Real Space Maps of Structural Correlations in Quantum Materials

Raymond Osborn, Materials Science Division, Argonne National Laboratory

Recent advances in synchrotron x-ray instrumentation now allow the collection of large volumes of diffuse scattering in crystalline materials with high efficiency. Reciprocal space volumes comprising tens of thousands of Brillouin zones can be collected in under 30 minutes, allowing the evolution of structural correlations with, *e.g.*, temperature, to be studied in unprecedented detail. The large size of the data sets and the speed of their collection represent both a challenge and an opportunity. The challenge is to perform data reduction rapidly while the experiment is underway, for which we have developed an automated Python workflow that I will briefly describe. The opportunity is that such comprehensive data sets enable completely new ways of accelerating scientific interpretation. In this talk, I will describe two approaches we have been pursuing. The first is unsupervised machine learning, which uses the temperature dependence of different contributions to the scattering to identify rapidly both structural order parameters and their associated fluctuations, while the measurements are still under way. The second is 3D-ΔPDF, pioneered by Weber and colleagues at ETH Zürich, which reveals structural correlations directly, displaying only the probabilities of interatomic vectors that deviate from the average structure, without the need for large-scale simulations. In $Sr_3Rh_4Sn_{13}$, we were able to track structural distortions both above *and* below the structural phase transition, providing a new way of rigorously confirming order-disorder behavior. We were also able to show how a first-order electronic phase transition in Mo_xVO_2 was stabilized by two-dimensional structural distortions, an unusual example of long-range electronic order being triggered by short-range structural fluctuations.

Work supported by the U.S. DOE, Office of Science, Basic Energy Sciences, Materials Science and Engineering Division.