Stephan Rosenkranz et al., Struct Chem Crystallogr Commun, 3:2
DOI: 10.21767/2470-9905-C1-002

Recent developments in single crystal diffuse scattering: Imaging nanoscale disorder in reciprocal space

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Correlated defects are responsible for the functional properties of many materials that underpin energy-related technologies. Single-crystal diffuse scattering using x-rays or neutrons offers a powerful probe of such short-range order in crystalline lattices, but its use has been limited by the experimental challenge of collecting data over a sufficiently large volume of reciprocal space and the theoretical challenge of modeling the results. However, instrumental and computational advances at both x-ray and neutron sources now allow the efficient measurement and rapid transformation of reciprocal space data into three-dimensional pair distribution functions, providing model-independent images of nanoscale disorder in real space. We discuss how these recent developments of efficient methods of measuring single crystal diffuse scattering provide new insights into cation disorder in electrode materials. Large volumes of measured diffuse scattering in reciprocal space are transformed into 3D difference pair distribution functions (3D-ΔPDF) that image defect-defect correlations in real space, allowing a model-independent view of short-range order. We demonstrate this with data on β-$Na_xV_2O_5$ with x=0.2 and 0.4 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 no long-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 directly reveal 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.

Work at Argonne National Laboratory was supported by the U.S. Department of Energy, Office of Science, Materials Sciences and Engineering. Research conducted at ORNL's Spallation Neutron Source was sponsored by the Scientific User Facilities Division, Office of Basic Energy Sciences, US. Department of Energy. Research conducted at the Cornell High Energy Synchrotron Source (CHESS) was supported by the NSF & NIH/NIGMS via NSF award DMR-1332208.

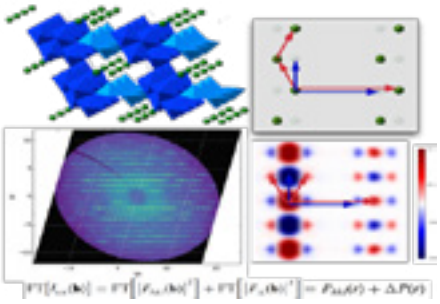

Figure 1: Transformation of the measured diffuse scattering from $Na_xV_2O_5$ and the 3D-ΔPDF transform, which reveals the freezing of the Na ions in a zigzag chain.

Biography

Stephan Rosenkranz is a Senior Physicist in Materials Science Division at Argonne National Laboratory, USA. He has completed his Ph.D. in Physics in 1996 at ETH Zurich. His Diploma in experimental physics in 1992 at ETH Zurich. His research interest is on Structure and dynamics of strongly correlated systems, in particular the role of phase competition in generating complex phenomena. Investigation of long-range order and excitations and short-range correlations and fluctuations due to the presence of ground states with competing order.

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