Supporting Information for Influence of Gold Nano-bipyramid Dimensions on Strong Coupling with Excitons of Monolayer MoS_2

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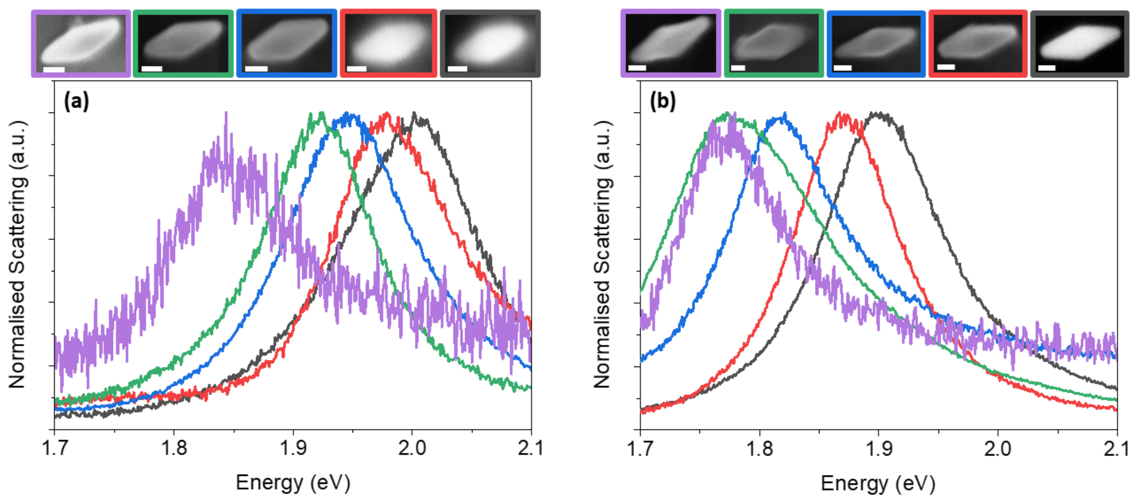

Figure S1. Experimental Scattering Spectra of (a) 80 nm and (b) 100 nm long bipyramids on a SiO_2/Si substrate. Corresponding SEM images are shown above. A large range of longitudinal plasmon energies are shown for each size, achieved by varying the aspect ratio. Increasing the length of the bipyramid is also shown to red-shift the plasmon resonance.

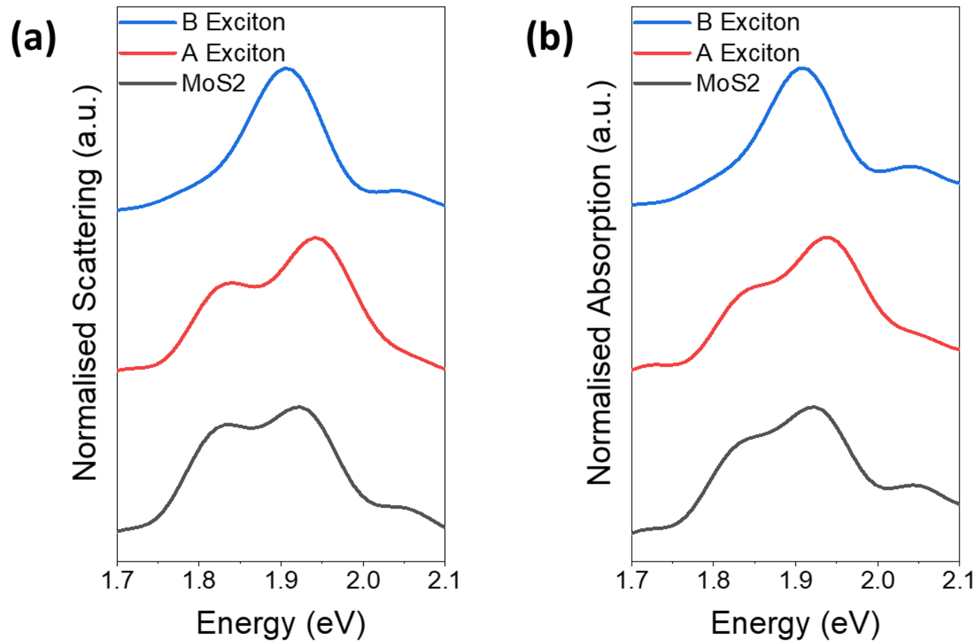

Figure S2. Simulated (a) scattering and (b) absorption spectra comparing hybrid system with a bipyramid on MoS_2 (black), MoS_2 with only the A exciton present (red) and MoS_2 with only the B exciton present (blue). It is clear that the A exciton dominates the coupling, but the B exciton has some contribution, resulting in a lower peak at a higher energy.

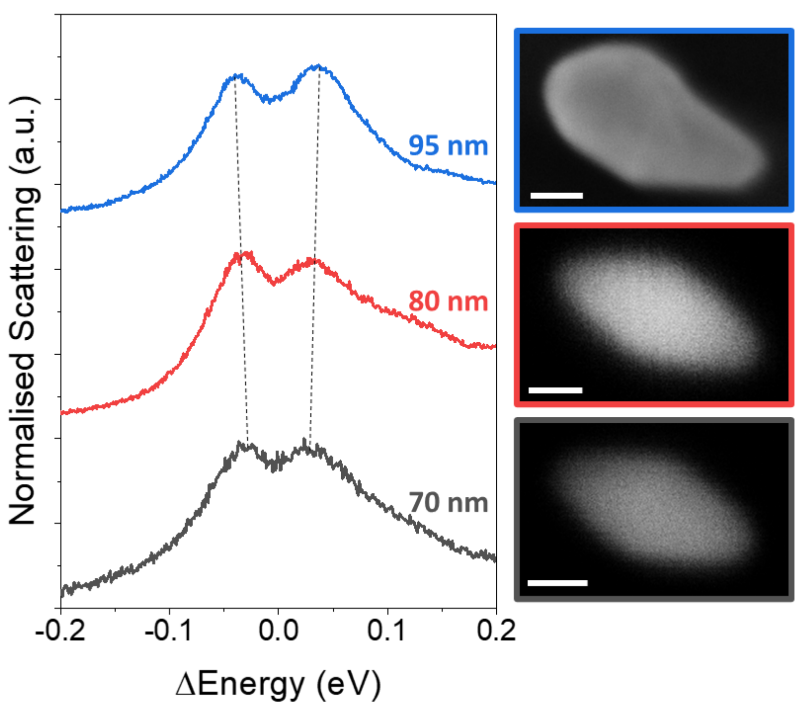

Figure S3. Example spectra of bipyramids sized 70 – 95 nm coupled to MoS_2 with zero detuning between the plasmon and exciton. X-axis shows the detuning from the energy of the original plasmon and exciton of the system.

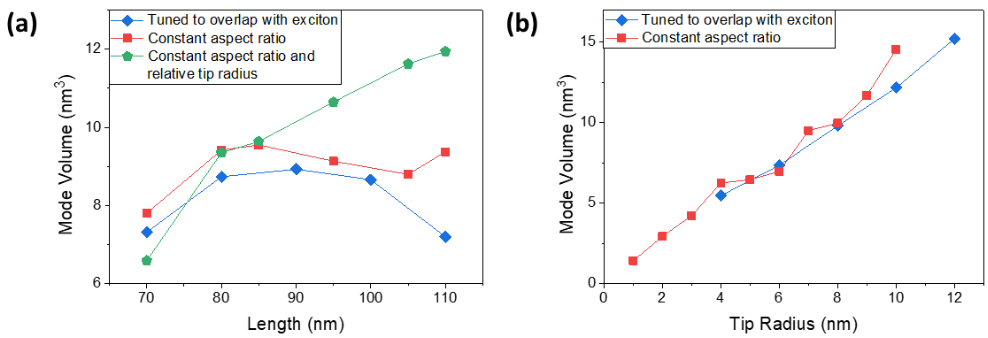

Figure S4: Mode Volume dependence for (a) the overall length of a bipyramid and (b) the tip radius of a bipyramid. The mode volume was taken from equation 5 in the main text and calculated over the volume of the MoS_2 layer underneath the entire bipyramid. The above data was taken from simulations of the same bipyramids as in figure 4 in the main text. The mode volume increases with the length of the bipyramid, except for when the tip radius is made smaller in proportion to the rest of the bipyramid, or when the aspect ratio is reduced to keep the plasmon at the same energy, as shown in (a). A strong increase in mode volume is shown for bipyramids of increasing tip radius in (b).

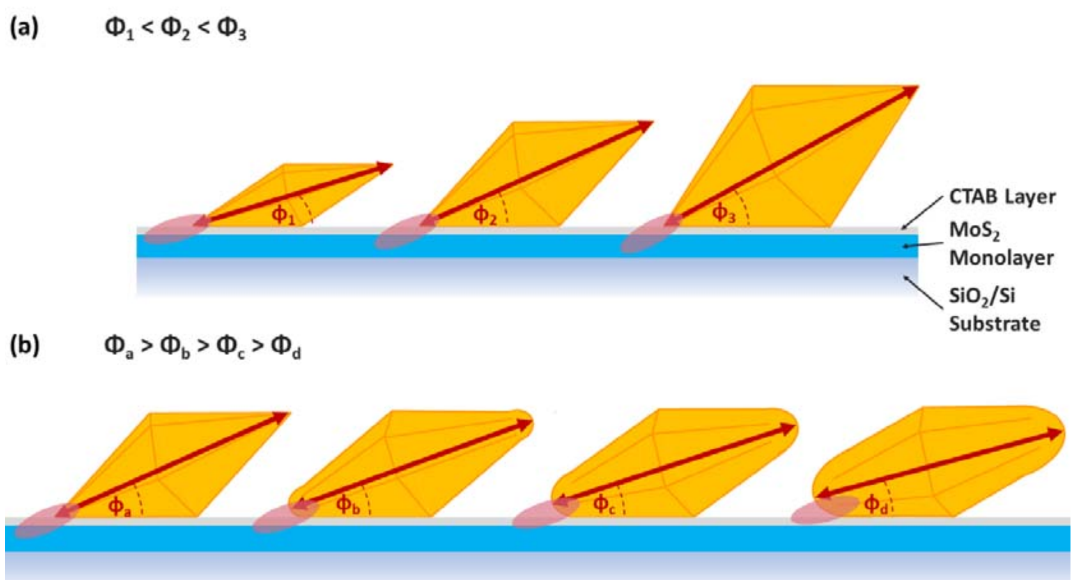

Figure S5: (a) Sketch of bipyramids of increasing length with aspect ratio tuned to have a plasmon at the same energy as the A exciton in MoS_2. The electric field is shown to overlap more directly with the MoS_2 for the larger bipyramids, thus decreasing the mode volume of the resonator. (b) Sketch of bipyramids with the same lengths and aspect ratios but different tip radii. The electric field is shown to overlap more directly with the MoS2 for the bipyramids with smaller tip radii, thus decreasing the mode volume of the resonator.

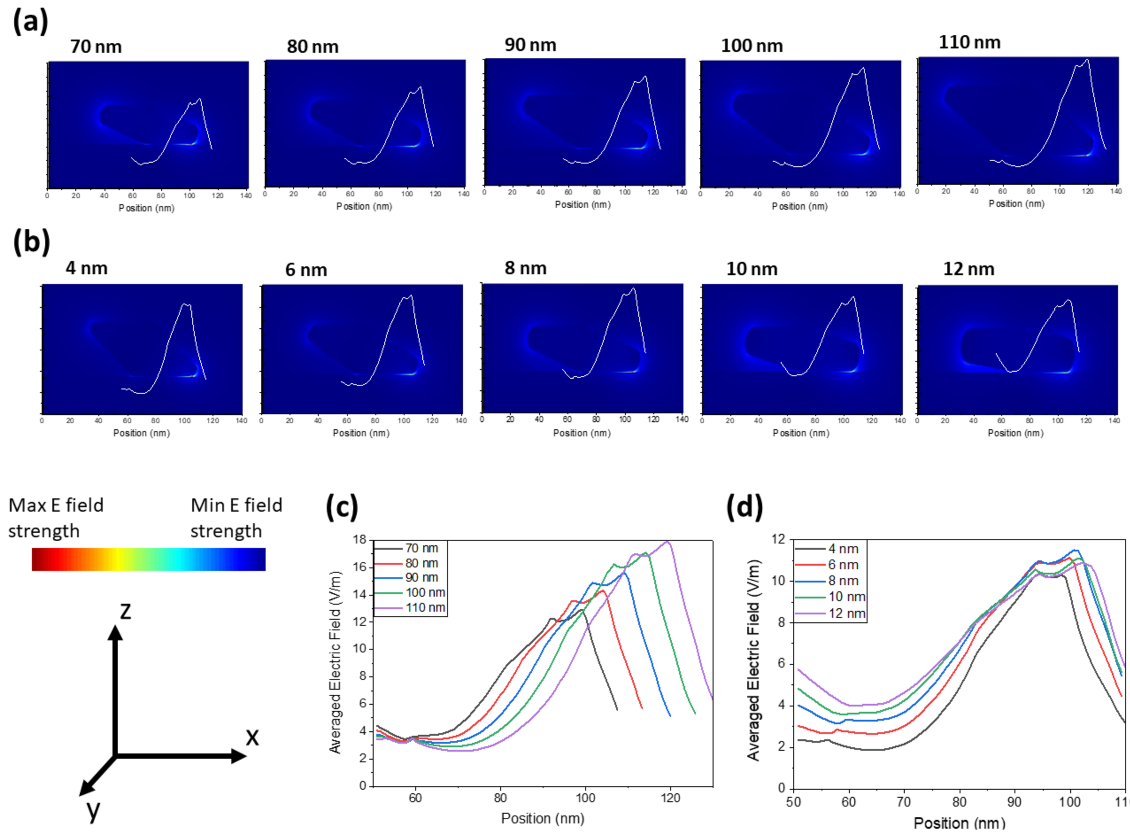

Figure S6. (a) and (b) Electric field maps of bipyramids, coupled to MoS_2, of (a) varying length and (b) varying tip radius, in the central slice through the bipyramid in the x-z plane. They correspond to the blue curves in figures 4 and S5, being tuned to overlap with the exciton energy. The overlay is the average electric field over the y-z plane for that segment. The corresponding average electric field plots from (a) and (b) are plotted again in (c) and (d) respectively. In (c) the FWHM increases marginally from 25 nm to 26.5 nm for bipyramids sized 70 to 110 nm, indicating a small mode volume increase for larger bipyramids. In (d) the FWHM increases significantly for the bipyramids with larger tip radii from 20.5 nm to 27 nm for tip radii changing from 4 to 12 nm. Therefore, the tip radii have a significant effect on the mode volume.

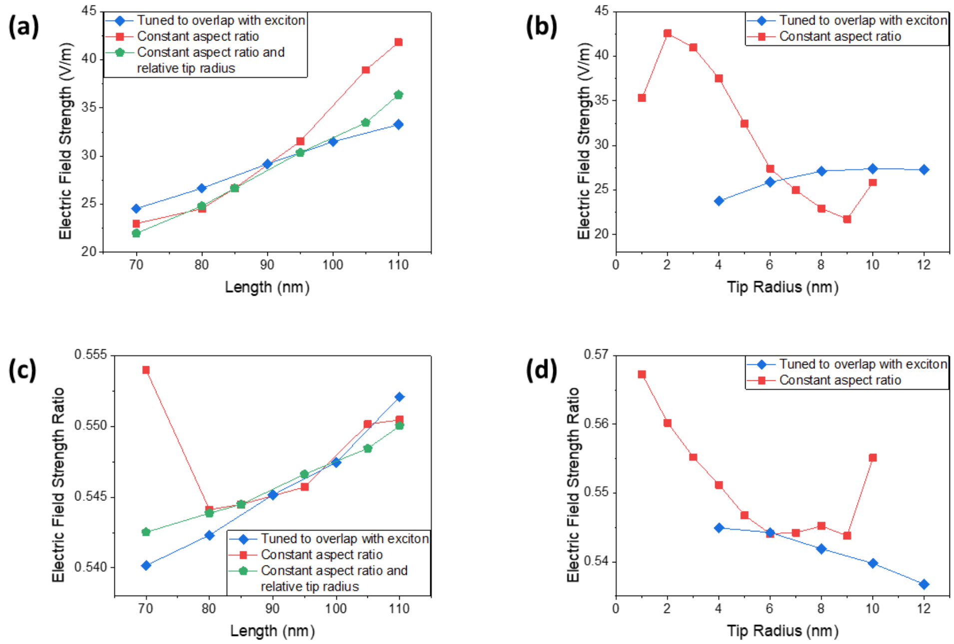

Figure S7. (a) and (b) Average electric field strength within CTAB and MoS_2 layers directly under the interacting bipyramid tip on the substrate. The results show that the overall electric field strength is higher for longer bipyramids and bipyramids with sharper tips. The averaged electric field strength shows a slight increase for the bipyramids tuned to overlap with the exciton in (b) due to the increased modal volume. (c) and (d) Ratio of electric field strength within the MoS_2 layer to that within the CTAB layer. This shows that the overlap between the field and the MoS_2 is larger for longer bipyramids and bipyramids with sharp tips. Outliers are the bipyramids with plasmons far in energy from the A exciton in MoS_2 as described in the main text.

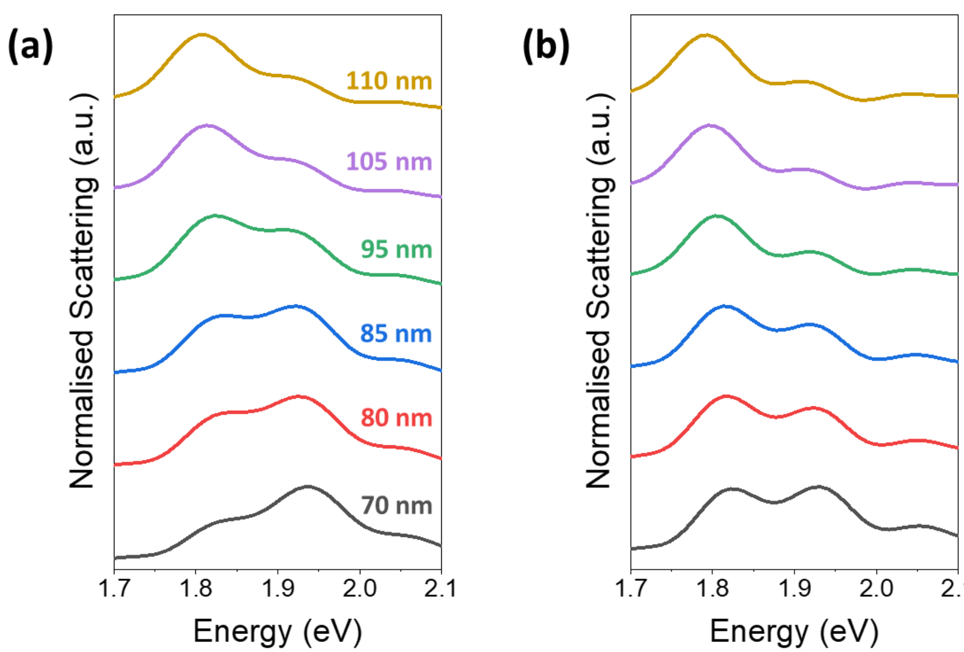

Figure S8. Scattering spectra for bipyramids coupled to MoS_2 modelled as cones attached at their base (a) and similar bipyramids modelled with a 10-facet geometry. The spectra are similar, showing that both models are valid to approximate the experimental results.
