

## Supporting Information

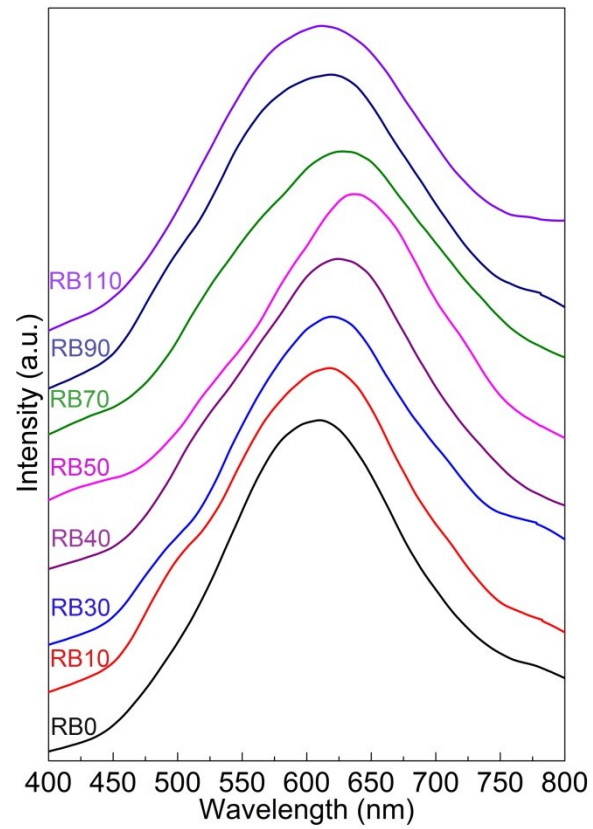
### Effects of Rotation-angle on Surface Plasmon Coupling of Nanoprisms

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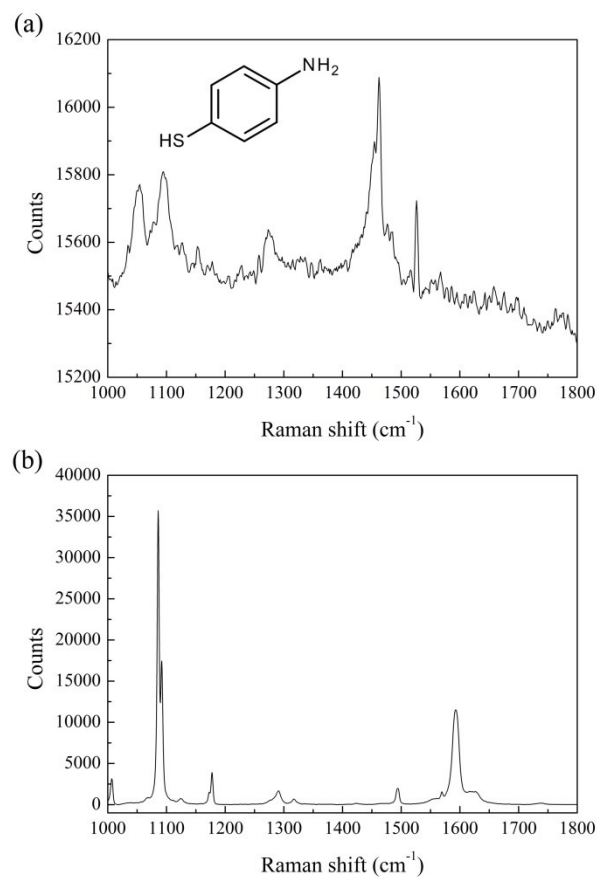
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## SCATTERING SPECTROSCOPY



**Figure S1.** Scattering spectra of selected samples of rotated bowtie. The corresponding wavelength of scattering peak as a function of the rotation angle is shown in Figure 4b. Due to the radiation damping effect of  $E$  field and the limitation of the instrumentation, the two modes (tip mode and edge mode) become an unresolvable peak in the scattering spectra.

## RAMAN SPECTRA OF *p*-ATP



**Figure S2.** The Raman spectra of *p*-ATP with excitation laser of (a) 532 nm and (b) 633 nm. For excitation of 532 nm, the peak around 1460 cm<sup>-1</sup> shows highest response, on the other hand, for excitation of 633 nm, the peak around 1070 cm<sup>-1</sup> is most sensitive to Raman spectroscopy.

**Table S1.** Raman intensity (in counts) of 633-nm-excited samples with Stokes shift of 1070  $\text{cm}^{-1}$ .

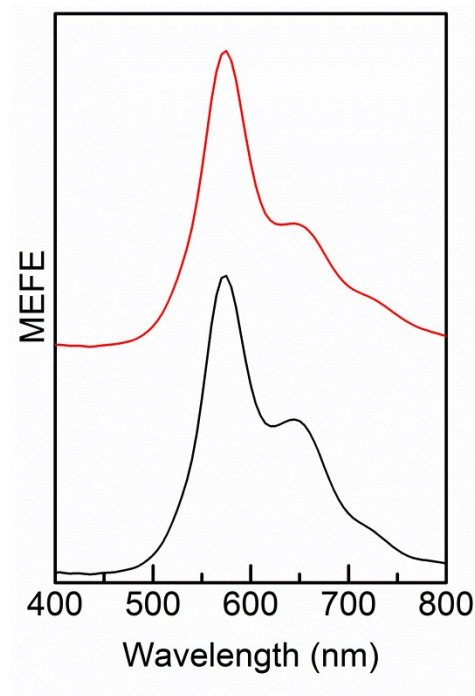
Sample \ No.	1	2	3	4	5	6
RB0	1326.84	1267.15	1248.50	1229.46	1213.14	1245.81
RB10	1482.36	1322.28	1334.88	1267.99	1271.06	1212.35
RB20	1345.50	1396.39	1274.73	1254.61	1257.18	1256.69
RB30	1398.26	1487.64	1426.74	1313.85	1295.77	1289.20
RB40	1523.28	1430.78	1531.22	1589.55	1458.77	1411.10
RB50	1817.34	1893.42	1827.55	1993.50	1801.82	1899.66
RB60	1311.47	1381.52	1395.54	1395.53	1426.09	1422.73
RB70	2670.16	2633.19	2526.02	2525.88	2448.63	2889.60
RB80	2382.86	2279.71	2215.98	2184.23	2334.46	2270.28
RB90	1709.26	1713.23	1734.25	1754.33	1736.44	1778.70
RB100	1668.34	1686.01	1660.90	1659.63	1610.13	1628.61
RB110	1538.80	1531.45	1573.59	1576.20	1525.75	1523.43
RB120	1376.21	1431.45	1411.00	1417.30	1382.32	1406.56

**Table S2.** Raman intensity (in counts) of 532-nm-excited samples with Stokes shift of 1460  $\text{cm}^{-1}$ .

Sample \ No.	1	2	3	4	5	6
RB0	192	196	192	191	199	196
RB10	173	171	171	184	175	171
RB20	158	158	152	152	166	158
RB30	212	230	215	210	200	200
RB40	165	158	164	167	161	165
RB50	172	171	175	171	175	166
RB60	242	241	229	224	221	222
RB70	183	181	179	181	189	183
RB80	173	169	170	174	176	177
RB90	148	147	150	147	146	154
RB100	138	140	137	149	140	141
RB110	126	118	128	136	121	113
RB120	107	117	110	109	117	135

### Size effect of rotated bowtie nanostructures

Size effect is also an important issue in the plasmon coupling of dimers, especially for the highly sensitive bowtie nanostructures. As the size increases, a red-shift in the resonance wavelength would occur. These phenomena could be discussed using the restoring energy. As the size becomes larger, the restoring force of the dipole moment in the nanostructure becomes weaker due to the larger distance between the charges at opposite interfaces of the particles, and thus results in lowering of the resonance frequency and red-shift in the resonance wavelength [1–4]. These phenomena could also be applied to the case of rotated bowties. For example, the spectra of RB70 for the rotated bowties with circumradius of 100 nm (black line) and circumradius of 105 nm (red line) are shown in Fig. S3. Both the tip-mode and edge-mode resonances show a slight red-shift in the resonance wavelength as the circumradius (and hence the size) increases.



**Figure S3.** The spectra of RB70 with circumradius of 100 nm (black line) and circumradius of 105 nm (red line). Both the tip-mode and edge-mode resonances show a slight red-shift in the resonance wavelength as the circumradius increases.

### References:

1. Peng Yang, Herve' Portale's, and Marie-Paule Pilen; Identification of Multipolar Surface Plasmon Resonances in Triangular Silver Nanoprisms with Very High Aspect Ratios Using the DDA Method. *J. Phys. Chem. C* 2009, 113, 11597–11604
2. Maier, S. A., *Plasmonics: Fundamentals and Applications: Fundamentals and Applications*. Springer: 2007.
3. Jeffrey N. Anker, W. Paige Hall, Olga Lyandres, Nilam C. Shah, Jing Zhao and Richard P. Van Duyne; Biosensing with plasmonic nanosensors. *Nature materials*, vol. 7, JUNE 2008
4. Amanda J. Haes, Christy L. Haynes and Richard P. Van Duyne, Nanosphere Lithography: Self-Assembled Photonic and Magnetic Materials; *MRS Proceedings* , Volume 636, 2000