Retrieving sub-angstrom resolution from low order dynamical diffraction intensities

Grigory Kornilov[1], Sam Fairman[1], Sherjeel Shabih[1], Dr. Benedikt Haas[1], Dr. Niklas Dellby[2], Adnan Hammud[3], Dr. Zbigniew Galazka[4], Prof. Christoph Koch[1]
[1]Department of Physics, Humboldt-Universität zu Berlin, Berlin, Germany, [2]Bruker AXS (formerly Nion Co. R&D), Kirkland, USA, [3]Fritz Haber Institute of the Max Planck Society, Berlin, Germany, [4]Leibniz-Institut für Kristallzüchtung, Berlin, Germany

Background incl. aims
While dynamical scattering has typically been viewed as a hindrance when attempting to retrieve an unknown crystal structure from electron diffraction data, it can also be of direct benefit since it encodes phase information in the intensity of the electron wave function exiting the crystal [1]. For example, recent advances in 3D electron diffraction have utilized dynamical effects to complement and significantly improve kinematical refinement methods [2]. Building on prior work by Feng et al. [3] we have developed a structure retrieval method which, using experimental large-angle-rocking-beam electron diffraction (LARBED) data, is not only capable of reconstructing complex crystal structures, but also shows that dynamical scattering can be directly used to increase the resolution of a reconstruction beyond what would be achievable under assumption of purely kinematical conditions.

Methods
The approach by Feng et al. [3], which was demonstrated on a rather simple structure ($SrTiO_3$), used a conjugate gradient method to minimize the sum of squared differences between an experimental LARBED pattern and one simulated via the Bloch wave formalism from reconstructed structure factors. We expanded on this method by switching to the adaptive moment estimation (ADAM) optimization algorithm as well as introducing analytical calculation of gradients of the scattering matrix. Significant improvements were achieved by utilizing a series expansion of the scattering matrix [4] to develop a weighting scheme which selectively prioritizes matching those diffraction intensities which have a dominating linear dependence on the structure factors of the crystal. Experimental measurements were performed using a Dectris ELA detector mounted to the end of the IRIS spectrometer of a Nion HERMES microscope, allowing zero-loss filtering with the energy-selecting EELS beam trap. The results presented below were achieved using a roughly 40nm thick lamella of β-Ga_2O_3 which was prepared with the FIB technique from a bulk crystal grown by the Czochralski method [5].

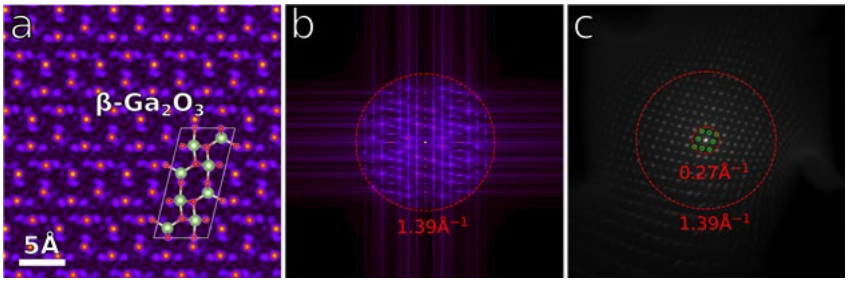
Results

Figure a) shows the 2D projected potential of β-Ga2O3 in [010] zone axis, generated from reconstructed structure factors, with the theoretical structure overlaid on top. The structure factors were refined from 1129 diffraction patterns of an experimental LARBED tilt series, collected with an accelerating voltage of 200 kV and a maximum tilt angle of 100 mrad. Figure b) shows the Fourier transform of the reconstructed potential with the red circle indicating the range of visible frequency components (image is not to scale with a)). Figure c) shows the mean of all experimental diffraction patterns collected in the LARBED measurement. While many excited beams are visible, only the first 8 (indicated by green circles) were actually used in the reconstruction of the crystal structure. Our method reaches a sub-angstrom resolution of 1.39 Å-1, which is over 5 times finer than the maximum resolution of 0.27 Å-1 which one obtains when only using the structure factors corresponding to these 8 beams, as in kinematic scattering theory. The reconstructed potential shows good agreement with reference data, further validating this result. In addition to the crystal structure, a specimen thickness of 36.9 nm was also retrieved. The reconstruction was performed ab-initio, i.e. without initializing it with any reasonable starting guess. Besides a small regularization term in the loss function to aid convergence, no further constraints were placed on reconstructed structure factors and no prior information about the crystal, except for its lattice parameters (which can also be extracted from the diffraction data), were used.

Conclusion

In summary, we have directly utilized experimental dynamical diffraction data to perform an ab-initio reconstruction of a crystal at a much higher resolution than would be possible with purely kinematical methods. While we show the best result in this text, our method has successfully been tested on experimental data from further crystals, such as BaTiO3 and Al2O3. An interesting consequence of these findings is that it may be possible to reconstruct a 3D structure from 2D rocking curve data, since there is no fundamental difference in how structure factors corresponding to different reciprocal lattice vectors influence the diffraction pattern produced by the crystal. Preliminary testing on simulated data appears to be promising in this regard and will be further discussed.

Graphic:



Keywords:

dynamical scattering, crystallography, electron diffraction

Reference:

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