

Edge mode coupling within a plasmonic nanoparticle - Supporting Information

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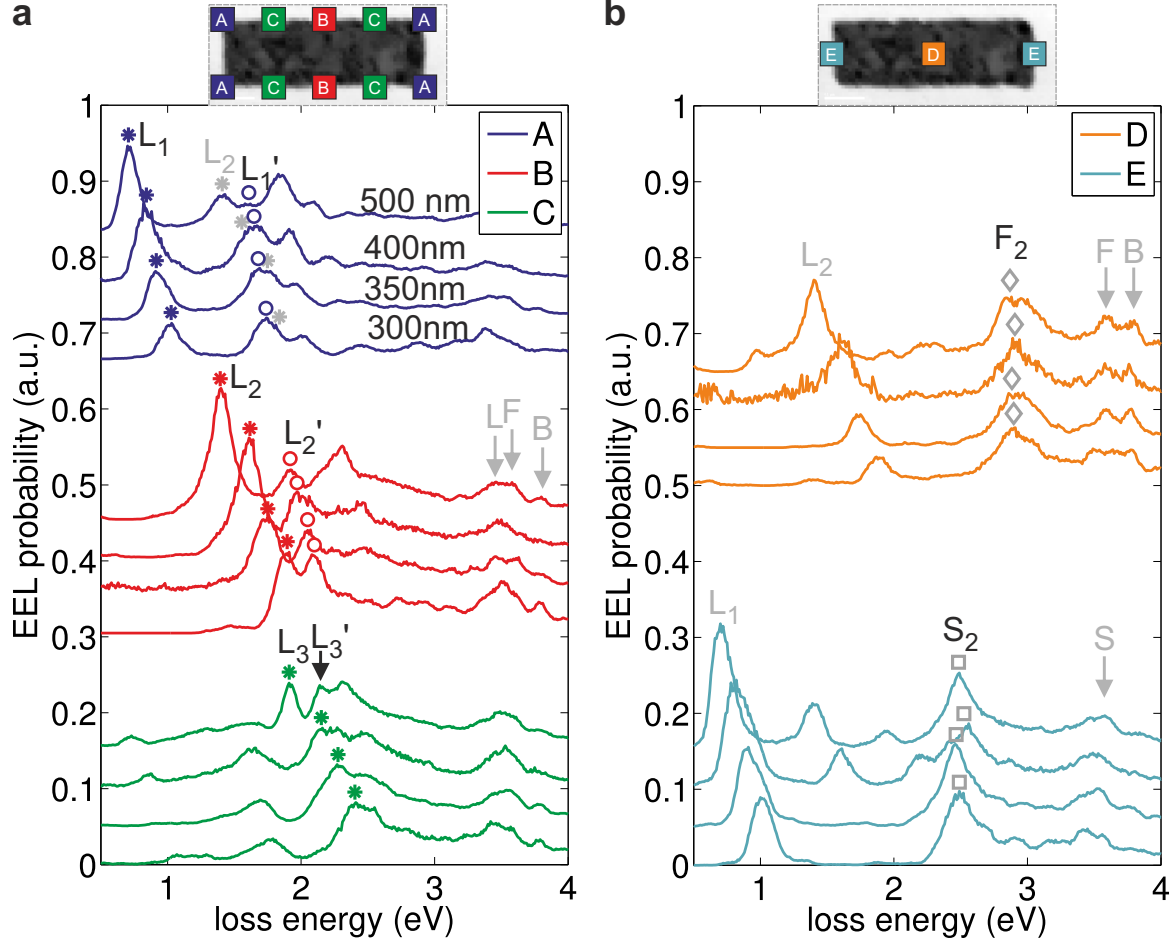
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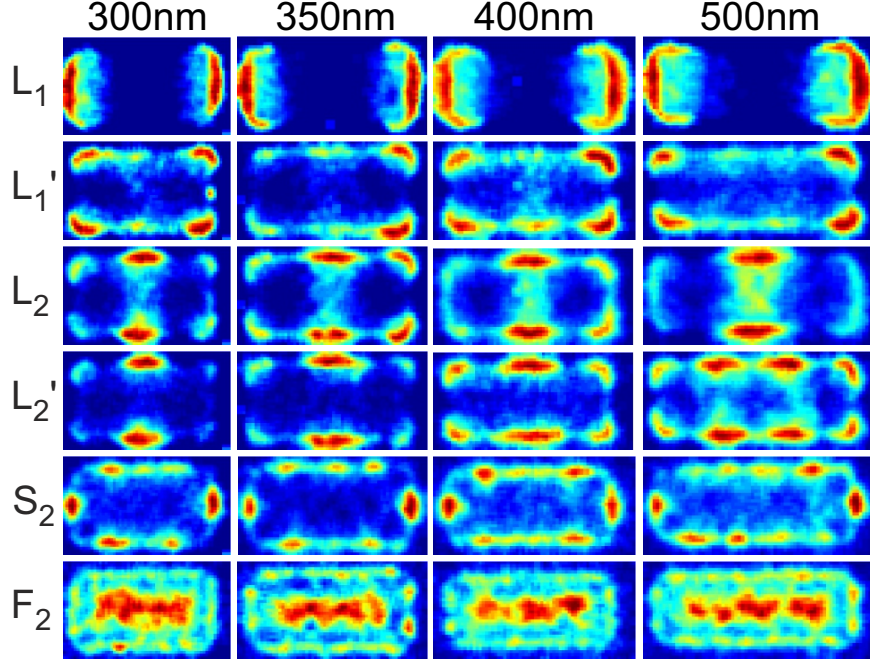
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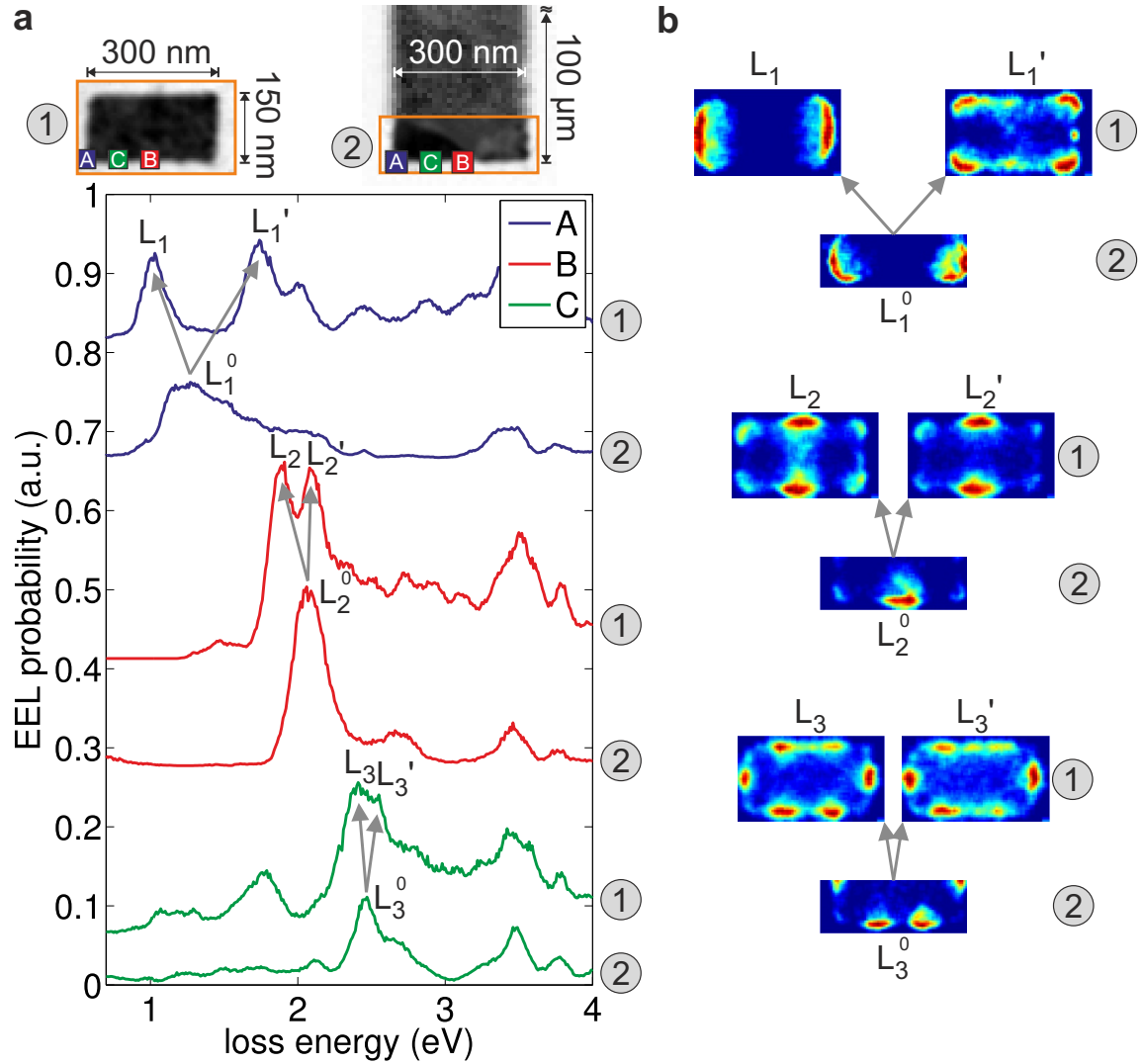
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Supplementary Figure 1: Deconvolved EEL spectra of edge and film modes for cuboids with lengths of 300, 350, 400 and 500 nm (compare Figure 1 in the main text). (a) Spectra taken along the long cuboid edge, the respective areas are indicated by the letters in the inset electron micrograph (data from like letters are averaged). L_x and L'_x ($x=1,2,3$) are the edge modes along the long cuboid axis of first to third order, L is the corresponding asymptotic edge mode. (b) Spectra taken along the short edge and from the cuboid center. S_2 is the second order edge mode along the short cuboid axis, F_2 is the second order film mode along the short cuboid axis and S is the corresponding asymptotic edge mode. In both panels, the EEL peaks are indicated by the same symbols as used in Figure 4 in the main text. F is the asymptotic film mode and B is the bulk plasmon mode.



Supplementary Figure 2: Measured EEL maps for cuboids with lengths of 300, 350, 400 and 500 nm. EEL maps for edge modes L_1 , L'_1 , L_2 and L'_2 along the long cuboid axes and edge and film modes S_2 and F_2 along the short cuboid axis. The center loss energies of the used 100 meV energy window correspond to the peak energies in Figure 4 in the main text and in Supplementary Figure 1, with the exception of the L'_1 maps for the cuboid lengths 350, 400 and 500 nm. In these cases, the maps are generated at 0.1 eV lower (350 nm), 0.15 eV higher (400 nm) and 0.2 eV higher energies (500 nm) than the corresponding peak energies (compare Supplementary Figure 1). This is done to separate the spectrally close mode patterns of the L'_1 and L_2 modes. We note that this was not possible for the L'_2 map for the cuboid with a length of 500 nm, due to the identical peak energy of the L_3 mode that dominates the observed pattern. However, the locally extracted spectrum (top red curve in Supplementary Figure 1) strongly hints towards the existence of the specific L'_2 mode. The distinct EEL map patterns for each mode, independent of the cuboid length, corroborate our mode assignments.



Supplementary Figure 3: Comparison of coupled and uncoupled plasmon modes along a 300 nm edge. (a) Deconvolved EEL spectra taken along the long cuboid edge with a length of 300 nm, the respective areas are indicated by the letters in the inset electron micrograph (1), highlighting the mode splitting of the edge modes L_1^0 - L_3^0 into L_1 - L_3 and L_1' - L_3' (gray arrows) due to plasmon coupling of the opposing edges. These spectra are compared to deconvolved EEL spectra taken along an edge of similar length but much larger width (100 μ m instead of 150 nm), thus hindering plasmon coupling and therefore mode splitting (2). (b) EEL maps extracted at loss energies corresponding to modes L_1 - L_3 and L_1' - L_3' (1) and L_1^0 - L_3^0 (2) respectively, as given by the highlighted peaks in (a). The energy window of each EEL map is 100 meV.