Supporting Information

Exploring the Benefits of Electron Tomography to Characterize the Precise Morphology of Core Shell Au@Ag Nanoparticles and its Implications for their Plasmonic Properties

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Experimental details: Segmentation from the reconstructed volume

Segmentation was performed to decompose the tomogram into its structural components (gold and silver components) by identifying the sets of voxels (3D pixels) that constitute them. A manual segmentation was undertaken, which consisted of assigning the structural features using visualization tools. In our case, the relatively high contrast of the HAADF-STEM images, in which Au is more intense (Z=79) than Ag (Z=47), allowed us to apply a simple density threshold and manual tracing. The obtained surface optimally matches the boundaries of the reconstructed orthogonal slices (orthoslices), where the resolution is highest.

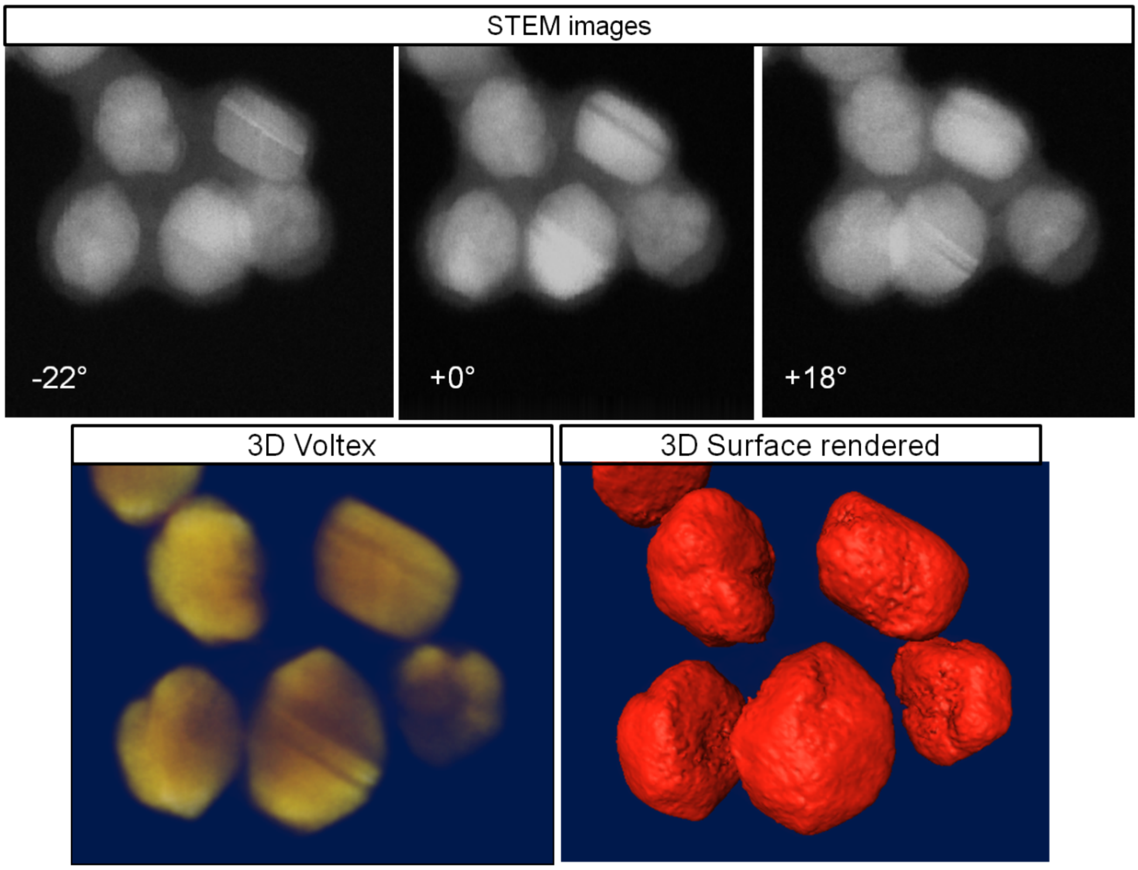

Figure S1. *Top row*: HAADF images of Au@Ag core-shell nanoparticles showing different contrast at different specimen tilt angles, acquired using an inner detector semi-angle of 35 mrad. In these images Au-core nanoparticles exhibit different contrast at different specimen tilt angles: dark lines across the particles arise from linear twin domains. *Bottom row*: 3D Voltex and Surface rendered representations for the Au cores. The twin boundaries remain visible on the 3D reconstructions, as it can be noticed from the 3DVoltex visualization, but the outer morphology quality, a parameter addressed in the presented work, is not affected.

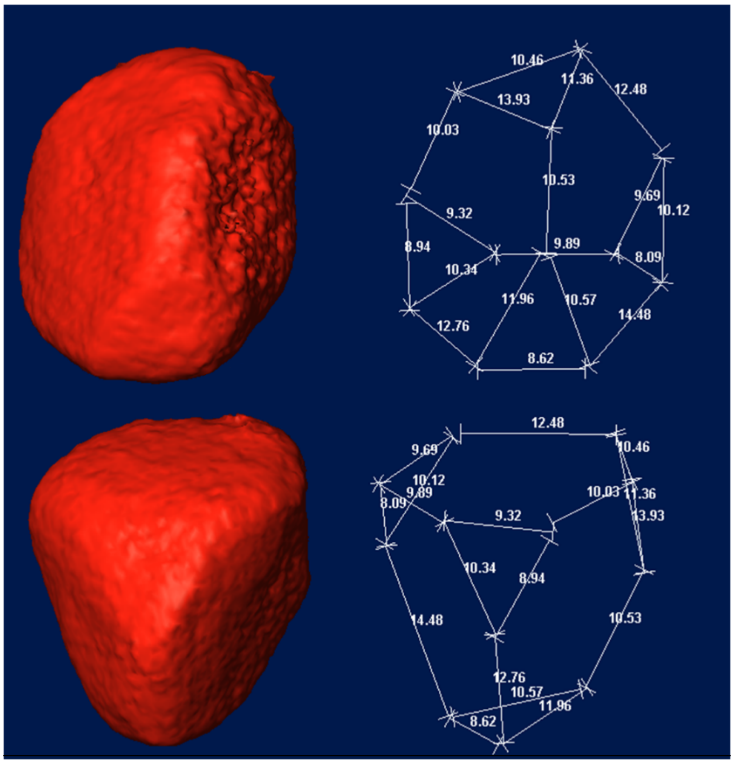

Figure S2. Nanometrology of the anisotropic Au NP core, denoted as reconstructed irregular polyhedral core (RIPC) within the text, obtained from the HAADF-STEM electron tomography analysis used as input for the DDA electrodynamics simulations.

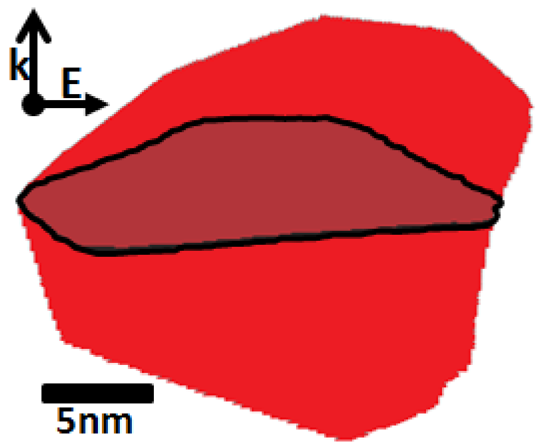

Figure S3. Schematic view of the plane where the electric field components and the field enhancement were calculated in the anisotropic Au NP and Ag@Au NP with an average Ag shell thickness of 2.3 nm.