Invited Lecture

Short-Range Order and 3D-ΔPDF in $V_{1-x}Mo_xO_2$

M. Krogstad[1]

[1]*Argonne National Laboratory, 9700 S. Cass Ave., Lemont, IL, USA*

krogstad@anl.gov

A cornerstone of condensed matter physics is the determination of crystal structure, consisting of perfect translational symmetry over long range, which produces sharp Bragg peaks in x-ray scattering. However, locally correlated deviations from the crystal structure will produce diffuse scattering that is relatively weak and broad compared to Bragg peaks. While not every material displays interesting diffuse scattering, local structure is crucial to understanding some phenomena, including relaxor ferroelectrics, colossal magnetoresistance, and ionic conductors.

Diffuse scattering requires a large dynamic range in detection and is best measured via broad reciprocal space surveys. This can be achieved using high-energy x-rays and a fast area detector, using continuous rotation of a single crystal to quickly map out a broad region of reciprocal space with high sensitivity. The resulting data is suitable for producing a three-dimensional pair distribution function (3D-PDF, or 3D-ΔPDF if Bragg scattering is removed), a real space picture of the deviations of the average structure that can be interpreted using known interatomic vectors. The approach is also helpful in finding weak structural peaks and for characterizing sample quality while being fast enough to produce composition/temperature phase diagrams in a few days.

As an example of this approach, high-energy diffuse x-ray scattering measurements were performed on a single crystal of the $V_{0.81}Mo_{0.19}O_2$ at Sector 6-ID-D at the Advanced Photon Source [1], which displays an insulator-metal transition around 150 K. In its low-temperature insulating phase, relatively sharp rods are observed in reciprocal space [Figure 1], indicating two-dimensional ordering of atomic displacements from the average crystallographic structure. The strong ordering within the planes can be clearly in the resulting 3D-ΔPDF. The slight oscillation of these rods about the [110] direction in reciprocal space can be explained by weak, inherently frustrated coupling between the ordered planes, also observable in the 3D-ΔPDF. Such fragile embedded order is predicted by an Ising-like ferrodistortive model proposed by Lovorn and Sarker [2], and the measured short-range order provides new insight to the this model and the connection between atomic and electronic structure in insulator-metal transitions. A combination of 3D-ΔPDF analysis, structural simulations, and direct calculation of diffuse scattering intensities are all demonstrated here, each supporting this model and clarifying the structure of this material.

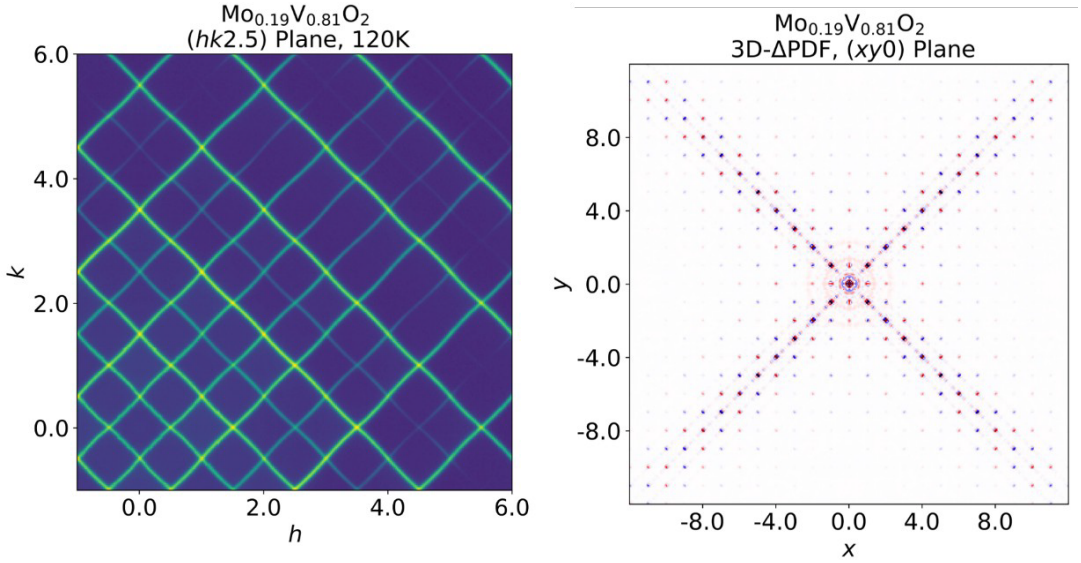

Figure 1. Diffuse scattering (left) and 3D-PDF (right) from $V_{0.81}Mo_{0.19}O_2$.

[1] Davenport, M., Krogstad, M., Whitt, L., Hu, C., Douglas, T., Ni, N., Rosenkranz, S., Osborn, R., &. Allred, J. (2021). *Phys. Rev. Lett.* **127**, 125501.

[2] Sarker, S. & Lovorn, T. (2020). *Phys. Rev. Lett.* **82**, 149902.

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