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Solving atomic structures by combining analysis of PDF pattern with DFT calculations in machine learning enhanced global optimization

Wilke Dononelli

Determination of crystal structures of nano-crystalline, quantum-crystalline, or amorphous compounds is a great challenge in solid states chemistry and physics. Structural analysis using atomic pair distribution function (**PDF**) of X-ray or neutron total scattering data has the potential to become the most efficient method in this field. Unfortunately, for structure refinements using this method, an initial starting model for the atomic positions is needed, but not available in many cases. To solve this problem, we have recently introduced an algorithm [1] that is able to determine the crystal structure of an unknown compound by means of an on-the-fly trained machine learning (**ML**) model that combines density functional calculations (**DFT**) with comparison of calculated and measured PDFs for global optimization. In our original work, we showed, that the algorithm is able to predict even meta-stable point defects in spinel structures.
Here, we will give a short introduction of the method followed by a case example. $H_2W_2O_7$ and $D_2W_2O_7$ are a metastable material synthesized via selective etching of the Aurivillius-related $Bi_2W_2O_9$ [2] with HCl and DCl, respectively. As shown by Wang et al. [3] this material has the potential to be used as an electrode for high-power proton-based energy storage. Due to the meta-stable nature of the material global optimization of the atomic structure relying on DFT calculations leads to a wrong layer structure. Only combining PDF measurements from X-ray and neutron scattering with DFT calculations leads to the correct layer structure similar to Aurivillius structure of the precursor.

[1] M. Kløve, S. Sommer, B. B. Iversen, B. Hammer and W. Dononelli, Solving atomic structures from PDF patterns with machine learning enhanced global optimization, 2022, submitted. preprint: http://www.quantumchemistry.eu/dftpdf/

N. Lefeld, M. M. Murshed, P. Bottke, Th. M. Gesing, Synthesis and characterizations of $H_2W_2O_7 \cdot nH_2O$ layered tungstates hosting hydrogen storage and conduction. 16th European Conference on Solid State Chemistry, 23. - 26.7.2017 Glasgow, UK.

Wang, R., et al, Fast Proton Insertion in Layered $H_2W_2O_7$ via Selective Etching of an Aurivillius Phase. Adv. Energy Mater. 2021, 11, 2003335.

Primary author(s) : Dr. DONONELLI, Wilke (University Bremen, MAPEX Center for Materials and Processes and Bremen Center for Computational Materials Science)

Co-author(s) : KLØVE, Magnus (Center for Integrated Materials Research, Department of Chemistry and Interdisciplinary Nanoscience Center (iNANO), Aarhus University); Mr. LEFELD, Niels (Universität Bremen, Chemische Kristallographie fester Stoffe, Institut für Anorganische Chemie und Kristallographie); Prof. GESING, Thorsten M. (Universität Bremen, Chemische Kristallographie fester Stoffe, Institut für Anorganische Chemie und Kristallographie); Prof. IVERSEN, Bo B. (Center for Integrated Materials Research, Department of Chemistry and Interdisciplinary Nanoscience Center (iNANO), Aarhus University)