MS112.O01

Accurate charge densities from powder X-Ray diffraction

Mads Ry Vogel Jørgensen[1], Kasper Tolborg[2], Hidetaka Kasai[3], Jacob Becker[2], Ann-Christin Dippel[4], Jens Als-Nielsen[5], Bo Brummerstedt Iversen[2]

[1]*Department Of Chemistry, Aarhus University & MAX IV Laboratory, Aarhus C, Denmark,* [2]*Center for Materials Crystallography, Department of Chemistry and iNANO, Aarhus University, Aarhus C, Denmark,* [3]*Faculty of Pure and Applied Sciences, CiRfSE and TIMS, University of Tsukuba, 1-1-1 Tennodai, Tsukuba, Ibaraki, 305-8571, Japan,* [4]*PETRA III, Deutsches Elektronen-Synchrotron - DESY, Hamburg, Germany,* [5]*Niels Bohr Institute, University of Copenhagen, Copenhagen, Denmark*
E-mail: mads@inano.au.dk

In recent years powder X-ray diffraction (PXRD) has proven to be a valuable alternative to single crystal X-ray diffraction for determining the electron density distributions (EDD) in high symmetry inorganic materials including subtle deformation in the core electron density. This was made possible by performing diffraction measurements in vacuum using high energy X-rays at a synchrotron radiation facility [1]. Some of the traditional systematic errors in single crystal diffraction, such as extinction and absorption are significantly reduced, or more easily corrected, in powder diffraction. However, there are some intrinsic issues when using powder diffraction, namely peak overlap and background modelling. By using a combined Hansen-Coppens (HC) multipole-Rietveld approach and minimizing the background signal by conducting the experiment in vacuum it is possible to extract very accurate structure factors. Applying the HC model allows for a more accurate partitioning of overlapping intensities and thus removes the bias towards the independent atom model.

The scope of this project is to obtain the best possible PXRD data for experimental EDD modelling. To facilitate this, we have in collaboration with JJ X-ray A/S, built a series of custom diffractometers that can be set up at almost any beam line. The key feature of the diffractometer is that the sample and direct beam are in vacuum, thus minimizing the air scattering. The original vacuum diffractometer had a sample to detector distance of 300 mm. This instrument has been shown to yield data of exceptionally high quality matching the Pendellösung data, but extending to much higher resolution [2]. Data obtained with this instrument has been used to study the core electron deformation in diamond, silicon and cubic boron nitride.

To permit studies of more complex materials we have recently commissioned a new version of the diffractometer, the 'Aarhus Vacuum Imaging plate Diffractometer' – AVID [3]. This instrument has improved both the peak width and the signal-to-background ratio by a factor of three. This has been possible by increasing the sample to detector distance to 1200 mm. Benchmark data on Si at 100 K, extending to $\sin(\theta)/\lambda \sim 2.4$ Å^{-1}, shows that the low order reflections are of a comparable quality and the high order data far surpass the quality of the structure factors from the old setup.

Here we review both method, the technical development of the instruments, and the results obtained.

[1] Jørgensen, M. R. V., Hathwar, V. R., Bindzus, N., et al. (2014). IUCrJ 1, 267-280.
[2] Wahlberg, N., Bindzus, N., Bjerg, L., et al. (2016). Acta Cryst. A 72, 28-35
[3] Tolborg, K., Jørgensen, M. R. V., Christensen, S., et al. Submitted to Acta Cryst. B.
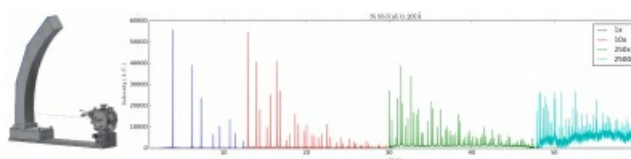
Keywords: Electron density, Synchrotron radiation, Powder X-ray diffraction