Physics, University of Washington, Seattle, WA 98195

Quantum optical nonlinearities have received growing interest for their key role in quantum information science, quantum simulations, and other quantum technologies. Unfortunately, most materials exhibit very weak optical nonlinearities, virtually non-existent at the single photon level. Nano-scale optical resonators can store light for a long period of time in cubic-wavelength scale volume, and thus present a unique opportunity to enhance the light-matter interaction. Additionally, breakthroughs in materials science allow us to engineer inherently strong nonlinear materials. In this talk, I will present our theoretical and experimental efforts in nonlinear nanophotonics, integrated with atomically thin 2D materials, specifically transition metal dichalcogenides and solution-processed quantum dots. By confining both light and matter in the wavelength scale, we aim to reach the nonlinear regime, where single photons start repelling each other. I will also elaborate on the possibility of scaling this platform to multiple single photon quantum nodes with the possibility of creating a correlated quantum fluid of light.

WK3

Quantum Nanophotonics: Controlling Photons with Spin on a Semiconductor Chip

Edo Waks
University of Maryland, College Park, MD 20742

Interactions between light and matter lie at the heart of optical communication and information technology. Nanophotonic devices enhance light-matter interactions by confining photons to small mode volumes, enabling optical information processing at low energies. In the strong coupling regime these interactions are sufficiently large to achieve a nonlinear response with a single photon. Such single-photon nonlinearities are highly desirable for photonic quantum information where atoms mediate interactions between photonic qubits. In this talk, I will describe our effort to develop and coherently control quantum dots strongly coupled to photonic crystals. Quantum dots are semiconductor “artificial atoms” that can act as efficient photon emitters and quantum memories. By embedding them in a photonic crystal cavity that spatially confines photons to less than a cubic wavelength, we can attain the strong coupling regime. I will discuss how we can exploit this regime to achieve a quantum transistor, where a single spin trapped in the quantum dot can flip a photon polarization and a single photon can flip

the spin-state, realizing a fundamental quantum interaction. I will then describe how this device can implement a single photon transistor, where a single photon can switch an optical signal composed of as many as 30 photons. Finally, I will discuss our approach to scale these devices to larger systems and couple them to novel photonic structures exhibiting topological properties.

WK3
Quantum Oscillations in Kondo Insulators

Lu Li
University of Michigan, Ann Arbor, MI 48109

In metals, orbital motions of conduction electrons are quantized in magnetic fields, which is manifested by quantum oscillations in electrical resistivity. This Landau quantization is generally absent in insulators, in which all the electrons are localized. Here we report a notable exception in an insulator—ytterbium dodecaboride (YbB12). The resistivity of YbB12, despite much larger than that of usual metals, exhibits profound quantum oscillations under intense magnetic fields. The large effective masses indicate strong correlation effects between electrons. Our result is the first discovery of quantum oscillations in the electrical resistivity of a strongly correlated insulator, and it will bring crucial insight to the understanding of the ground state in gapped Kondo systems.

WK3
Nanophotonics in the Flatland: Engineering Novel Photonic Platforms from 2D Materials to Metadevices and Metamaterials

Koray Aydin
Electrical Engineering and Computer Science, Northwestern University, Evanston, IL 60208

Nanophotonic materials and devices facilitate strong light-matter interactions at subwavelength scales, thus providing unique opportunities to control and manipulate photons. In this talk, I will present novel approaches in controlling light-matter interactions at the nanoscale. I will discuss visible frequency metasurfaces broadband phase control and anomalous reflection, spectrum splitting using metallic metasurfaces enabled by phase engineering at the subwavelength scale. I will also present two different approaches for obtaining narrow-band resonant absorption filters at visible wavelengths. First structure is based on the surface lattice resonances in periodic nanowire and nanoring arrays fabricated on a reflecting metallic substrate. Enhanced photoluminescence enhancement from a single monolayer MoS_2 via plasmonic nanostructures will also be discussed. 2D layered materials received great attention due to their unique optical, electrical and mechanical properties; however, due to their thickness, light-matter interactions is rather weak. We utilize plasmonic nanostructures to strongly enhance electric fields locally at subwavelength scales therefore facilitating increased light emission and absorption in 2D semiconducting materials. Finally, I will present inverse-designed broadband electromagnetic metadevices that can bend and focus light using thin polymeric structured metamaterials at millimeter-wave and optical frequencies.

WK3
Electronic Structure and Quantum Optics of Carbon Nanotube Defects

Han Htoon
Los Alamos National Laboratory, Los Alamos, NM 87545

Materials scientists have been utilizing incorporation of impurities and defects as a powerful tool for introduction of optically active quantum states that enable a wide range of novel functionalities. Nitrogen and silicon vacancy centers introduced into diamond, for example, enable quantum technology applications ranging from ultrasensitive sensing of electric/magnetic fields to eavesdropping-proof communication and quantum computing. Recent studies have shown that such introduction of quantum states is also possible in semiconducting single-walled carbon nanotubes (SWCNTs) through low-level covalent attachment of various chemical functional groups such as ether/epoxide and a variety of monovalent and divalent alkyl and aryl functionalities. Here in this talk, I will review our recent single nanotube low temperature PL spectroscopy and quantum optic experiments revealing their unique electronic structure [1,2] and single photon emission [3,4] properties that in some aspect can rival or even exceed to those demonstrated in diamond NV centers.

[1] X. Ma, L. Adamska, H. Yamaguchi, S.E. Yalcin, S. Tretiak, S.K. Doorn, and H. Htoon (2014). “Electronic Structure and Chemical Nature of Oxygen Dopant States in Carbon Nanotubes.” *ACS Nano* **8**: 10782.

[2] X. He, B.J. Gifford, N.F. Hartmann, R. Ihly, X. Ma, S.V. Kilina, Y. Luo, K. Shayan, S. Strauf, J.L. Blackburn, S. Tretiak, S.K. Doorn, and H. Htoon (2017). “Low-Temperature Single Carbon Nanotube Spectroscopy of sp3 Quantum Defects.” *ACS Nano* 10.1021/acsnano.7b03022.

[3] X. He, N.F. Hartmann, X. Ma, Y. Kim, R. Ihly, J.L. Blackburn, W. Gao, J. Kono, Y. Yomogida, and A. Hirano (2017). “Tunable room-temperature single-photon emission at telecom wavelengths from sp 3 defects in carbon nanotubes.” *Nat. Photonics* **11**: 577.

[4] X. Ma, N.F. Hartmann, J.K.S. Baldwin, S.K. Doorn, and H. Htoon (2015). “Room Temperature Single-Photon Generation from Solitary Dopants of Carbon Nanotubes. “Nat. *Nanotechnol.* **10**: 671.

WK3

Direct Evidence of Ferromagnetism in a Quantum Anomalous Hall System

Weida Wu
Rutgers University, New Brunswick, NJ 08901

Quantum anomalous Hall (QAH) systems are of great fundamental interest and of potential application (e.g., quantum computation) because of dissipationless conduction without external magnetic field. The QAHE has been realized in magnetically doped topological insulator thin films. There is experimental evidence of chiral Majorana fermion modes in QAH-superconductor heterostructure. However, full quantization requires extremely low temperature (<50 mK) in the initial works, though it was significantly improved with modulation doping or co-doping of magnetic elements. Improved ferromagnetism was indicated in these thin films, yet a direct evidence of long-range ferromagnetic order is lacking. In this talk, I will present direct visualization of long-range ferromagnetic order in thin films of Cr and V co-doped $(Bi,Sb)_2Te_3$ using low-temperature magnetic force microscopy with *in situ* transport. The magnetization reversal process reveals a typical ferromagnetic domain behavior (i.e., domain nucleation and domain wall propagation) in contrast to much weaker magnetic signals observed in the end members, possibly due to superparamagnetic behavior observed in Cr doped TI films. The gate dependence of magnetic reversal process confirms that the long range ferromagnetic order is robust against bulk carrier modulation. The observed long-range ferromagnetic order resolves one of the major challenges in QAH systems, and paves the way to high-temperature dissipationless conduction by exploring magnetic topological insulators.

WK3

Bulk Topological Superconductors, Gap Structure, and Effect of Electron Scattering

Ulrich Welp
Argonne National Laboratory, Argonne, IL 60439

Doped topological insulators such as Bi_2Se_3 represent to date the most promising basis for the realization of bulk topological superconductors. Such topological superconductors may serve as platform for quantum computing utilizing the non-Abelian braiding statistics of Majorana zero modes. Here, we present studies of the thermodynamic, magneto-transport and structural characteristics of $Nb_xBi_2Se_3$ and $Sr_xBi_2Se_3$. The results of specific heat and magnetization measurements show that superconductivity in these materials is a bulk property. Both display a surprisingly large two-fold in-plane asymmetry of the superconducting state, which is not expected, considering their trigonal crystal structure but which can be accounted for in the topological odd-parity nematic superconducting Eu state. Indeed, the low-temperature variation of the penetration depth and the robustness against electron scattering give evidence for the nematic nodal Δ_{4x} gap structure.

Tuesday, May 8

APS Workshop 4
Frontiers of Materials Research with Single-crystal Total Scattering

Location: Building 402, Room E1100/1200

Organizers: Stephan Rosenkranz (MSD) and Douglas Robinson (APS)

Many physical properties of crystalline materials are strongly enhanced or even driven by local disorder and short-range correlations. These include ionic conduction, thermoelectricity, ferroelectric relaxor behavior, unconventional superconductivity, colossal magnetoresistance, and many more. Their microscopic understanding requires accurate measurements of the total scattering comprising both Bragg peaks from the long-range average order and diffuse scattering from deviations from that average, which includes short-range correlations as well as extended short-range order resulting from defect-defect interactions. Measurements over a large three-dimensional volume of reciprocal space, with sufficient resolution to separate diffuse from Bragg scattering and sufficient dynamic range to include both, are necessary to accurately test models of complex disorder, whether obtained by the use of phenomenological potentials or short-range-order parameters or by *ab initio* methods. New high-energy detectors, such as the recently acquired 2M CdTe Pilatus detector, will also enable the transformation of diffuse scattering into 3D Pair Distribution Functions, which provide model-independent information on the length scales of defect correlations.

This workshop provides a forum for presenting recent developments in instrumentation and analysis that make this technique a powerful tool to investigate complex disorder. It will bring together experts from a wide range of scientific topics to present recent results and the potential impact improved single crystal total scattering instrumentation would have on many fields of great importance to future energy applications.

9:00 – 9:15	Stephan Rosenkranz (Materials Science Division, Argonne National Laboratory) and Doug Robinson (Advanced Photon Source, Argonne National Laboratory) *Introduction*
9:15 – 9:45	Richard Welberry (Research School of Chemistry, Australian National University) *Single Crystal Diffuse Scattering*
9:45 – 10:15	Thomas Weber (ETH Zurich) *The 3D-ΔPDF Method for Analysing Single Crystal Diffuse Scattering*
10:15 – 10:45	Break
10:45 – 11:15	Branton Campbell (Brigham Young University) *Symmetry-mode Analysis of Local Structure in Complex Solids*
11:15 – 11:40	Jacob Ruff (Cornell University) *Towards Comprehensive Diffraction Studies of Crystals and Films Using High-energy X-rays*
11:40 – 12:05	Ray Osborn (Argonne National Laboratory) *First Results Obtained with the New Pilatus 2M CdTe Detector at APS*
12:05 – 1:30	Lunch
1:30 – 2:00	Marek Pasciak (Institute of Physics of the Czech Academy of Sciences) *Diffuse Scattering in Dielectric Materials: $BaTiO_3$, $PbZrO_3$ and Relaxor Ferroelectrics*
2:00 – 2:30	Bruce Gaulin (McMaster University) *Magnetic and Charge Diffuse Scattering from Geometrically Frustrated Magnets*
2:30 – 3:00	Alan I. Goldman (Ames Laboratory and Iowa State University) *Short-range Magnetic Correlations in Quasicrystalline i-Tb-Cd*
3:00 – 3:30	Break

3:30 – 4:00	Olivier Delaire (Duke University) *Diffuse Scattering to Study Phonons near Lattice Instabilities and Ordering Transitions*
4:00 – 4:30	Matt Krogstad (Argonne National Laboratory) *Order-disorder Transition in $Na_xV_2O_5$*
4:30 – 5:00	Doug Robinson (Argonne National Laboratory) *Current and Future Capabilities for Diffuse Scattering at 6-ID-D*
5:00 – 5:30	General Discussion
5:30	Adjourn

WK4

Single Crystal Diffuse Scattering

Richard Welberry
Research School of Chemistry, Australian National University, Canberra ACT 0200, Australia

Both SCDS (single crystal diffuse scattering) and PDF (pair distribution function) analyses make use of the same total scattering that is obtained in diffraction experiments (both Bragg scattering and diffuse scattering), but they do so in very different ways. In this presentation, I will concentrate on the SCDS case, on which my research has concentrated throughout my career.

Diffuse scattering—the coherently scattered intensity that is not localised on the reciprocal lattice—contains a wealth of information about the local order (order on the nanoscale) in crystalline materials. Since molecules and atoms will respond most strongly to their local chemical environments, it is a valuable tool in understanding how structure leads to properties. However, at present its collection and analysis are relatively specialised. Monte Carlo (MC) computer simulation of a model structure has become a powerful and well-accepted technique for aiding the interpretation and analysis of diffuse scattering patterns. Its great strength is its flexibility—as long as an MC energy can be defined, a model can be developed and tested. At one extreme a very simplified model may be useful in demonstrating particular qualitative effects, while at the other a quantitative and very detailed description of disordered structures can be obtained.

For SCDS, scattering data can be measured over the whole three-dimensional diffraction space and there is no orientational averaging. SCDS thus accesses the maximum amount of very detailed scattering information. To set against this is the fact that it is restricted to situations where single crystals are available. Moreover SCDS experiments and analyses are lengthy and complex.

WK4

The 3D-ΔPDF Method for Analysing Single Crystal Diffuse Scattering

Thomas Weber
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The 3D-ΔPDF is the Fourier transform of the diffuse part of single crystal scattering [1]. The resulting pattern provides information about real structure properties that are not represented by the average structure. Substitutional disorder, correlated displacements or size effect distortions show typical fingerprints that are easily identified and quantitatively analysed [1,2].

The single crystal approach has several advantages compared to the well-established powder PDF method. First, the three-dimensional information allows extracting information that is not accessible by powder diffraction. The intrinsic angular projection in powder experiments leads to information loss along the angular directions and to severe overlappings at long direct and reciprocal space distances. As a consequence, one is restricted to relatively simple disorder models. The 3D-ΔPDF has no such limitations. Pair correlations exceeding a distance of 100 Angstroms may be routinely accessed [3] and full 3D information is preserved. Even complex problems such as phasonic or structural disorder in quasicrystals can be successfully analysed [4–6]. Second, the relatively easy separation of Bragg and diffuse scattering in single crystal diffraction patterns allows investigation of disorder without being biased by systematic or statistical errors from the strong Bragg peaks (e.g., extinction, multiple scattering, detector saturation). Even though the relative error of Bragg scattering is usually much smaller, its integral error may significantly exceed the total diffuse intensities. Since the average structure is usually well known before the real structure is investigated it is advantageous to eliminate Bragg peaks from the diffraction pattern before diffuse scattering is analysed.

Despite its obvious advantages the 3D-ΔPDF approach is not yet as mature as the powder PDF method. One

restriction, apart from computational challenges, is the limited experimental accessibility of high-Q diffraction data, which particularly complicates the understanding of small atomic shifts or anharmonicity. The development of high energy beamlines optimized for diffuse scattering experiments will certainly help to fill this gap.

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WK4

Symmetry-mode Analysis of Local Structure in Complex Solids

Branton J. Campbell and Harold T. Stokes

Department of Physics and Astronomy, Brigham Young University, Provo, UT 84602

Symmetry-lowering phase transitions often result in a large number of new structural degrees of freedom, which can be challenging to characterize experimentally. Recasting such a problem in terms of group-theoretical symmetry modes can dramatically reduce effective structural complexity without actually reducing the number of parameters involved. A symmetry mode is a pattern of changes within the crystal (a linear combination of traditional structural parameters) associated with a single irreducible matrix representation of the crystal's symmetry group. Because nature tends to employ the smallest possible number of symmetry modes in a phase transition, this description is highly compact. To determine the low-symmetry structure, one can first identify which symmetry modes are active below the transition, and then simply turn off the others (i.e., fix their amplitudes at zero). Symmetry modes can be used to treat atomic displacements, magnetic moments, polyhedral rotations, ADPs, occupational orderings, lattice strains, or any other types of crystalline order parameters. Though symmetry modes have been primarily used to characterize long-range order, they have recently been used to reduce complexity in both small-box and big-box local-structure models. In this presentation, we'll describe the symmetry mode parameter set, highlight user-friendly tools for obtaining symmetry-mode descriptions of any phase transition, review some recently examples in the literature, and discuss the use of symmetry-mode models with single-crystal diffuse scattering data.

WK4

Towards Comprehensive Diffraction Studies of Crystals and Films Using High-energy X-rays

Jacob Ruff

Cornell High Energy Synchrotron Source (CHESS), Cornell University, Ithaca, NY 14853

Efficient, comprehensive surveys of reciprocal space offer an important new vista on the physics of materials, which has long been lacking. This talk will briefly review ongoing efforts at CHESS to collect such data for an assortment of important systems. We will discuss the advantages and drawbacks of several schemes for rapid collection of "total" x-ray scattering from crystals and films. Data collected using a Pilatus6M with silicon sensor at high energy will be highlighted, as well as preliminary results with medipix3 systems using higher-Z materials. Time permitting, we will also discuss the proposed <QM>^{2} beamline which is being developed as part of the CHESS-U upgrade, and which aims to facilitate rapid, comprehensive surveys of charge and magnetic scattering from quantum materials.

WK4

First Results from the New Pilatus 2M CdTe Detector at APS

Ray Osborn[1], Matthew Krogstad[1], Stephan Rosenkranz[1], Douglas Robinson[2], Guy Jennings[2], and Justin Wozniak[3]

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We have been developing high-throughput methods of measuring diffuse scattering with high-energy x-rays that utilize the latest advances in fast area detectors. We use the sample rotation method commonly used in crystallography, collecting frames at 10 Hz every 0.1° over a full 360° rotation in shutter-less mode. These frames are then transformed into reciprocal space using the APS CCTW package. These may also be transformed into real space to produce 3D ΔPDF "images" using the punch-and-fill method pioneered by Weber and colleagues at ETH Zürich. With large reciprocal space volumes collected in under 30 minutes, it is possible to collect the temperature dependence of total scattering in a sample in a few hours, allowing a number of ordering phenomena, such as order-disorder transitions and charge-density-waves, to be explored in detail, both above and below the transitions. I will present the first results of using these techniques with the Pilatus 2M CdTe detector on Sector 6-ID-D on a range of different samples.

WK4

Diffuse Scattering in Dielectric Materials: $BaTiO_3$, $PbZrO_3$ and Relaxor Ferroelectrics

Marek Paściak
Institute of Physics of the Czech Academy of Sciences, Prague, Czech Republic

Modern dielectric materials are mostly based on oxides with the perovskite structure, the framework which, being relatively simple, offers broad chemical flexibility and hence possibility of fine-tuning of material properties. Such engineering often aims at reaching structural heterogeneity on a nanoscopic level that leads to short to medium-ranged polar fluctuations and high electro-mechanical coupling. Therefore diffuse scattering is frequently observed in dielectric materials and its interpretation brings valuable insights into understanding of how local polarization is distributed.

This has been the case of relaxor ferroelectrics (e.g., $PbMg_{1/3}Nb_{2/3}O_3$ - PMN) for which characteristic diffuse scattering observed down to lowest temperatures [1] has been related to so called polar nanoregions which are deemed responsible for special dielectric behaviour of relaxors. The talk will review our work exploring the relationship of diffuse scattering, chemical ordering [2] and polar nanoregions.

Another example described in the talk will be that of $PbZrO_3$ (PZO), an antiferroelectric material, the end member of a technologically important solid solution $PbZr_xTi_{1-x}O_3$. PZO has a peculiar phase transition with three lattice modes being involved: antiparallel displacements of Pb atoms and two rotation (tilting) modes of oxygen octahedra. Detailed diffuse scattering analysis allows one to identify the signals related to the important instabilities and discuss the mechanism behind a simultaneous action of these distortions [3].

Finally some attention will be paid to the classical $BaTiO_3$. In our recent work [4] the narrow sheets of diffuse scattering, related to anisotropic longitudinal correlations of Ti ions, are shown to be caused entirely by the overdamped anharmonic soft phonon branch. This finding demonstrates that the occurrence of diffuse scattering agrees with a displacive picture of the cubic phase of this textbook ferroelectric material.

Some methodological aspects of the interpretation of diffuse scattering in dielectric materials will be also discussed with the emphasis on large-scale molecular dynamics simulations giving access to both time and space resolution of short-range polar correlations.

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WK4

Magnetic and Charge Diffuse Scattering from Geometrically Frustrated Magnets

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Magnetic materials based on triangles and tetrahedra have long been a playground for the physics of exotic ground states stabilized by geometrical frustration. The three dimensional cubic pyrochlores of the form $A_2B_2O_7$ have figured prominently, as both the A and B sublattices, independently, decorate a three dimensional network of corner sharing tetrahedra in this family of materials. We have focussed neutron and x-ray scattering studies on rare earth pyrochlores with magnetic trivalent rare earth ions at the A site, and (typically) non-magnetic tetravalent transition metal ions such as Ti^{4+} at the B site. More recently we have also been interested in the role of quenched disorder on the resulting exotic ground states. Studies of solid-solutions of the form $Tb_2Ti_{(1-x)}Sn_{(x)}O_7$ and the compound Tb_2NbScO_7 have been carried out. Understanding the magnetic and charge correlations in these systems requires both sophisticated x-ray and neutron scattering. If time permits, I will also discuss recent results on the triangular lattice system $YbMgGaO_4$, which features triangular nets of magnetic Yb^{3+} ions in the presence of charged disorder due to random positions of Mg^{2+} and Ga^{3+} within this structure.

WK4

Short-range Magnetic Correlations in Quasicrystalline i-Tb-Cd

Alan I. Goldman
Iowa State University and Ames Laboratory, Ames, IA 50011

To date, all of the known quasicrystals with moment bearing elements exhibit frustration and spin glass behavior at low temperatures, even though there is abundant theoretical evidence that long-range magnetic order is possible. Employing neutron scattering methods, we have identified the local moment configurations on the Tsai-type icosahedral clusters of i-Tb-Cd that provide insight into the underlying frustration inherent to these novel systems. Inelastic neutron scattering measurements, on CNCS at ORNL, show that the Tb moments are Ising-like, directed along the fivefold axes of the icosahedral clusters. Our elastic neutron scattering

measurements of the magnetic diffuse scattering, performed on CNCS and CORELLI, reveal significant structure consistent with short range magnetic correlations on these clusters. Using a simple spin Hamiltonian, we identified the lowest energy spin configurations for Ising-like moments on a single icosahedron comprised of Tb^{3+} ions at the vertices, which lead to scattering patterns in substantial agreement with the magnetic diffuse scattering.

WK4
Diffuse Scattering to Study Phonons near Lattice Instabilities and Ordering Transitions

Olivier Delaire
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A detailed understanding of atomic dynamics is of broad interest for the design of efficient energy materials, for example to establish reliable microscopic models of thermal transport and thermodynamics. Lattice instabilities and ordering transitions impact functional properties in many materials, including ferroelectrics, thermoelectrics, phase-change materials, and superconductors. Understanding the effects of lattice modulations, defects, and short-range order on the phonons is key to controlling the functional properties of these materials, and diffuse scattering experiments are essential in these investigations.

X-ray diffuse scattering complements spectroscopy experiments with either x-rays or neutrons, providing detailed information about phonon dispersions throughout reciprocal space. I will describe how we use these tools to obtain quantitative information on phonon mean-free-paths, revealing dominant scattering mechanisms, including anharmonicity, electron-phonon coupling, and scattering by defects or nanostructures. Such microscopic information about the microscopic origins of thermal transport provides important insights to design more efficient thermoelectric materials. In addition, first-principles simulations of the atomic dynamics enable the rationalization of extensive experimental datasets. In particular, *ab initio* molecular dynamics simulations at finite-temperature allow us to capture striking effects of anharmonicity.

In this presentation, I will present results from our investigations of phonon transport in several important thermoelectric materials ($AgSbTe_2$, $AgBiSe_2$, PbTe) [1–3]. Further, phonons can provide a large contribution to the entropy of solid-solid phase-transitions, and I will discuss several cases where our investigations of anharmonic phonons lead to a better understanding of phase-stability, highlighting recent results on thermodynamics of the metal-insulator transition in VO_2 [4] and in ferroelectric oxides [5].

O. Delaire acknowledges funding from U.S. Department of Energy, Office of Basic Energy Sciences, Materials Science and Engineering Division, through the Office of Science Early Career program, and as part of the S3TEC EFRC.

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WK4
Order-disorder Transition in $Na_xV_2O_5$

Matthew Krogstad[1], Raymond Osborn[1], John Vaughey[1], Jacob Ruff[2], and Stephan Rosenkranz[1]

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[2] Cornell High Energy Synchrotron Source (CHESS), Cornell University, Ithaca, NY 14853

Efficient methods of measuring single crystal diffuse scattering using synchrotron x-rays have been developed and can provide new insights into short-range cation ordering in electrode materials. Alongside simulation and analytical methods of looking at such data, the 3D pair distribution function (3D-ΔPDF) can clearly display occupational correlations in real space, allowing a model-independent view of short-range order. This is demonstrated with data on β-$Na_xV_2O_5$, with x=0.2 and x=0.4, collected over the temperature range 100K to 500K. The sodium intercalants partially occupy sites on two-rung ladders penetrating the framework of vanadium oxide pyramids and octahedra, with only short-range order at room temperature and above. However, at x=0.4, the length scale of sodium-sodium correlations increases significantly below 200K with the emergence of forbidden Bragg peaks below an order-disorder transition. The 3D-ΔPDF show that the sodium ions occupy alternate sites on each ladder rung with a zig-zag configuration that is in phase with neighboring ladders. The growth in the length scale of sodium-sodium correlations with decreasing temperature is clearly seen in real space images that allow a quantitative determination of the interionic interactions that impede ionic mobility.

WK4

Current and Future Capabilities for Diffuse Scattering at 6-ID-D

Doug Robinson
Advanced Photon Source, Argonne National Laboratory, Argonne, IL 60439

Presently supported high-energy diffuse scattering methods at 6-ID-D include classical diffractometer-mounted point detector, static area detector / oscillating sample, and small to medium sized diffractometer-mounted area detectors step scanned through selected volumes of reciprocal space. Remapping software includes custom user-written code as well as x-ray utilities and the APS supported RSMap3D. Sample environments include various closed cycle cryostats covering the range from ca. 2K to 800K, thermoelectric stages from -30C to 250C, furnaces to 1200C, and magnetic fields to ca. 0.5T. Diamond anvil cells are available from the MM group. We borrow detectors and Oxford Cryostream systems as needed.

Broadly determined by user demand, future capabilities might include trajectory scanning using diffractometer-mounted area detectors, long-travel motorized stages to position and orient the larger area detectors, high temperature gas jet sample heating, lower temperature environments with lower backgrounds, Bragg peak masks machine-produced on demand, focusing optics for smaller samples, and more user-friendly 3D simulation and fitting software. Input from workshop participants on priorities is encouraged.

Tuesday, May 8

APS/CNM Workshop 5
In Situ Rheology, SAXS, and XPCS for the Study of Soft Matter

Location: Buiding 446, Advanced Protein Characterization Facility Conference Room

Organizers: Xiao-Min Lin (NST), Suresh Narayanan and Alec Sandy (APS)

Soft matter is a class of materials that includes complex fluids such as polymers, liquid crystals, and colloidal suspensions. The presence of hierarchical structures from micrometer to nanometer scales instills in these materials highly unusual structural and dynamical properties, especially when they are deformed and under flow in response to external stress. These studies are typically carried out using separate measurements of macroscopic mechanical properties using rheology and microstructure using SAXS/SANS.

In situ rheology and SAXS measurements offer unique insight into such materials that would otherwise remain unknown. A recent development in this area is the use of time-resolved SAXS and XPCS to study dynamics at the nanoscale while simultaneously measuring rheological properties using a rheometer. This development opens up new avenues where stress relaxation at the nanoscale can be tracked using coherent x-ray speckles and connected to the microstructure and bulk properties.

While there are a handful of *in situ* rheology-SAXS/SANS capabilities across the globe, the APS and CNM now provide unique capabilities at Sector 8-ID to simultaneously study rheology, microstructure, and dynamics in soft materials at a single location.

This workshop will bring together a team of experts in these areas of research with the aim of identifying forefront areas of research exploiting these unique capabilities.

9:15 – 9:30	Alec Sandy and Suresh Narayanan (Argonne National Laboratory) *Opening Remarks*
9:30 – 10:00	Ronald Larson (University of Michigan) *Multi-scale Modeling of the Structure and Rheology of Surfactant Solutions and Polymer-colloidal Networks*
10:00 – 10:30	Xiao-Min Lin (Argonne National Laboratory) *Unraveling the Role of Order-to-disorder Transition in Dense Colloidal Solution by* In Situ *Small-angle X-ray Scattering*
10:30 – 10:45	Break
10:45 – 11:15	James Harden (University of Ottawa) *Rheo-XPCS Studies of Shear Induced Rejuvenation, Nano-plasticity, and Stress Relaxation in Soft Glassy Materials*
11:15 – 11:45	James Swan (Massachusetts Institute of Technology) *Fast Methods for Simulating the Dynamics of Soft Materials*
11:45 – 12:15	Matthew Helgeson (University of California Santa Barbara) *Toward* In Situ *Morphology Characterization of Complex Fluids under Arbitrary Processing Flows*
12:15 – 1:30	Lunch
1:30 – 2:00	Simon Rogers (University of Illinois at Urbana-Champaign) *Coupling Large-amplitude Oscillatory Shear Stress Measurements to Microstructure Changes via* In Situ *Rheo-scattering*

2:00 – 2:30	Chris Sorensen (Kansas State University) *Studies of Shear Effects on Aggregation and Gelation*
2:30 – 2:45	Break
2:45 – 3:15	Jacinta Conrad (University of Houston) *Transport of Interacting Nanoparticles in Complex Polymeric Fluids*
3:15 – 3:45	Subramanian Ramakrishnan (Florida State University) *Universal Scaling of Quench-dependent Dynamics in Intermediate Concentration Colloidal Gels*
3:45 – 4:15	Xiang Cheng (University of Minnesota) *From Flocking Birds to Swarming Bacteria: Study of the Dynamics of Active Fluids Using Fast Confocal Rheometry*
4:15 – 4:20	Closing Remarks
4:20	Adjourn

WK5

Multi-scale Modeling of the Structure and Rheology of Surfactant Solutions and Polymer-colloidal Networks

Ronald G. Larson
Department of Chemical Engineering, University of Michigan, Ann Arbor, MI 48109

Surfactant solutions andlatex colloidal solutions containing end-functionalized telechelic polymers self-assemble in water into a variety of structures, including long, entangled micelles as well as polymer gels and colloid/polymer gels that are important for consumer products, paints, and drug release formulations. The complex structures of these solutions require multi-scale modeling that includes atomistic, and coarse-grained molecular simulations, as well as colloidal scale simulations to connect rheological properties to composition. We show examples of such modeling and how its predictions can be compared to rheological and scattering data.

WK5

Unraveling the Role of Order-to-disorder Transition in Dense Colloidal solution by *In Situ* Small-angle X-ray Scattering

Xiao-Min Lin
Center for Nanoscale Materials, Argonne National Laboratory, Argonne, IL 60439

In situ synchrotron small angle x-ray scattering (SAXS) can reveal unprecedented structural details in dense colloidal nanoparticle solutions. These information are valuable to understand the physical properties of the entire system, in particular how it responds to macroscopic mechanical deformation. Using *in situ* SAXS, we demonstrate that an order-to-disorder transition leads to a dynamic shear thickening in a lower stress regime than the standard steady shear thickening. We show that the order-to-disorder transition is controlled by strain, which is distinguishably different from steady shear thickening which is a stress related phenomenon. The appearance of this two-step shear thinning and thickening transition is also influenced by particle size, monodispersity and measurement conditions (i.e., oscillatory shear vs. steady shear). Our results demonstrate conclusively that the order-to-disorder transition induced thickening is completely unrelated to the mechanism that drives the steady shear thickening.

WK5

Rheo-XPCS Studies of Shear Induced Rejuvenation, Nano-plasticity, and Stress Relaxation in Soft Glassy Materials

James L. Harden
Physics Department, University of Ottawa, Ottawa ON K1N 6N5, Canada

This talk presents an overview of x-ray photon correlation spectroscopy under shear (Rheo-XPCS) as a method for studying the behaviour of soft glassy nanostructured materials in response to applied deformation. We present studies of three soft glassy solids (concentrated nanocolloidal networks, clay gels, and jammed suspensions of charged silica nanoparticles) subjected to different programmes of *in situ* shear strain that provide insight into particle rearrangements at the nanometer scale and their connection to macroscopic mechanical behaviour of the materials. Particular examples of reversible local yielding, mechanical rejuvenation and over-aging, and protracted anisotropic microscopic relaxation following step-strains will be discussed.

WK5

Fast Methods for Simulating the Dynamics of Soft Materials

James Swan
Massachusetts Institute of Technology, Cambridge, MA 02139

In this talk, I will discuss newly developed computational methods for Brownian Dynamics simulations of soft materials. These Brownian dynamics techniques utilize a new decomposition of the linear operator governing the microhydrodynamics of suspended objects to enable rapid sampling of thermal fluctuations. Simulations of up to 2 million particles are possible on modern GPU hardware, and these methods avoid many of the complications and inaccuracies present in explicit solvent methods such as dissipative particle dynamics and multi-particle collision dynamics. I will show how such Brownian Dynamics methods are suitable for generating dynamics across large length scales, long time scales, and far from equilibrium, which should be of interest to researchers employing XPCS in experimental studies of soft materials.

WK5

Toward *In Situ* Morphology Characterization of Complex Fluids under Arbitrary Processing Flows

Matthew E. Helgeson
Department of Chemical Engineering, University of California Santa Barbara, Santa Barbara, CA 93106

In situ small angle neutron scattering under flow (flow-SANS) has become a critical tool for measuring and formulating processing-structure-property relationships of polymeric fluids. However, sample environments and associated measurement methods for flow-SANS/SAXS have largely limited these measurements to steady state flows and simple rheometric deformations (pure shearing or elongation) that fail to capture the complex nonlinear and time-varying deformations encountered during polymer processing. Recently, significant advances in neutron detection as well as the design of new fluidic devices have opened up new capabilities for probing complex, time-varying deformations that more reliably emulate real processing flows. Here, we will summarize the key advances leading to these capabilities, and illustrate their usefulness with two examples involving the flow-induced structuring in polymer nanocomposites. In the first example, we use time-resolved rheo-SANS, involving simultaneous flow-SANS and rheological measurement, to explore the kinetics and mechanics of shear-induced aggregation of nanoparticle suspensions in associative polymers during startup and cessation. The results show that, over a wide range of conditions, clustering is dominated by competition of hydrodynamic interactions and Brownian motion of the dispersed nanoparticles, rather than by polymer normal stresses as originally proposed. In the second, we use a newly developed fluidic four-roll mill (FFoRM) in order to probe how the flow-induced alignment of rodlike nanoparticles in polymer solutions depends on the type of applied deformation.

WK5

Coupling Large-amplitude Oscillatory Shear Stress Measurements to Microstructure Changes via *In Situ* Rheo-scattering

Simon A. Rogers
Department of Chemical and Biomolecular Engineering, University of Illinois at Urbana-Champaign, Champaign, IL 61801

Large-amplitude oscillatory shear (LAOS) has become a popular technique for probing soft materials far from equilibrium by accessing their nonlinear rheological responses. In such tests, the stress-strain relationships cannot be described by linear differential equations with constant coefficients, and so new methods must be used to provide meaning to the measured results. Over the past two decades, a number of different mathematical formalisms have been proposed for gaining physical meaning from LAOS tests, including Fourier transformation, describing the stress decomposition by Chebyshev polynomials, and the sequence of physical processes methods. Each of these methods provides a different set of interpretations for the same experiment. An important question is which formalism, if any, is right?

In this presentation, I will introduce the most popular methods for understanding the rheological responses to LAOS experiments and then discuss the integral role of time-resolved rheo-scattering studies, which provide molecular-level information regarding microstructural rearrangements. The development of time-resolved rheo-scattering experiments has provided deep insight into how soft materials respond to deformations that take them far from equilibrium. These insights are calling into question the adequacy of some of the traditional analysis methods, while providing unique datasets by which new concepts can be tested.

WK5

Studies of Shear Effects on Aggregation and Gelation

C.M. Sorensen
Department of Physics, Kansas State University, Manhattan, KS 66506

This paper will describe light scattering and rheological studies of colloid and polymer solution aggregation and gelation under shear. Shear is found to enhance the aggregation rate and decrease the gel time. Shear also

uncovers the percolated superaggregate structure in the nascent particulate gel. For polymer gels it can act as a state variable for the behavior of the viscosity.

WK5

Transport of Interacting Nanoparticles in Complex Polymeric Fluids

Ryan Poling-Skutvik, Ramanan Krishnamoorti, and Jacinta C. Conrad

Department of Chemical and Biomolecular Engineering, University of Houston, Houston, TX 77004

Transport of nanoparticles through complex fluids is essential for environment remediation, nanocomposite processing, and targeted drug delivery. Because nanoparticles are of comparable size to the heterogeneities present in many complex fluids, their dynamics decouple from the bulk fluid properties. Thus, nanoparticles can transport many orders of magnitude faster than expected. To understand the underlying physics of transport in this size regime, we measure the dynamics of nanoparticles moving through polymer solutions, which serve as model complex fluids with well-controlled and tunable heterogeneities. On short time scales, the nanoparticles move subdiffusively through the heterogeneous solutions, coupling to the segmental and center-of-mass dynamics of the polymer coils. On long time scales, the nanoparticle dynamics deviate from Stokes-Einstein predictions and depend on particle size and polymer concentration. Nanoparticle dynamics in polymer solutions are further modified through surface modifications, specifically electrostatic charges or grafted polymers. Electrostatic charges lead to long-range interactions between the particles in organic solvents without disrupting the structure or dynamics of the surrounding polymer solution. The long-range interparticle interactions slow nanoparticle dynamics across the interparticle distance, even though the nanoparticle dynamics are subdiffusive and coupled to the polymer relaxations. Grafted polymers help to stabilize the nanoparticles in complex fluids and lead to soft physical interactions between the grafted particles and the surrounding polymer chains with implications on their transport behavior. Our work illustrates that tuning the nanoparticle size and modifying the particle surface chemistry grants excellent control over the transport properties of nanoparticles through complex media.

WK5

Universal Scaling of Quench-dependent Dynamics in Intermediate Concentration Colloidal Gels

D. Bahadur[1], S. Ramakrishnan[1], Q. Zhang[2], A. Sandy[2], and R. Leheny[3]

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We have examined the formation and dissolution of gels composed of intermediate volume fraction nanoparticles with temperature-dependent short-range attraction using small-angle xray scattering (SAXS), x-ray photon correlation spectroscopy (XPCS), and rheology to obtain nanoscale and macroscale sensitivity to structure and dynamics. Gel formation after temperature quenches to the vicinity of the rheologically-determined gel temperature T_{gel} was characterized via the slow-down of dynamics observed in the intensity autocorrelation functions (g_2) as a function of quench depth ($\Delta T = T - T_{gel}$), wave vector, and formation time (t_f). We find self-similarity in the slow-down of dynamics that maps the wave-vector-dependent dynamics at a particular ΔT and t_f to that at other ΔTs and t_fs via an effective scaling temperature, T_s. A single T_s applies to a broad range of ΔT and t_f but does depend on the particle size. The rate of dynamical evolution implied by the scaling is a far stronger function of ΔT than the attraction strength between colloids. We interpret this strong temperature dependence as collective particle motion possibly arising from rearrangements of energetically-favored locally structures observed in confocal microscopy and via simulations.

WK5

From Flocking Birds to Swarming Bacteria: Study of the Dynamics of Active Fluids Using Fast Confocal Rheometry

Xiang Cheng

Chemical Engineering and Materials Science, University of Minnesota, Minneapolis, MN 55455

Active fluids are a novel class of non-equilibrium complex fluids with examples across a wide range of biological and physical systems such as flocking animals, swarming microorganisms, vibrated granular rods, and suspensions of synthetic colloidal swimmers. Different from familiar non-equilibrium systems where free energy is injected from boundaries, an active fluid is a dispersion of large numbers of self-propelled units, which convert the ambient/internal free energy and maintain non-equilibrium steady states

at microscopic scales. Due to this distinct feature, active fluids exhibit fascinating and unusual behaviors unseen in conventional complex fluids. Here, combining high-speed confocal microscopy, holographic imaging, *in situ* rheological measurements and biochemical engineering, we experimentally investigate the dynamics of active fluids. In particular, we use *E. coli* suspensions as our model system and illustrate three unique properties of active fluids (i.e., (i) abnormal rheology, (ii) enhanced diffusion of passive tracers and (iii) emergence of collective swarming). Using theoretical tools of fluid mechanics and statistical mechanics, we develop a quantitative understanding of these interesting behaviors. Our study illustrates the general organizing principles of active fluids that can be exploited for designing “smart” fluids with controllable fluid properties. Our results also shed new light on fundamental transport processes in microbiological systems.

Tuesday, May 8

CNM Workshop 6
Tribology of 2D Materials: From Nanoscale to Macroscale

Location: Building 440, Room A105/106

Organizers: Anirudha V. Sumant and Subramanian Sankaranarayanan (CNM), Ali Erdemir (ES)

The purpose of this workshop is to bring together experts from academia, national labs and industry to discuss the latest developments in the tribology of 2D materials from nano/microscale to meso-macroscale. The emphasis will be on uncovering fundamental mechanisms of wear/friction at atomic scales and finding a link to translate at the macroscale. Another aspect includes showcasing CNM's unique multifunctional tribometer to the tribology community and fostering new collaborations through the CNM user proposal mechanism.

Workshop topics:

- Superlubricity using 2D materials
- Physical/chemical interactions between 2D material layers and bulk substrates
- Theory/simulations for understanding mechanisms of sliding/shearing
- 2D materials as solid lubricants
- 2D materials as additives in composites and in liquid lubricants
- Advances in tribological properties measurements techniques
- Tribological properties of 2D materials and the mesoscopic link between nano and macro scales
- Environmental considerations

The CNM has expertise and state-of-the-art facilities in the synthesis, functionalization and characterization of 2D materials with tribological properties, with particular emphasis on understanding from nano-to-micro-to-macro scale. Applications may include new solid lubricant materials for NEMS/MEMS (nano-micro) to moving electrical contacts, sliding/rolling, rotating and bearings, etc. (meso-macro). This workshop will include topics covering fundamental studies on the development of 2D materials and their systematic characterization to understanding their structural, chemical, mechanical, and tribological properties. Also welcome are theoretical and modeling approaches that can provide in-depth understanding of tribo-physical and chemical interactions with the substrate and interfaces under severe contact pressure and shear stress; issues related to tribo-chemical changes, and substrate interactions. This workshop is expected to provide an excellent platform for academics, scientists, and students to exchange ideas, foster collaborations with user facilities at Argonne and embark upon new challenges in nanoscience.

Session 1: Physical/Chemical Interactions between 2D Materials Layers (Session Chair: Anirudha Sumant)

8:50 – 9:00	Anirudha V. Sumant (Center for Nanoscale Materials) *Welcome and Introduction*
9:00 – 9:30	James Batteas (Texas A&M University) *Controlling Friction, Energy Dissipation, and Chemical Reactivity in 2D Nanomaterials*
9:30 – 10:00	Graham Cross (Trinity College) *Self-assembly of Graphene Ribbons on a Substrate at the Micrometer Scale*
10:00 – 10:30	Break

Session 2: Theory/Simulations Studies on Understanding Friction and Mechanics at the Interface (Session Chair: Subramanian Sankaranarayanan)

10:30 – 11:00 Horacio Espinosa (Northwestern University)
Intra and Interlayer Mechanical Properties of Graphene Oxide

11:00 – 11:30 Izabela Szlufarska (University of Wisconsin-Madison)
Multi-scale Model of Time Dependent Friction

11:30 – 12:00 Srilok Srinivasan (Argonne National Laboratory)
Effect of Graphene Wrapped Nanodiamond vs. Graphitized Nanodiamond on the Superlubricity (contributed talk)

12:00 – 1:30 Lunch

Session 3: 2D Lubricants as an Additives in Oil and in Composites (Session Chair: Diana Berman)

1:45 – 2:15 Jiaxing Huang (Northwestern University)
Self-dispersed Crumpled Graphene Balls in Oil for Friction and Wear Reduction

2:15 – 2:45 Elena Polyakova (Graphene Laboratories, Inc.)
An Update on Development of Graphene-enhanced Composite Materials

2:45 – 3:05 Kalyan Mutyala (Argonne National Laboratory)
Iron Nanoparticle-driven Tribochemistry Leading to the Superlubric Sliding Interfaces (contributed talk)

3:05 – 3:30 Break

Session 4: 2D Materials as Solid Lubricants (Session Chair: Ali Erdemir)

3:30 – 4:00 Subramanian Sankaranarayanan (Argonne National Laboratory)
Machine Learnt Models for 2-D Materials and Their Tribological Applications

4:00 – 4:30 Diana Berman (University of North Texas)
Mechanisms for Controlling Friction and New Approaches for Achieving Superlubricity Regime in 2D Materials

4:30 Ali Erdemir (Argonne National Laboratory)
Wrap-up and Closing Remarks

Optional: Tour to show multifunctional tribometer facility at CNM

WK6

Controlling of Friction, Energy Dissipation, and Chemical Reactivity in 2D Nanomaterials

Meagan Elinksi, Zhuotong Liu, Maelani Negrito, Nathaniel Hawthorne, and James Batteas
Departments of Chemistry and Materials Science and Engineering, Texas A&M University, College Station, TX 77843

Control of friction and wear is a ubiquitous challenge in numerous machined interfaces ranging from biomedical implants, to engines, to nano- and micro-scaled electromechanical systems (MEMS) devices. A key challenge in developing boundary lubrication schemes for such systems is how to reduce wear at the rough surfaces of such devices, where nanoscaled asperities dominate the interfacial contacts. The robust mechanical properties and general chemical inertness of two-dimensional (2D) nanomaterials, such as graphene and MoS_2, has made them of interest for modifying surface frictional properties. While single layer graphene and MoS_2 can readily adapt to surface structure on the atomic scale, when deposited on substrates with nanoscopic roughness of ~10 nm rms (as is common in many machined interfaces) a conformal coating generally cannot be fully formed, due to competition between adhesion to the substrate nanoscopic asperities and the bending rigidity of the material. This often leaves a mixture of supported and unsupported regions which respond differently to applied load, with spatial variations in mechanical properties and chemical bonding. Increased strain in these materials on rough surfaces also makes them susceptible to chemical reactions that can

lead to tribochemical breakdown. Here, we describe a combination of AFM nanomechanical and confocal Raman microspectroscopy studies of graphene and MoS_2 on silica surfaces with controlled nanoscopic roughness, to examine the how this impacts their frictional properties and alters their tribochemical reactivity, where strain dependent reactions can be driven by applied forces.

WK6

Self-assembly of Graphene Ribbons on a Substrate at the Micrometre Scale

Graham L.W. Cross

AMBER and CRANN Institutes and School of Physics, Trinity College, Dublin 2, Ireland

We discuss the large-scale self-assembly of graphene ribbons we recently discovered for surface adhered sheets exfoliated onto silicon oxide substrates [1]. Directed folding of flap-like structures seeds growth of long ribbons that spontaneously peel and tear the sheet as they slide in superlubricous fashion [2]. We observe ribbon growth up to 20 micrometers in size in ambient conditions. Measurement of ribbon velocity versus width in a slow growth regime reveals a logarithmic dependence consistent with thermally-activated bond dissociation. Our analysis suggests this form of self-assembly may be a general phenomenon common to a large class of 2D materials.

In this paper we present a theoretical treatment of the phenomena based an energy minimization analysis technique [3] originally applied to macroscopic tearing of adhered sheets. Our simple model predicts ribbon length and taper angle based on a balance of forces acting on two crack tips. The analysis predicts a driving force proportional to ribbon width based on self-affinity of the graphene relative to the substrate, and suggests the phenome may be general for any thermally activated sheet.

[1] Annett, J. and Cross, G.L.W. (2016). "Self-assembly of graphene ribbons by spontaneous self-tearing and peeling from a substrate." *Nature* **535**: 271–275.

[2] Liu, Z. et al. (2012). "Observation of microscale superlubricity in graphite." *Physical Review Letters* **108**: 205503.

[3] Hamm, E., Reis, P., LeBlanc, M., Roman, B. and Cerda, E. (2008). "Tearing as a test for mechanical characterization of thin adhesive films." *Nature Materials* **7**: 386–390.

WK6

Intra and Interlayer Mechanical Properties of Graphene Oxide

Horacio D. Espinosa

James and Nancy Farley Professor of Mechanical Engineering, Northwestern University, Evanston, IL 60208

In the past decade, there has been a major thrust to synthesize low dimensional materials exhibiting unique and outstanding physical properties. These nanomaterials are envisioned as building blocks for the next generation of lightweight materials, electronics, sensors, and energy systems. In these applications, identification of intra- and inter-layer mechanical properties is essential. However, such endeavor has proven challenging from both experimental and modeling perspectives. In this presentation, progress in nanoscale mechanical experimentation and modeling will be summarized. We will discuss the mechanical properties of graphene oxide, a material presenting functional groups ideal for synthesis of multilayer nanocomposites and membrane filtration films. The effect of chemical functional group type and density on monolayer toughness, stiffness, and strength will be ascertained using a combination of nanomechanics experiments and molecular modeling. Pathways for achieving a several-fold increase in the toughness of graphene oxide monolayers together with AFM adhesion and friction studies between GO monolayers will be presented and their implication on the design of devices analyzed.

WK6

Multi-scale Model of Time Dependent Friction

Izabela Szlufarska[1], Zhuohan Li[1], and Yun Liu[2]

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

[2] Apple, Inc., Cupertino, CA 95014

We have developed a multi-scale simulation approach to investigate the effects of surface chemistry and surface roughness on the origin of static friction, which is a highly debated topic in the field of tribology. This approach combines density functional theory calculations of chemical reactions at interfaces, kinetic Monte Carlo simulations that extend simulation time scales to those that can be accessed in experiments, and boundary element method to extend simulation length scale and investigate multi-asperity contacts. For most solid/solid contacts, static friction increases logarithmically with time, a phenomenon known as aging. We discovered molecular mechanisms that are responsible for aging of silica/silica contacts in the presence of water and that are based purely on interfacial chemistry. By combing simulations with atomic force

microscopy (AFM) experiments we identified the effects of contact pressure on both friction and aging, showing that aging is accelerated when the contact pressure is increased. Our model shows a quantitative agreement with AFM experiments on load-dependence of aging in single-asperity contacts. In addition, our model predicts that aging is a non-monotonic function of temperature and therefore the temperature can be used to control this phenomenon. Finally, we make predictions of how chemical aging will scale to large rough surfaces and how it will depend on the nature of the contact roughness.

Y. Liu and I. Szlufarska (2012). "Chemical origins of frictional aging." *Phys. Rev. Lett.* **109**: 186102.

K. Tian, N.N. Gosvami, D.L. Goldsby, Y. Liu, I. Szlufarska, and R. Carpick (2017). "Load and time dependence of interfacial chemical bond-induced friction at the nanoscale." *Phys. Rev. Lett.* **118**: 076103.

Z. Li and I. Szlufarska. "Multi-physics nature of frictional aging," (submitted).

Z. Li, L. Pastewka, and I. Szlufarska. "Chemical aging of large-scale randomly rough contacts," (to be submitted).

WK6
Effect of Graphene Wrapped Nanodiamond vs. Graphitized Nanodiamond on the Superlubricity

Srilok Srinivasan[1], Diana Berman[1], Subramanian Sankaranarayan[1], Ali Erdemir[2], and Anirudha V. Sumant[1]

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

[2] Energy Systems Division, Argonne National Laboratory, Argonne, IL 60439

Reducing the friction between moving mechanical parts has direct implications in variety of applications ranging from micromechanical systems to large-scale automotive and mechanical systems and can help save huge amounts of energy, which would otherwise be lost due to frictional dissipation. Macroscale superlubricity has been demonstrated between diamond like carbon (DLC) and nanodiamond (ND) in the presence of graphene flakes dispersed on SiO_2 or silicon surface in dry atmosphere. Atomistic simulations show that graphene flakes wrap around the ND at the sliding interface leading to superlubric behavior. While the graphene wrapped ND has a low contact area and results in an incommensurate contact, a nanodiamond with similar template can be synthesized by graphitizing the outer surface of the ND resulting in a diamond core and graphitized shell. The experimental results demonstrated at least 10 times higher friction in case of graphitized nanodiamond. Here, we elucidate the mechanism of energy dissipation by comparing the frictional forces between the DLC/graphene wrapped-ND interface and DLC/graphitized-ND interface using atomistic simulations. Our results explain the higher friction observed experimentally in case of graphitized nanodiamond on the basis of interaction of wrapped graphene which allow easy rotation of ND while in case of graphitized ND, chemical bonding between graphitized carbon and core ND prevent such rotation resulting in increased work and thus higher friction.

WK6
Self-dispersed Crumpled Graphene Balls in Oil for Friction and Wear Reduction

Jiaxing Huang

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

Aggregation is a major problem for ultrafine particle additives in lubricant oil because it reduces the effective particle concentrations, prevents particles from entering the contact area of working surfaces, and leads to unstable tribological performance. Molecular ligands can help the particles to disperse, but they tend to degrade under the harsh tribological conditions. Therefore, self-dispersed particles without the need for surfactant are highly desirable. Here I will discuss such type of ultrafine particles made of crumpled, paper-ball-like graphene, which indeed can self-disperse in lubricant oil, and exhibit stable tribological performances.

WK6
An Update on Development of Graphene-enhanced Composite Materials

Elena Polyakova

Graphene Laboratories, Inc., Calverton, NY 11933

Graphene materials completely dispersed in various polymer matrices represent the first commercially feasible use of graphene. In this talk, we focus on our current developments in high-performance graphene composites based on both thermoplastic and thermoset resins. One of our first graphene products is PLA-based conductive filament with volume resistivity as low as 0.6 Ω cm. This material is compatible with most of the commercial FDM printers and suitable for creating conductive traces and sensors. In addition, we have created a line of graphene composites and masterbatches that can then be diluted for further processing into parts by extrusion or injection molding. Another area of significant progress in functional graphene materials is the launch of several graphene-enhanced conductive adhesives distributed under trademark G6-EPOXY™ (visit www.g6-epoxy.com for the full list of specifications). We have developed metal-free carbon-based epoxy with volume resistivity below 5 Ω cm, making it one of the best metal-free conductive adhesives currently available in the market.

We have also developed hybrid epoxies (highly electrically conductive adhesives) with a proprietary formulated blend of graphene, silver fillers and other additives. With the development of this proprietary formulation, we have not only guaranteed the electrical conductivity of these epoxies to be at par with the existing commercial products but have also successfully managed to substantially reduce the percentage of silver content in the epoxies. The critical belief behind reducing silver content had two major points: 1) improving the mechanical properties and ease of processing and 2) keeping the cost price of the product low, thereby making it more commercially viable and attractive for a variety of applications.

WK6

Iron Nanoparticle-driven Tribochemistry Leading to the Superlubric Sliding Interfaces

Kalyan C. Mutyala[1], Diana Berman[1], Srilok Srinivasan[1], Subramanian K.R.S. Sankaranarayanan[1], Ali Erdemir[2], Elena Shevchenko[1], and Anirudha V. Sumant[1]

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

[2] Energy Systems Division, Argonne National Laboratory, Argonne, IL 60439

High contact pressure and shear during relative movement of the sliding interfaces result in the formation of a tribofilm, which often dominates the friction and wear performance of such interfaces. However, the nature of the tribofilm formed as well as how it influences friction and wear remain poorly understood and pose uncertainties in predicting tribo-performance. We demonstrate that iron nanoparticles in the presence of graphene at the sliding interface undergo tribochemical reaction under high contact pressures leading to the formation of onion-like-carbon nanostructures (OLCs). We show that such tribochemical reaction is observed for pure iron nanoparticles and depends sensitively on the core-shell chemistry of the nanoparticle. In case of iron nanoparticles with oxidized shell, the tribochemical reaction seems to be hampered by the presence of an oxide shell that prevents diffusion of carbon into the iron nanoparticle and hence the formation of OLCs. Formation of OLCs facilitate superlubricity demonstrating interesting interplay of stress-induced tribochemistry at the tribological interface.

WK6

Machine Learnt Models for 2D Materials and Their Tribological Applications

Subramanian Sankaranarayanan

Argonne National Laboratory, Argonne, IL 60439

Recent advances in machine learning (ML) and data science algorithms, along with high performance computing (HPC) capabilities, present unique opportunities to address cutting edge problems in material science. In parallel, modern supercomputers together with the availability of highly scalable atomistic simulation codes have begun to revolutionize the modeling and computational analyses of materials. Molecular dynamics (MD) is one such powerful technique, which has a broad user base. There are various flavors of MD from the highly accurate *ab initio* molecular dynamics (AIMD) to semi-empirical atomistic MD to more computational efficient coarse-grained (CG) models. There is still a substantial gap between AIMD, which are computationally intractable for large systems, and those based on classical force fields (atomistic and CG) that rely on pre-defined functional form for inter/intra atomic/bead interactions, which inherently limits their ability to capture physics for complex interfaces. To bridge this gap, we have developed a machine-learning framework that combines the accuracy and flexibility of electronic structure calculations with the speed of classical potentials. This framework enables accurate prediction of inter-atomic forces and thereby allow high fidelity dynamical and statistical simulations of tribological interfaces, properties and functionalities of new 2D lubricant materials, as well as pathways and mechanisms of their in operando synthesis and assembly. Our framework has the potential to achieve paradigm-changing breakthroughs in materials design for tribological applications. It would ultimately lead to next generation lubricant materials and technology that provide innovative solutions to pressing environmental and energy challenges facing our nation.

WK6

Mechanisms for Controlling Friction and New Approaches for Achieving Superlubricity Regime in 2D Materials

D. Berman[1], A. Erdemir[2], and A.V. Sumant[3]

[1] Materials Science and Engineering, University of North Texas, Denton, TX 76207

[2] Engineering Systems Division, Argonne National Laboratory, Argonne, IL 60439

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

Friction is an important aspect of many areas of everyday life. Varieties in mechanical systems from nano to macroscale and efforts to minimize energy losses intersect with challenges of controlling the friction. The popularity of recently discovered 2D materials and their usefulness for multiple applications enabled understanding the friction at a more fundamental level and opened new routes for manipulating friction to superlubricity or near zero friction values.

Here, we review the basic mechanisms that complies the frictional energy dissipation, such as wear, molecular deformation, thermal effect, electronic effect, bonding, environment and chemistry, phonons, and structural effect. We present the case studies highlighting how these mechanisms are controlled in 2D materials. Finally, we highlight recent advances in implementing 2D materials for friction reduction to superlubricity across scales from nano- up to macroscale contacts. We show that nanoscale superlubricity mechanisms originating from the formation of frictionless nanoscale systems, as in case of graphene-nanodiamond scrolls and carbon nanoonion structures, can be successfully transferred to macroscale effects. Development of 2D materials opened a new pathway for manipulating friction, which makes superlubric friction today's reality.

[1] D. Berman, et al. (2018). "Approaches for Achieving Superlubricity in Two-Dimensional Materials," *ACS Nano* **12**(3): 2122–2137.

[2] D. Berman, et al. (2015). Special issue in *Diamond and Related Materials* **54**: 91.

[3] D. Berman, et al. (2014). *Materials Today* **17**: 31–42.

[4] D. Berman, et al. (2015). *Science* **348**: 1118–1122.

Wednesday, May 9

APS Workshop 7
High-resolution 3D X-ray Imaging

Location: Building 401, Room A1100

Organizers: Si Chen, Doga Gursoy, and Vincent de Andrade (APS)

Many science areas have been revolutionized by the use of high-resolution x-ray tomography. Both the structural information and elemental/chemical or magnetic distribution can be revealed in 3D in a non-destructive fashion. These studies have been further advanced by combining tomography with the development of scanning nanoprobes and full-field systems like transmission x-ray microscopy (TXM) or projection microscopes, with spatial resolution routinely operated at sub-100 nm and expected to approach sub-10 nm in the case of coherent diffraction imaging techniques after the upgrade of the storage rings with the multi-bend achromat lattice at the APS and other light sources. As the spatial resolution approaches nanoscale, new challenges arise due to the limitations in various aspects, including instrument stability, measurement precision, rotation stage runout, and shallow depth of field of focusing optics. Both instrumentation and novel methods in data collection and processing are essential to realize the potential of high-resolution 3D imaging. This workshop will serve to strengthen collaborations among the synchrotron x-ray nano-imaging community to elevate high-resolution 3D x-ray imaging beyond demonstration stage and make it practical for routine user operations.

8:30 – 8:40	Opening Remarks
8:40 – 9:10	Evgeny Nazaretski (NSLS-II, Brookhaven National Laboratory) *Hard X-ray Imaging Approaching 10 nm Spatial Resolution: Instrumentational Challenges and Solutions*
9:10 – 9:40	Jan Garrevoet (Deutsches Elektronen-Synchrotron) *Spectroscopic X-ray Imaging at the Nanometer Length Scale*
9:40 – 10:00	Nestor Zaluzec (Argonne National Laboratory) *Hyperspectral Imaging and Tomography Using Electron-optical Beam Lines*
10:00 – 10:30	Break
10:30 – 11:00	Yuan-Hung (Mike) Lo (University of California, Los Angeles) *3D Coherent X-ray Diffractive Imaging of Biological Structures*
11:00 – 11:30	Mingyuan Ge (NSLS-II, Brookhaven National Laboratory) *Quantitative Analysis of Fluorescent Nanotomograpy and Recent Progress of the TXM Beamline at NSLS-II*
11:30 – 12:00	Ke Yuan (Argonne National Laboratory) *Investigation of Metal-mineral Interactions by Transmission X-ray Microscopy*
12:00 – 1:30	Lunch
1:30 – 2:00	Young-Sang Yu (Advanced Light Source) *Three-dimensional Localization of Nanoscale Battery Reactions Using Soft X-ray Tomography*
2:00 – 2:30	Peng Li (University of Sheffield) *Ptychographic Tomography*
2:30 – 3:00	Stefano Marchesini (Advance Light Source) *Ptycho-tomography: Algorithms and Challenges*
3:00 – 3:30	Break

3:30 – 4:00	Junjing Deng (Argonne National Laboratory) *High-resolution Fast Ptychography with Photon-efficient Scanning*
4:00 – 4:30	Vincent de Andrade (Argonne National Laboratory) *Fast X-ray Nano-tomography with Full-field Techniques*
4:30	David Vine (Sigray) *Sigray UltimaXRM: Designing a Next-generation X-ray Microscope for On-demand Nanoscale Tomography*

WK7

Hard X-ray Imaging Approaching 10 nm Spatial Resolution: Instrumentational Challenges and Solutions

E. Nazaretski
NSLS-II, Brookhaven National Laboratory, Upton, NY 11973

The hard x-ray nanoprobe (HXN) beamline at NSLS-II has been designed and constructed to enable imaging experiments with unprecedented spatial resolution and detection sensitivity. The HXN x-ray microscope is a key instrument for the beamline, providing a suite of experimental capabilities which includes scanning fluorescence, diffraction, differential phase contrast and ptychography utilizing multilayer Laue lenses (MLL) and zone plates (ZP) as nanofocusing optics. During this presentation, different phases of the x-ray microscope development process will be reviewed. Various prototype systems designed and constructed prior to completion of the HXN-microscope will be discussed. Experimental data demonstrating ~11 × 12 nm^2 spatial resolution imaging using MLL optics will be presented. I will discuss instrumentational challenges associated with high spatial resolution 2D and 3D imaging and will outline future development plans.

WK7

Spectroscopic X-ray imaging at the Nanometer Length Scale

Jan Garrevoet
Deutsches Elektronen-Synchrotron (DESY) Notkestraße 85, 22607 Hamburg, Germany

The latest developments in x-ray optics and x-ray detector technology, open new avenues for the determination of 2D/3D elemental distribution and speciation within the investigated samples, both on the microscopic as on the nanoscopic scale. The P06 hard x-ray micro/nano-probe is specialised in several imaging techniques, which make use of these new technologies.

The nanoprobe experiment utilises several different optics, depending on the needed incident energy, for focusing the coherent part of the x-ray beam to typical beam sizes of 50–100 nm. It is designed for scanning coherent x-ray diffraction microscopy (ptychography) [1], but is used also for nano-XRF and nano-XRD imaging.

The Microprobe is a versatile experiment for scanning x-ray microscopy with x-ray fluorescence, x-ray absorption spectroscopy and x-ray diffraction contrasts. A KB system focusses a beam of some 10^{10} photons/s down to 300 nm focus size in the energy range 5–21 keV.

Compound refractive lenses (CRL's) are used at higher energies up to 80 keV. Advanced detector technology, namely the Maia x-ray fluorescence detector and the EIGER X 4M hybid photon counting detector, enable on-the-fly scanning schemes with millisecond dwell times per scan pixel. The ability to collect megapixel images in less than an hour facilitates series of 2D images for full 3D multimodal-tomography, spectro-microscopy, time-resolved *in situ* microscopy or other multi-dimensional microscopic experiments.

Several examples will be shown covering, XRF-XRD tomography on bone, XRF-ptychography tomography on material science samples, and others [2].

[1] A. Schropp, R. Hoppe, J. Patommel, D. Samberg, F. Seiboth, S. Stephan, G. Wellenreuther, G. Falkenberg, and C.G. Schroer (2012). *Appl. Phys. Lett.* **100**: 253112.

[2] S. Kalirai, U. Boesenberg, G. Falkenberg, F. Meirer, and B.M. Weckhuysen (2015). *ChemCatChem* **7**: 3674–3682.

WK7

Hyperspectral Imaging and Tomography Using Electron-optical Beam Lines

Nestor J. Zaluzec
Photon Sciences Division, Argonne National Laboratory, Argonne, IL 60439

Electron-optical beams complement the capabilities of x-ray beam lines for high spatial resolution imaging, diffraction, and spectroscopy for characterization of today's challenging problems. In this presentation, we will discuss the current capabilities and future prospects which can be accomplished by correlative and complementary investigations at Argonne.

WK7

3D Coherent X-ray Diffractive Imaging of Biological Structures

Yuan-Hung Lo[1,2], Junjing Deng[3], Marcus Gallagher-Jones[4], Si Chen[3], Alan Pryor Jr.[1], Qiaoling Jin[5], Young Pyo Hong[5], Stefan Vogt[3], Chris Jacobsen[3,5,6], and Jianwei Miao[1]

[1] Department of Physics and Astronomy and California NanoSystems Institute, University of California, Los Angeles, CA 90095

[2] Department of Bioengineering, University of California, Los Angeles, CA 90095

[3] Advanced Photon Source, Argonne National Laboratory, Argonne, IL 60439

[4] Department of Chemistry and Biochemistry, UCLA-DOE Institute for Genomics and Proteomics, Los Angeles, CA 90095

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

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

As far back as 1860, Lionel Beale demonstrated the power of microscopy by using a basic microscope to observe changes in nuclear size and shape in cells from pharyngeal cancer patients. Nowadays, high resolution bioimaging is becoming an increasingly important tool for life scientists to visualize and decipher the intricate machineries of life. In the realm of mesoscale imaging, which spans the length scale greater than the molecular machinery and smaller than a single cell, coherent diffractive imaging [1] (CDI) and its scanning version, termed ptychography [2,3], emerge as an effective tool for visualizing cellular ultrastructure and tracking targeted small molecules, organelles and biomarkers [4]. X-ray's short wavelength and high penetration depth enables 3D imaging of whole cells with tens of nanometer resolution [5], while circumventing the need for time-consuming sample pre-processing steps such as fluorescent labelling, chemical staining or sectioning. Moreover, the technique complements scanning transmission x-ray microscopy and x-ray fluorescence microscopy to offer rich, correlative information on sub-cellular organization and chemical composition. Together, they have enabled multiscale 3D imaging of nanoparticle internalization in intact cells [6] and correlative structural imaging and organelle identification of un-labelled cells [7,8]. As the tool continues to advance, it will find useful applications in biology and medicine in areas such as nanotoxciology, biomineralization, bioenergy and environmental systems. In this talk, I will provide an overview of ptychography and a recently developed iterative Fourier-based tomography algorithm, GENFIRE [9,10], as well as highlight recent advances in high resolution x-ray 3D imaging in the context of biology. I will also discuss current challenges and future directions of the field.

[1] J. Miao et al. (1999). *Nature* **400**: 342–344.

[2] J.M. Rodenburg et al. (2007). *Phys. Rev. Lett.* **98**: 034801.

[3] P. Thibault et al. (2008). *Science* **321**: 379–382.

[4] J. Miao, T. Ishikawa, I.K. Robinson, and M.M. Murnane (2015). *Science* **348**: 530–535.

[5] J. Rodriguez et al. (2015). *IUCrJ* **2**: 575–583.

[6] M. Gallagher-Jones et al. (2017). *Sci. Rep.* **7**: 4757.

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

[8] J. Deng et al. (2015). *PNAS* **112**: 2314–2319.

[9] Y. Yang et al. (2017). *Nature* **542**: 75–79.

[10] A.J. Pryor et al. (2017). *Sci. Rep.* **7**: 10409.

WK7

Quantitative Analysis of Fluorescent Nanotomograpy and Recent Progress of the TXM Beamline at NSLS-II

Mingyuan Ge, Xiaojing Huang, Hanfei Yan, Yong S. Chu, Evgeny Nazaretski, David Scott Coburn, and Wah-Keat Lee

NSLS-II, Brookhaven National Laboratory, Upton, NY 11973

Fluorescent nanotomography available at the hard x-ray nanoprobe (HXN, NSLS-II) can provide 3D nanoscale elemental distribution with high spacial resolution. However, accurate quantification of 3D elemental concentration is hampered by a well-known self-absorption problem, and it is particular severe for the low energy fluorescence x-rays. Correction of self-absorption is a non-trivial task and requires an iterative and three-dimensional solution. In the first part of the presentation, I will describe an approach using experimental data taken from mixed ionic ceramic membrane samples and demonstrate how accurate absorption correction leads to a discovery of a new material phase in this material system. In the second part of the presentation, I will report on the recent progress of a newly constructed TXM beamline (18-ID) at NSLS-II. This home-designed full-field microscope is capable of 2D and 3D imaging with 30 nm resolution. Fast tomography is realized, which is benchmarked to finish a 3D scan within 1 minute.

WK7

Investigation of Metal-mineral Interactions by Transmission X-ray Microscopy

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Interactions of aqueous metal ions with mineral surfaces play a critical role in understanding the mobility of metals in subsurface environments, templating growth of secondary mineral phases, and top-down design of complex surface structures. Here, we showed an example of calcite interactions with Pb^{2+} and Fe^{2+} ions respectively during the mineral dissolution reaction [1–3]. In Pb-calcite interactions, calcite exhibits non-classical surface features characterized as micro pyramids developed spontaneously in acidic Pb-bearing solutions. Subsequent pseudomorphic growth of a cerussite ($PbCO_3$) phase was observed, where nucleation occurred entirely within a pore space created by dissolution at the calcite/substrate interface. Cerussite was separated from the calcite by pores of less than 200 nm as resolved by the transmission x-ray microscopy (TXM), consistent with an interface-coupled dissolution-precipitation mechanism. In the study of Fe-calcite interactions, formation of iron oxyhydroxide layers having distinct morphologies was observed during the dissolution of calcite in acidic Fe(II)-rich solutions. A pseudomorphic lepidocrocite (γ-FeOOH) shell together with multiple iron oxyhydroxide layers encapsulated within the shell was imaged by optical and transmission x-ray microscopies. Subsequent detachment of the lepidocrocite film from the dissolving calcite surface yielded a freestanding pseudomorphic iron oxyhydroxide micro box. In both cases, TXM provided the key morphological information in understanding the kinetics of mineral dissolution and formation mechanism of complex structures at the mineral-water interface.

This material is based on work supported by the U.S. Department of Energy, Office of Basic Energy Sciences, Division of Chemical Sciences, Geosciences, and Biosciences through Argonne National Laboratory. Argonne is a U.S. Department of Energy laboratory managed by UChicago Argonne, LLC, under Contract No. DE-AC02-06CH11357. 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. 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.

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WK7

Three-dimensional Localization of Nanoscale Battery Reactions Using Soft X-ray Tomography

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Techniques capable of analyzing chemical states at high spatial resolution are essential for elucidating the complex phenomena at the nanoscale that underpin materials' properties. For example, battery function is determined by the efficiency and reversibility of the electrochemical phase transformations at solid electrodes, creating the need to accurately define relationships between chemistry, mechanics and morphology. Conventional x-ray imaging methods are well suited to probe chemical states in bulk matter, but they are also limited in spatial resolution to a few tens of nanometers by the x-ray optics. Furthermore, bulk x-ray diffraction can unambiguously differentiate between two-phase and metastable single-phase delithiation pathways but it cannot map heterogeneities in the spatial distribution of such states. In turn, electron-based techniques achieve very high spatial resolution and can provide three-dimensional (3D) quantification of the chemical state10, but they also suffer from diffraction contrast effects and non-linearities for material thicknesses greater than the mean-free-path of inelastic scattering. Soft x-ray ptychography has recently narrowed the gap in spatial resolution while retaining high sensitivity to chemical states and penetration through functional volumes of matter [1]. If data is only collected along one two-dimensional (2D) projection, the analysis of complex systems becomes problematic because of the likelihood of overlapping material with differing chemical components. This problem is readily solved by the use of x-ray based computed tomography, but the quantification of chemical states in three dimensions by conventional methods comes with limited spatial resolution, which is currently, at best, 30 nm.

In this study, soft x-ray tomography combined with ptychography, developed at beamline 5.3.2.1 at the

Advanced Light Source (Berkeley, CA) is applied to assess the chemical and morphological consequences of electrochemical delithiation of aggregated nano-sized lithium iron phosphate ($LiFePO_4$) plates. Our work provides quantitative analysis of oxidation states in 3-dimensions at the 10 nm scale [2]. After segmentation, statistical analysis of the chemical phase for any given individual particle was achieved, and information was extracted of the particle interior. The ability to visualize the nanoscale chemical state distribution over functional volumes will revolutionize the study of energy storage materials and enable the design of optimized morphologies for the next generation of devices.

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WK7
Ptychographic Tomography

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Ptychography is a scanning coherent diffraction imaging technique. It makes use of multiple diffraction patterns, measured as a coherent finite illumination (also called the probe) scans over a set of positions on a specimen, and where the adjacent illuminated areas partially overlap [1]. Provided this overlap is sufficient, the redundancy it introduces allows iterative algorithms to phase back the measurements and reconstruct complex images of the specimen. Moreover, the information redundancy is so large that it allows, thanks to various algorithmic developments, the reconstruction of the illumination function, the scan positions, the source coherence function, etc. Since it eliminates the requirement for objective lenses, the achievable resolution is in principle only limited by the wavelength and the diffraction angle collected by the detector. Its ability to provide quantitative phase images makes ptychography a very attractive imaging technique for many modalities, from electron to photon beams and from the THz to the x-ray regime [2].

With the high penetration power of x-rays, ptychography can be naturally extended to a 3D imaging technique by combination with computed tomography. This is referred to as ptychographic tomography [3]. Ptychographic tomography is able to provide non-destructive 3D volumetric structure information of a sample at sub-10nm resolution. In this talk, I will give a detailed introduction to this imaging technique with emphasis on some key steps in the combination of ptychography and tomography [4]. Some perspectives regarding future development of 3D ptychography will be briefly given as well.

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WK7
Ptycho-tomography: Algorithms and Challenges

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Ptychography has enabled in recent years nanometer resolution, macroscopic field of view, capability to retrieve chemical, orbital, electronic, or magnetic contrast, by reconstruction of both the phase (refractive index) and attenuation coefficients of a sample. The ptycho-tomography phase retrieval problem in the linear regime is in principle similar to two-dimensional ptychography. In practice, three-dimensional tomographic reconstruction can be challenging because the phase of the tomographic projection may be wrapped around, sample drifts are almost inevitable at high resolution, measurements may have outliers, a fluctuating background, or may be restricted to a limited angular range of sample rotations. Here, we discuss the algorithmic and computational challenges aimed at ptycho-tomography reconstructions and present our current solution. (Joint work with Huibin Chang and Pablo Enfedaque.)

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High-resolution Fast Ptychography with Photon-efficient Scanning

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The Advanced Photon Source Upgrade (APS-U) will provide more than 100 times higher coherent flux, which will enable a revolution in x-ray microscopy. To make efficient use of the tremendous increased flux and achieve the highest possible spatial resolution, novel hardware designs and advanced control techniques for fast high-precision scanning need to be developed for the next generation x-ray microscopes. We describe our recent developments of fly-scan ptychography on the Velociprobe [1] (a prototype instrument to prove instrumentation concepts critical for the APS-U) and the Bionanoprobe [2]. Unlike current commonly used fly-scan schemes in which only one axis is fly-scanned [3], both axes in our 2D scan are continuously moved throughout the scan, and the use of high bandwidth interferometric measurements enables accurate recording of positions where the data is taken. The extended fly-scan ptychography provides high photon-efficient use (almost no positioning overheads) and makes possible for arbitrary fly-scan trajectories. This fly-scan concept can also be applied for other motion axes, such as the rotation axis for 3D imaging. By using continuous scanning, novel data analysis, and a new generation of synchrotron light source, it will enable 3D high-throughput high-resolution imaging.

The work on the Velociprobe was supported by Argonne's Laboratory Directed Research and Development (LDRD, 2015-153-R2). 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.

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WK7

Fast X-ray Nano-tomography with Full-field Techniques

V. De Andrade, A. Deriy, M. Wojcik, S. Bean, D. Shu, T. Mooney, K. Peterson, A. Glowacki, D. Gürsoy, T. Bicer, X. Yang, M. Wolfman, N. Kasthuri, and F. De Carlo

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Transmission x-ray microscope (TXM) and projection microscope are two well-established full-field imaging techniques in the synchrotron community. For the last 10 years, *ex situ* but also *in situ* nano-tomography experiments have been routinely performed with isotropic spatial resolution around 50 nm. At sector 32-ID of APS, a new in-house TXM has been designed with the desire to combine performance and flexibility for sample environment. The instrument benefits from the in-house development of cutting-edge x-ray optics, mechanical components and a suite of software to push the limit of 3D nano-imaging while reducing total x-ray dose. It operates either with a moderate spatial resolution (60 nm) and large field of view (~60 μm) or at very high spatial resolution (16 nm) at a smaller field of view (~10 μm).

In this talk, we will give a brief overview of the most innovative applications that emerged from the TXM at 32-ID in the fields of battery research, metallurgy and neurosciences. In addition, technical challenges the TXM is facing as well as improvement plans will be discussed. At last, we will draw a quick comparison between TXM and projection microscopes as the one currently in construction at 32-ID.

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Sigray UltimaXRM: Designing a Next-generation X-ray Microscope for On-demand Nanoscale Tomography

David Vine

Sigray, Concord, CA 94520

Three-dimensional x-ray microscopy is an essential tool for non-destructively characterizing specimens at the nanoscale and uniquely enables *in situ* studies on microstructural evolution. Once the exclusive domain of synchrotron beamlines, nanoscale x-ray microscopes are a popular tool for the research laboratory that appreciates the combination of high spatial resolution with the throughput gains that come with on-demand access.

The novel Sigray UltimaXRM™ is the highest resolution commercial 3D x-ray microscope, and reduces the capability gap between synchrotron and sealed tubes source based x-ray systems by enabling energy tunability. The system pairs our high-brilliance multi-energy microfocus source featuring up to four x-ray target

materials that allow the user to tune the x-ray energy to the sample under study. A proprietary high efficiency condenser and parfocal objective lens for each target makes switching between energies simple and fast without the need for time-consuming refocusing. Each imaging mode is available with absorption and phase contrast with best-in-class 40 nm spatial resolution mode, and a large field-of-view mode for easy alignment.

This presentation will outline the technical advances that underpin the performance of the UltimaXRM, including efforts to control and ameliorate the effects of temperature variation, stage stability and runout.

Wednesday, May 9

APS/CNM Workshop 8
Tipping X-ray – Comprehensive Nanoscale Characterization with Multimodal X-ray Imaging

Location: Building 401, Room A5000

Organizers: Haidan Wen and Volker Rose (APS)

A deeper understanding of energy conversion and transport in real-world materials and devices requires multimodal imaging capabilities to visualize rich, localized physical and chemical processes. These processes can be delicate and need to be measured *in situ* to capture the evolution of nanoscale objects, ranging from correlated electronic phases in quantum materials to defects at electrochemical interfaces. To meet this challenge, major instrumentation initiatives that integrate scanning near-field optical microscopy and scanning tunneling microscopy with x-ray beamlines (SX-STM) are under rapid development at the Advanced Photon Source. These new instruments open up opportunities to investigate localized nanoscopic phenomena with unprecedented comprehensive information including chemical sensitivity, local structure distortion, and electronic heterogeneities. The world's first user program in SX-STM will soon become available at the new XTIP beamline. This workshop will gather world-leading researchers in the field of nanoscale characterization to identify new scientific opportunities, cultivate user community, and form international collaborations.

9:00 – 9: 15	Haidan Wen and Volker Rose (Argonne National Laboratory) *Welcome and Workshop Charge*
9:15 – 9:20	Jonathan Lang and Stefan Vogt (Argonne National Laboratory) *Introductory Remarks*
9:20 – 9:40	Qian Li (Argonne National Laboratory) *Development of a Time-resolved Multimodal Imaging Platform*
9:40 – 10:00	Nozomi Shirato (Argonne National Laboratory) *XTIP: A Dedicated New APS Beamline for Synchrotron X-ray Scanning Tunneling Microscopy*
10:00 – 10:30	Break
10:30 – 11:00	Jeremy Levy (University of Pittsburgh) *One-dimensional Transport at the $LaAlO_3/SrTiO_3$ Interface*
11:00 – 11:30	Sergei Kalinin (Oak Ridge National Laboratory) *Deep Learning in Atomically Resolved Imaging: From Mechanisms of Solid State Reactions to E-beam Atomic Fabrications*
11:30 – 12:00	Mengkun Liu (Stony Brook University) *Recent Development in Scattering-type Scanning Near-field Optical Microscope*
12:00 – 1:30	Lunch
1:30 – 2:00	Tyler Cocker (Michigan State University) *Ultrafast Terahertz Microscopy: From Near Fields to Single Atoms*
2:00 – 2:30	Woei Wu Larry Pai (National Taiwan University) *Spectro-microscopy at Nanoscale with Combined STM and Synchrotron Techniques–Case Studies of Charge Density Wave Visualization and Novel Instrumentation*
2:30 – 3:00	TeYu Chien (University of Wyoming) *Cross-sectional Scanning Tunneling Microscopy and Spectroscopy for Complex Oxide Interfaces and Beyond*
3:00 – 3:30	Break

3:30 – 4:00 Didier Tonneau (Aix-Marseille Université – CINaM)
Visible Nanophotonics for High Lateral Resolution X-ray-based Techniques

4:00 – 4:30 Eli Rotenberg (Lawrence Berkeley National Laboratory)
MAESTRO: Investigating the Electronic Structure of Materials with Multimodal Growth and Characterization at the ALS

4:30 – 5:30 Panel Discussion
How Can X-ray Help? Scientific Drivers and Technical Wish List

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Development of a Time-resolved Multimodal Imaging Platform

Qian Li and Haidan Wen
Advance Photon Source, Argonne National Laboratory, Argonne, IL 60439

In the past decades, scanning probe microscopy (SPM) and scanning near-field optical microscopy (SNOM) have been developed to probe a broad spectrum of physical properties with unparalleled spatial and temporal resolutions. However, unlike scattering methods, SPM/SNOM rarely provide direct information about the structures underlying probed response. This has caused a number of unanswered questions for condensed matter systems, which are characteristic of mesoscopic heterogeneities in a spontaneous and/or excited nature. Here at the Advanced Photon Source (APS), we have started an initiative to develop a platform that integrates SPM/SNOM with focused x-ray diffraction microscopy, which allows imaging of nonequilibrium states utilizing ultrafast lasers and picosecond-scale pulsed x-rays from the APS synchrotron. In this talk, I will briefly review the current development status, and discuss new technical leads and potential scientific applications. Finally, I will present a case study where we explored the structural mechanisms of mechanical switching in ferroelectric thin films. We observed enhanced diffuse scattering with well-defined spatial correlations in the pressure-scanned films and attributed it to Huang scattering due to defect redistributions, providing structural insights into mechanically induced ferroelectric switching.

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XTIP: A Dedicated New APS Beamline for Synchrotron X-ray Scanning Tunneling Microscopy

Nozomi Shirato[1], Mike Fisher[2], Ruben Reininger[2], Saw Wai Hla[1,3], and Volker Rose[2]
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Synchrotron x-ray scanning tunneling microscopy (SX-STM) utilizes an atomically sharp tip as a detector and illuminates a tip-sample junction with high intensity monochromatic x-ray beam to collect x-ray induced signals. The recent studies have shown that the technique has elemental sensitivity at the atomic limit and probes local magnetic properties utilizing polarized beams. At Argonne, currently, a first dedicated beamline for SX-STM is under construction in Advanced Photon Source. The soft x-ray beamline can produce a photon flux of 10^{11}-10^{13} photons/sec at 1 keV over the energy range of 400 to 1600 eV. The beamline will be equipped with linear and circular polarizers and focusing capabilities down to 10 μm. A spherical grating monochromator has a resolving power better than 4000. By utilizing the beamline, one can explore chemical, magnetic and electronic properties of nanoscale materials.

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One-dimensional Transport at the $LaAlO_3$/$SrTiO_3$ Interface

Jeremy Levy
Department of Physics and Astronomy, University of Pittsburgh, Pittsburgh, PA 15260
Pittsburgh Quantum Institute, Pittsburgh, PA 15260

The study of strongly correlated electronic systems and the development of quantum transport in nanoelectronic devices have followed distinct, mostly non-overlapping paths. Electronic correlations of complex materials lead to emergent properties such as

superconductivity, magnetism, and Mott insulator phases. Nanoelectronics generally starts with far simpler materials (e.g., carbon-based or semiconductors) and derives functionality from doping and spatial confinement to two or fewer spatial dimensions. In the last decade, these two fields have begun to overlap. The development of new growth techniques for complex oxides have enabled new families of heterostructures which can be electrostatically gated between insulating, ferromagnetic, conducting and superconducting phases. In my own research, we use a scanning probe to "write" and "erase" conducting nanostructures at the $LaAlO_3/SrTiO_3$ interface. The process is similar to that of an Etch-a-Sketch toy, but with a precision of two nanometers. A wide variety of nanoscale devices have already been demonstrated, including nanowires, nanoscale photodetectors, THz emitters and detectors, tunnel junctions, diodes, field-effect transistors, single-electron transistors, superconducting nanostructures and ballistic electron waveguides. In this talk, I will focus on the physics of quasi-one-dimensional conductive structures formed at the $LaAlO_3/SrTiO_3$, where we observe quantized ballistic transport and intrinsically one-dimensional superconducting behavior. These one-dimensional building blocks may form the basis for novel technologies, including a platform for complex-oxide-based quantum computation and quantum simulation.

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Deep Learning in Atomically Resolved Imaging: From Mechanisms of Solid State Reactions to E-beam Atomic Fabrications

Sergei V. Kalinin

The Institute for Functional Imaging of Materials, Materials Sciences and Technology Division, and the Center for Nanophase Materials Sciences, Oak Ridge National Laboratory, Oak Ridge, TN 37831

Understanding elementary mechanisms behind solid-state phase transformations and reactions is the key to optimizing desired functional properties of many technologically relevant materials. Recent advances in scanning transmission electron microscopy (STEM) allows the real-time visualization of solid-state transformations in materials, including those induced by an electron beam and temperature, with atomic resolution. However, despite the ever-expanding capabilities for high-resolution data acquisition, the inferred information about kinetics and thermodynamics of the process and single defect dynamics and interactions is minimal, due to the inherent limitations of manual *ex situ* analysis of the collected volumes of data and lack of feedback to theory.

In this presentation, I discuss the research activity coordinated by the Institute for Functional Imaging of Materials (IFIM) aimed at bridging imaging and theory via big data technologies to enable design of new materials with tailored functionalities. In this presentation, I will illustrate several examples of using deep learning networks and inverse modelling to extract materials specific physics from imaging in STM and STEM. For atomistic systems, we developed a deep learning framework for dynamic STEM imaging that is trained to find the structures (defects) that break a crystal lattice periodicity and apply it for mapping solid state reactions and transformations in layered WS_2 doped with Mo. This framework allows extracting thousands of lattice defects from raw STEM data (single images and movies) in a matter of seconds, which are then classified into different categories using unsupervised clustering methods. We further expanded our framework to extract parameters of diffusion for the sulfur vacancies and analyzed transition probabilities associated with switching between different configurations of defect complexes consisting of Mo dopant and sulfur vacancy, providing insight into point defect dynamics and reactions. This approach is universal and its application to beam induced reactions allows mapping chemical transformation pathways in solids at the atomic level. Several additional examples of deep-learning analytics of molecular systems, theory-assisted image recognition, and extraction of mesoscopic and atomistic physical parameters will be illustrated.

Finally, incorporation of the real time feedback in electron microscopy opens the pathway towards the use of atomically focused beam of scanning transmission electron microscope to control and direct matter on atomic scales. I will introduce several examples of beam-induced fabrication on atomic level, and demonstrate how beam control, rapid image analytics, and image- and ptychography based feedback allows for controlling matter on atomic level.

This research is supported by and performed at the Center for Nanophase Materials Sciences, sponsored at Oak Ridge National Laboratory by the Scientific User Facilities Division, BES DOE.

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Recent Development in Scattering-type Scanning Near-field Optical Microscope

Mengkun Liu

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Over the past decade, optical near-field techniques, especially the scattering-type scanning near-field optical microscope (s-SNOM), have undergone tremendous development. This is partly due to the ever-increasing need for the exploration of the nano-world using new tools and partly due to the many technical advances in laser and scanning probe techniques. I will use this opportunity to report the recent advances in the IR and THz near-field microscopy and spectroscopy technology and discuss

their applications in the nanoscale electrodynamics in strongly correlated electron materials. Specifically, I will discuss the details of modeling scanning near-field infrared microscopy and its application in understanding the mesoscopic insulator to metal phase transitions in VO_2 and Ca_2RuO_4 with over a broad spectral range (350 cm^{-1} to 2500 cm^{-1}). I will also discuss the future development of near-field scanning microscope including the cryogenic capabilities and its coupling to ultrafast pump-probe spectroscopy including both IR and THz frequency range. These developments set the stage for future spectroscopic investigations to access the fundamental properties of complex materials at the nanoscale.

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Ultrafast Terahertz Microscopy: From Near Fields to Single Atoms

Tyler L. Cocker
Michigan State University, East Lansing, MI 48824

A new experimental frontier has recently emerged with the potential to significantly impact physics, chemistry, materials science, and biology: the regime of ultrafast time resolution and ultrasmall spatial resolution. This is the domain in which single atoms, molecules, and electronic orbitals move. It also corresponds, on larger scales, to the territory of low-energy elementary excitations such as plasmons, phonons, and interlevel transitions in excitons. These processes are of particular importance for nanomaterial functionality and typically survive for only femtoseconds to picoseconds after photoexcitation.

In this talk, I will show how these diverse dynamics can be studied with new techniques that combine terahertz technology with scanning probe microscopy. First, I will describe how ultrafast near-field microscopy has been employed to perform sub-cycle spectroscopy of single nanoparticles [1], reveal hidden structure in correlated electron systems [2], and resolve transient interface polaritons in van der Waals heterostructures [3]. Then I will discuss the development of a related technique: lightwave-driven terahertz scanning tunneling microscopy [4,5]. In this novel approach, the oscillating electric field of a phase-stable, few-cycle light pulse at an atomically sharp tip can be used to remove a single electron from a single molecular orbital within a time window faster than an oscillation cycle of the terahertz wave. I will show how this technique has been used to take ultrafast snapshot images of the electron density in single molecular orbitals and watch the motion of a single molecule for the first time [5].

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WK8

Spectro-microscopy at Nanoscale with Combined STM and Synchrotron Techniques – Case Studies of Charge Density Wave Visualization and Novel Instrumentation

Dr. Woei Wu Pai
Center for Condensed Matter Sciences, National Taiwan University, Taiwan, R.O.C. 106

Combining synchrotron-based techniques with scanning tunneling microscopy/spectroscopy is a promising route to elucidate materials properties at nanoscale. Here I describe our two recent studies in this direction. In one case, we combine ARPES and STM/STS to study anomalous behaviors of charge density wave (CDW) in transition metal dichalcogenides when they are thinned to monolayers. In 1T-$TiTe_2$, in which no CDW is observed in bulk, a (2x2) CDW with a pseudogap and a transition temperature of ~92 K develops only in monolayers. In 1T-VSe_2, the monolayer form has a twice higher CDW transition temperature than its bulk value and also the CDW periodicity dramatically changes. In another case, we have been developing a new STM setup incorporating radio frequency reflectometry and related electronics (e.g., radio-frequency reflectrometry STM, RFSTM). One advantage of RFSTM is that the processed RF reflection signal exhibits the same behavior as tunneling current but is not affected by external electron flux impinging the tunneling junction. This opens the possibility of high-resolution chemical mapping by RFSTM when combined with synchrotron x-ray absorption spectroscopy. Some preliminary test data of the RFSTM operation will be discussed.

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Cross-sectional Scanning Tunneling Microscopy and Spectroscopy for Complex Oxide Interfaces and Beyond

TeYu Chien
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Owing to the discovery of novel interfacial phenoman, interfacial physics has been attract plenty attention recently. In particular, complex oxide interfaces have exhibited wide range of novel interfacial physical properties not seen in the bulk counterparts. Unlike traditional metals and semiconductors, where electrons are treated as electron gas (weakly interactions), complex

oxides exhibit a highly coupled environment for charge, spin, orbital and lattice to have strong interactions. These highly coupled interactions provide a unique environment for many novel functionalities, such as superconductivity, ferromagnetism, ferroelectricity, coloso-magnetoresistance, and multiferroics. These highly coupled interactions are also, on the other hand, the reason that the properties of these materials are difficulty to be predicted. In this talk, I will introduce an experimental technique—cross-sectional scanning tunneling microscopy and spectroscopy (XSTM/S)—for studying complex oxide interfaces [1–7]. In the end, I will also discuss the future of this technique for interfacial physics beyond complex oxides [7–13].

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WK8

Visible Nanophotonics for High Lateral Resolution X-ray-based Techniques

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X-ray spectroscopies are powerful techniques for material analysis. These techniques allow to characterize material composition at very high accuracy and to define atom environments at ultimate accuracy of sub-A resolution. Unfortunatly, these techniques cannot provide accurate x-ray beam probe positionning on a peculiar area or nano-object of the surface.

Scanning probe microscopy (SPM) are surface analysis techniques allowing to achieve the ultimate atomic lateral resolution for imaging application. Unfortunately, these techniques do not allow to define the apriori material composition.

Our team has worked on the combination of both techniques and has shown the feasibility of simultaneous material chemical mapping and surface topography at sub-50 nm lateral resolution in both analysis. Our tool is based on local visible luminescence collection of the sample under x-ray illumination using a sharp pulled optical fibre. The fiber is simultaneously used as probe of a shear force microscope. We will present the first experiments on nano-XEOL analysis, highlighting the high potential of SPM coupling with x-ray spectrometry.

The results obtained suggest that luminescent nano-clusters could be efficiently be used as x-ray detectors operating at high lateral and time resolution. We presently develop a new generation of x-ray nano-sensor based on a luminescent cluster grafted at the extremity of a sharp optical fiber. The cluster luminescence is collected by a photon counter through the optical fiber. By adding a nano-antenna at the extremity of the fiber to increase the collection yield, we have shown that sub micrometer lateral resolution x-ray sensor can be achieved. The sensor qualification tests have been performed using our low power laboratory source (Rh target, 35 kV, 800 µA). Working in synchrotron environment, due to the significantly higher brightness of the source, a nanometer lateral resolution is expected. This open the way of a new technique for XSW (x-ray free standing waves) detection.

WK8

MAESTRO: Investigating the Electronic Structure of Materials with Multimodal Growth and Characterization at the ALS

Eli Rotenberg
Lawrence Berkeley National Laboratory, Berkeley, CA 94720

MAESTRO, the Microscopic and Electronic Structure Observatory, is a new synchrotron based facility for the study of *in situ* prepared materials, including oxides, 2D van der waals material, semiconductors, metals, and surfaces. This experimental system fuses powerful sample preparation tools (glovebox, MBE, PLD) with state of the art photoemission end stations (μARPES, PEEM, nanoARPES)—all of which are connected through an automated UHV transfer system. A particularly novel feature of MAESTRO is its nanoARPES setup. This technique mates the merits of state of the art angle resolved photoemission (ARPES) with spatial resolution presently less than 120 nm, with an eventual goal of less than 50 nm, bringing *k* - and energy resolved electronic contrast on the nano- and mesoscale within reach.

This talk will show highlights of the electronic structure of *in situ* grown oxides such as CuO, VO_2, as well as chalcogenides like WS_2. Although these studies are on (nominally) homogeneous systems, prospects for new results from inhomogeneous systems when studied with nano-probe ARPES will be discussed. As an example, I will show recent results on the spatially-resolved electronic structure of hybrid systems composed of monolayers of 2D chalcogenides mechanically transferred onto TiO_2 or STO.

Wednesday, May 9

CNM Workshop 9
Nanoscience for Quantum Science: Developing, Characterizing, and Harnessing Optically Active Defects

Location: Building 440, Room A105/106

Organizers: Jeffrey Guest, Stephen Gray, and Xuedan Ma (CNM)

Systems for storing and manipulating quantum information, particularly optically addressable spin systems, are intrinsically nanoscale objects; they are structured and function on nanometer length scales. However, experimental efforts have largely avoided nanoscience approaches to understanding and controlling such systems due to the additional complexity introduced, relying instead on defects buried inside 'perfect' crystals or laser-cooled ions trapped in vacuum far from surfaces. This workshop will explore emerging efforts and untapped potential in this overlapping space, particularly where nanoscience characterization, synthesis, fabrication and theory tools can help understand and control the underlying physics determining the behavior, the governing interactions, and the de-coherence mechanisms of these quantum systems.

For example, the local environment of optically-active defects is known to play an important role in determining the energetics and coupling of electronic and spin states to each other and the local strained lattice; the ability to probe this environment directly will be possible with nanoscale x-ray probes. Additionally, surfaces are typically eschewed by the quantum science community. However, they are a powerful platform for atomic scale development and an intrinsic part of any nanoscale quantum sensor. Questions this workshop would like to answer are whether there are fundamental limitations to coherence properties of quantum states at or near surfaces and what can surface science approaches do to reach these limits. To date, defects explored by the quantum science community have largely been naturally occurring, but combined with improved theoretical approaches, new developments in synthesis and atomic scale control by scanning probes has opened up new frontiers in artificial nanostructures. Hybrid quantum systems are likely to play a critical role in converting long-lived quantum information into 'flying' qubits for quantum communication; engineering systems for this purpose is likely to require developments in materials science and the harnessing of nanofabrication capabilities, for example in nanomechanical systems that couple optically-active defects to solid-state phonons. Theoretical efforts that marry atomic-scale understanding of quantum properties to mesoscopic behaviors and de-coherence physics will also play a pivotal role.

8:30 – 8:40	Introductory Remarks
8:40 – 9:30	David Awschalom (University of Chicago/Argonne National Laboratory) *Controlling Defect Spin States with Photons, Magnons, and Phonons*
9:30 – 10:10	Alec Jenkins (University of California Santa Barbara) *The NV Center in Diamond: A Versatile Quantum Sensing Technology*
10:10 – 10:40	Break
10:40 – 11:20	Cyrus Dreyer (Rutgers University) *Carrier Capture and Recombination at Point Defects from First Principles*
11:20 – 12:00	Giulia Galli (University of Chicago/Argonne National Laboratory) *Harnessing Point Defects in Solids and Nanostructures*
12:00 – 1:40	Lunch
1:40 – 2:20	Jay Gupta (Ohio State University) *Tunable Control over Individual Defects in Semiconductors at the Atomic Scale via STM*
2:20 – 3:00	Greg Fuchs (Cornell University) *The Electronic and Vibronic Properties of Single Photon Emitters in Hexagonal Boron Nitride*

3:00 – 3:30	Break
3:30 – 4:10	Ajit Srivastava (Emory University) *Phonon-photon Entanglement in WSe_2 Quantum Dots*
4:10 – 4:50	Tian Zhong (University of Chicago) *A Nanophotonic Platform Integrating Quantum Memories and Single Rare-earth Ions*
4:50 – 5:00	Closing Remarks

WK9

Controlling Defect Spin States with Photons, Magnons, and Phonons

David D. Awschalom

University of Chicago, Chicago, IL 60637

Argonne National Laboratory, Argonne, IL 60439

There is a growing interest in exploiting the quantum properties of electronic and nuclear spins for the manipulation and storage of quantum information. Current efforts embrace materials with incorporated defects, whose unique electronic and nuclear spin states allow the processing of information because of their explicitly quantum nature. Here we focus on recent developments in manipulating and connecting spins in both silicon carbide (SiC) and diamond. We find that defect-based electronic states in SiC can be isolated at the single spin level [1,2] with surprisingly long spin coherence times and high-fidelity control within a wafer-scale material operating at near-telecom wavelengths. Similarly, the spin-photon interface in diamond offers an opportunity to implement all-optical quantum spin gates [3] for quantum information processing. Moreover, we present pathways for connecting spins using magnons and phonons. Hybrid spin-magnon systems use ferromagnetic spin-wave modes to perform long-range coherent control of spins in diamond using surface magnons in YIG thin-films [4]. The magnetic modes amplify the oscillating field of the microwave source by more than two orders of magnitude, thereby efficiently driving remote spin states. In addition, fabricated surface acoustic wave resonators exploit both the piezoelectric and isotropic phonon properties of SiC to create Autler-Townes splittings, universal ground state spin control, and mechanically drive coherent Rabi oscillations of magnetically forbidden spin transitions [5].

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WK9

The NV Center in Diamond: A Versatile Quantum Sensing Technology

Alec Jenkins

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The nitrogen vacancy (NV) center in diamond is an atomic-scale defect that exhibits remarkably coherent quantum properties in a uniquely accessible way: at room temperature, in ambient conditions, and even immersed in biological environments. NV centers are being explored for a variety of quantum technologies, including quantum sensing and quantum information processing. In this talk, I introduce the physics and materials science behind the success of the NV center as a quantum sensor. I highlight some of the major achievements of NV-based quantum sensors and I present a versatile imaging platform where we have incorporated NV centers into a scanning probe microscope. I discuss our imaging of vortices in superconductors [1] and nanoscale conductivity, and then focus on our measurements of skyrmions in thin film magnetic multilayers. Skyrmions have been proposed as the basis of high density and low power memory devices, but before practical devices based on skyrmions can be realized, new materials must be designed that maximize skyrmion current-driven velocities, minimize the depinning current densities, and host nanoscale skyrmions at length scales competitive with existing technologies. I present our studies of skyrmion magnetic structure and domain wall pinning, demonstrating the NV as a useful tool in the development of skyrmion-based technologies. I also outline the material challenges facing the widespread use of NV centers in quantum applications, including the mitigation of surface-induced quantum decoherence. To that end, I present work in the Jayich Lab in which the NV center is used to spectroscopically probe sources of surface and bulk-related decoherence [2,3].

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WK9

Carrier Capture and Recombination at Point Defects from First Principles

Cyrus Dreyer

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Point defects play an important role in determining the properties of materials and devices, and are an exciting platform for quantum applications in computing, communication, and metrology. In order to identify detrimental defects and exploit useful ones, a quantitative understanding of how defects interact with free carriers in the host material is required. In particular, radiative and nonradiative capture of carriers at defects are key processes that govern the defects' role as recombination centers or carrier traps, their optical signature in photoluminescence, and the ability to manipulate their charge states. In this talk, I will give an overview of how we understand carrier capture and recombination at defects, and how we can determine rates and other properties from first principles, density-functional theory calculations. I will give examples for various defects in III-nitride semiconductors.

WK9

Harnessing Point Defects in Solids and Nanostructures

Giulia Galli

University of Chicago, Chicago, IL 60637

We will present first principles calculations of defective solids and nanostructures of interest for quantum information science applications.

WK9

Tunable Control over Individual Defects in Semiconductors at the Atomic Scale via STM

Jay Gupta

Department of Physics, Ohio State University

The scaling of electronic devices such as transistors to nanometer dimensions requires more precise control of individual dopants in semiconductor nanostructures, as statistical fluctuations can impact device performance and functionality. Toward this end, the scanning tunneling microscope (STM) is emerging as a useful tool for its capabilities of atomic manipulation, imaging and tunneling spectroscopy. I will discuss our STM studies of acceptors, adatoms and other defects on III-V semiconductor surfaces [1–6]. We have recently extended methods first developed for magnetic acceptors in GaAs (e.g., Co, Mn) to rare earth atoms (Er). Surprisingly, we find that Er adsorbs in three distinct states upon deposition *in situ* at low temperature (5K). Perturbation by the STM tip switches Er adatoms from metastable to stable adsorption states, and can also form Er acceptors by substitution for surface layer Ga atoms. Tunneling spectroscopy reveals distinct states, which evolve systematically for Er in different interstitial and substitutional environments. These experiments allow us to compare the hybridization of the relatively isolated 4f orbitals in Er to the more well studied 3d orbitals in the transition metals. I will also discuss our recent work on InSb(110), where we discovered pronounced surface-tunable conductance from individual adatoms over 100 nm^2 areas. Here the STM tip was used to deposit individual adatoms on the surface. These adatoms are positively charged when the tip is far from the adatom, but switch to a neutral state when the tip approaches. This results in a suppression of surface conductance by 100x, evident as an apparent 'crater' feature in STM topographic images. These features are unusually sensitive to the tunneling conditions (voltage, set current, tip sharpness), and can also be tuned by optical illumination. Unlike other recent tip-induced dopant ionization studies, the effect here is suggestive of a competing rates model, whereby the ratio of filling/draining rates of the adatom charge transition level is tuned by proximity of the STM tip. The prominence of these features is attributed to the very shallow nature of the residual donors in InSb, which allows sensitive tuning of the surface conductance over relatively large areas by individual atoms. These studies show that tunable control over single dopants in semiconductors is becoming a realistic route for next-generation classical- and quantum-based information technologies.

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WK9

The Electronic and Vibronic Properties of Single Photon Emitters in Hexagonal Boron Nitride

Gregory D. Fuchs

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Isolated point defects in wide bandgap materials can serve as sources of single photons for applications in quantum optics, precision sensing, and quantum information processing. Along with the discovery of new material hosts of "quantum defects", there is a corresponding opportunity to uncover new functionality if we can understand defect states and properties. The recent observation of bright, photo-stable, and spectrally-narrow single photon emission from defects in hexagonal boron nitride (h-BN) offers a new toolbox for quantum optics because of the possibilities for easily integrating single-photon sources into van der Waals heterostructures for electrical gating, and into pre-patterned optical structures for optical enhancement.

I will present our investigations of h-BN defects with the goal of understanding their fundamental electronic and vibronic properties. We first perform a survey of optically isolated h-BN defects and we observe zero-phonon line (ZPL) energies in a range that exceeds 500 meV. Next we study the temperature dependence of the ZPL spectral width and amplitude. We find that the results are consistent with a lattice vibration model that considers piezoelectric coupling with phonons in the defects two-dimensional h-BN sheet [1].

We also examine the mechanisms of optical absorption and emission within the Huang-Rhys model. We find that if the energy difference ΔE between the exciting laser energy and the ZPL energy is less than 200 meV, the largest phonon energy in h-BN, then the polarization axes for absorption and emission are nearly always aligned as expected. However, if ΔE is greater than 200 meV, the polarization axes for absorption and emission can be misaligned by any angle between 0 and 90°. This observation reveals the presence of two optical absorption mechanisms for h-BN defects—either through direct, phonon-mediated optical absorption between or through indirect optical absorption via an intermediate electronic state [2]. These results provide new insights into the electronic and vibronic processes within the optical cycle of single photon emitters hosted in a 2-dimensional insulator.

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WK9

Phonon-photon Entanglement in WSe_2 Quantum Dots

Ajit Srivastava

Emory University, Atlanta, Georgia 30322

Monolayer transition metal dichalcogenides (TMDs), such as WSe_2, are atomically thin semiconductors with a "valley" degree of freedom, which can be optically addressed, thus opening up exciting possibilities for "valleytronics". Recently, naturally occurring single quantum emitters, believed to be excitons trapped in shallow potentials, were reported in TMDs. They seem to inherit the valley degree of freedom from the host TMD and owing to their longer lifetimes, appear promising for quantum information processing applications.

In this talk, I will begin by highlighting some unique properties of TMDs excitons which result from the off-Gamma-point origin of the constituent single particle electronic states. After describing the basic properties of quantum dots in TMDs, I will present evidence for quantum entanglement between chiral phonons of the 2D host and single photons emitted from the quantum dots. Finally, I will discuss our future plans for implementing a dynamically tunable array of qubits in pristine TMDs which can serve as an ideal platform for quantum information processing applications and also for understanding fundamental many-body physics.

WK9

A Nanophotonic Platform Integrating Quantum Memories and Single Rare-earth Ions

Tian Zhong

University of Chicago, Chicago, IL 60637

The integration of rare-earth ions in an on-chip photonic platform will enable quantum repeaters and scalable quantum networks. While ensemble-based quantum memories have been realized, isolating single rare-earth ions as either single photon source or spin qubits remains an outstanding challenge due to rare-earth's weak photoluminescence and ultranarrow linewidth.

In this talk, I will describe a nanophotonic platform consisting of yttrium orthovanadate (YVO) photonic crystal nanobeam resonators coupled to a mesoscopic ensemble of neodymium (Nd) ions. The cavity acts as a memory at the center of the inhomogeneous line when prepared with efficient spectral hole burning; meanwhile, it permits addressing of single Nd ions at the far tails of the inhomogeneous distribution, showing near-transform-limited, high-purity single photon emission. Integrating both ensemble and single emitter quantum systems, this nanoscale quantum device could pave the way for scalable quantum networks and long-distance quantum communications.

Wednesday, May 10

APS Workshop 10
Applications of Synchrotron X-ray Techniques for Studying Metal Additive Manufacturing

Location: Building 446, Advanced Protein Characterization Facility Conference Room

Organizers: Tao Sun, Andrew Chuang, Xianghui Xiao, Ruqing Xu, Yang Ren, and Kamel Fezzaa (APS)

Additive manufacturing (AM, a.k.a. 3D printing) refers to a suite of transformative technologies that build three-dimensional objects by adding materials layer by layer based on digital design. In particular, metal AM has found many applications in the fields of biomedical, aerospace, automobile, and defense. Compared with conventional metal manufacturing techniques, AM exhibits many unique advantages, including short design-to-market, short supply chain, on-site and on-demand spares and tools manufacturing, less consumption of energy, and less generation of material waste. More importantly, AM largely eliminates tooling constraints, and gives us the freedom to design and build parts with complex geometries and improved performance. Metal AM has developed rapidly in the last three decades, thanks to substantial investments in the technology from both public and private groups worldwide. Numerous 3D printer manufacturers have emerged, and the technique maturation was seemingly reached. However, a precise control of microstructures and properties of additively manufactured products remains challenging, and the metallic materials we can use for AM are still very limited. Therefore, solving the fundamental material problems holds the key to unleash AM's full potential to revolutionize the way we build metal parts.

There are a few AM techniques for printing metallic materials. Except binder jetting, all other metal AM techniques involve depositing thermal energy to the sample with laser or electron sources. In a typical build process, the laser or electron beam heats up the sample locally to temperatures higher than the melting or even boiling temperatures of metals. In the meanwhile, the heating and cooling rates are in the order 106 K/s and above, and the local thermal gradient can easily reach 103 K/mm. Such extreme thermal conditions allow us to fabricate materials with very unique and favorable microstructures that we could never achieve before; but on the other hand, a variety of defects exist in AM materials, including porosity, cracks, residual stress, undesired grain structures and non-equilibrium phases.

Synchrotron x-ray techniques are among the most versatile and effective techniques for characterizing materials microstructures and their evolution in various processes and conditions. The APS has seen more and more users from the AM community in the recent years, who take the advantages of the superior hard x-ray source and sophisticated beamline instruments the APS affords. With state-of-the-art high-energy x-ray diffraction, Laue microscopy, computed tomography, high-speed imaging techniques, scientists start to address many critical material issues in AM associated with the feed stocks, build processes, and end products. This workshop aims to bring together AM experts from industry, government labs, and academia for a full-day discussion. It will serve as a venue for the AM user community to present their observations and theoretical advances from synchrotron experiments, identify the major challenges associated with AM materials, and propose desired beamline instruments and capabilities for future study.

Program Session Chair: Tao Sun

8:30 – 8:40 Tao Sun (Argonne National Laboratory)
Welcome and Introduction

8:40 – 9:00 Aaron Greco (Argonne National Laboratory)
Argonne Additive Manufacturing Initiative

High-speed Imaging Session Chair: Kamel Fezzaa

9:00 – 9:30 John Barnes (The Barnes Group Advisors)
Additive Manufacturing Is Making Our Future

9:30 – 10:00 Lianyi Chen (Missouri University of Science and Technology)
Characterizing the Dynamics of Laser Powder Bed Fusion Additive Manufacturing Processes by High-speed X-ray Imaging/Diffraction

10:00 – 10:30 Break

Tomography Session Chair: Xianghui Xiao

10:30 – 11:00 Jian Cao (Northwestern University)
In situ *Characterization of Directed Energy Deposition Process*

11:00 – 11:30 Christopher Tassone (SLAC National Accelerator Laboratory)
Tracking the Evolution of Phase, Microstructure, and Meso-structure Using High Speed in situ *X-ray Diffraction, and X-ray Imaging of Metallic Alloys during Selective Laser Melting Additive Manufacturing*

11:30 – 12:00 Hahn Choo (University of Tennessee)
The Effect of Volumetric Energy Density on Defect Characteristics in a 3-D Printed Stainless Steel Alloy

12:00 – 1:30 Lunch

Diffraction Session Chair: Ruqing Xu

1:30 – 2:00 Uta Ruett (Advanced Photon Source)
Perspectives for Insight to AM with High-energy X-rays at Sector 11

2:00 – 2:30 Lyle Levine (National Institute of Standards and Technology)
Synchrotron X-ray and Neutron Based Benchmark Measurements for Additive Manufacturing

2:30 – 3:00 Xin Sun (Oak Ridge National Laboratory)
High-energy X-ray in Qualifying Additive Manufacturing Parts for Energy Applications

3:00 – 3:30 Break

Diffraction and Microscopy Session Chair: Andrew Chuang

3:30 – 4:00 Don Brown (Los Alamos National Laboratory)
Using Neutron and High Energy X-ray Diffraction to Probe Additively Manufactured Materials over a Range of Length and Time Scales

4:00 – 4:30 Paul Shade (Air Force Research Laboratory)
Materials Informed Data Driven Design of Additive Structures

4:30 – 5:00 General Discussion

WK10
Additive Manufacturing is Making Our Future

John Barnes
The Barnes Group Advisors, Pittsburgh, PA 15201

Additive manufacturing has progressed to a stage as a legitimate manufacturing technology. The infrastructure around the technology continues to develop and grow each week. Companies around the world are now developing an AM Strategy whether it directly or indirectly impacts them. More and more materials producers are trying to understand how they get involved. Industrial manufacturers are assessing their ability to use the technology to be faster, better or more affordable whilst trying to understand if it is a “make/buy” for them.

Analytical firms are turning their codes to help automate, simulate and analyze AM. In short, the AM world is now very diverse, very dynamic and very real. This talk will describe activities which may not be getting the attention they deserve.

WK10

Characterizing the Dynamics of Laser Powder Bed Fusion Additive Manufacturing Processes by High-speed X-ray Imaging/Diffraction

Lianyi Chen[1] and Tao Sun[2]

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[2] X-ray Science Division, Advanced Photon Source, Argonne National Laboratory, Argonne, IL 60439

Laser powder bed fusion is a major additive manufacturing technique for producing complex-shaped metal parts by selective melting successive layers of metal powders using a laser beam. Understanding the physics of laser powder bed fusion (L-PBF) processes is critical for establishing location-specific processing-microstructure-property relationships. The non-transparency of metals to visible light and the highly localized (tens of micrometers) and very short (tens of microseconds) interaction of a laser beam with metal powders during L-PBF pose a huge challenge to the characterization and understanding of this process. The detailed physics of the L-PBF process and the mechanisms of defect formation and microstructure evolution are still not clear. In this talk, I will present our research on characterizing the dynamics of laser powder bed fusion additive manufacturing processes by high-energy high-speed x-ray imaging/diffraction. The dynamics of powder spreading, powder spattering, melt pool evolution, pore evolution and solidification will be discussed. The results obtained in this work are important for establishing the location-specific processing-microstructure relationships in L-PBF of metals.

WK10

In situ Characterization of Directed Energy Deposition Process

Jian Cao

Department of Mechanical Engineering, Northwestern University, Evanston, IL 60208

Directed energy deposition, or DED, is an additive manufacturing process that uses a high-powered laser to melt blown metallic powder, and introduces large gradients and sensitivity in thermal histories within a built component that lead to unique phase transformations, microstructures, residual stress and anisotropic mechanical behavior. Because of simultaneous re-melting of underlying layers and surrounding areas during the process, the DED-processed component undergoes directional solidification with grain coarsening in the build direction with the exception of the top layer. *In situ* monitoring using high-speed synchrotron imaging is used to capture the interaction of melt-pool and powder and the formation of porosity during build. Experimental techniques are coupled with thermal models to better predict for the thermal history at localized areas.

WK10

Tracking the Evolution of Phase, Microstructure, and Meso-structure Using High Speed *in situ* X-ray Diffraction, and X-ray Imaging of Metallic Alloys during Selective Laser Melting Additive Manufacturing

Vivek Thampy[1], A. Fong[1], A.M. Kiss[1], N.P. Calta[2], A.A. Martin[2], J. Wang[2], P.J. Depond[2], G.M. Guss[4], M.J. Kramer[3], T. van Buuren[2], I. Matthews[2], M.F. Toney[1], J.N. Weker[1], K.H. Stone[1], and C.J. Tassone[1]

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Metal additive manufacturing (AM) using Laser Powder Bed Fusion (LPBF) is an emerging technology that provides significant design flexibility relative to conventional manufacturing techniques and enables the production of highly complex parts at minimal additional cost. However, the underlying physics of powder melting and subsequent recrystallization is complex and remains poorly understood. We have developed and commissioned a portable single layer AM testbed to enable *in situ* x-ray diffraction, with 2 ms temporal and ~300 μm spatial resolution, and x-ray radiography, with 25 μs temporal and μm spatial resolution, during the AM build process in order to track the mesoscale structure, microstructure and phase of the powder melt as it crystalizes on re-solidification. Using Ti-6Al-4V, a widely used aerospace alloy, as a model system, we have directly observed the disappearance of the room temperature alpha-Ti phase upon melting, followed by the appearance of the high temperature beta-Ti phase immediately after re-solidification, and its subsequent transformation back into the alpha phase upon cooling through the beta transus. We have also followed the formation of sub-surface voids as a function of processing parameters, and used these observations to develop strategies to mitigate void formation. Furthermore, we establish the dependence of this crucial parameter on the printing parameters such as power density and depth from the powder substrate interface.

WK10

The Effect of Volumetric Energy Density on Defect Characteristics in a 3-D Printed Stainless Steel Alloy

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One of the key issues related to the microstructure quality of 3-D printed structural alloys is the presence of defects at the micron-length scale. Therefore, understanding the relationship between processing parameters and defect characteristics is of fundamental and technical importance. Moreover, the role of microscopic defects, such as pores and cracks, on the fatigue behavior of the 3-D printed engineering component is a critical issue. In this talk, the role of volumetric energy density (VED) in the development of microstructure and defect characteristics during the selective laser melting (SLM) 3-D printing process will be discussed using 316L stainless steel as a model system.

WK10

Perspectives for Insight to AM with High-energy X-rays at Sector 11

Uta Ruett, Yang Ren, Xiaoyi Zhang, and Tao Sun

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High-energy x-rays above 50 keV photon energy offer many unique possibilities for structural analysis of matter. For Additive Manufacturing (AM), the most important features are the high penetration into material and the ability of structural analysis of amorphous, nanostructured and liquid states by PDF analysis.

This presentation will outline possible options for the *in situ* studies of laser printing nowadays and in the future after the APS upgrade at sector 11. Beside the major application of *in situ* probing of phase formation and crystallinity during solidification processes in AM, time resolved measurements between laser melting and x-ray probing on time scales below msec will be discussed. The new challenges for AM printing metallic glasses and layers of different materials will be addressed.

WK10

Synchrotron X-ray and Neutron Based Benchmark Measurements for Additive Manufacturing

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Additive manufacturing (AM) of metal components is a rapidly expanding manufacturing paradigm that could revolutionize the design and production of complex, high-value metal parts. However, the extreme processing conditions create inhomogeneous materials that may include unexpected metallic phases, build defects, and high localized stresses at multiple length scales. We report synergistic synchrotron x-ray and neutron measurements of AM materials that are providing detailed microstructure and stress characterization that guide the development of quantitative AM process models. Example measurements include microbeam x-ray diffraction at 34ID-E, ultra-small-angle x-ray scattering (USAXS), SAXS and wide-angle x-ray scattering (WAXS) at 9ID-C, powder diffraction at 11BM, white-beam diffraction at CHESS (A2) and neutron residual stress measurements at the NIST Center for Neutron Research (NCNR). Many of these measurements are contributing to the ongoing Additive Manufacturing Benchmark Test series (AM-Bench) that has partnered with 57 national labs, companies, universities and organizations around the world. In addition, these measurement results are being used to validate high-performance computing AM process models developed jointly by Oak Ridge National Laboratory, Lawrence Livermore National Laboratory, and Los Alamos National Laboratory through the Transforming Additive Manufacturing through Exascale Simulation Project (ExaAM), a use case for the DOE-funded Exascale Computing Project.

WK10

High Energy X-ray in Qualifying Additive Manufacturing Parts for Energy Applications

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Additive Manufacturing (AM) has attracted growing attentions in the energy sector as one of the top breakthrough technologies. Beyond rapid prototyping, AM has been demonstrated as a viable manufacturing technology in making parts with complex geometries that are not possible to make with conventional processes. It also has the advantages of short lead time, energy efficiency manufacturing as well as cost and weight savings.

Despite the growing interests, one of the main technical barriers for the faster adoption of various AM technologies in manufacturing industry lies in the area of part qualification. This is because the conventional manufacturing processes part qualification typically goes back to materials qualifications, and homogeneous materials are assumed and used for fabrication. For a given material and a given geometry, however, one of the key features of the additively manufactured parts are the process parameters/conditions—and location-dependent microstructures and defect structures. Hence the conventional materials qualification process is no longer applicable to additively manufactured parts.

We propose a mechanism-based simulation framework coupled with high throughput experiment at APS to accelerate the qualification process of additively manufactured part in energy applications. Much work still needs to be performed for a complete hierarchical validation and calibration approach across different length scales in predicting the part performance (quantities of interest) with a quantified confidence level. We present some results where *in situ* experiments with high energy x-ray at APS have been used to decipher microstructure features and to determine material modeling parameters in order to predict properties at the part scale.

WK10

Using Neutron and High Energy X-ray Diffraction to Probe Additively Manufactured Materials over a Range of Length and Time Scales

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The last decade has seen tremendous advances in the ability of 3rd generation synchrotron x-ray sources to probe microstructure at unprecedented length and time scales under unique environments that simulate manufacturing conditions. Concurrently, manufacturing is undergoing a revolution as investments are made in advanced manufacturing techniques, such as additive manufacture. It is natural that advanced manufacturing techniques should couple with advanced *in situ* characterization techniques in order to accelerate the process of qualification of products for critical applications.

This talk will present our efforts to characterize the relationship between additive manufacture process and the subsequent deposition microstructure, which in turn controls the as-built component properties. As an example of studying the effect of processing on microstructure, high energy x-ray diffraction has been used to monitor microstructural evolution *in situ* during additive manufacture of 304L stainless steel and Ti-6Al-4V with sub-second time resolution and sub 0.1mm spatial resolution. Specifically, the evolution of phase fractions, liquid and multiple solid phases, phases stresses, and texture is monitored following solidification and subsequent rapid cooling.

WK10

Materials Informed Data Driven Design of Additive Structures

Paul Shade

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Metal additive manufacturing (AM) presents both extreme potential and concern for component design. The ability to spatially tailor processing conditions opens the door to sophisticated designs with heterogeneous materials and locally optimized properties; yet, accounting for this heterogeneity, before exploiting it, requires the ability to link processing to structure and structure to properties/performance at a local scale. In this presentation, an effort to address these issues through the AFRL AM Modeling Challenge Series will be described. Highly-pedigreed datasets will be generated that include detailed process descriptions, advanced characterization, and mechanical testing. These datasets will be made available to the AM community in process-structure and structure-property material modeling challenge problems.

Thursday, May 10

WKS
User Workshop for Micromanipulator Use in Diamond Anvil Cell Loading and Other Applications

Location: Building 402, Room E1100/1200

Organizers: Steven M. Barnett (Barnett Technical Services), Ross Hrubiak (HP-CAT), and Vitali Prakapenka (GSECARS)

The study of material physics under high pressure provides valuable information on the properties of these materials at deep pressures within the Earth (or in other environments). A critical component of these studies is the Diamond Anvil Call (DAC), where materials under study are placed between two diamonds and a gasket to hold the material in place. For some studies, the samples and other materials need to be placed between the diamonds within limited space with sub-micron resolution. More recently, positioning of pressure calibrates, pressure mediating fluids, and the desire to attach additional sensors (e.g., electrodes) has required even more precise control of sampling position.

Over the past four years, the community of researchers who utilize DACs in their research have started to utilize a micromanipulator manufactured by Micro Support in their research. In some instances, user facilities provide a central resource for DAC loading. In other locations, research is performed independently of the user facilities.

This workshop will facilitate the collaborative interaction between scientists throughout the U.S. to share best practices in their micro-manipulation work. Through this Workshop, users will learn about valuable techniques from others who practice the art and discuss future developments to provide for additional sampling modalities that will allow for extensions of their research.

The morning session will be composed of presentations on best practices, followed by an afternoon session with hands-on practice on equipment that will be brought to the workshop. Speakers will also be encouraged to bring their experimental modules to demonstrate these capabilities and future developments.

This workshop will bring together scientists from throughout the U.S. to share best practices in their micro-manipulation work as well as others who with to receive hands-on experience with these tools.

Through this Workshop, users will learn about valuable techniques from others who practice the art and discuss future developments that will allow for extensions of their work/research.

8:30 – 9:15	Welcome Coffee
9:15 – 9:30	Welcome from Steve Barnett
9:30 – 9:45	Vitali Prakapenka (GSECARS) *GSECARS Programs*
9:45 – 10:00	Ross Hrubiak (HP-CAT) *HP-CAT Programs*
10:00 – 10:20	Break
10:20 – 10:40	Christopher Snead – NASA
10:40 – 12:00	Steve Barnett *New Accessories to Come and Open Discussion*
12:00 – 1:00	Lunch
1:00	Hands-on Workshop/practical Sessions with Micro Support Axis Pro Tools at GSECARS and HP-CAT Facilities