

## Supporting Information

### Spectroscopy, Imaging and Modeling of Individual Gold Decahedra

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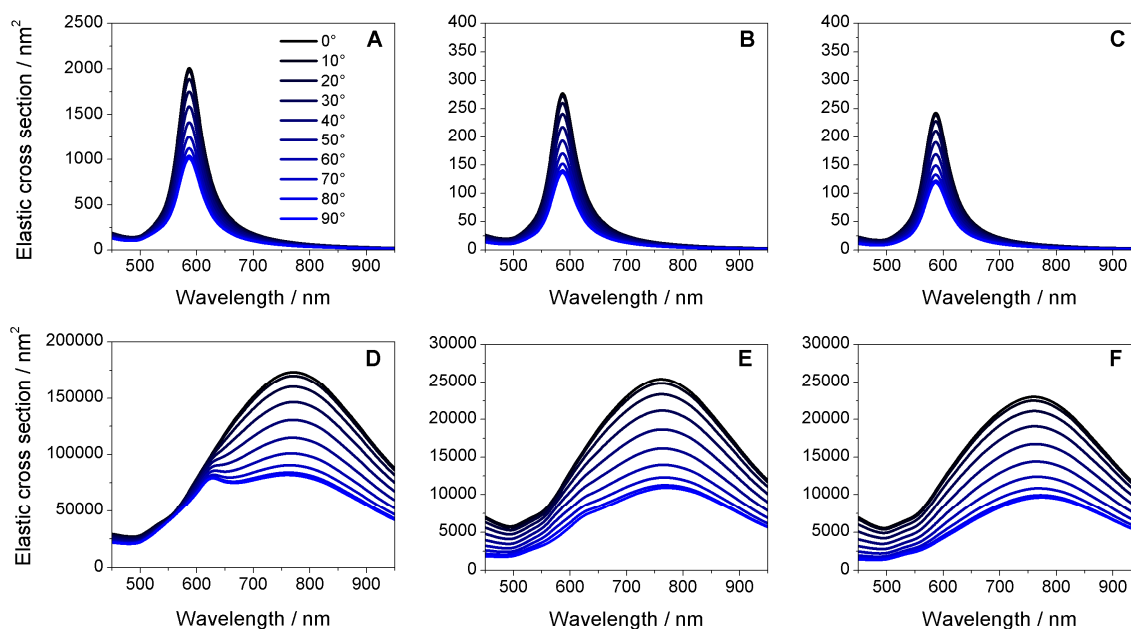
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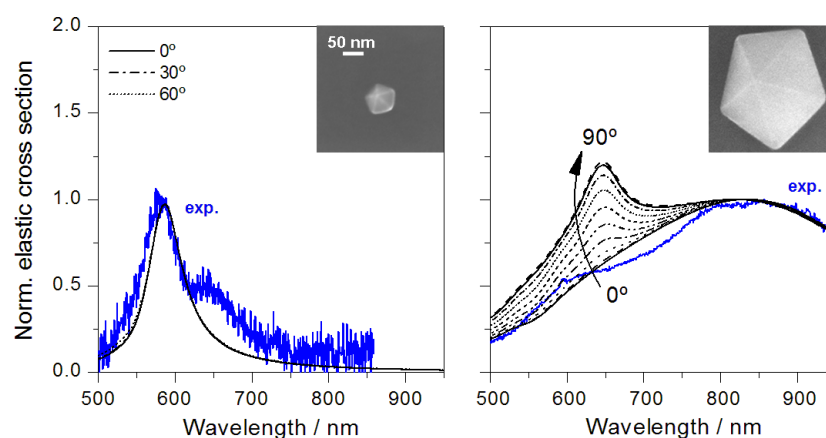
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**Figure S1.** Calculated (BEM-3D) elastic scattering of a 44 nm (A-C) and a 150 nm (D-F) gold decahedron. The elastic scattering was averaged over all scattering directions (A and D); over the collection angles of our DFM setup (B and E) or at the scattering angle determined by the central direction of collection angles in the DFM (C and F). The particles are oriented on the

substrate as shown in Fig. 2B. The angles of the incident light are indicated in the legend. As can be observed, integration over all scattering directions vs. integration over the actual collection angles in our DFM setup increases the magnitude of the elastic cross section for both sizes. However, it does not produce any further change in the spectrum of the smaller decahedron, although it does cause a slight blue shift/red shift (depending on the angle of the incident light) and a decrease in the relative quadrupole intensity, in the case of the larger particle. This effect is more dramatic when the elastic cross section is acquired at a single angle determined by the central direction of collection angles in the dark-field microscope.



**Figure S2.** Calculated (BEM-3D) angle-dependent scattering spectra of the decahedra shown in the SEM micrographs (experimental spectrum in blue). The side lengths are: 44 nm (left) and 170 nm (right). The angle of the incident light is indicated in the legend. All the spectra are normalized at their respective azimuthal dipole wavelength.