Diffuse Scattering in Single Crystals

Empa
Materials Science and Technology

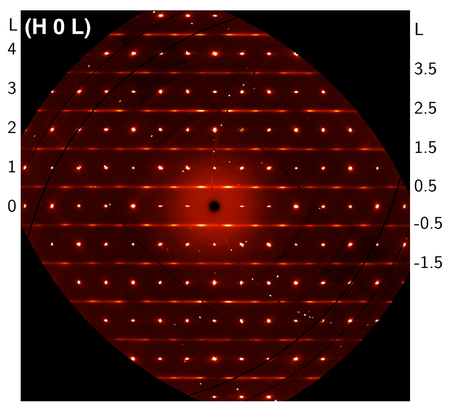
X-ray and Neutron Diffuse Scattering (XDS, NDS) provide $\boldsymbol{k}$-space data on atomic pair correlation functions and thus on deviations from the average periodic structure.

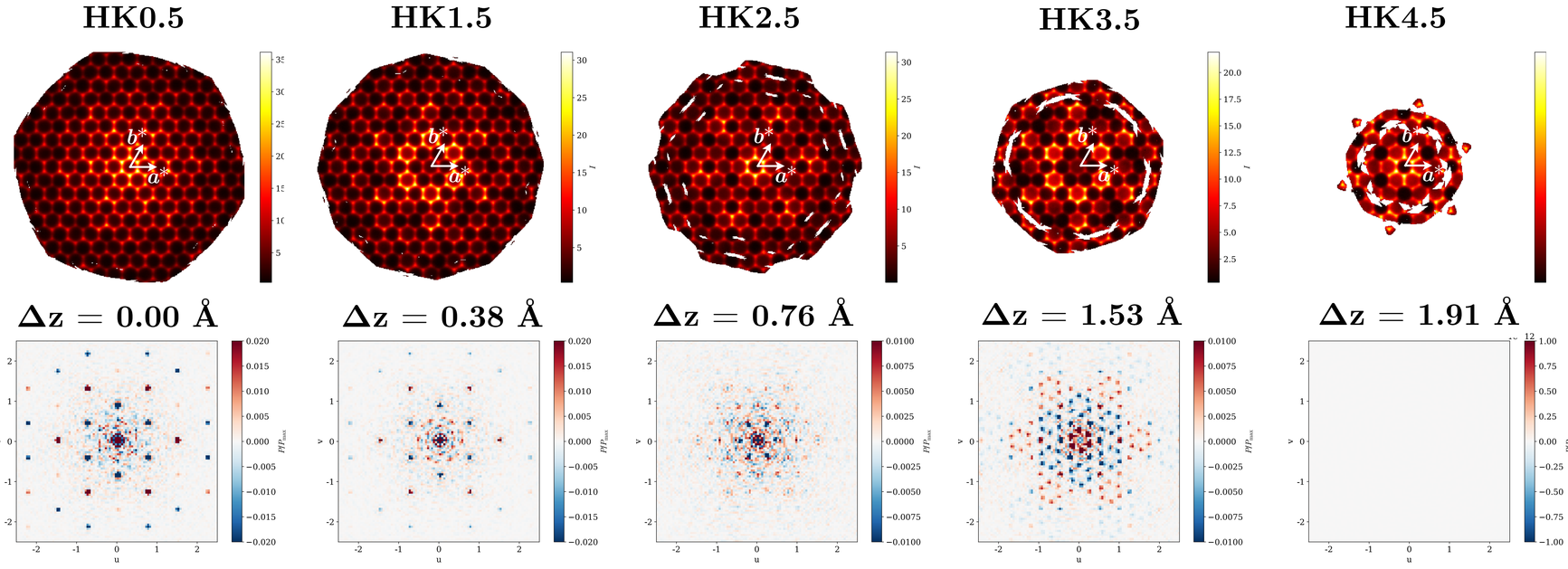
Using high quality datasets it is possible to analyse the diffuse scattering component and extract statistically reliable data from the atomic pair correlation functions: that is about the deviations from the average periodic structure of the crystalline lattice, identify the various types of disorder and derive an abstract model of interatomic vectors. By means of Monte Carlo simulations it is possible then to build and optimize large model crystals providing a specific atomistic disorder model, whose Fourier transform fits to the experimental diffuse scattering.

Experiment

Synchrotron single crystal X-ray diffraction SNBL@ESRF dataset 100 K

$NaLaF_4$

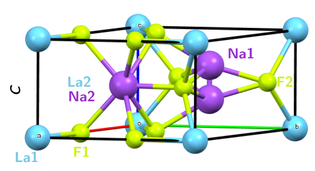




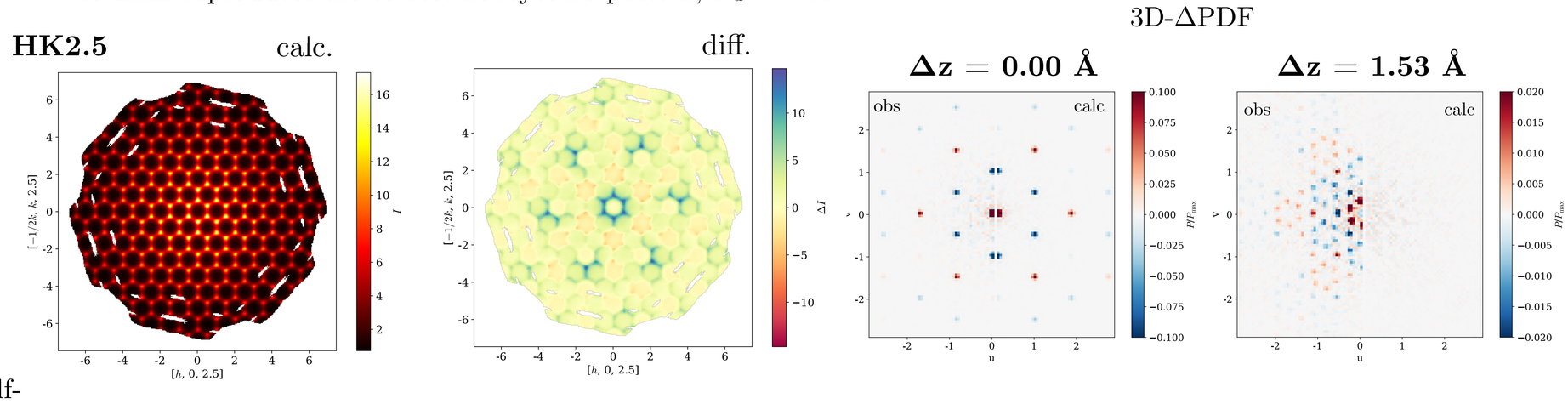
Modelling the disorder

La2/Na2 occupational disorder modelled with Ising parameter, $p = 1.00 \pm 0.04$, between La2-Na2 next neighbour columns reproduces the correct honeycomb pattern, $R_w = 0.42$.

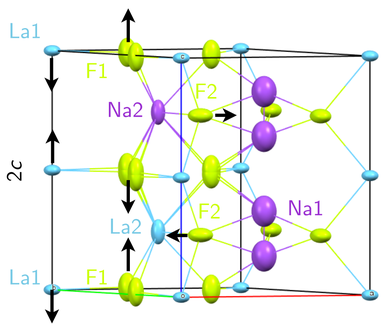
average structure from Bragg peaks $P\bar{6}$



two Ln-sites:
La1 fully ordered
La2/2, Na2/2 disordered

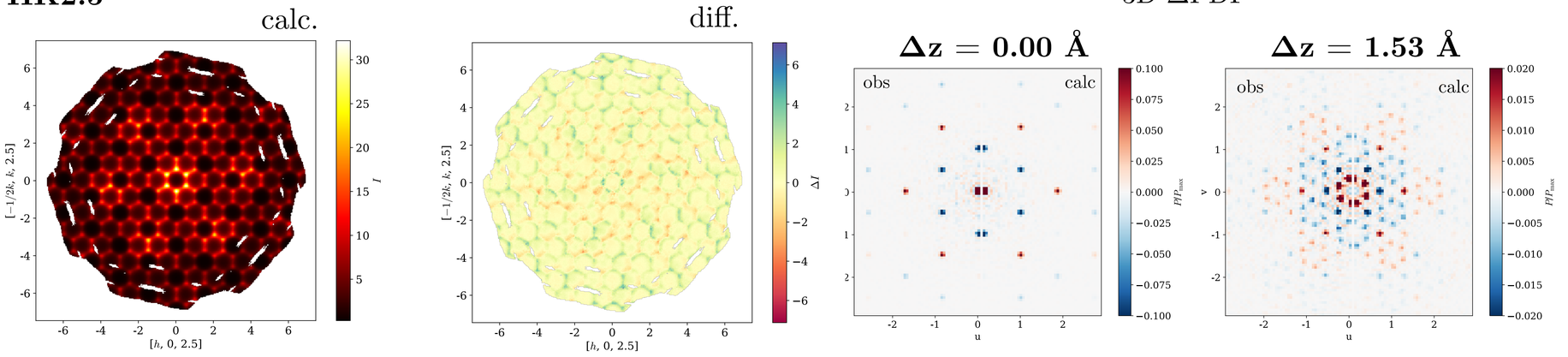


The sharp horizontal lines at half-integer L are produced by a real structure with:



- Columns with Ln...Na...Ln... strictly alternating along c
- Translational symmetry $2c$

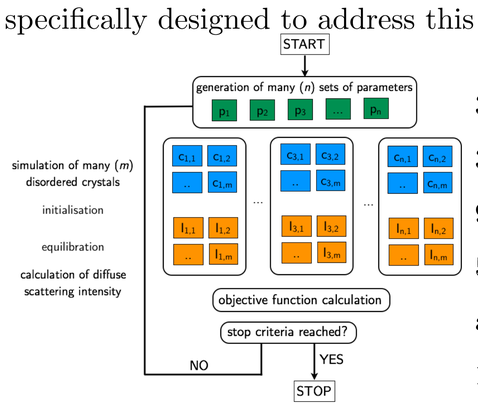
F1 positional disorder along c reproduces some of the diffuse scattering intensity modulations; with La1 positional disorder along c the empty-filled honeycomb pattern is reproduced. Optimisation of La2/Na2 ADP U_{33} adjusts I_{calc} to the proper $\boldsymbol{q}$-dependence, $R_w = 0.21$.



La1, F1 and F2 are displaced from their average position by a Coulomb-type interaction with La2-Na2 columns, given the specific symmetry conditions of their sites, La1 and F1 shift along the c-axis while F2 shifts in the ab-plane (F2 modelling not shown).

High Performance Global Optimisation - population based method

The program ZODS (Zurich Oak Ridge Disorder Simulation) is an expandable, easy-to-use, High Performance Computing program which can run both on desktop and clusters specifically designed to address this kind of problems



36 crystals “clones”

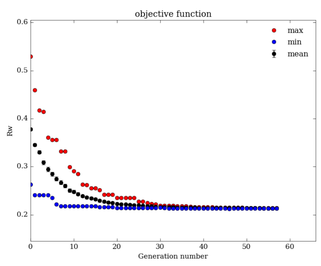
300 x 300 unit cells each

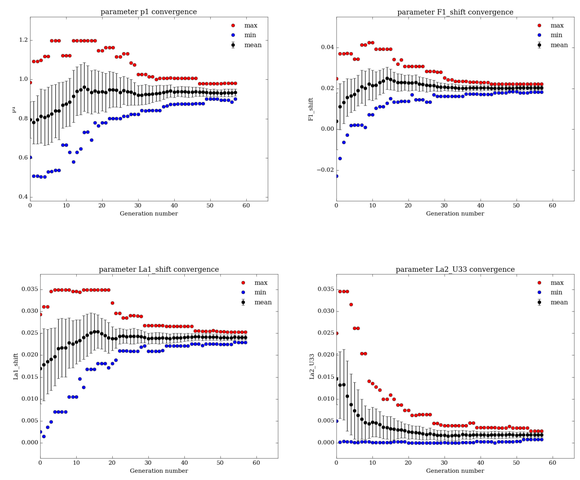
900 lots for intensity calculation

51 parameters (gene) sets

approx. 60 generations

1836 computing units (“cores”) on PizDaint@CSCS





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[2] T. Weber and A. Simonov, *Zeit. für Krist.*, (2012) 227, 238-247; A. Simonov, T. Weber, and W. Steurer, *J. App. Cryst.* (2014) 47, 1146-1152.

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