

Electronic Supplementary Information

Detecting the Spatial Rearrangement of Individual Gold Nanoparticle Heterodimers

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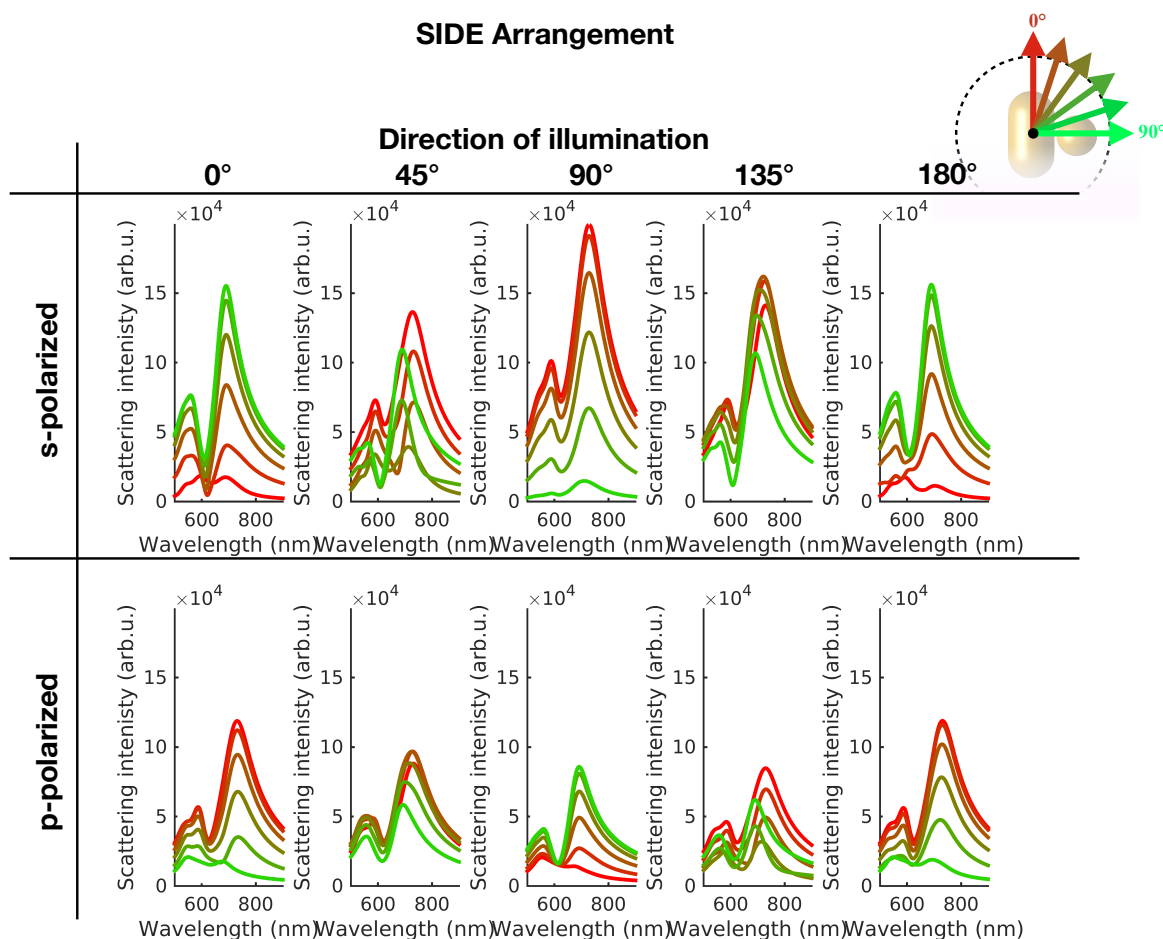


Figure S1: Simulated polarization resolved spectra for the side-arrangement. Line color changes with analyzer angle from red (0°) to green (90°). L(rod) = 123 nm; D(rod) = 66 nm; D(particle) = 58 nm; Relative displacement = 0.012; Gap between particles = 1.5 nm; Angle of incidence = 57.5°.

TOP Arrangement

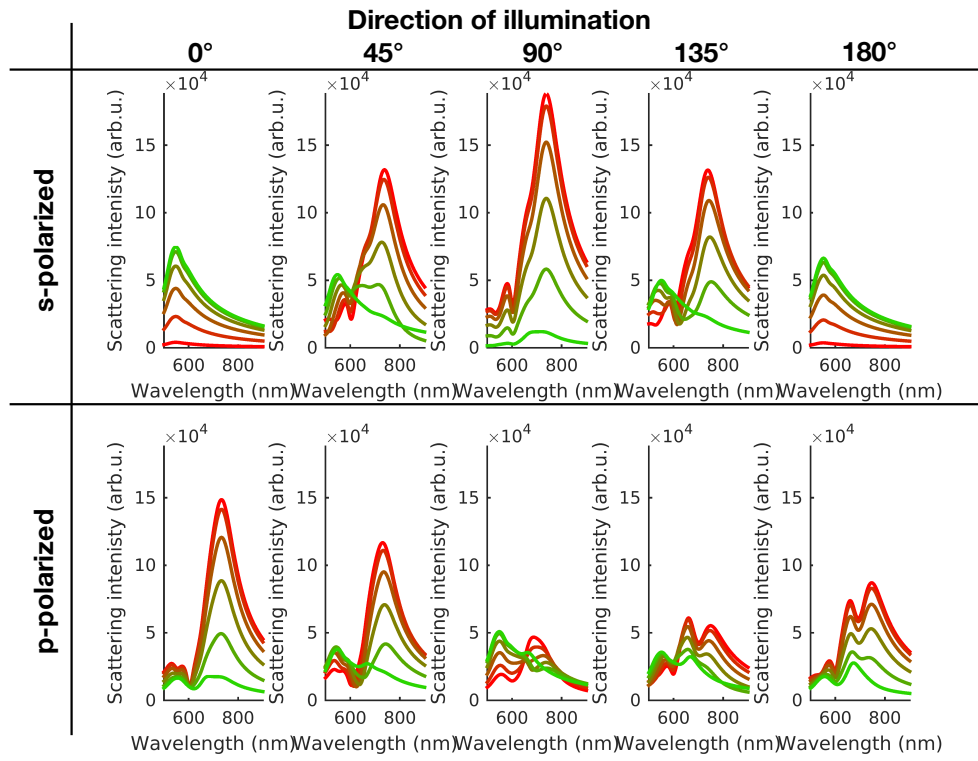


Figure S2: Simulated polarization resolved spectra for the top-arrangement. Line color changes with analyzer angle from red (0°) to green (90°). $L(\text{rod}) = 122 \text{ nm}$; $D(\text{rod}) = 64 \text{ nm}$; $D(\text{particle}) = 47 \text{ nm}$; Relative displacement = 0.112; Gap between particles = 1.5 nm; Angle of incidence = 57.5° .

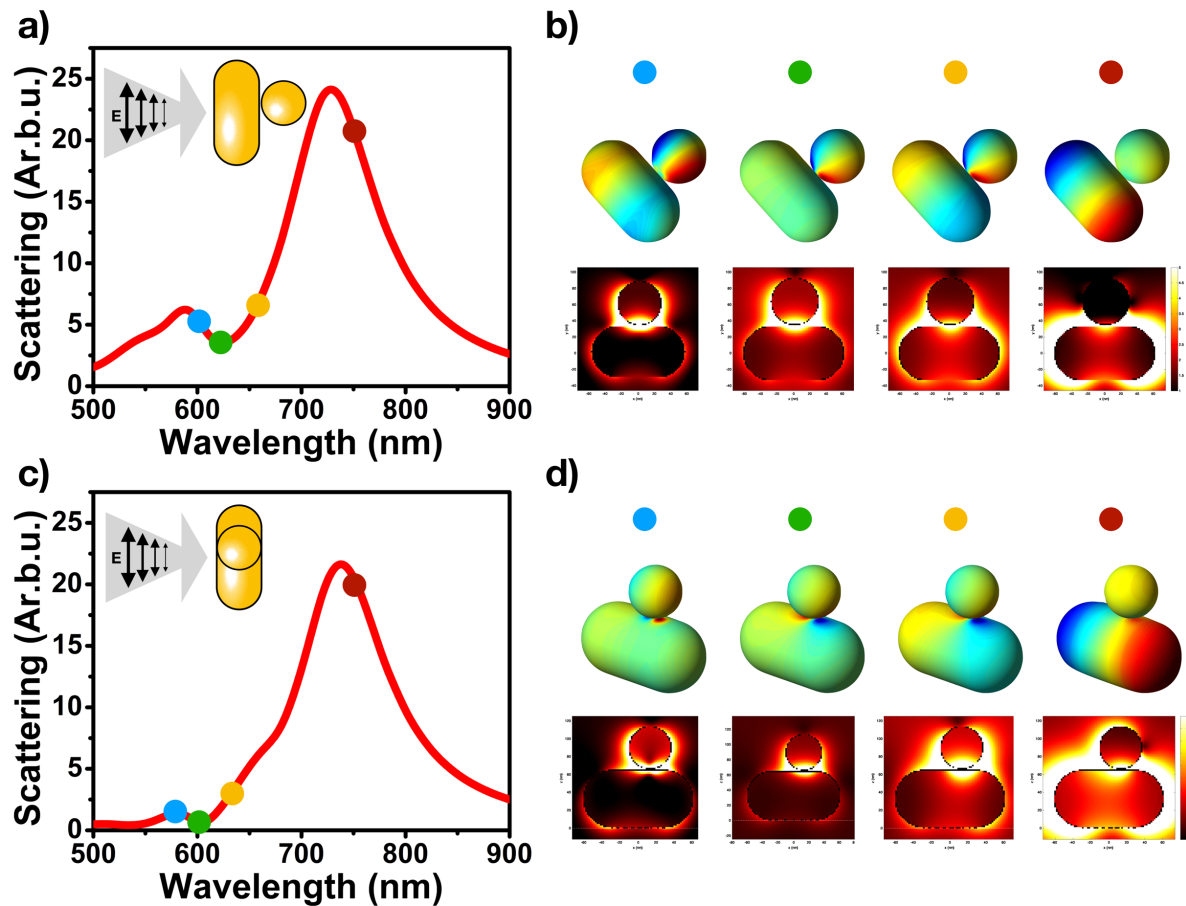


Figure S3: Simulated scattering spectrum (a), phase information (surface charge plots) and near-field maps (b) calculated for the side arrangement at different wavelengths marked by the coloured dot. Same for the top-arrangement (c-d). Excitation is longitudinal. The phase information clearly shows that above the resonance of the nanorod (red dot), the phases on the two particles are matched. On the blue side of the rod longitudinal resonance (orange dot), however, the phase on the rod becomes the opposite, leading to a diminishing dipole at the dip (green dot). At the blue side of the dip (blue dot), the interaction of the sphere dipole with a rod quadrupolar mode, on the red side a sphere-dipole, rod-dipole interaction can be anticipated. The near-field cut planes are located the center of the structure in the XY and the XZ planes for the side and top-arrangements, respectively.

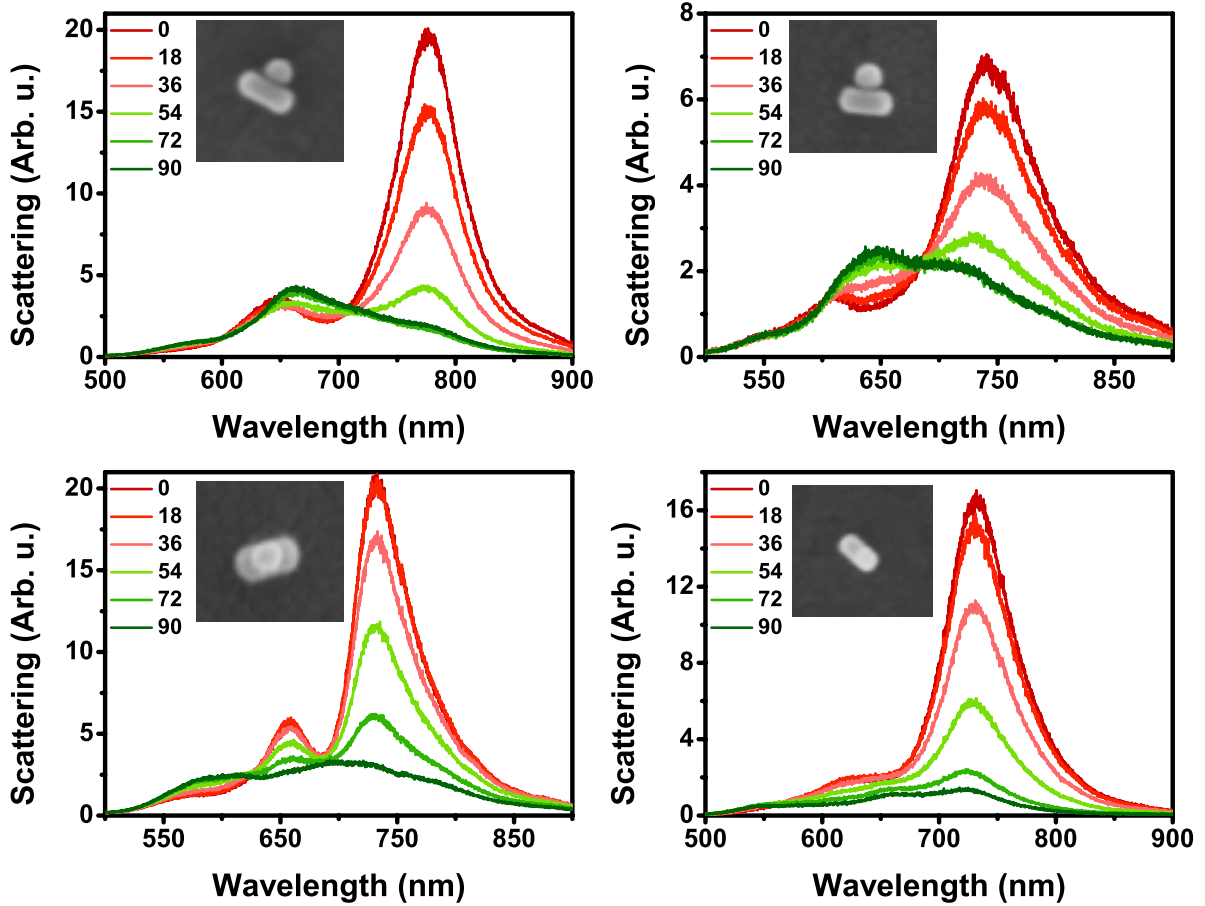


Figure S4: Additional measured scattering spectra of heterodimers showing side (top row) and top-arrangement (bottom row) at different analyser angles. The insets show the SEM image of the corresponding heterodimers.

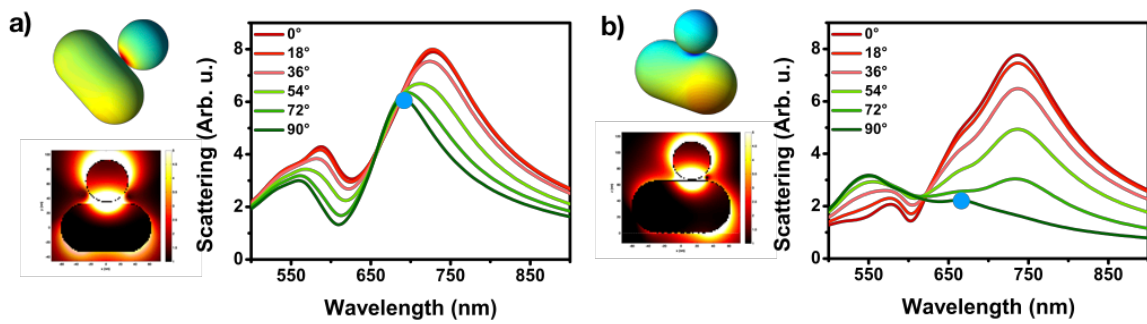


Figure S5: Surface charge density and near-field maps for the side (a) and top-arrangements (b) calculated at the spectral positions marked by the blue dot

HD1 - SIDE Arrangement

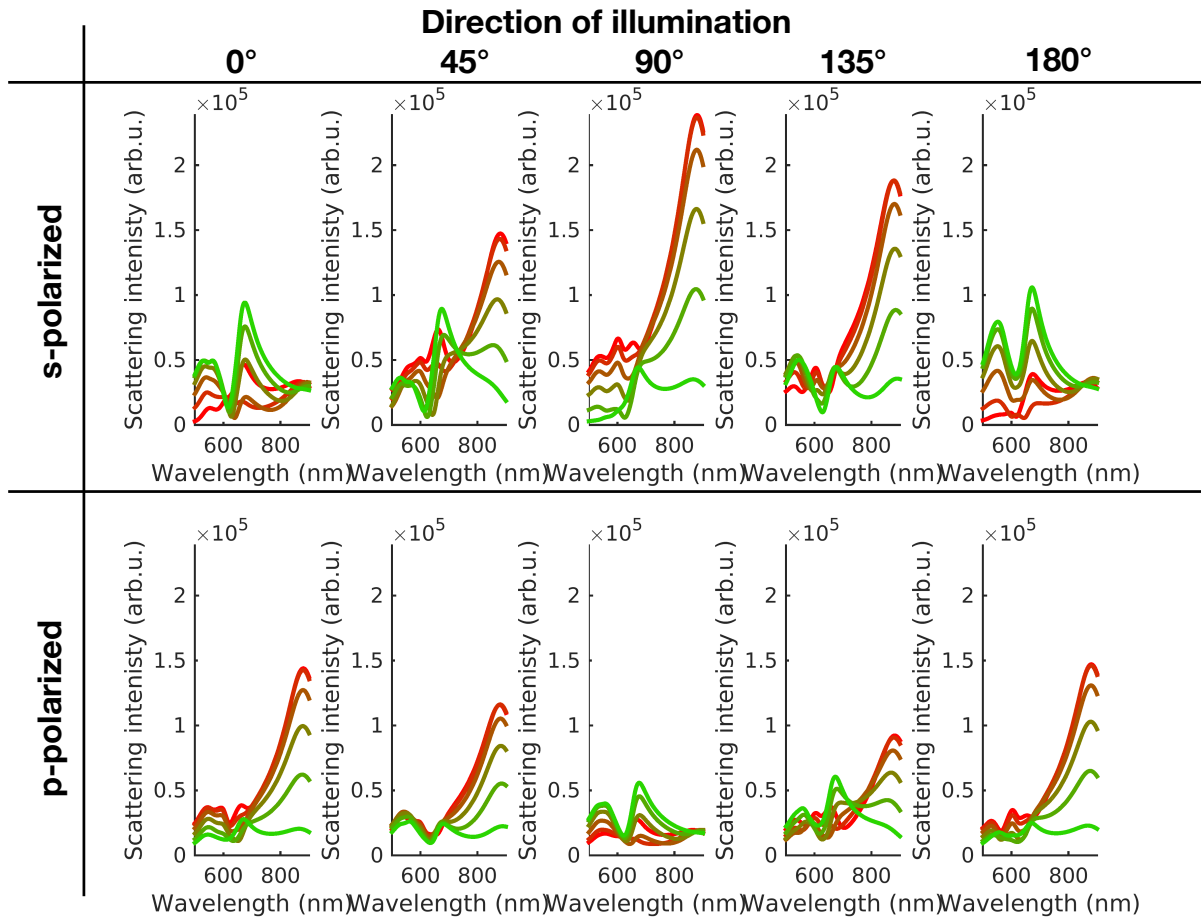


Figure S6: Simulated spectra of HD1. Line color changes with analyzer angle from red (0°) to green (90°). Geometric parameters: $L(\text{rod}) = 150 \text{ nm}$; $D(\text{rod}) = 56 \text{ nm}$; $D(\text{particle}) = 49 \text{ nm}$; Relative displacement = 0.236; Gap between particles = 1.5 nm, Gap to substrate = 1 nm.

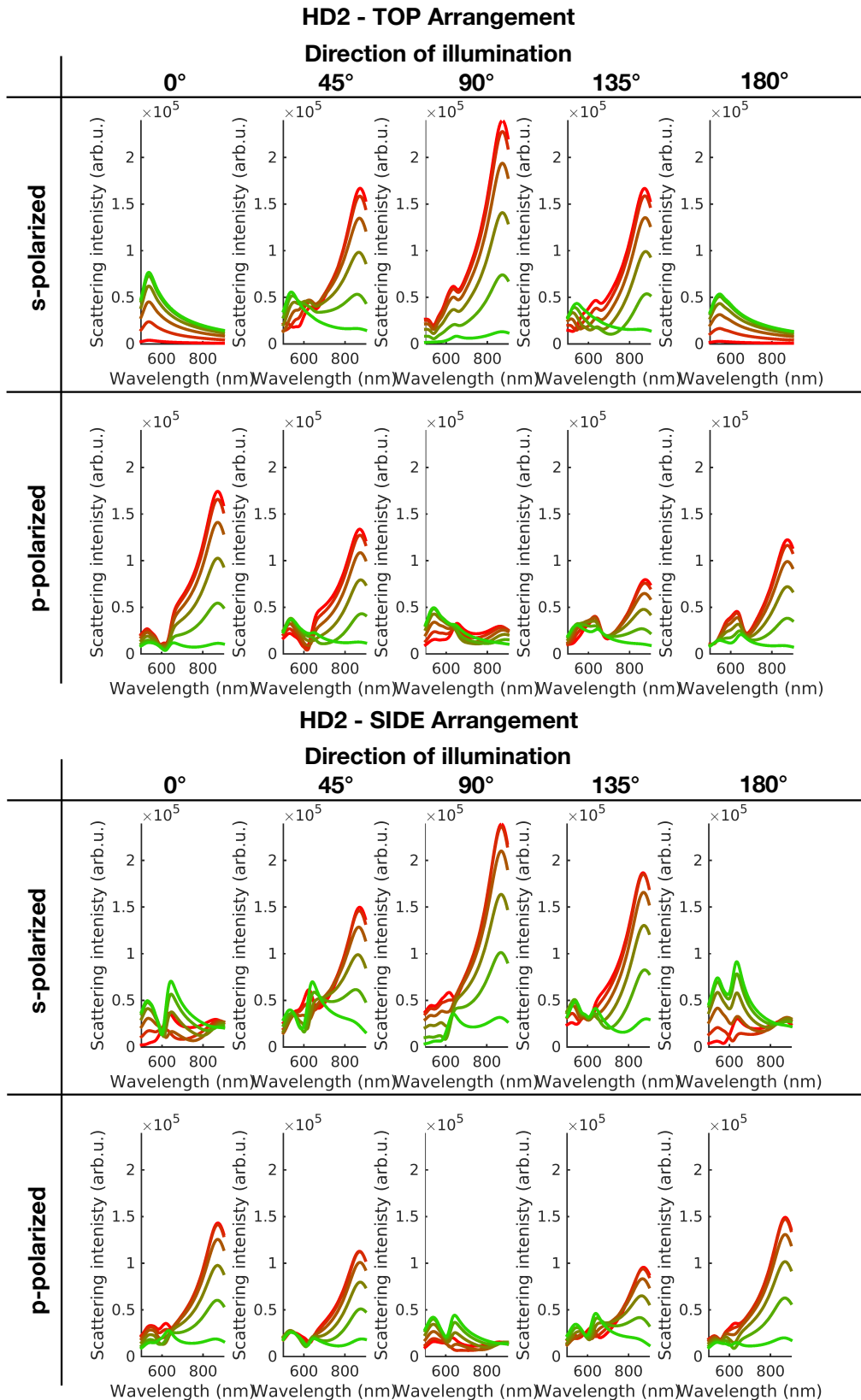


Figure S7: Simulated spectra of HD2. For the top-arrangement the relative particle displacement was set to be the same as for the side-arrangement. Line color changes with analyzer angle form red (0°) to green (90°). Geometric parameters: $L(\text{rod}) = 147 \text{ nm}$; $D(\text{rod}) = 56 \text{ nm}$; $D(\text{particle}) = 43 \text{ nm}$; Relative displacement = 0.411; Gap between particles = 1.5 nm; Gap to substrate: 1 nm.