BIO Web of Conferences **129**, 02014 (2024) https://doi.org/10.1051/bioconf/202412902014
EMC 2024

Momentum-resolved STEM Tomography of Gold-Silver core-shell Nanoparticles

Sebastian Sturm[1], Dr. Felizitas Kirner[2], Prof. Dr. Elena Sturm[2], Prof. Dr. Knut Müller-Caspary[1]

[1]Ludwig-Maximilians-Universität München, Department of Chemistry, Munich, Germany, [2]Ludwig-Maximilians-Universität München, Department of Earth and Environmental Sciences, Munich, Germany

Background

Precisely tailored Gold-Silver core-shell (Ag@Au) Nanoparticles (NPs) can have outstanding optical properties due to localized surface plasmon resonances, making them suitable candidates for advanced senors or for application in photothermal therapy [1]. Exact control over the nanocrystal growth however, demands for suitable analytic methods to confirm the quality and reproducibility of the synthesis. Facetted core-shell NPs like this especially benefit from techniques like Electron Tomography and STEM Tomography in particular, which are able to reconstruct their nanoscopic structures in all three dimensions, via acquisition of a tilt series of projections [2].

Depending on the detectors used, STEM offers a great variety of suitable signals for these projections [3]. For example the commonly used Bright-field (BF), annular dark-field (ADF) and high-angle annular dark-field (HAADF) signals each cover rings with different radii, corresponding to certain magnitudes in momentum space. However, the exact radii of the detector in momentum space additionally vary with the chosen camera length and the choice of those radii in the experiment has significant impact on the fidelity of the tomographic reconstruction, since most techniques rely on the linearity of the signal during projection and might even have different optima for detecting different materials. To understand the impact of the detector choice and to overcome its limitations, Momentum-resolved STEM (MR-STEM or 4D-STEM) has been applied, where for each scan position of a 2D scan raster an individual 2D diffraction pattern is recorded with the help of a fast direct electron detector and signals for each magnitude of momentum have been extracted from this data set. This allows to apply and compare arbitrary virtual detectors post-experimentally. In combination this enables MR-STEM Tomography, where the best suitable signal with respect to its performance in tomographic reconstruction of the Ag@Au NPs can be determined in hindsight.

Methods

The MR STEM experiments were conducted using a probe corrected FEI Titan Themis operated at 300kV and equipped with a Quantum Detectors MerlinEM direct electron detector (based on Medipix 3).
Acquisition of the 4D tilt series has been done using an in-house developed software written in python, utilizing the FEI Scripting COM interface. A 4D tilt series with a tilt range of -75° .. +72.5° in 2.5° steps has been recorded with 256 x 256 scan points and 256 x 256 pixel diffraction patterns at an semi-convergence angle of 18mrad. For reconstruction, a WSIRT reconstruction algorithm has been used with 5 to 10 iterations.

Results

The performance of different detector radii in momentum space in regards of tomographic reconstruction has been investigated on reconstructions of selected slices. Additionally an magnitude of momentum resolved WSIRT reconstruction of the whole volume has been explored, leading to a 4D dataset (x, y, z,|k|), which can then be evaluated e.g. using Principal component analysis (PCA). Furthermore the second moment in momentum space has been calculated, where ring segments with radius k have been weighted with k^2, summing up from a chosen minimum radius to a cut-of radius. Figure 1 exemplary shows the reconstructed Second Moment tomogram (c) of four self-assembled Ag@Au NPs (b), clearly depicting the octahedrally faceted Au cores encapsulated by truncated cubically faceted Ag shells.

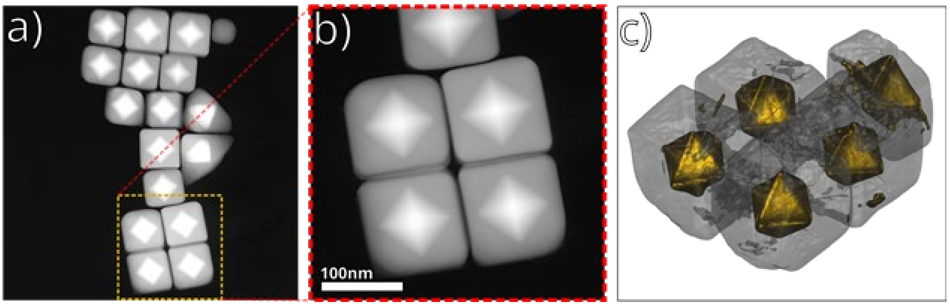
Conclusion

Momentum-resolved STEM has been explored to find the most suitable signal for STEM Tomography, comparing different ADF / HAADF signals. For the Ag@Au NPs, the second moment has proven to quickly converge to similar results as the HAADF signal reconstructions, if the outer detector radius is chosen large enough, however the less strict dependence on the inner radius might still be advantageous for some applications.

Figure 1:
Momentum Resolved STEM Tomography of Gold-Silver Core-Shell Nanoparticles:

a) HAADF STEM overview image
b) HAADF STEM image of the region of interest, highlighted in (a)
c) Second moment tomogram as an example of momentum resolved STEM tomography.

Graphic:



Keywords:

STEM Tomography, 4D STEM

Reference:

[1] J SCHULTZ, et al., Adv. Optical Mater. 2021
[2] M WEYLAND, et al. Materials Today, 2004,
[3] H ROBERT, et al. Ultramicroscopy, 2022,