Electronic Supplementary Material (ESI) for Nanoscale

SUPPORTING INFORMATION

Electron Tomography and 3D Molecular Simulations of Platinum Nanocrystals

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SI 1: Reconstruction method

The 3D reconstructions were computed from the tilt series of BF-STEM and HAADF-STEM images by using the algebraic reconstruction technique (ART). This algorithm uses an iterative scheme which consists in re-projecting a computed reconstruction along the tilt directions to compare with the initial projections and the result is a difference tomogram. The procedure is successively applied until the convergence is reached and a sufficient signal-to-noise ratio (SNR) is achieved. In our case, no visible improvement was observed after 15 iterations (Figure SI 1A). Compared to the analytical methods such as weighted back-projection (WPB), the benefits of the ART technique are that it provides a reconstruction where the SNR is considerable higher and the missing wedge artefacts are less visible (Figure SI 1B, C).

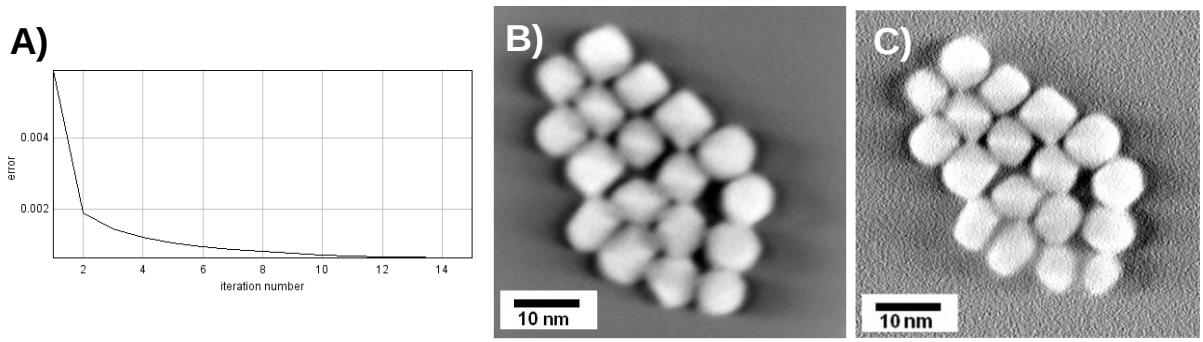

Figure SI1 A) Dependence of the mean absolute error between the initial and re-projected images on the number of iterations. B), C) Cross-sectional views extracted from the tomographic reconstructions of the analyzed group of nanoparticles, obtained by using the ART (B) and WBP (C) reconstruction algorithms.

SI 2: Comparison between the Bright Field (BF) and High Angle Annular Dark Field (HAADF) reconstructions obtained by electron tomography in the STEM mode.

As can be observed on Figure A, the BF and HAADF reconstructions are very similar except for the typical inverted contrast between BF and HAADF. This strong similarity is reinforced by comparing the NCs edges obtained from an image segmentation process based on a threshold sorting procedure of the voxels as a function of their grey-level intensities. Knowing that the two reconstructions are spatially correlated, it can be very useful sometimes to perform a combined analysis, as in the case of inhomogeneous materials made of compounds with high and low atomic numbers elements. Actually, it is well known that the HAADF mode is sensitive rather to high Z elements; in this case, one of the possible solutions for the 3D analysis of specimens made of high and low Z elements is to compensate the lack of sensitivity of HAADF to low Z compounds by the simultaneous analysis of the BF reconstruction.

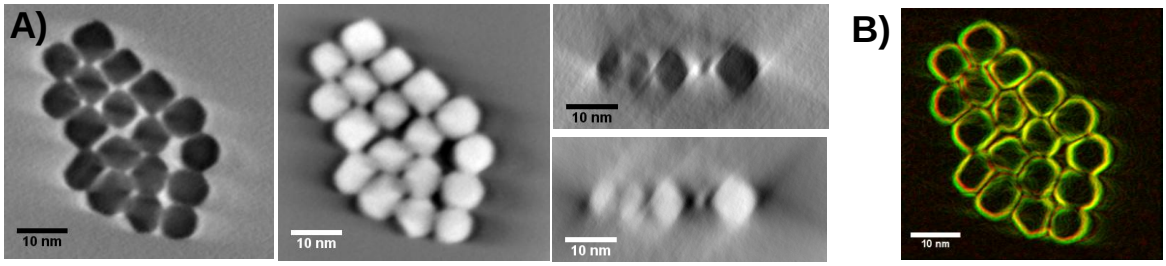

Figure SI2 A) Longitudinal and transversal sections taken at the same depth and orientation through the two spatially correlated BF-STEM and HAADF-STEM reconstructions. B) Left: spatial extensions of the Pt NCs obtained through an edge detection procedure applied to the BF (in red) and HAADF (in green) slices presented in A).

SI 3: Equilibrium shapes of Pt nanocrystals with sizes of 3 and 7 nm, as deduced by molecular dynamics simulations

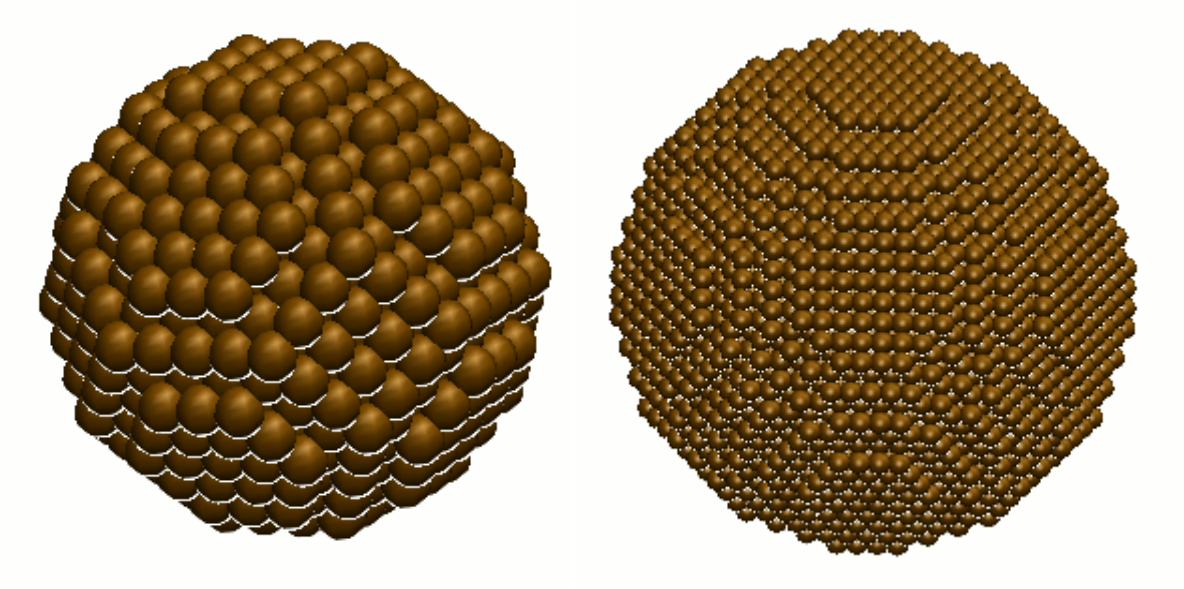

Figure SI3 Two snapshots extracted from molecular dynamic calculations for two Pt nanoparticles with size of 3 nm (left) and 7 nm (right). A spherical shape was used as initial condition for both nanoparticles. The computing interval was 6 ns and the annealing temperature 1700 K. The resulting 3D atomic arrangements show that the equilibrium shapes at this temperature are close to a truncated octahedron.

SI 4: Direct evidence of the improvement of the signal-to-noise ratio (SNR) in a tomographic reconstruction compared to the individual images.

Figure A shows some typical initial projections from the BF tilt series acquired in the STEM mode on a 5 nm-sized nanoparticle Pt assembly, with the equivalent projections computed from the reconstructed volume. Of course, the SNR is better in the calculated projections, thanks to the redundancy of the information coming from a series of projections of the object.[i] This can be specifically analyzed through the corresponding line profiles drawn on two typical equivalent projections (Figure B).

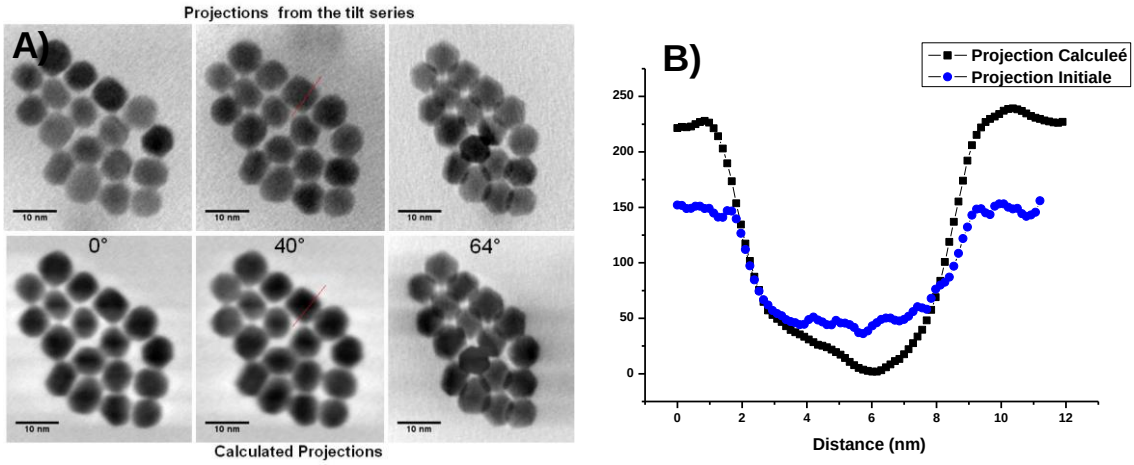

Figure SI4 A) Some typical initial projections (at 0°, 40° and 60° tilt angle) of a monolayer sample, compared to the corresponding projections obtained by projecting the reconstruction volume along the same directions. B) Intensity profiles corresponding to the red lines.

However, it is worth to note that, though the SNR is higher in the re-projections of the tomogram, these computed projections are not necessarily more reliable than the initial projections. The reason is the presence of the missing wedge which yields to a rather bad definition of the object along the electron beam direction. As a consequence, the benefit of considering in the analysis the computed projections instead of the initial 2D images is justified especially for the projections that are calculated along a direction close to the electron beam axis.

[i] Hart, R.G., Electron microscopy of unstained biological material: the polytropic montage, *Science* **1968**, 159, 1464–1467.