APS Workshop 7: X-ray Diffraction Coherent Techniques Applied to Chemistry and Materials Science

Thursday, May 8, Morning

8:20 – 8:30 Workshop Organizers
Opening Remarks

8:30 – 9:00 Stephan O. Hruszkewycz (Materials Science, Argonne National Laboratory)
Using Coherent Hard X-rays to Enable Imaging of Nanoscale Structural Heterogeneity in Materials

9:00 – 9:30 Ian Robinson (London Centre for Nanotechnology, University College London)
Functional Strains and Domains in Crystalline Materials

9:30 – 10:00 Amélie Rochet (Brazilian Synchrotron Light Laboratory and Brazilian Center for Research in Energy and Materials)
Investigating Nano-Zeolite Dynamics with Coherent X-ray Scattering Methods

10:00 – 10:20 Break

10:20 – 10:50 Andrej Singer (Cornell University)
Operando *Bragg Coherent Diffraction Imaging in Battery Materials*

10:50 – 11:10 Ross Harder (X-ray Science, Argonne National Laboratory)
Bragg Coherent Diffraction Imaging in the APS-U Era

11:10 – 12:00 Coherent Beamline Visits

12:00 Adjourn Day One

Friday, May 9, Morning

8:00 – 8:10 Workshop Organizers
Opening Remarks

8:10 – 8:40 Mathew Cherukara (X-ray Science, Argonne National Laboratory)
High Performance Computing and AI-enabled Coherent Materials Characterization and Experimental Automation

8:40 – 9:10 G. Brian Stephenson (Materials Science, Argonne National Laboratory)
High-speed XPCS Studies of Composition Fluctuations in Liquids

9:10 – 9:40 Roopali Kukreja (Materials Science and Engineering, University of California-Davis)
Critical Slowdown of Fluctuations in the Vicinity of Metal Insulator Transition in Rare Earth Nickelates

9:40 – 10:00 Eric M. Dufresne (Advanced Photon Source, Argonne National Laboratory)
The APS-U 8-ID XPCS Beamline and its Instruments

10:00 – 10:20 Break

10:20 – 10:50 Hanfei Yan (National Synchrotron Light Source II, Brookhaven National Laboratory)
X-ray Tomography at the Deep Nanoscale with Coherent Diffractive Imaging

10:50 – 11:20 Paul Fenter (Chemical Sciences and Engineering, Argonne National Laboratory)
Coherent X-ray Interfacial Imaging: Progress and Prospects

11:20 – 11:40 Hua Zhou (X-ray Science, Argonne National Laboratory)
Coherent High-energy X-rays Advancing the Quantum and Energy Frontiers

11:40 – 12:00 Workshop Organizers
Final Remarks and Discussion

12:00 Adjourn

Using Coherent Hard X-rays to Enable Imaging of Nanoscale Structural Heterogeneity in Materials

Stephan Hruszkewycz[1]

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

In this talk, we will cover the basic principles governing x-ray Bragg coherent diffraction imaging at modern synchrotron light sources that enable structural heterogeneities of crystalline materials to be imaged at the nanoscale. This capability, combined with the penetrating power of hard x-rays, provides an opportunity to image the response of materials in environments pertinent to applications and revealing of fundamental connections between nanoscale distributions of lattice strain, defects, and domain boundaries and materials properties. The methods of single-particle Bragg coherent diffraction imaging and thin film Bragg ptychography will be discussed, along with examples of these methods in imaging strain evolution in catalytic nanocrystals and domain morphology in ferroelectric thin films. We will also discuss utilizing coherence in the very hard x-ray regime (>50keV) for coherent imaging of buried volumes within the bulk, a possibility now afforded by the APS Upgrade.

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

Functional Strains and Domains in Crystalline Materials

Ian Robinson[1,2]

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From x-ray diffraction measurements, the "microstrain" is defined by the classical Williamson-Hall analysis of neutron or x-ray powder diffraction data. With the development of strain-sensitive imaging methods, such as Bragg coherent diffraction imaging (BCDI), we can now spatially resolve the strain field and add detail to the understanding of strain. Nanoparticles of Barium Titanate perform three times better than macroscopic materials in supercapacitors, causing a flurry of interest to discover how the material worked. While classical XRD shows the material is cubic, x-ray pair distribution function measurements clearly show the local structure is lower symmetry than cubic. This apparent inconsistency is resolved by examining 3D BCDI images of selected nanocrystals, which show the existence of ~50 nm-sized internal domains, interpreted as tetragonal twins, which cause the average crystalline structure to appear cubic [1]. The ability of these twin boundaries to migrate under the influence of electric fields explains the dielectric anomaly for the nanocrystalline phase [2]. In other examples, a striped array domain structure was identified in magnetite microcrystals, which is driven by the structural phase transition at low temperature [3]. Finally, a pronounced phase domain structure was found to be introduced in solid-state electrolyte materials, which may account for their functional improvement in ion conductivity.

[1] "Structural investigation of the metastability of barium titanate nanoparticles grown under hydrothermal conditions", Ana F. Suzana, Sizhan Liu, Jiecheng Diao, Longlong Wu, Tadesse A. Assefa, Ross Harder, Wonsuk Cha, and Ian K. Robinson, Advanced Functional Materials 2208012 (2023)
[2] Electric Field Driven Domain Wall Dynamics in BaTiO3 Nanoparticles, Jialun Liu, David Yang, Ana F. Suzana, Steven J. Leake, Ian K. Robinson, Physical Review B 111 054101 (2025)
[3] Symmetry Breaking during Low-Temperature Domain Formation in micron-sized Magnetite Crystals Yue Dong, David Yang, Jialun Liu, Aly Abdeldaim, Wei Wang and Ian Robinson, submitted to Physical Review B (2025)

Investigating Nano-Zeolite Dynamics with Coherent X-ray Scattering Methods

Amélie Rochet[1,2], Florian Meneau[1,2], Luiza M. Manente[1,2], and Gabriel de Biasi Bafero[1]

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[2]Institute of Chemistry, University of Campinas (UNICAMP), Campinas, Brazil

In order to improve catalysts efficiency, a better understanding of the correlation between three-dimensional displacement fields and the catalytic activity of nanocrystals is essential. Bragg coherent x-ray diffraction imaging (BCDI) enables the determination of lattice strain and defects dynamics at the surface and inner core of nanocrystals. BCDI benefits from the use of coherent hard x-rays to unveil changes at the single nanoparticle level under *operando* conditions with a spatial resolution of the order of 10s of nanometers and picometer sensitivity for lattice displacements. In this work, I will present how BCDI in combination with mass spectrometry can reveal nanoscale peculiarities tuning the catalytic properties of nanomaterials. In particular, I will discuss the highly dynamic 3D strain distribution of nano-ZSM-5 catalysts (300–400 nm) under ethanol upgrading reaction conditions.

I will also show some recent results of chemistry and materials science imaging and dynamics probed by CDI and XPCS at the Cateretê Beamline, the coherent x-ray scattering beamline at the Brazilian Synchrotron Light National Laboratory, Sirius.

Financial support from MCTI, CNPq and the São Paulo Research Foundation - FAPESP (#2021/06876-1) are gratefully acknowledged.

Operando Bragg Coherent Diffraction Imaging in Battery Materials

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Discontinuous solid-solid phase transformations impact rechargeable battery electrode properties. We will present our work using *operando* Bragg coherent diffractive imaging (BCDI) to study phase transformations in lithium-ion battery materials. We will discuss the nucleation and growth processes observed within a particle during operation. Our results show an evolution from curved coherent to planar semi-coherent interfaces controlled by dislocation dynamics. We will address the relationship between interface propagation and transformation kinetics during battery cycling. I will also discuss structure-selective x-ray spectroscopy, which allows us to link the long-range order with redox reactions in layered oxides during intercalation. This work demonstrates how *operando* techniques provide opportunities to study nanoscale phase transformations under various conditions.

Bragg Coherent Diffraction Imaging in the APS-U era

Ross Harder[1] and Wonsuk Cha[1]

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

Bragg Coherent Diffraction Imaging (BCDI) was born at the Advanced Photon Source (APS). Subsequently, beamline 34-ID-C was established as a dedicated facility for early development of the technique and later as a strong instrument for scientific studies using BCDI. This presentation will briefly highlight some of the scientific results from 34-ID-C and discuss both the scientific and technical plans for the new ATOMIC beamline.

This research used resources of the Advanced Photon Source, a U.S. Department of Energy (DOE) Office of Science user facility at Argonne National Laboratory and is based on research supported by the U.S. DOE Office of Science-Basic Energy Sciences, under Contract No. DE-AC02-06CH11357.

High Performance Computing and AI-enabled Coherent Materials Characterization and Experimental Automation

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The capabilities provided by next generation light sources along with the development of new characterization techniques and detector advances are revolutionizing materials characterization by providing the ability to perform scale-bridging, multi-modal materials characterization under *in-situ* and *operando* conditions. For example, providing the ability to image in 3D large fields of view (~mm^3) at high resolution (<10 nm), while simultaneously acquiring information about structure, strain, elemental composition, oxidation state, photovoltaic response etc.

However, these novel capabilities dramatically increase the complexity and volume of data generated, particularly for coherent techniques. Conventional data processing and analysis methods become infeasible in the face of such large and varied data streams. The use of AI/ML methods is becoming indispensable for real-time analysis, data abstraction and decision making at advanced, high-data rate instruments. I will describe how high-performance computing (HPC) along with AI on edge devices enables real-time data analysis and self-driving experiments, creating the next generation of AI-powered materials characterization tools.

[1] Prince, Michael H., et al. "Opportunities for retrieval and tool augmented large language models in scientific facilities." *npj Computational Materials* 10.1 (2024): 251.
[2] Babu, Anakha V., et al. "Deep learning at the edge enables real-time streaming ptychographic imaging." *Nature Communications* 14.1 (2023): 7059.
[3] Kandel, Saugat, et al. "Demonstration of an AI-driven workflow for autonomous high-resolution scanning microscopy." *Nature Communications* 14.1 (2023): 5501.
[4] Cherukara, Mathew J., et al. "AI-enabled high-resolution scanning coherent diffraction imaging." *Applied Physics Letters* 117.4 (2020): 044103.

High-speed XPCS Studies of Composition Fluctuations in Liquids

G. Brian Stephenson[1], Allison Peroutka[2], Dina Sheyfer[3], Jyotsana Lal[2], Qingteng Zhang[3], Eric M. Dufresne[3], Suresh Narayanan[3], and Michael Servis[2]

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

High-speed x-ray photon correlation spectroscopy (XPCS) using new coherent x-ray sources and fast detectors opens new avenues to explore fluctuation dynamics in fluids. We have been studying systems relevant to liquid-liquid extraction processes, where an organic solution of extractant molecules is used to separate ions from an aqueous solution by formation of nanoscale molecular complexes. The organic phase exhibits incipient phase separation, and critical fluctuations play a key role in the structure of the molecular complexes [1-5]. Here we present XPCS studies of microsecond timescale composition fluctuations within 5 K of the critical temperature T_C, carried out at APS beamline 8-ID. With the 500 times higher coherent x-ray flux that is now available from the APS Upgrade and higher-speed detectors such as the TEMPUS [6], it should be possible to observe fluctuation dynamics much further away from T_C. This will enable exploration of the crossover from Ising to mean-field behavior, as well as the changes in dynamics expected at the Widom line (the locus of fluctuation maxima extending from the critical point into the single-phase region). We will present preliminary progress along these lines, as well as behavior calculated from models.

Work supported by U.S. Department of Energy (DOE), Office of Science, Office of Basic Energy Sciences, Chemical Sciences, Geosciences, and Biosciences Division, Separation Science Program, under Contract DE-AC02-06CH11357. This research used resources of beamlines 12-ID-C and 8-ID-I at the Advanced Photon Source, a DOE Office of Science User Facility operated by Argonne National Laboratory.

[1] D. Sheyfer et al., *Phys. Rev. Lett*. 125, 125504 (2020).
[2] M. Servis et al., *J. Phys. Chem. Lett.* 12, 5807-5812 (2021).
[3] D. Sheyfer et al., *J. Phys. Chem. B* 126, 12, 2420-2429 (2022).
[4] B. L. Bonnet et al., *Phys. Chem. Chem. Phys.* 25, 16389-16403 (2023).
[5] T. Rahman et al., *J. Mol. Liq.* 393, 123625 (2024).
[6] J. Correa, et al., *J. Synchrotron Rad.* 31, 1209 (2024)

Critical Slowdown of Fluctuations in the Vicinity of Metal Insulator Transition in Rare Earth Nickelates

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Rare earth nickelates RNiO3 display a metal to insulator transition (MIT), which is accompanied by a magnetic transition, charge ordering, and a crystal structure change from orthorhombic low temperature to monoclinic high temperature state. While this system has been widely studied, the nature of fluctuation across the transition, and the associated time- and length-scales is not known. Spontaneous fluctuations are important component for stabilizing topological magnetic structures such as skyrmions in quantum materials. However, the dynamic susceptibility of the nickelates remains relatively unexplored, and the role played by nanoscale phenomena such as domain-wall formation and motion, local strain fields and phase separation in underlying pathways of MIT is not well-understood. In our work, we focus on understanding the role of nanoscale heterogeneities and their fluctuations in rare earth nickelates by employing x-ray photon correlation spectroscopy (XPCS). Our XPCS measurements on $NdNiO_3$ thin films show complex evolution of fluctuations dependent upon temperature and wavevector q. We also observe unexpected non-equilibrium dynamics and suggests a new approach to understanding these materials.

The APS-U 8-ID XPCS Beamline and its Instruments

Eric M. Dufresne[1], Dana Capatina[1], Miaoqi Chu[1], Matt Highland[1], Zhang Jiang[1], Luca Rebuffi[1], Xianbo Shi[1], Deming Shu[1], Joe Strzalka[1], Kevin Wakefield[1], Qingteng Zhang[1], Zhan Zhang[1], and Suresh Narayanan[1]

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The Advanced Photon Source (APS) recently upgraded its storage ring to a Multibend Achromat with first beam in 2024. The 8-ID X-ray Photon Correlation Spectroscopy (XPCS) beamline was selected as a Featured Beamline of APS-U and two instruments for small- and wide-angle XPCS have been commissioned, and first experiments are taking place. They provide hard x-ray coherent beams on samples from 0.3 to 30 μm with detectors capable of sampling fluctuations from 1 μs to 1000s with large megapixel cameras and high throughput. This talk will describe the beamline specifications, instruments, sample environments, and current status.

This research used resources of the Advanced Photon Source, a U.S. Department of Energy (DOE) Office of Science user facility at Argonne National Laboratory and is based on research supported by the U.S. DOE Office of Science-Basic Energy Sciences, under Contract No. DE-AC02-06CH11357.

X-ray Tomography at the Deep Nanoscale with Coherent Diffractive Imaging

Hanfei Yan[1], Zirui Gao[1], Weihe Xu[1], Wei Xu[1], Evgeny Nazaretski[1], Randy Smith[1], Ajith Pattammattel[1], Xiaojing Huang[1], Yong Chu[1], Dmitri Gavrilov[1], Aaron Michelson[2], Oleg Gang[2], Fang Lu[2], Chonghang Zhao[1], Sukho Kongtawong[1], Yoshiteru Hidaka[1], Guimei Wang[1], Yuke Tian[1], and Kiman Ha[1]

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X-ray ptychography is a coherent diffractive imaging technique that utilizes phase-retrieval algorithms to surpass the resolution limit imposed by focusing optics. It has become a widely used technique available at synchrotron facilities. At the hard x-ray nanoprobe beamline (HXN) of National Synchrotron Light Source II, we have employed ptychography to achieve three-dimensional imaging at the deep nanoscale [1]. This method has proven to be a powerful tool for studying defects in nanoparticle superlattices [2], which are challenging to image by other means. Additionally, probe retrieval enables super-resolution fluorescence imaging through deconvolution and upscaling, facilitating correlative investigations.

In this talk, we will present our recent instrumentation developments that enable high-speed ptychographic scans in the kHz range [3] and discuss the associated challenges, including data processing, beam stability requirements, and low signal-to-noise ratio. We will outline our strategies to address these challenges, such as developing real-time reconstruction methods and implementing a fast source-feedback system based on x-ray beam position monitoring. Finally, we will introduce machine-learning-aided tomography reconstruction with limited-angle projections and spectro-tomography for chemical imaging in 3D. Together with ptychography, these techniques enable smart, fast, and multimodal nano-tomography at HXN.

[1] Michelson, A. et al. Three-dimensional visualization of nanoparticle lattices and multimaterial frameworks. Science 376, 203-207 (2022).
[2] Fang, L. et al. Octo-Diamond Lattice of Nanoscale Tetrahedra with Interchanging Chiral Motifs, Nature Materials, in press.
[3] Xu, W. et al. A versatile high-speed x-ray microscope for sub-10 nm imaging. Review of Scientific Instruments 95, 113705 (2024).

Coherent X-ray Interfacial Imaging: Progress and Prospects

Paul Fenter[1]

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

A robust understanding of interfacial reactivity (e.g., crystal growth, dissolution) that includes both the intrinsic and extrinsic controls over reactivity is a long-term goal of interfacial science, contrasting classical factors such as epitaxy and growth modes with the role of defects and reactive transport. I will describe our work to leverage the coherence of fourth generation x-ray sources to enable a new interfacial imaging modality. This approach extends the established approach of Bragg coherent diffraction imaging (BCDI) to interfaces through reciprocal space maps of coherent x-ray reflectivity (e.g., including both the crystal truncation rod and diffuse scattering). I will describe the conceptual basis and interpretation of coherent interfacial imaging interfaces and thin films [1] along with recent results in demonstrating the successful imaging of interfacial topography. Comparison with BCDI and incoherent interfacial imaging approaches [2], along with the prospects and challenges of this approach, will be discussed.

This work includes significant contributions from Irene Calvo Almazan, Anusheela Das, Ana Suzana and other collaborators. Data were obtained at multiple beamlines (APS-12-ID-D, ESRF-ID1, APSU-8-ID-E). This work was funded by the U.S. Department of Energy, Office of Science, Office of Basic Energy Sciences, Geosciences program under Contract DE-AC02-06CH11357 (FWP# 57814) to UChicago Argonne, LLC as operator of Argonne National Laboratory.

[1] P. Fenter, I. Calvo Almazan, “Imaging Interfacial Topography with Coherent X-ray Reflectivity”, *Physical Review B*, 109, 094112 (1-13) (2024).
[2] P. Fenter, C. Park, and Z. Zhang, Y. Wang “Observation of Subnanometre-high Surface Topography with X-ray Reflection Phase-Contrast Microscopy”, Nature Physics 2(10) 700-704 (2006).

Coherent High-energy X-rays Advancing the Quantum and Energy Frontiers

Hua Zhou[1]

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The Advanced Photon Source Upgrade (APS-U) has ushered in a new era of high-coherence, high-energy x-ray capabilities, establishing APS as a premier facility for *in-situ* and *operando* surface and coherent x-ray experiments. This talk will introduce the coherent high-energy x-ray (CHEX) beamline at 28-ID, a state-of-the-art feature beamline designed to push the frontiers of quantum materials, energy technologies, and microelectronics.

CHEX combines advanced coherent x-ray techniques, including coherent diffraction imaging and x-ray photon correlation spectroscopy, with a highly flexible experimental setup. Featuring four branches, two undulators, and six experimental stations—up to four operable simultaneously—CHEX provides an extensive energy range and enables long-term *in-situ* and *operando* studies as well as rapid, high-throughput experiments. Researchers can harness its exceptional coherence and brightness to explore synthesis mechanisms, dynamic material transformations, and nanoscale structural evolution under realistic conditions. This talk will highlight the unique scientific opportunities enabled by CHEX, including real-time probing of synthesis processes previously inaccessible to coherent x-ray studies, investigations of emerging materials for neuromorphic computing, and advancements in electrochemical systems (e.g., batteries and catalysts). The presentation hopes to foster discussion on collaborative research directions and innovative experimental approaches that CHEX makes possible in the post-APS-U era.