## MS26 O1

**Precise Estimation of Structure Parameters from Electron Microscopy Images** Sandra Van Aert, Sara Bals, Gustaaf Van Tendeloo, Dirk Van Dyck *Department of Physics, University of Antwerp, Antwerp, Belgium.*
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Electron microscopy allows to precisely determine unknown structure parameters. For example, the precision with which atom positions can be determined from electron microscopy images is comparable to the precision obtained from X-ray powder diffraction data. Furthermore, as a result of the strong interaction of electrons with the material under study, it is possible to obtain this information on a more local scale. However, in order to precisely determine unknown structure parameters, a quantitative, model-based method is required. The aim of this contribution is to introduce such a method and to demonstrate its practical applicability by means of recent experimental examples.

The key to precise structure determination is the availability of an expectation model, which is a, usually physics based, model describing the expectations of the image pixel values. In fact, the expectation model includes all the ingredients needed to perform a computer simulation of the images and it is parametric in the quantities of interest, such as the atom positions. Next, the unknown parameters of the model are estimated using statistical parameter estimation theory. This is done by optimizing a criterion of goodness of fit, such as the likelihood function or the least squares sum. This criterion quantifies the similarity between the observations and the expectation model. The parameters for which this criterion is optimal, represent the estimated parameters. A comprehensive report of this model-based method can be found in [1-2].

In one of the experimental examples which will be shown, the atom column positions of a new ceramic compound, $Bi_4Mn_{1/3}W_{2/3}O_8Cl$, have been determined with a precision in the picometer range [3]. Therefore, exit wave reconstruction combined with statistical parameter estimation has been used. The phase of the reconstructed exit wave reveals the light oxygen atoms in the presence of heavier atoms. Although the phase of the exit wave is often considered as the final result, it has been used as a starting point for quantitative refinement of the atom column positions. Therefore, a model has been proposed to describe the expectations of the pixel values in the reconstructed phase. The unknown parameters of this model, including the atom column positions, have been estimated in the least squares sense. Next, mean interatomic distances and their corresponding standard deviations have been calculated from the estimated atom column positions. The standard deviation, being a measure of the precision, ranges from 3 to 10 pm. Furthermore, a good agreement has been found when comparing these results with X-ray powder diffraction data. It is also found that projected distances beyond the information limit of the microscope (110 pm) can be determined with picometer range precision.

[1] den Dekker, A.J.; Van Aert, S.; van den Bos, A.; Van Dyck, D. Ultramicrosocpy, 2005, 104, 83.
[2] Van Aert, S.; den Dekker, A.J.; van den Bos, A.; Van Dyck, D.; Chen, J.H. Ultramicroscopy, 2005, 104, 107.
[3] Bals, S.; Van Aert, S.; Van Tendeloo, G.; Ávila-Brande, D. Physical Review Letters, 2006, 96, 096106 (1-4).

## MS26 O2

**Symmetry study of $PbZr_{1-x}Ti_xO_3$ by Convergent-Beam Electron Diffraction** Roland Schierholz[a], Hartmut Fuess[a], Yoichiro Ogata[b] Kenji Tsuda[b], Masami Terauchi[b]
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$PbZr_{1-x}Ti_xO_{3-}$ ceramics exhibit the highest values in piezoelectric constants close to the morphotropic phase boundary (MPB). At this boundary the crystal structure changes with composition and the material contains minute domain structures. These were reported to affect X-ray diffraction profiles the same way as a lower crystallographic symmetry [1].

To analyse the crystal symmetry of $PbZr_{1-x}Ti_xO_3$ over the MPB convergent-beam electron diffraction (CBED) was used. CBED enables us to determine local symmetries of a few nm-size specimen area. TEM-samples with compositions x = 0.40, 0.45, 0.46, and 0.48 were prepared by using ion-milling instrument (ion-slicer). CBED experiments were mainly conducted by using a transmission electron microscope JEM 2010 equipped with $LaB_6$.

To judgement on symmetry is based on projected Whole pattern (proj. WP) symmetry whole pattern (WP) symmetry including higher order reflections, as well as the symmetry of Dark field Patterns (DP's). The results for the two morphotropic compositions x = 0.45 and 0.46 show evidence for a monoclinic structure first proposed by Noheda *et al.* [2] based on fitting to X-ray powder diffraction profile.

For the x = 0.46 composition also heating experiments were performed and at ~300 °C tetragonal symmetry was observed. This result is again consistent with X-ray powder diffraction experiments by Noheda *et al.* [3].

For the compositions on the tetragonal x = 0.48 and on the rhombohedral side x = 0.40 of the MPB symmetries of CBED-patterns were not always clear as expected for the above named crystallographic symmetries of single crystalline materials of non-solid-solution systems. Since a breakdown of symmetry in CBED-patterns can be caused by several other influences, an interpretation of the results is not straightforward. Ideas for these influences and how to recognize some of them will be discussed.

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