APS Workshop 6: Training Workshop on Combining Reverse Monte Carlo Analysis of X-ray Scattering and Extended X-ray Absorption

Tuesday, May 6, Afternoon

1:30 – 1:35 Workshop Organizers
Opening Remarks

1:35 – 1:55 Shelly D. Kelly (X-ray Science, Argonne National Laboratory)
Trends in Extended X-ray Absorption Fine Structure Spectroscopy

1:55 – 2:15 Byeongdu Lee (X-ray Science, Argonne National Laboratory)
Small Angle Scattering from the Dilute Solution of Nanoparticles

2:15 – 2:35 Markus Winterer (Faculty of Engineering and Center for Nano-integration Duisburg-Essen [CENIDE], University of Duisburg-Essen)
Combining Reverse Monte Carlo Analysis of X-ray Scattering and Extended X-ray Absorption Spectroscopy

2:35 – 2:45 Break

2:45 – 4:00 Markus Winterer (Faculty of Engineering and Center for Nano-integration Duisburg-Essen [CENIDE], University of Duisburg-Essen)
Combining Reverse Monte Carlo Analysis of X-ray Scattering and Extended X-ray Absorption Spectroscopy

4:00 – 4:30 Chengjun Sun (Advanced Photon Source, Argonne National Laboratory)
Panel Discussion: Application of Combining Reverse Monte Carlo Analysis and Future Potential of Integrating with AI/ML

4:30 Adjourn

Trends in Extended X-ray Absorption Fine Structure Spectroscopy

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Advanced spectroscopies are planned for the new APS-U beamline S-25. This beamline will leverage the APS upgrade, featuring brighter beams with reduced divergence, to enhance x-ray collection using our 100mm and 300mm KB mirror systems for advanced spectroscopy and imaging. Alongside x-ray absorption measurements of dilute systems, the spectroscopy group has commissioned two new spectrometers – one for high energy resolution fluorescence detection (HERFD) utilizing a 3-crystal spectrometer, and another large crystal array spectrometer for simultaneous multiple-edge x-ray emission spectroscopy. We are also in the final design stage of a new x-ray Raman scattering microscope, which will enable measurements of low energy edges such as C/O/N using 10 KeV X-rays for *in-situ* and *operando* studies. Additionally, the spectroscopy group is at the forefront of developing Bluesky/Orphyd controls integrated with EPICS, paving the way for automation with a user-friendly interface. These advancements will be discussed.

Small Angle Scattering from the Dilute Solution of Nanoparticles

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Small-angle x-ray scattering (SAXS), which probes the lower q region (typically below 1 $Å^{-1}$), provides low-resolution structural information such as the size and shape of nanoparticles. While SAXS-derived parameters can constrain reverse Monte Carlo simulations, efforts have also been made to analyze full scattering curves, incorporating both SAXS and wide-angle x-ray scattering (WAXS). In this talk, I will briefly discuss the mathematical connection between SAXS and WAXS for nanoparticles and highlight recent advances in computational methods for SAXS analysis in dilute nanoparticle solutions.

Combining Reverse Monte Carlo Analysis of X-ray Scattering and Extended X-ray Absorption Spectroscopy

Markus Winterer[1]

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Extended x-ray absorption fine structure (EXAFS) spectra contain information about the local, molecular type structure, whereas x-ray diffraction (XRD) data reveal the periodic structure or long-range order (crystal structure) of materials. Variations in local and periodic structure greatly influence materials properties and related applications. However, data analysis often is performed independently for EXAFS spectra and diffraction data even if measured simultaneously.

RMC simulations enable the analysis of x-ray scattering data as well as EXAFS spectra data via partial pair distribution (pPDF) functions obtained from a physical, structural model. In case of nanoparticles and scattering data this approach suffers from the termination of the pPDFs due to the finite size of the particles. This produces artifacts in the computed scattering intensity due to the long-range probing distance of scattering which are eliminated using the Debye scattering equation (DSE) [1]. Simultaneous refinement of XS data and EXAFS spectra of small nanoparticles are thus enabled using a mutual structural model. This method allows the self-consistent extraction of complementary information on local structure contained in EXAFS and long-range order in XS data. However, refinement of raw diffraction data for crystals of larger domain size (larger than about 10 nm or 50.000 atoms) are difficult. A Rietveld code embedded into the RMC code and feedback of the essential structural information between both refinement paths enables the coupled refinement of diffraction and EXAFS data are in this case [3].

We will introduce the combined analysis of x-ray scattering and extended x-ray absorption spectra using nanocrystalline $LaFeO_3$ [1] and SnO_2 [2,3] as model systems, provide examples, and demonstrate the use of the code.

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[1] M. Winterer and J. Geiß, *Combining reverse Monte Carlo analysis of X-ray scattering and extended X-ray absorption fine structure spectra of very small nanoparticles*, J. Appl. Cryst. 56 (2023) pp. 7; doi.org/10.1107/S1600576722010858.
[2] V. Mackert, T. Winter, S. Jackson, R. Kalia, A. Levish, S. Lukic, J. Geiss, and M. Winterer, V*ery Small Nanocrystalline Tin Dioxide Particles: Local-, Crystal-, and Micro-Structure*, J. Phys. Chem. C 127 (2023) 17389–17405, 16p.; doi.org/10.1021/acs.jpcc.3c02110.
[3] M. Winterer, *Coupling Rietveld refinement of X-ray diffraction data and reverse Monte Carlo analysis of extended X-ray absorption fine structure spectra*, J. Mat. Res. (2025); doi.org/10.1557/s43578-025-01545-3.

Panel Discussion: Application of Combining Reverse Monte Carlo Analysis and Future Potential of Integrating with AI/ML

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The integration of reverse Monte Carlo (RMC) analysis with x-ray absorption fine structure (XAFS) and x-ray scattering techniques represents a promising frontier in materials characterization. This combination allows for more precise modeling of disordered systems by leveraging the strengths of each method. Future applications are expected to enhance our understanding of complex materials, offering unprecedented insights into their structural dynamics and properties; furthermore, we will also explore the future opportunity of AI and machine learning (ML) for data analysis with potential of application reverse RMC.