Supporting Inofrmation

Optical Characterization of Gold Nanoblock Dimers: From Capacitive Coupling to Charge Transfer Plasmons and Rod Modes

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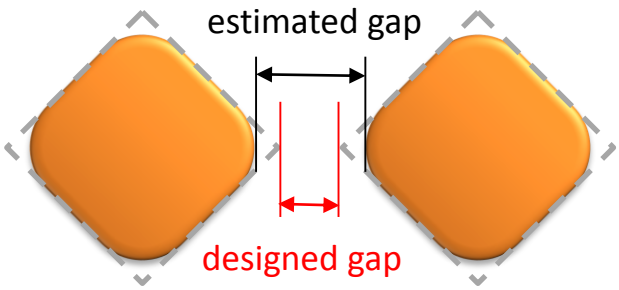

Figure S1. Designed and estimated geometries of nanoblock dimers. While the nanoblock dimers were designed as the grey dashed outlines indicate with a designed gap as given by the red arrow, the corners of the fabricated structures (yellow blocks) were actually round due to the EBL process. We estimated a physical gap size as given by the black arrow based on comparison with simulations. A comparison between designed and estimated gap sizes is in Table S1.

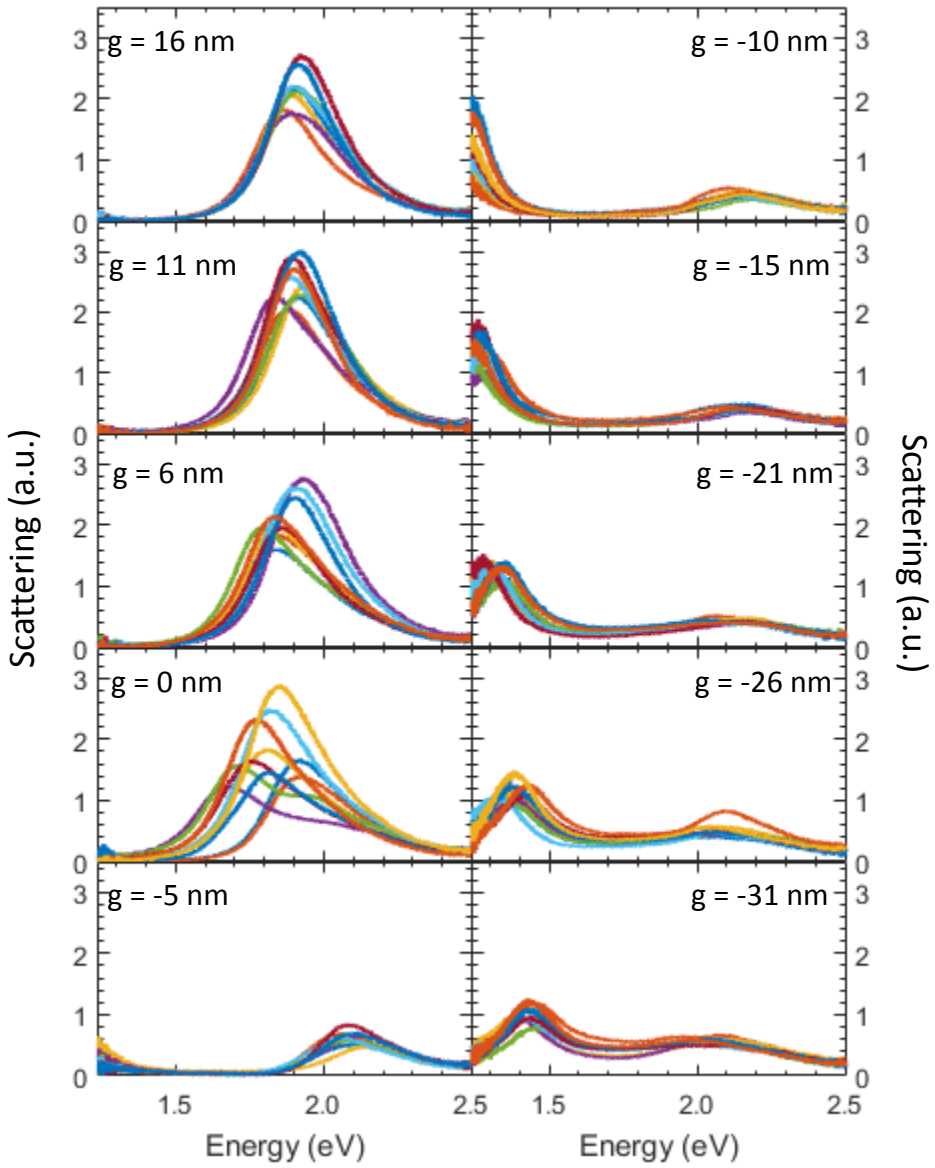

Figure S2. Single-particle scattering spectra of square nanoblock dimers with longitudinal polarization. The edge length of the nanoblock was 80 nm and the gap sizes (*g*) are indicated in each panel. The large heterogeneity among the individual spectra with a gap size of 0 nm is indicative of a significant structural variety of the gap size and geometry.

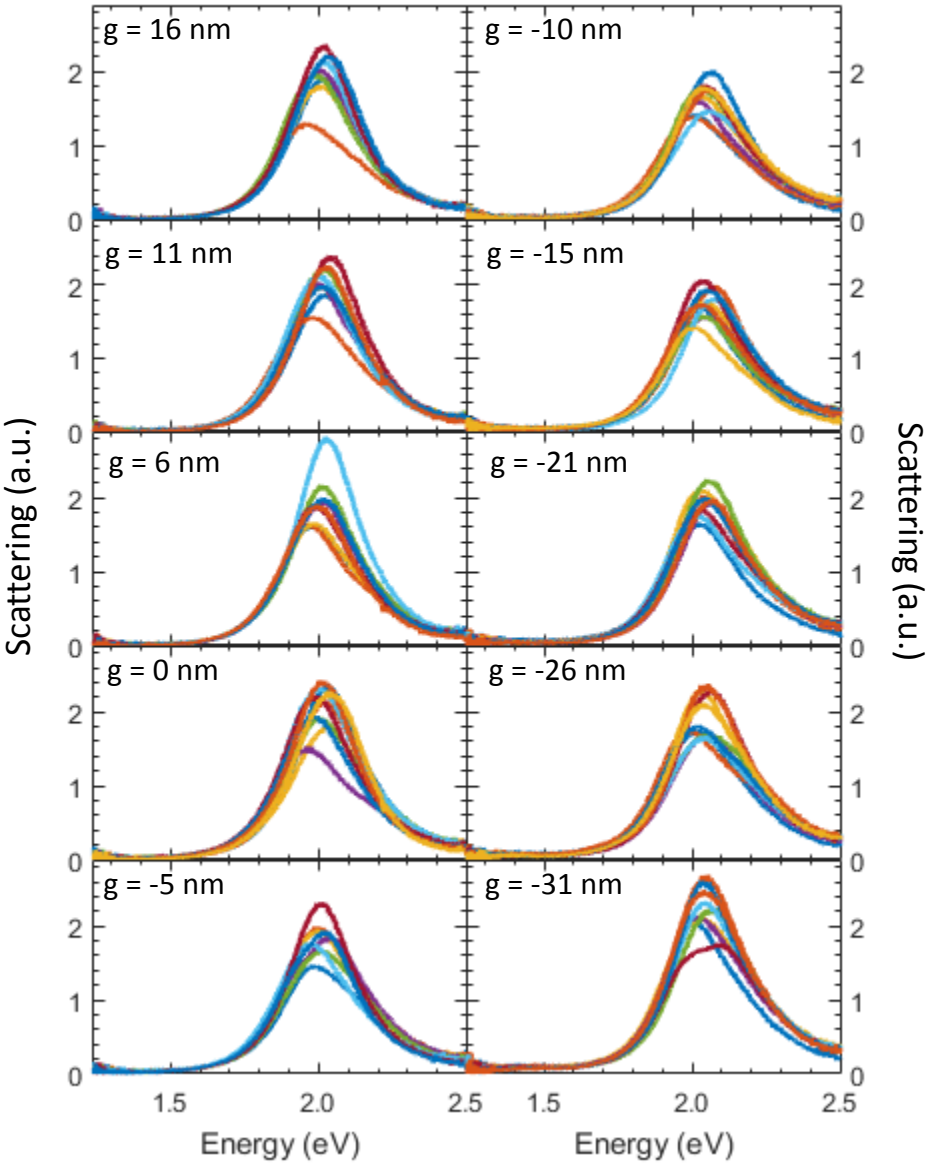

Figure S3. Single-particle scattering spectra of square nanoblock dimers with transverse polarization. The edge length of the nanoblock was 80 nm and the gap sizes (*g*) are indicated in each panel.

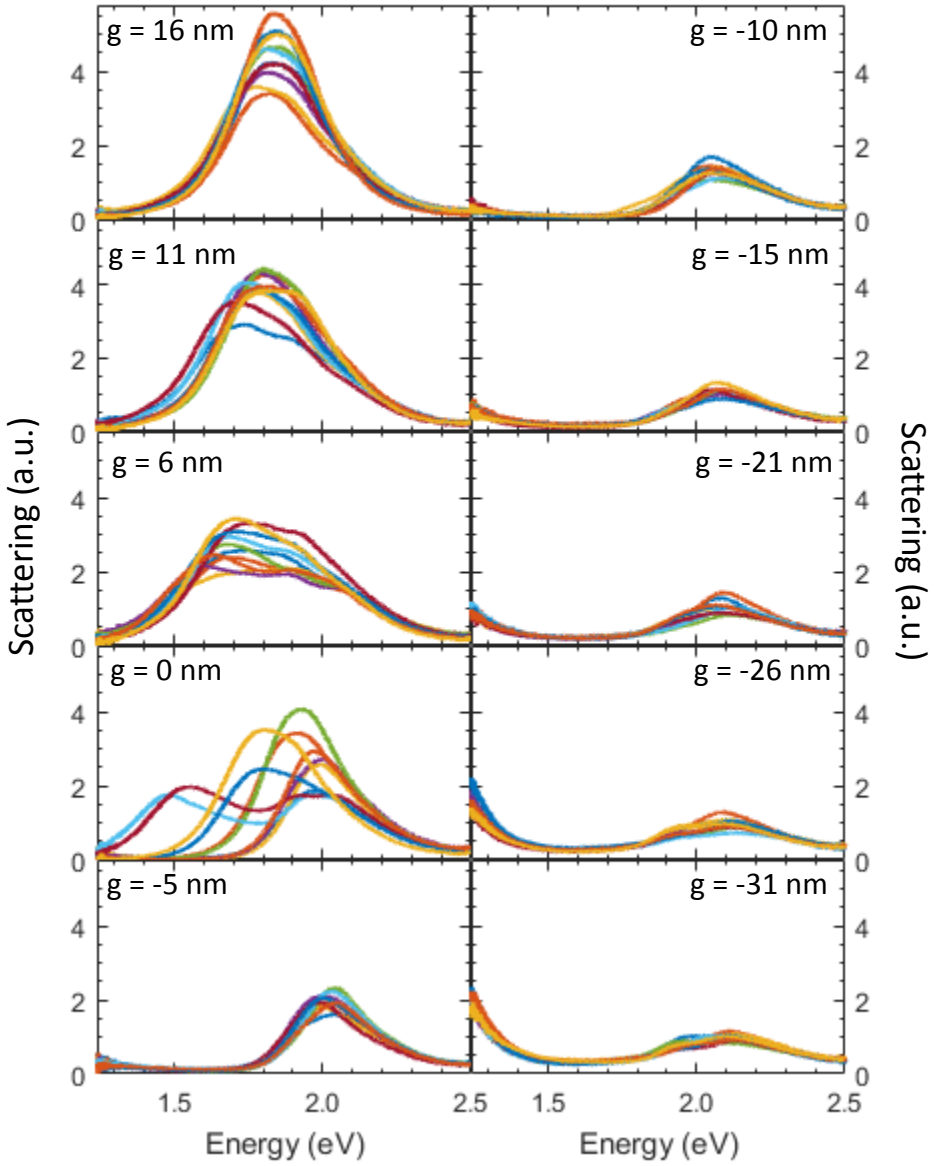

Figure S4. Single-particle scattering spectra of square nanoblock dimers with longitudinal polarization. The edge length of the nanoblock was 100 nm and the gap sizes (*g*) are indicated in each panel. The large heterogeneity among the individual spectra with a gap size of 0 nm is indicative of a significant structural variety of the gap size and geometry.

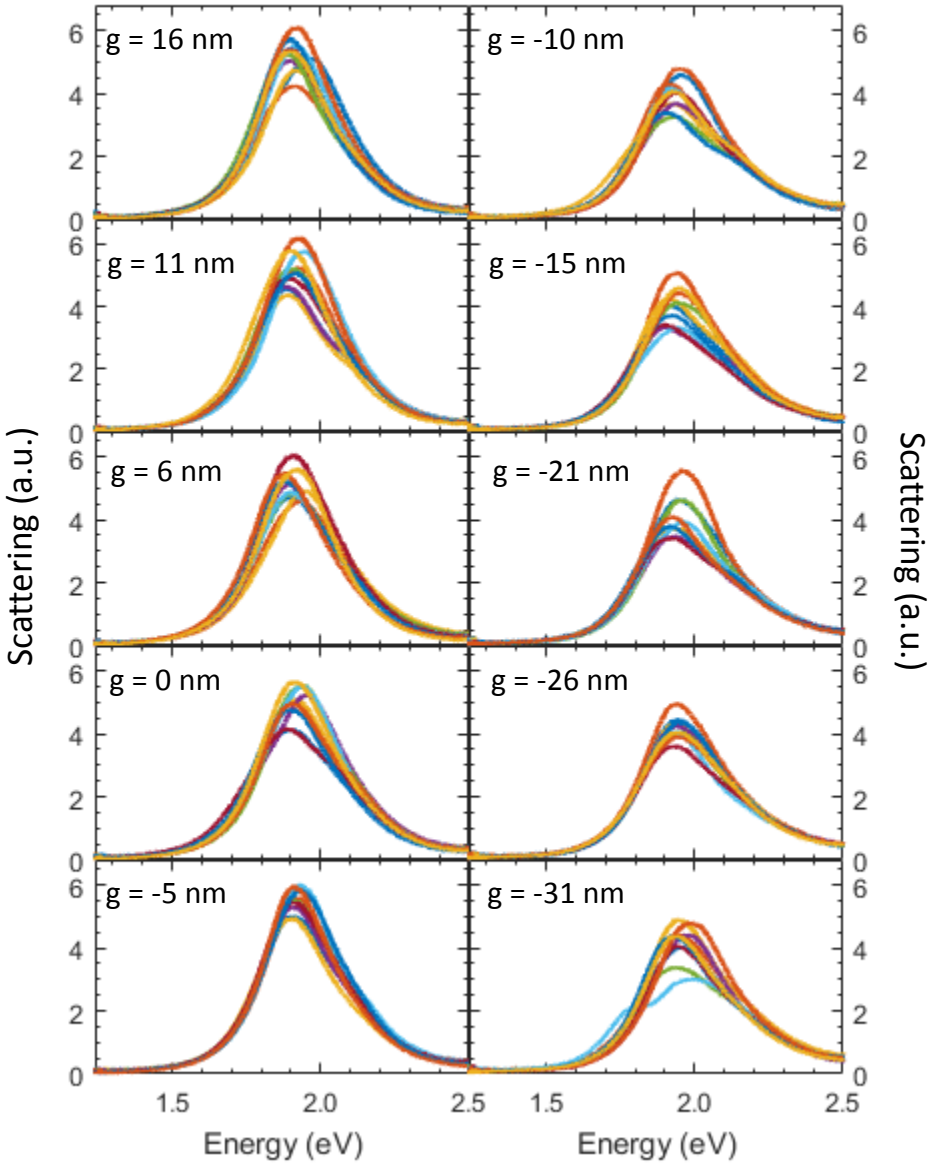

Figure S5. Single-particle scattering spectra of square nanoblock dimers with transverse polarization. The edge length of the nanoblock was 100 nm and the gap sizes (*g*) are indicated in each panel.

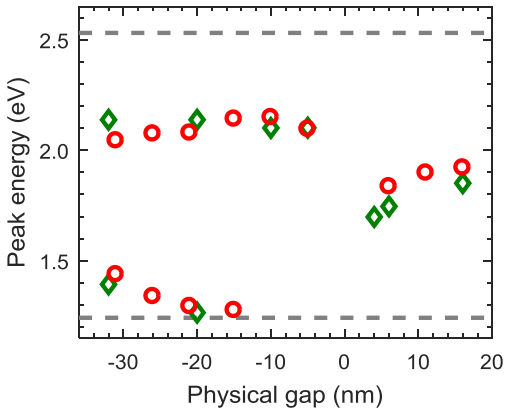

Figure S6. Scattering peak energy evolution of L-modes as a function of gap size. All the peaks in the spectral window, as indicated by the grey dashed lines, are included. Spectra for some dimers have multiple peaks, which are all included here. Green diamonds: calculated. Red circles: experimental.

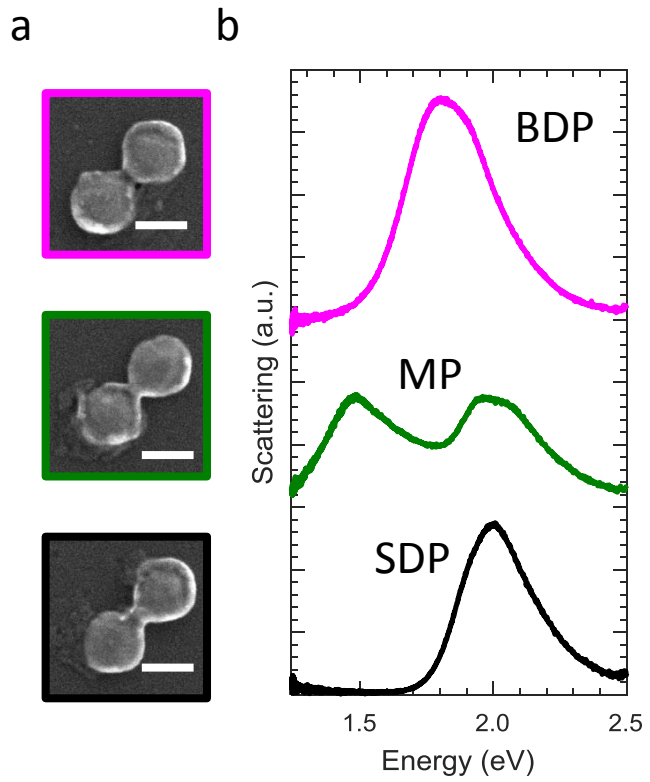

Figure S7. Another example illustrating the variety seen in single-particle scattering spectra of nanoblock dimers for the transition from capacitive coupling to conductive contact. (a) Correlated SEM images (scale bars: 100 nm) and (b) experimental scattering spectra of dimers with a gap of 0 nm and a nanoblock edge length of 100 nm.

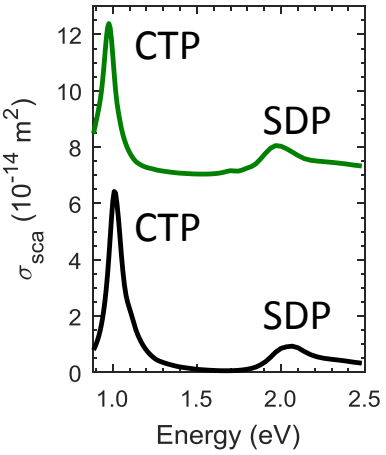

Figure S8. Simulated scattering spectra of nanoblock dimers with a length of 80 nm and gaps of -1 (top) and -2 nm (bottom) shown here for an extended spectral window. The spectra are the same as given in Figure 3. An offset is applied for visualization.

Table S1. Comparison between designed and estimated gap sizes. Unit: nm.

Designed	12	7	2	-4	-9	-14	-19	-25	-30	-35
Estimated	16	11	6	0	-5	-10	-15	-21	-26	-31