Supplemental Material for "Fine-tuning of polariton energies in



tailored plasmon cavities and J-aggregates hybrid system"

Figure S1. Absorption spectrum of TDBC monomer. Corresponding dye concentration is 0.03 μ M. TDBC monomers possess a broad absorption band at 522 nm, and the linewidth reached as high as 170 meV.



Figure S2. Experimental structure tuning results. The UPB is tuned with an accuracy of 2.5 nm (10.3 meV) in a range of 17.6 nm (328.8 meV). The LPB is tuned with an accuracy of 6.4 nm (20.5 meV) in a range of 44.7 nm (143.3 meV).



Figure S3. A set of extinction spectra of nanorods@J-aggregates with increasing solvent refractive index. The gray dashed lined are guide to compare the intensity viriation of the hybrid polariton states.



Figure S4. Correctted dispersion of the hybrid polariton states. The simulated result fits well with the coupled oscillator model after considering the dieletric coating layer.



Figure S5. The curvature of the edge of Au@Ag nanorods with different Ag shell thickness. **a.** Capsule nanorod (the Ag shell is ~3 nm). **b.** Quadrate Au@Ag nanorod (Ag shell thickness: ~8 nm in the axial direction, ~15 nm in the radial direction)



Figure S6. The variation of the J-aggregates' absorption peak amplitude in different solvent environments.

Synthesis Au@Ag nanorods coating with J-aggregates. The as-prepared Au@Ag nanoparticles were centrifuged (6000 rpm/min, 10min) and redispersed in deionized water. The extinction of the Au@Ag nanoparticles were controlled at the same strength. After that, 3 μ M TDBC dye molecules were added into 1 ml Au@Ag nanoparticles solution and shaken gently. 5 minutes later, Au@Ag nanoparticles were coating with J-aggregates layer. By changing the volume of TDBC, we can control the number of J-aggregates on the Au@Ag nanoparticles. The thickness of J-aggregates is about 1 nm.

Optical characterization. The extinction spectrums were measured by a J-1500 Circular Dichroism Spectrometer (JASCO, Japan) in a cuvette with 10mm light path. The TEM images were taken by a JEM-2100F TEM machine (JEOL Inc., Japan).