Optimal experiment design for atom-counting from 4D scanning transmission electron microscopy

Aberration-corrected scanning transmission electron microscopy (STEM) has become a powerful technique for materials characterisation of complex nanostructures. In combination with quantitative methods reliable structural and chemical information can be obtained in two dimensions (2D) as well as in three dimensions (3D) from experimental images. In a quantitative framework, images are treated as datasets from which structure parameters are determined by comparison with image simulations or by using parameter estimation based methods. In order to obtain precise atom counts along the viewing direction, scattering cross-sections are very useful. These cross-sections describe the total scattered intensity for each atomic column.

So far, atom-counting has mainly been used for homogeneous nanomaterials. In order to extend this method to heterogeneous materials, the 4D STEM technique will be explored, where a fast pixelated detector is used to record the full 2D diffraction pattern as a function of the position of the probe position. The availability of 4D STEM datasets can be used to create and analyse multiple 2D STEM images providing independent information concerning thickness, composition and ordering of the atoms along the viewing direction.

In order to determine the limits to the precision with which this information can be estimated from multiple 2D STEM images, the principles of statistical detection theory will be used. Here, the probability of error is computed for detecting both the number and type of atoms in a binary atomic column. We will derive for this system the optimal angles of two independent regions in the detector plane from which the 2D STEM images are created.