Complex Nanofeatures in Crystals

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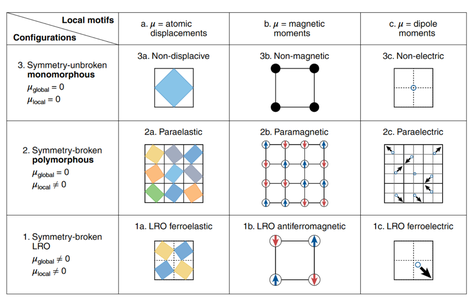
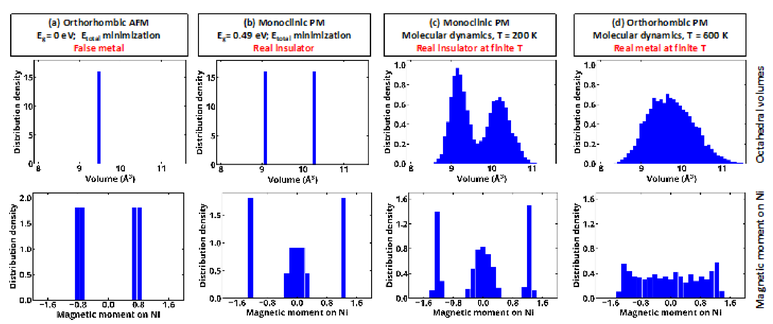
Intrinsic broken local symmetry



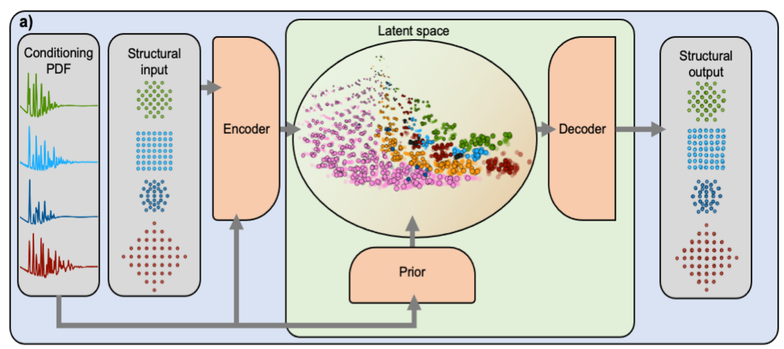
- The novel polymorphous-DFT (polyDFT) searches for materials with a spontaneous *local* symmetry breaking (i.e., polymorphous) property
- Local symmetry breaking is measured using total scattering atomic pair distribution function (PDF) analysis.
- Zunger, A. Nat. Comput. Sci. 2, 529-532 2022

$YNiO_3$ is spin polymorphous



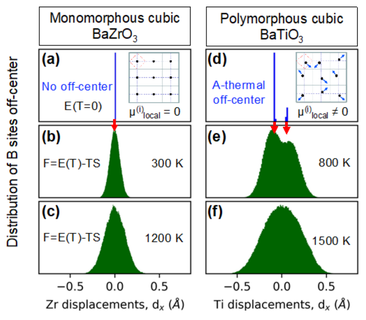
- $YNiO_3$ has a metal-insulator transition on cooling
- from polyDFT: magnetic polymorphism opens a gap in the insulating phase
- structural polymorphism only enhances but does not drive this behavior
- O.I. Malyi and A. Zunger, Phys. Rev. Mater. 7, 044409 (2023). 10.1103/PhysRevMaterials.7.044409

Towards GenAI structure solution

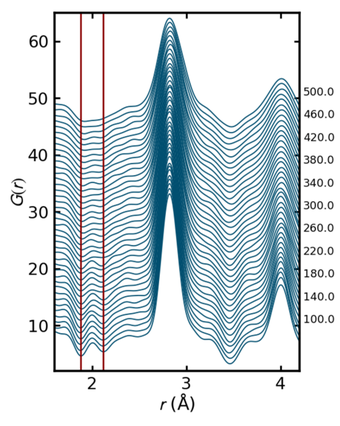


- A convolutional variable autoencoder was used to find structure given a PDF as input
- works well for close-packed metallic nanoparticles
- E.T.S. Kjær, A.S. Anker, M.N. Weng, S.J.L. Billinge, R. Selvan, and K.M.Ø. Jensen, Digital Discovery 2 (2023), pp. 69–80. doi: 10.1039/D2DD00086E.

$BaTiO_3$ is polymorphous

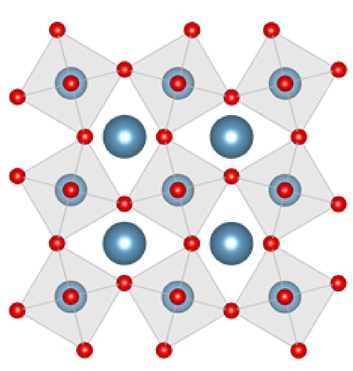


- polyDFT predicts nominally cubic $BaTiO_3$ to be distorted locally.
- Experimental PDFs verify this behavior with clearly bimodal peaks at 500 K (the peaks appear negative due to the negative neutron scattering length of Ti)

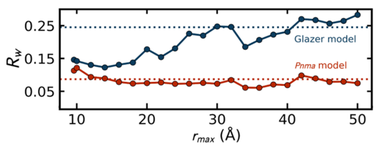


X.-G. Zhao, O. I. Malyi, S. J. L. Billinge, A. Zunger Phys. Rev. B 105, 224108 (2022)

Geometric modelling

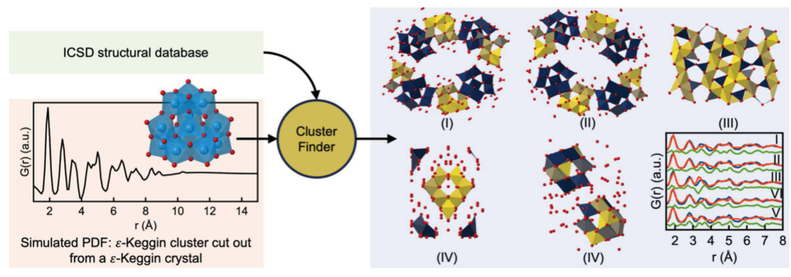


- geometric models of collective rotational distortions are developed and fit to data from polymorphous samples
- The fits allow the separation of rigid and non-rigid contributions to the distortions as evidenced by the difference in fit quality between the non-rigid space-group modeling vs. the rigid Glazer tilt model



S. Skjaervoe, Martin A. Karlsen, Riccardo Comin, Simon J. L. Billinge, arXiv:2203.00127 (2022).

Find clusters within structures



- Given a measured PDF from a small cluster in solution, what is the cluster?
- Assume it exists within a known structure, search structural databases for candidate structures and prune them to find the sub-cluster
- example is a keggin cluster
- A.S. Anker, U. Friis-Jensen, F.L. Johansen, S.J.L. Billinge, and K.M.Ø. Jensen, Acta Crystallogr. A 80.2 (2023), pp. 213–220. doi: 10.1107/S2053273324001116.