**Three-dimensional atomic models from a single projection using Z-contrast imaging: verification by electron tomography and opportunities**

De Backer, A.[1], Jones, L.[2,3], Lobato, I.[1], Altantzis, T.[1], Nellist, P.D.[4], Bals, S.[1] and Van Aert, S.[1]

[1] EMAT, University of Antwerp, Belgium, Belgium, [2] Department of Materials, University of Oxford, United Kingdom, Ireland, [3] Advanced Microscopy Laboratory (AML), School of Physics & CRANN, Trinity College Dublin, the University of Dublin, Ireland, Ireland, [4] Department of Materials, University of Oxford, United Kingdom, United Kingdom

In order to fully exploit structure - property relations of nanomaterials, a three-dimensional (3D) characterization at the atomic scale is often required. In recent years, the resolution of electron tomography has reached the atomic scale [1,2]. However, for a successful 3D reconstruction using electron tomography, there are several limitations. Electron tomography requires multiple exposures, which is not always feasible in practice. For example, *in situ* experiments or the characterization of beam sensitive materials at the atomic scale are nearly impossible using electron tomography. In order to overcome these limitations, a newly developed alternative method is presented where the 3D atomic structure is reconstructed from a single projection [3,4,5].

The method combines atoms-counting [6,7] in annular dark field (ADF) scanning transmission electron microscopy (STEM) with prior knowledge about a material's crystal structure. The atom counts are used to generate an initial 3D configuration by positioning the atoms in each atomic column, parallel to the beam direction, symmetrically around a central plane. An energy minimization can be applied to obtain a relaxed 3D model of the nanostructure.

We validate, at the atomic scale, this single projection reconstruction approach against state-of-the-art compressive sensing electron tomography [2]. For the tomography approach, three projection ADF STEM images of a Au nanorod have been acquired along different major zone axes. In Fig. 1, a comparison is shown between the two atomic resolution reconstruction methods. An excellent visual match of the overall morphology of the nanorod has been found.

Atom-counting combined with energy minimization opens up new possibilities for the 3D characterization of materials where electron tomography cannot be applied. Perhaps the biggest potential scope for this is where *in situ* holders simply do not exist with sufficient tilt ranges for tomography. We illustrate the utility of the proposed approach for imaging a Au nanodumbbell on an *in situ* heating holder. The nanodumbbell was heated up to 330 °C and underwent a morphological transformation. The 3D atomic resolution reconstructions of the nanodumbbell and nanorod, before and after heating respectively, are shown in Fig. 2. From the reconstructions, the surface facets can be clearly observed for the entire tip of the nanodumbbell and nanorod and a significant increase of low index facets is observed after heating. We expect these measured properties to be of critical importance for the study of catalysis.

In conclusion, an impressive agreement was found between our new atom-counting/energy minimization approach and state-of-the-art compressive sensing electron tomography. This method opens up the possibility for the study of beam-sensitive materials, 2D self-assembled structures [8], and where *in situ* hardware makes tomography impossible.

[1] Van Aert et al., Nature 470 (2011) p 374

[2] Goris et al., Nature Materials 11 (2012) p 930

[3] Bals et al., Nature Communications 3 (2012) p 897

[4] Jones et al., Nano Letters 14 (2014) p 6336

[5] De Backer et al, Nanoscale 9 (2017) p 8791

[6] Van Aert et al., Physical Review B 87 (2013) 064107

[7] De Backer et al., Ultramicroscopy 134 (2013) p 23

[8] Geuchies et al., Nature Materials 15 (2016) p 1248

The authors gratefully acknowledge funding from the Research Foundation Flanders (FWO, Belgium) through project fundings (G.0369.15N, G.0368.15N, WO.010.16N) and postdoctoral grants to T.A. and A.D.B. S.B. acknowledges financial support from the European Research Council (Starting Grant No. COLOURATOM 335078). Funding from the European Union Seventh Framework Programme under Grant Agreement 312483 - ESTEEM2 (Integrated Infrastructure Initiative-I3) is acknowledged. The authors would also like to thank Luis Liz-Marzán, Marek Grzelczak, and Ana Sánchez-Iglesias for sample provision.

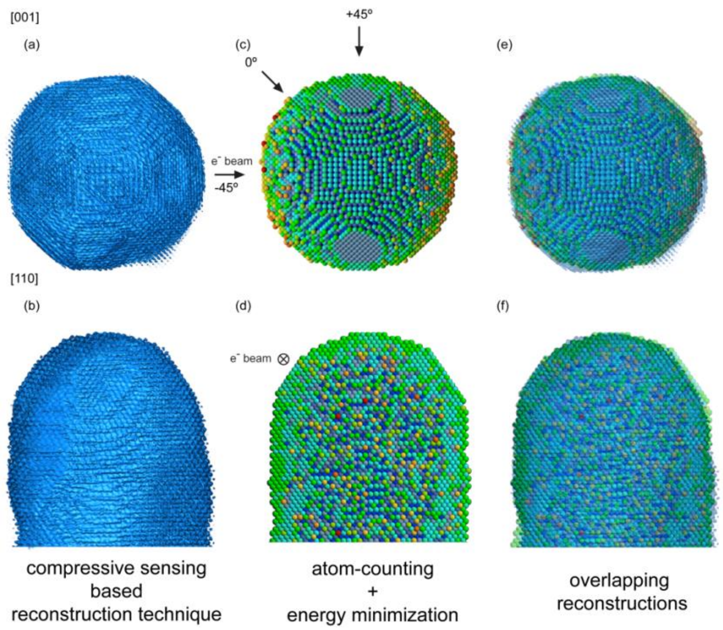


**Figure 1. Comparison of the atom-counting/energy minimization and atomic resolution tomography reconstruction.** (a,b) Compressive sensing based reconstruction of a Au nanorod viewed along [001] and [110] direction. (c,d) Reconstruction based on atom-counting and energy minimization using a single projection image (the coloring of the atoms indicates the nearest-neighbor coordination, from 1 in red to 12 in dark blue). (e,f) Overlap of the reconstructions shown in (a,c) and (b,d).

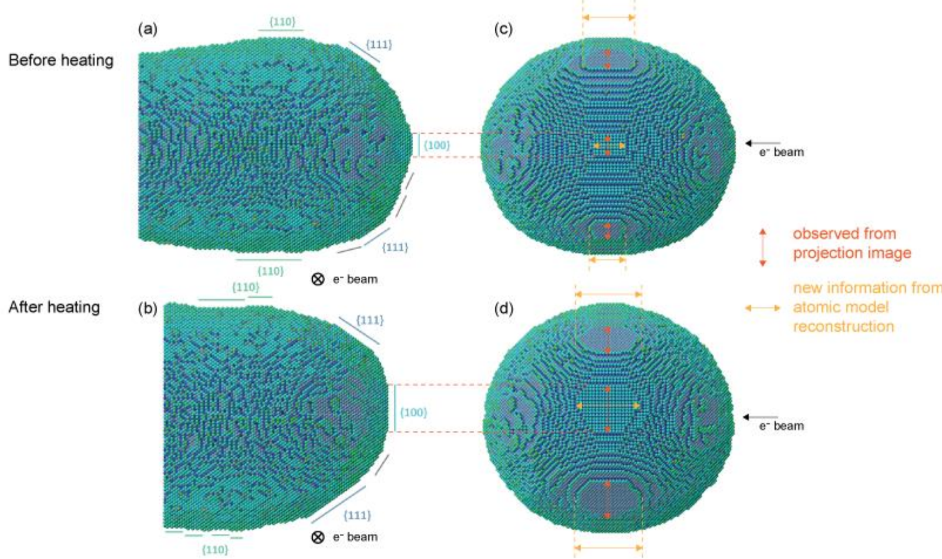


**Figure 2. 3D atomic models before and after heating along different viewing directions.** (a,c) Au dumbbell before heating, (b,d) Au rod after heating. The coloring of the Au atoms determines the nearest neighbor coordination.