# Recent developments in measuring and analysing large 3D volumes of scattering data to investigate the role of complex disorder on crystalline materials properties

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Many advanced functional properties of crystalline materials derive from complex disorder and short-range correlations that emerge from a subtle balance among competing interactions involving spin, charge, orbital, and strain degrees of freedom. Materials that harbor such disorder generally exhibit strongly enhanced responses, with electronic, magnetic, optical, and thermal properties that are extremely sensitive to perturbations such as magnetic or electric fields and are of considerable importance for future applications. Obtaining a detailed understanding of such complex disorder is required to control and exploit these unusual patterns that persist within short-range ordered states in order to access functional responses inaccessible to conventional, long-range ordered materials. Diffuse scattering is a powerful probe of such complex disorder and when measured from single crystals over large 3D volumes of reciprocal space provides detailed information regarding the existence and morphology of local distortions, as well as defect–defect correlations, i.e., the tendency for defects to cluster into nanoscale ordered structures [1,2].

Recent developments in instrumental advances now allow efficient measurements of single crystal diffraction data over large volumes of reciprocal space using synchrotron x-rays or neutrons. In the case of the latter, dedicated instrumentation, in particular the Corelli instrument at the Spallation Neutron Source, enables measurements of such volumes with elastic discrimination [3]. The value of combining the complementarity of neutrons and x-rays of such measurements over large space of temperature and compositions will be demonstrated on recent investigations that provide new insight on the relation of local order to material properties in relaxor ferroelectrics [4]. While analyzing diffuse scattering data and obtaining detailed models of the underlying structural motifs remains challenging, the availability of comprehensive scattering intensities over large 3D volumes enables new ways of analyzing the data, by utilizing the 3D-ΔPDF method [5]. This method allows to derive for example, direct, model free reconstructions of ionic correlations [6], which are essential to the properties of many energy materials, as well as magnetic correlations in frustrated magnets [7]. Recent advances in Machine Learning methods further provide invaluable and new, rapid insight into the information contained in these large data sets, in particular when measured over varying experimental parameters such as temperature or external fields [8].

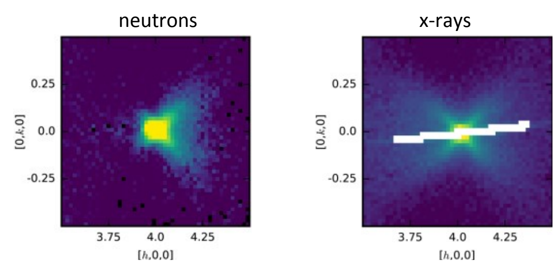


**Figure 1**. The shape difference of the diffuse butterflies observed with neutrons and x-rays in a relaxor compound illuminates the important role of light elements in these compounds [4].

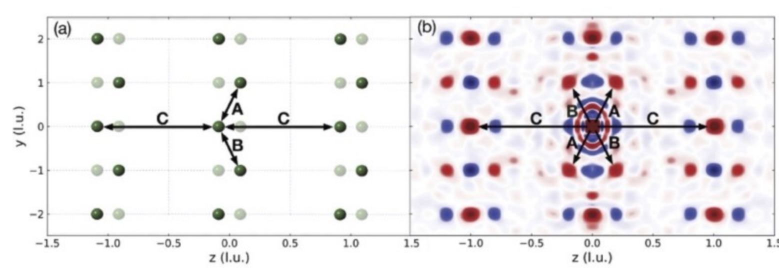


**Figure 2**. Real space model (a) of sodium correlations in $Na_{0.45}V_2O_5$ derived from the ΔPDF transform (b). The Na sites form two-leg ladders and the zig-zag model in (a) is derived by connecting occupied sites with more probable (red) vectors, ignoring neighboring sites with less probable (blue) vectors [6].

[1] Welberry,T.R. Weber, T. (2016) *Crystallography Reviews* **22**, 2-78
[2] see contributions in Issue Diffuse Scattering (2005). *Z. Kristallogr. Cryst. Mater.* **220**, Issue 12
[3] Ye, F., *et al*. (2018). *J. Appl. Cryst.*. **51**, 315 - 322.
[4] Krogstad, M.J., *et al*. (2018). *Nat. Mater.* **17**, 718 - 724.
[5] Weber, T., Simonov, A. (2012). *Z. Kristallogr.* **225**, 238.
[6] Krogstad, M.J, *et al*. (2020). *Nat. Mater.* **19**, 63 - 68.
[7] Roth, N., May, A.F., Ye, F., Chakoumakos, B.C., Iversen, B.B. (2018). *IUrJ.* **5**, 410.
[8] Venderley, J., *et al*. (2020). *Cond-Mat. Archiv* arXiv:2008.03275.

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