3D Imaging and Analysis

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Three-dimensional particle reconstruction by depth sectioning in scanning transmission electron microscopy

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Thanks to recent progress in aberration correction, atomic resolution is now possible in High Angle Annular Dark Field Scanning Transmission Electronic Microscopy (HAADF-STEM). Unfortunately this impressive resolution is limited to two-dimensional images and information on the axial structure of the sample is still missing. Recently several methods [1,2] have been proposed to obtain three-dimensional information from a tilt-series. While efficient, this technique is limited to the small volume of the sample that remains in the field of view for all tilts. Earlier, van Benthem *et al.* [3] proposed the use of depth sectioning, where several images of the same sample are taken at different focus. In this way vertical structures can be observed directly, like in three-dimensional light microscopy [4]. This technique is easy to implement on commercial microscopes and does not necessitate a specific sample preparation. Unfortunately the vertical size of the probe is of the order of several nm, thus far too large to distinguish individual atoms [4]. Moreover non-linear interaction between atoms are particularly important in the propagation direction, hindering a direct interpretation of the set of measurements [5]. Besides this drawback, we think that this technique might allow useful measurements, providing a suitable data treatment and using enough *a priori* knowledge on the sample. In this work, we study the possibility to measure the atom position in a gold nanoparticle using a three-dimensional image. We first study the precision that can be achieved using the so-called Cramér-Rao lower Bound (CRLB). This theoretical tool gives a lower bound on the variance with which parameters can be estimated unbiasedly and can be derived from a parametric statistical model defining the expectation as well as the noise statistics [6]. To compute the expectation values, we used a multislice simulation of a column of 10 gold atoms imaged by a HAADF detector assuming an aberration-corrected STEM microscope with a semi convergence angle of 22 mrad. We described the noise as a Poisson statistic. Our study proves that, even in this ideal case, the precision that can be reached when estimating the atom vertical position is larger than the inter-atom distance for realistic dose values. However, when estimating only the center-of-mass of the column a precision of a few pm is reachable (see Figure 1). This theoretical study proves that depth sectioning cannot be used to estimate the position of every single atoms of a nanoparticle but can be used to estimate the position of each column. Based on this result, we developed a procedure for reconstructing a gold nanoparticle from an HAADF-STEM depth sectioning three-dimensional image. We first estimate the lateral position and atom numbers of each column of the particle from one of the two-dimensional images [7]. We then simulate with an accurate multislice algorithm, a focal series of each type of atom columns present in the particle, assuming the inter-atomic distance as identical as in a perfect crystal. These templates are then matched to the nanoparticle's image using a maximum likelihood procedure. A numerical example of this procedure is shown in Figure 2.

1. S. Van Aert, K. J. Batenburg, M. D. Rossell, R. Erni and G. Van Tendeloo, Nature 470 (2011), pp. 374–377.
2. B. Goris, S. Bals, W. Van den Broek, E. Carbo-Argibay, S. Gomez-Grana, L. M. Liz-Marzan and G. Van Tendeloo, Nature Mat. 11 (2012), pp. 930–935.
3. K. van Benthem, A. R. Lupini, M. P. Oxley, S. D. Findlay, L. J. Allen and S. J. Pennycook, Ultramicroscopy 106 (2006), pp. 1062 – 1068.
4. W. Van den Broek, S. Van Aert, D. Van Dyck, Ultramicroscopy 110 (2010), pp. 548 – 554.
5. E. C. Cosgriff, P. D. Nellist, Ultramicroscopy 107 (2007), pp. 626 – 634.6A. den Dekker, S. Van Aert, A. van den Bos and D. Van Dyck, Ultramicroscopy 104 (2005), pp. 83 – 106.7S. Van Aert, A. De Backer, G. T. Martinez, B. Goris, S. Bals and G. Van Tendeloo, Phys. Rev. B, 87, (2013) p. 0641078 The authors acknowledge financial support from the Research Foundation Flanders (FWO) through project fundings G.0393.11, G.0064.10 and G.0374.13.

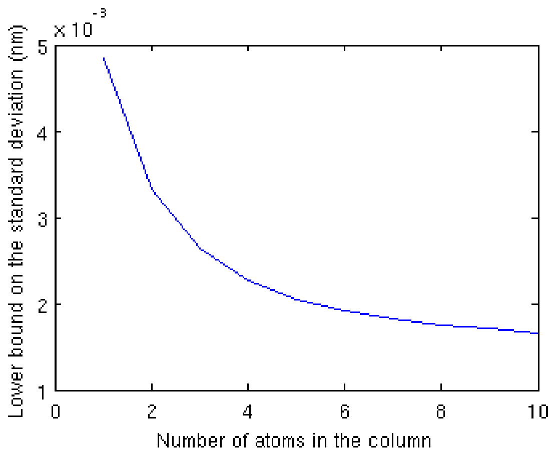

Figure 1. Lower bound on the standard deviation with which the vertical position of a gold atom column can be estimated from HAADF-STEM depth sectioning as a function of the number of atom in the column. The electron dose is 210 000 electrons, corresponding to a dwell time of 1 ms with a probe current of 40 pA.

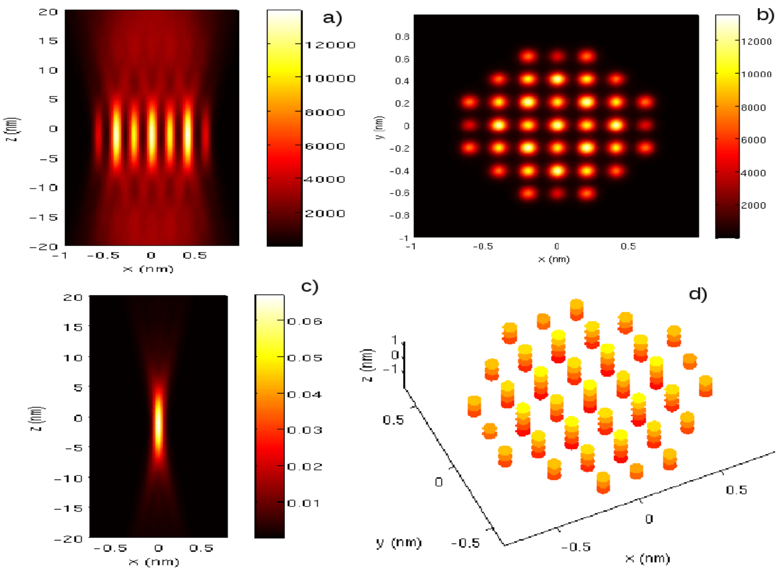

Figure 2. Particle reconstruction of an ellipsoidal gold nanoparticle from synthetic data of HAADF-STEM focal series. a) and b) (xz) and (xy) cut of the simulated depth sectioning of the nanoparticle. Colorbars scale in number of detected electrons. c) (xz) cut of the template for a column with 4 gold atoms. Colorbar scales in the detection probability for one electron dose. d) Reconstructed atom positions. Atoms with the same color are at the same vertical position.