Program

May 6, 2026

Time-Resolved Opportunities at the APS and Beyond: Upgraded Capabilities and Compact X-ray FEL Visions (WK#3 | 2026 APS/CNM user meeting)

Building 402 Lecture Hall
Argonne National Laboratory, 9700 S Cass Ave, Lemont, IL 60439

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Wed, May 6

8:30 AM

Opening remarks

Contribution | **Speaker:** Jonathan Lang (Argonne National Laboratory)

8:40 AM

8:40 AM

Time resolved PDF investigation of inhomogeneous melting in thin metal films

Contribution | **Speaker:** Ian Robinson (Brookhaven National Laboratory)

Description

We performed ultrafast time-resolved diffraction experiments at the x-ray free-electron laser (XFEL) facility in Pohang, Korea to study 300 nm polycrystalline thin films of gold evaporated onto silicon nitride windows, melted by a Ti-sapphire laser pulse [1]. A clear splitting of the (111) powder ring was found at certain fluences after a time delay of 20-100 ps. From the evolution of the x-ray diffraction lineshape, we established separate roles for the electron and phonon contributions in the melting dynamics, consistent with the prevailing 2-temperature model. We deduced that the laser energy is primarily taken up by the electrons, which becomes transmitted into the crystal lattice preferentially at the grain boundaries, converting to heat which diffuses into the grains and eventually melts them. The appearance of liquid was then tracked by pair-distribution function analysis and found to have a slight time dependence following melting [2]. We concluded that the melting process is highly heterogeneous, commencing at the domain boundaries [1]. The role of domain boundaries in the electrical and mechanical properties of crystals is known from electrical conductivity measurements and theoretical modelling. This model can be tested on other metals with focussed ultrafast lasers at high power levels which are sufficient to melt a thin film in a single shot.

[1] Tadesse A. Assefa, Yue Cao, Soham Banerjee, Sungwon Kim, Dongjin Kim, Sunam Kim, Jae Hyuk Lee, Sang-Youn Park, Intae Eom, Jaeku Park, Daewoog Nam, Sangsoo Kim, Sae Hwan Chun, Hyojung Hyun, Kyung Sook Kim, Pavol Juhas, Emil S. Bozin, Ming Lu, Changyong Song, Hyunjung Kim, Simon J. L. Billinge and Ian K. Robinson, Science Advances 6 eaax2445 (2020)

[2] Ian K. Robinson, Jack P. Griffiths, Robert Koch, Tadesse A. Assefa, Ana F. Suzana, Yue Cao, Sungwon Kim, Dongjin Kim, Heemin Lee, Sunam Kim, Jae Hyuk Lee, Sang-Youn Park, Intae Eom, Jaeku Park, Daewoog Nam, Sangsoo Kim, Sae Hwan Chun, Hyojung Hyun, Kyung sook Kim, Ming Lu, Changyong Song, Hyunjung Kim, Simon J. L. Billinge and Emil S. Bozin, IUCrJ 10 656–661 (2023

9:05 AM

9:05 AM

Ultrafast lattice dynamics of metal halide double perovskites by time-resolved X-ray probes

Contribution | **Speaker:** Peijun Guo (Yale)

Description

Developing light-emitting semiconductors with high photoluminescence quantum yield (PLQY) is important for energy-efficient solid-state lighting applications. A subset of solution-processed metal halide perovskites has been shown to exhibit intrinsic, broadband white-light emission with high PLQY from 10% to 90%. The intrinsic white-light emission is attributed to self-trapped excitons (STEs), where photo-excited electron-hole pairs are strongly coupled to local lattice deformations. I will discuss our efforts on understanding light-induced lattice dynamics underpinning STEs in a prototypical metal halide double perovskite using optical pump-probe spectroscopy, time-resolved photoluminescence spectroscopy, and optical-pump X-ray diffraction probe experiments. I will show that charge carrier recombination is asynchronous with lattice dynamics and recovery, and our hypothesis of a long-lived metastable structure with a recovery time of milliseconds. Additionally, I will present our recent work on using synchrotron x-ray micro-diffraction to discern the microstructural evolution of a newly observed spherulite phases from chiral 2D metal halide perovskites.

9:30 AM

9:30 AM

Stochastic and heterogeneous dynamics probed by time-resolved scattering

Contribution | **Speaker:** Alfred Zong (Stanford University)

Description

Photoexcitation by ultrashort laser pulses plays a crucial role in controlling reaction pathways, creating nonequilibrium material properties, and offering a microscopic view of complex dynamics at the molecular level. The photo-response following a laser pulse is, in general, non-identical between multiple exposures due to spatiotemporal fluctuations in a material or the stochastic nature of dynamical pathways. However, most ultrafast experiments using a stroboscopic pump-probe scheme struggle to distinguish intrinsic sample fluctuations from extrinsic apparatus noise, often missing seemingly random deviations from the averaged shot-to-shot response. Leveraging the stability and high photon-flux of time-resolved x-ray micro-diffraction at Beamline 7-ID-C at the Advanced Photon Source, we employed some established statistical tools to quantitatively characterize the stochastic behavior of the photoinduced dynamics in a solid-state lithium-based ionic conductor. By analyzing temporal evolutions of the lattice parameter of a single grain in a powder ensemble, we found that the sample responses after different shots contain random fluctuations that are, however, not independent. Instead, there is a correlation between the nonequilibrium lattice trajectories following adjacent laser shots with a characteristic “correlation length” of approximately 1,500 shots, which represents an energy barrier of ~0.4 eV for switching the photoinduced pathway, a value that is close to the activation energy of lithium ion diffusion [1]. I will conclude the talk by discussing new opportunities brought by this type of analysis to study fluctuations and explore photoinduced metastable states buried in oft-presumed random, uncorrelated stochastic dynamics.

[1] J. McClellan, A. Zong, et al., “Photoinduced correlations in stochastic dynamics of a solid-state ionic conductor,” Nature Communications, in press (2026)

9:55 AM

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Coffee Break

Break

10:15 AM

10:15 AM

Experimentally detecting subtle drivers in the reactivity of aqueous ferrocyanide

Contribution | **Speaker:** Anne Marie March (Argonne National Laboratory)

Description

We are using a multi-edge, time-resolved x-ray spectroscopy approach to investigate how the solvent environment controls reactivity in aqueous iron hexacyanide. This highly charged coordination complex ($[Fe(CN)_6]^{4-}$) exhibits useful redox behavior ($[Fe^{2+}(CN)_6]^{4-} \rightleftharpoons [Fe^{3+}(CN)_6]^{3-}$) alongside a minor ligand exchange reaction in which CN^- is released and replaced by water. We ask whether systematic tuning of the solvation shell (a mixture of water molecules and counterions) can control the quantum yield of this photoaquation channel. Specifically, do particular counterion identities and concentrations bias the fast, collective solvent response to photodissociated CN^- such that geminate recombination is complete, effectively restoring the starting complex? Conversely, are there conditions under which cage escape is maximized, setting the stage for
downstream bimolecular chemistry such as Prussian blue formation? Key to answering these questions is the
ability to characterize solvation shell structure and dynamics during transient reaction stages. We argue that multimodal time-resolved X-ray probes are well-poised to provide this information. This talk will present our efforts using XAS at the Fe K- and L-edges, counterion (K, Na, Ca) K-edges and Fe 1s XES. We anticipate that
probes at the ligand and solvent K-edge and solution scattering (WAXS and time-resolved PDF) will be powerful additions.

This work was supported by the US Department of Energy, Office of Science, Basic Energy Sciences, Chemical Sciences, Geosciences, and Biosciences Division.

10:40 AM

10:40 AM

AI-Driven Spectroscopic Characterization of Defects and Disorder

Contribution | **Speaker:** Mingda Li (Massachusetts Institute of Technology)

Description

Defects and structural disorder govern materials functionality, yet their quantitative, non-destructive characterization remains a major challenge. For defect configuration, we introduce DefectNet, a foundation model that predicts the chemical identity and concentration of multiple coexisting substitutional point defects directly from phonon density-of-states spectra. Trained on over 16,000 simulated spectra across 2,000 semiconductors, the model identifies up to six defect species over a wide concentration range and generalizes to unseen materials. Validation with experimental inelastic scattering data demonstrates accurate, transferable defect quantification from vibrational spectroscopy.

For defect dynamics, we develop an integrated experiment–theory–AI framework combining coherent x-ray photon correlation spectroscopy (XPCS) with theory-informed stochastic simulations and semi-supervised domain adaptation. The approach quantitatively extracts grain-boundary diffusivity, stiffness, and effective boundary concentration directly from measured two-time correlation functions, overcoming the domain gap between simulation and experiment. This framework enables robust characterization of slow, non-equilibrium grain-boundary relaxation in nanocrystalline silicon and provides a general route for bridging theory and experiment in complex spectroscopic measurements.

11:05 AM

11:05 AM

Upgraded pump-probe capabilities at the APS

Contribution | **Speaker:** Xiaoyi Zhang

Description

This brief presentation will highlight the Time-Resolved Research Group's new and enhanced capabilities developed to leverage the APS Upgrade, including pump-probe multimodal imaging and diffraction, grazing-incidence scattering, pair distribution function measurements, and double-laser-pump TR-XAS. Delivered immediately before the 40-minute discussion period in the morning session, it is intended to help frame conversation on how these tools can be most effectively applied to important scientific questions. A central goal is to inform the community about these capabilities while inviting feedback on priority research directions, experimental needs, and data challenges that can help guide the future development of time-resolved capabilities at the APS.

11:20 AM

11:20 AM

Discussion

Session

12:00 PM

12:00 PM

Lunch Break

Break

1:30 PM

1:30 PM

Introduction: A Vision for Argonne's Compact XFEL

Contribution | **Speaker:** Philippe Piot

Description

Argonne National Laboratory is exploring next-generation x-ray free-electron laser (XFEL) concepts to enable capabilities beyond existing facilities while complementing the Advanced Photon Source (APS) storage ring. In this early phase, the focus is on architectures that support multi-user operation and deliver versatile photon-pulse characteristics, including flexible temporal structure, wavelength tunability, and customizable pulse formats.

Advanced accelerator concepts under consideration include collinear wakefield acceleration and two-beam acceleration as potential pathways to compact, high-efficiency XFELs. Synergies with a possible evolution of the APS injection complex are also being explored to leverage existing infrastructure.

This contribution outlines the emerging vision and key directions and invites community input on photon-pulse requirements to help guide the development of a next-generation XFEL at Argonne.

1:50 PM

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Structured Light and X-Ray Pulse Innovation: New Frontiers in XFEL Capabilities

Contribution | **Speaker:** Agostino Marinelli

Description

X-ray free-electron lasers (XFELs) are the brightest sources of x-rays available, with a peak brightness that surpasses table-top harmonic sources and synchrotron radiation facilities by many orders of magnitude. A defining feature of XFELs is their inherent flexibility, which enables the control of the spectral, spatial, and temporal properties of the radiation and tailoring of the pulse properties to specific scientific experiments. Since the early days of LCLS, a vibrant XFEL research and development program has radically changed the way we do science with XFELs, developing new capabilities such as attosecond pump/probe methods, Terawatt pulses and seeding and self-seeding techniques.
In my talk I will give an overview of XFEL advances in the last decade and discuss ongoing and future R&D: from high-brightness cavity-based XFELs to wave-form controlled attosecond pulses.

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Generating the Fastest X-ray Pulses—Advances in Attosecond Inner-Shell X-ray Laser Science

Contribution | **Speaker:** Uwe Bergmann (University of Wisconsin, Madison)

Description

We present our progress in exploring the phenomenon of stimulated x-ray emission spectroscopy (S-XES) based on inner-shell lasing at 6 – 8 keV as a new spectroscopy tool, and as a new source of ultrafast hard x-ray pulses. We first discuss the principle of S-XES, and the experimental methods required for generating and measuring it. We then discuss recent results for spectroscopy applications of S-XES and its potential and challenges of providing enhanced electronic structure sensitivity. We then discuss applications of S-XES as a powerful new x-ray source for probing attosecond dynamics. Here we present the observation of strong lasing effects and a new technique, x-ray coherent attosecond pulse pair spectroscopy (X-CAPPS) enabling interferometry for probing phenomena in the 500 attosecond to 5 femtosecond time window. Finally, we describe the XFEL parameters that would be ideal for advancing the described research.

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Discussion

Session

3:00 PM

3:00 PM

Coffee Break

Break

3:20 PM

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Illuminating Chemical Dynamics: Advancing Reaction Pathways with Future XFEL Capabilities

Contribution | **Speaker:** Thomas Wolf (SLAC National Laboratory & Stanford University)

Description

X-ray free electron lasers (XFELs), with their unique source properties—including femtosecond and sub-femtosecond pulse durations, high peak brightness, and coherence—offer compelling opportunities for chemical research. These capabilities allow for the investigation of compounds undergoing photochemical reactions using site- and element-specific spectroscopy, and, in parallel, for following the corresponding structural changes in real-time via x-ray scattering. Furthermore, non-linear x-ray spectroscopy methods can provide unparalleled insights, for instance, into the chemical composition of interfaces. In this presentation, I will provide an overview of the Linac Coherent Light Source (LCLS), the world's first hard x-ray XFEL, and detail its ongoing upgrade. I will also review current directions in chemistry-related research at LCLS and highlight opportunities afforded by the capabilities of a future next-generation XFEL light source.

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Visualizing dynamics in quantum materials and devices: case for a compact XFEL

Contribution | **Speaker:** Yue Cao (Argonne National Laboratory)

Description

In this talk, I will discuss scientific opportunities at the proposed compact XFEL. These opportunities center around the ultrafast dynamics in quantum materials. These dynamics include changes in the electron wavefunction and density within atomic clusters or cages, as well as evolutions of charge and magnetic domains. In both cases, x-ray resonance and polarization will play a central role and yield insights complementary to that from harder x-ray sources. The interpretation of the experimental data will share similarities with the chemical sciences, while having challenges of its own. The talk will highlight progress from XFEL sources around the world in the last decade and discuss unique opportunities going forward.

The work at Argonne National Laboratory was supported by the U.S. Department of Energy, Office of Science, Basic Energy Sciences, Materials Science and Engineering Division, and through the Early Career Research Program.

4:10 PM

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Unraveling the Molecular Machinery of Life: Biology with XFELs and Next Generation XFELS

Contribution | **Speaker:** Lois Pollack (Cornell University)

Description

X-rays from XFEL sources have had enormous impact on structural biology both through crystallography and solution scattering. The new methods demonstrated at XFELs focus on measuring the structural dynamics of biological macromolecules; these motions enable life. The demonstration of serial femtosecond crystallography, where tens of thousands of small crystals are individually sampled by single, intense x-ray pulses just prior to their destruction, enabled measurements on room temperature (as opposed to frozen) samples e.g., [1]. The ability to gain useful diffraction from micron sized protein crystals also opened up the field of millisecond scale mix-and-inject serial crystallography [2], where the dynamics of reactions involving proteins, including the creation of reaction intermediates, could be followed with atomic resolution. Solution scattering also benefits from the high intensity of XFEL sources. Synchrotron small angle x-ray scattering has proven effective at determining the low-resolution structures of (randomly oriented) biological molecules in solution, including measurements on ensembles of structures or time-resolved studies that follow large scale conformational transitions such as folding. Wide angle x-ray scattering, WAXS has the potential to increase the spatial resolution of solution studies, but the signal strength can be hundreds to thousands of times weaker than SAXS. The high intensity of the XFEL sources now enables time-resolved WAXS studies e.g. [3, 4], increasing the spatial resolution of measurements of structural dynamics. The major theme of these experiments is use of XFEL x-rays to elucidate the structural dynamics of biomolecules, an essential complement to what is known about the structures of biomolecules. Next generation XFELs offer more pulses, to offset concerns about sample consumption as well as novel approaches for experiments on biological molecules.

1. Barends, T.R.M., B. Stauch, V. Cherezov, and I. Schlichting, Serial femtosecond crystallography. Nature Reviews Methods Primers, 2022. 2(1).
2. Pandey, S., et al., Observation of substrate diffusion and ligand binding in enzyme crystals using high-repetition-rate mix-and-inject serial crystallography. Iucrj, 2021. 8: p. 878-895.
3. Zielinski, K.A., et al., RNA structures and dynamics with A resolution revealed by x-ray free-electron lasers. Science Advances, 2023. 9(39).
4. Perera, S.M.D.C., et al., Time-Resolved Wide-Angle X-Ray Scattering Reveals Protein Quake in Rhodopsin Activation. Biophysical Journal, 2017. 112(3): p. 506a-507a

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Discussion: Closing Discussion

Session

5:00 PM