Advances in Single Crystal Neutron Diffraction

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Single crystal neutron diffraction is a premier tool to complement X-ray diffraction by being exquisitely sensitive to elusive elements like, hydrogen, boron, lithium, in heavier element frameworks or matrices. Furthermore, decisively distinguishing similar elements like carbon and nitrogen or iron and manganese in crystal structures. This is important to investigate battery and energy storage relevant materials and their components. It is also of interest for molecular arrangements, where hydrogen bonding is critical and small changes have major impacts, for example in small molecule medical compounds. Neutron diffraction can provide structure and kinetic information of slow changing processes by collecting data in steps, following the process. Fast changing processes can be investigated by stroboscopic measurements if they are readily reproduceable over the time of cycling or pulsing.
Deciphering the kinetic mechanism that drives structural change can be shown through stroboscopic cycling of a model material, potassium dihydrogen phosphate, through an alternating electric field. It has been shown, that the structure of potassium dihydrogen phosphate (KDP) orders and displays a short OH…O bond below the paraelectric- ferroelectric phase transition. OH…O is in the a-b plane and with applied electric field along the c-axis, hydrogen can be pulled or pushed to move between the oxygens. By applying an alternating high voltage electric field along the c-axis, the hydrogen switches between two phosphate oxygens at the same frequency. This allows to study the kinetics of the hydrogen switching. In this example, a voltage with frequencies between 0.1Hz and 10Hz was used to collect experimental data. Data were collected continuously in a stroboscopic mode, where statistically significant intensities are accumulated by repeated electric field cycling. Data were collected at the single crystal time-of-flight Laue neutron diffractometer TOPAZ, which gathers data in event mode. This allowed post-processing of the data and filtering of each event according to voltage value, which is saved as meta-data, and recombining events with the same voltage into a narrow voltage bin. A full, independent structure refinement of each bin allowed to sample the voltage cycle in fine steps. It showed intricate structure movements and adjustments that enabled the hydrogen switching. This manifested in anisotropic displacement parameters, atomic distances and changes in extinction.

We will show the results of event filtering of stroboscopic data and structure analysis.

Fancher C.M., Hoffmann C., Wang X.P., Daemen L.L., Schultz A.J., "Operando single crystal neutron diffraction reveals insight into the field response mechanisms in the hydrogen- bonded KH2PO4 ferroelectric", APL Materials, 9, 021111 (2021).

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