Supporting information for

Trace Detection of Chiral J-aggregated Molecules Adsorbed on Single Au Nanorods

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Calculation of the detectable number of molecules

The volume of a TDBC molecule is evaluated to be about 2 nm^3, which is based on the size of 2-2.5 nm and the thickness of 0.7-1.2 nm. The width of the Au NRs in our study is 40 nm. The aspect ratio of the Au NRs ranges from 1.34 to 2.46, and hence the volume of the TDBC shell is calculated to be 7.8×10^3 to 1.4×10^4 nm^3. Considering the volumes of the molecule and the TDBC shell, the number of TDBC molecules adsorbed on the Au NR are evaluated to be in a range from 3.9×10^3 to 7.1×10^3. Therefore, the detection limit, on the basis of our single-particle CDS spectroscopic results, is estimated to be on the order of 10^3 molecules.

We measured the CD spectrum of 30 μM TDBC molecules dispersed in 0.5 mM CTAB solution using a Chirascan-plus circular dichroism spectrometer from Applied Photophysics (Figure S13). The optical path is 10 mm and the spot area is about 4 mm^2. Then we calculated the number of molecules in the detection beam is about 7.2×10^{14}. The detection limit of the spectrometer is about 0.1 mdeg. Therefore, the lowest measurable number of TDBC molecules is 2.5×10^{12} using the CD spectrometer.

$$V = 4\ mm^2 \times 10\ mm = 40\ mm^3$$

$$N = 40\ mm^3 \times 30\ uM \times N_A = 7.2 \times 10^{14}$$

$$\frac{7.2 \times 10^{14}}{28.5\ mdeg} = \frac{x}{0.1\ mdeg}$$

$$x = 2.5 \times 10^{12}$$

These calculations suggest that TDBC molecules strongly coupled with the plasmonic nanocrystal can obtain an amplification factor in a range from 3.5×10^8 to 6.4×10^8 using single-particle CDS spectroscopy.

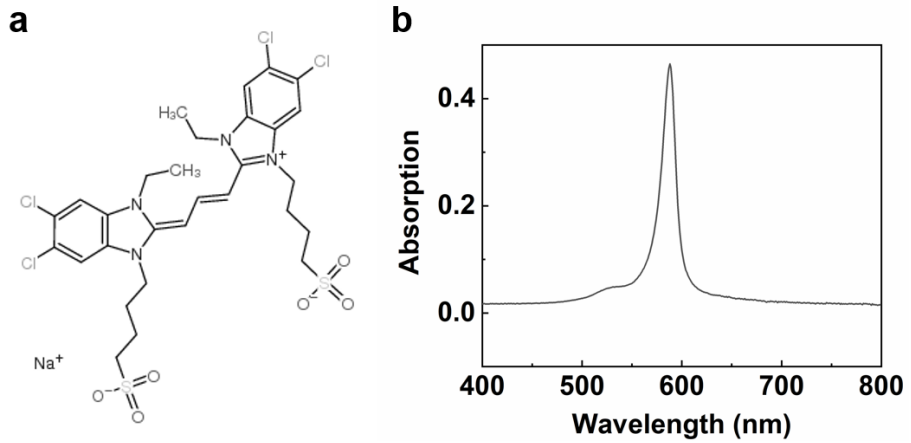

Figure S1. (a) Structure of the TDBC molecule. (b) Absorption spectrum of TDBC molecules. The TDBC molecules with concentration of 30 μM are dispersed in 0.5 mM CTAB solution.

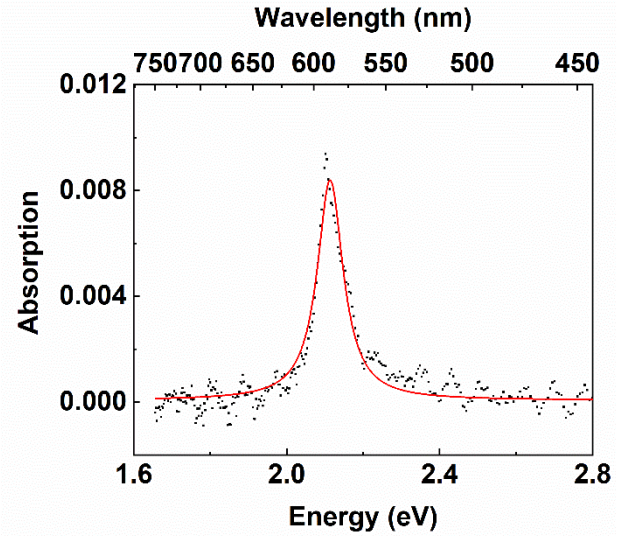

Figure S2. Absorption spectrum of the TDBC molecules adsorbed on a glass substrate functionalized by APTES (dots). The measured absorption peak is fitted with Lorentz function (red curve). The FWHM of the absorption peak is 83.0 ± 2.4 meV, obtained by the best fit.

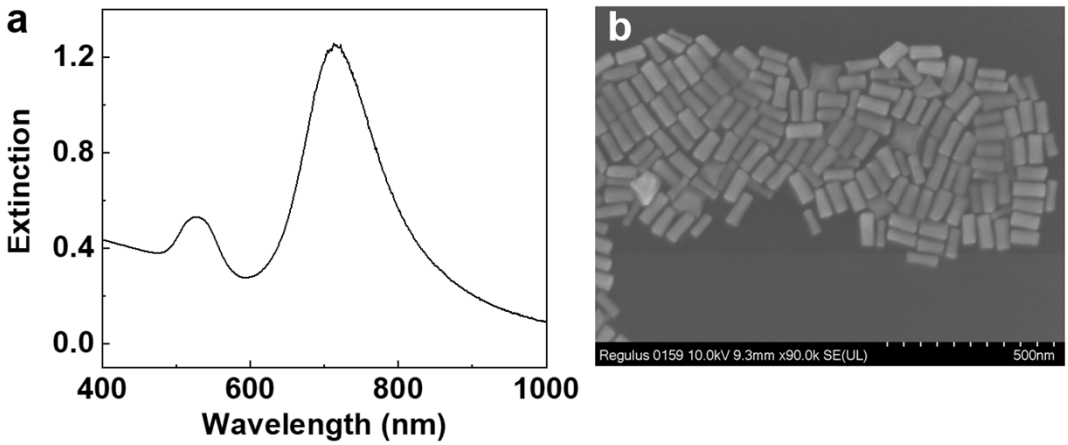

Figure S3. (a) Extinction spectra of the starting Au NRs. The Au NRs possess an ensemble LSPR wavelength of 715 nm. (b) SEM image of the starting Au NRs. The average length and width are 99.3 ± 4.0 and 40.7 ± 2.4 nm, respectively.

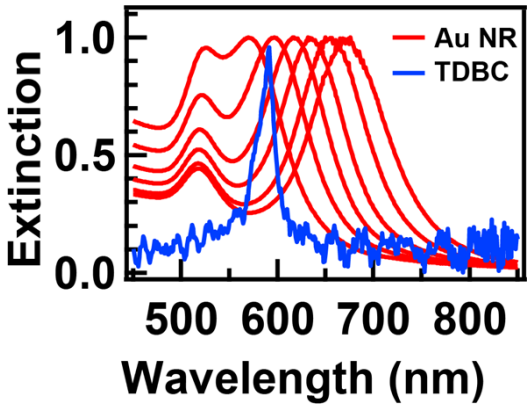

Figure S4. Extinction spectra of the 6 Au NR ensemble samples with various LSPR wavelengths at 570, 596, 616, 635, 653, and 670 nm (red curves, from left to right). The absorption spectrum of the TDBC J-aggregates adsorbed on the glass substrate is plotted for comparison (blue curve).

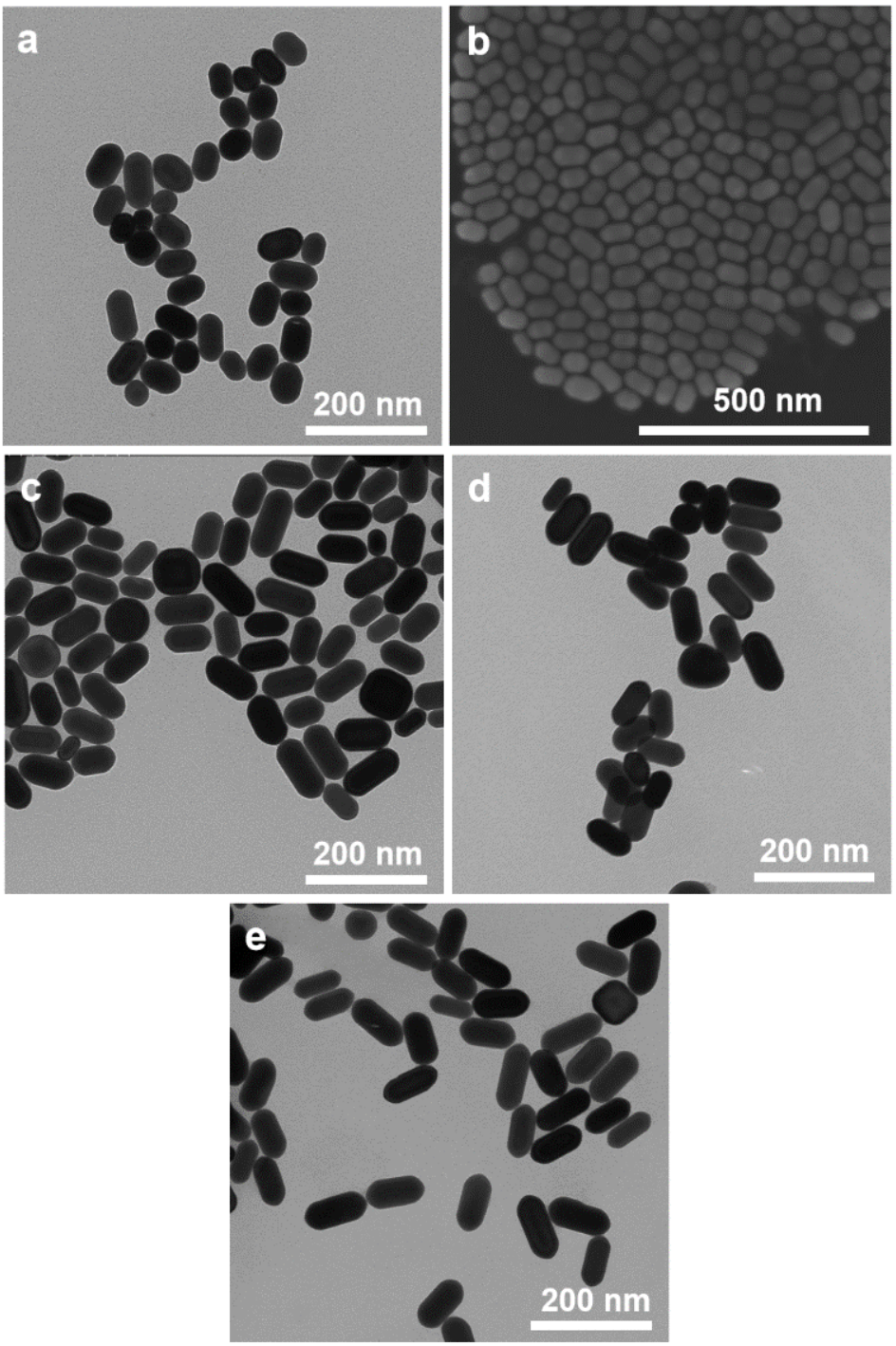

Figure S5. TEM and SEM images of the Au NR samples obtained by anisotropic oxidation. From (a) to (e), the Au NRs possess ensemble LSPR wavelengths of 570, 596, 616, 635, and 670 nm, respectively.

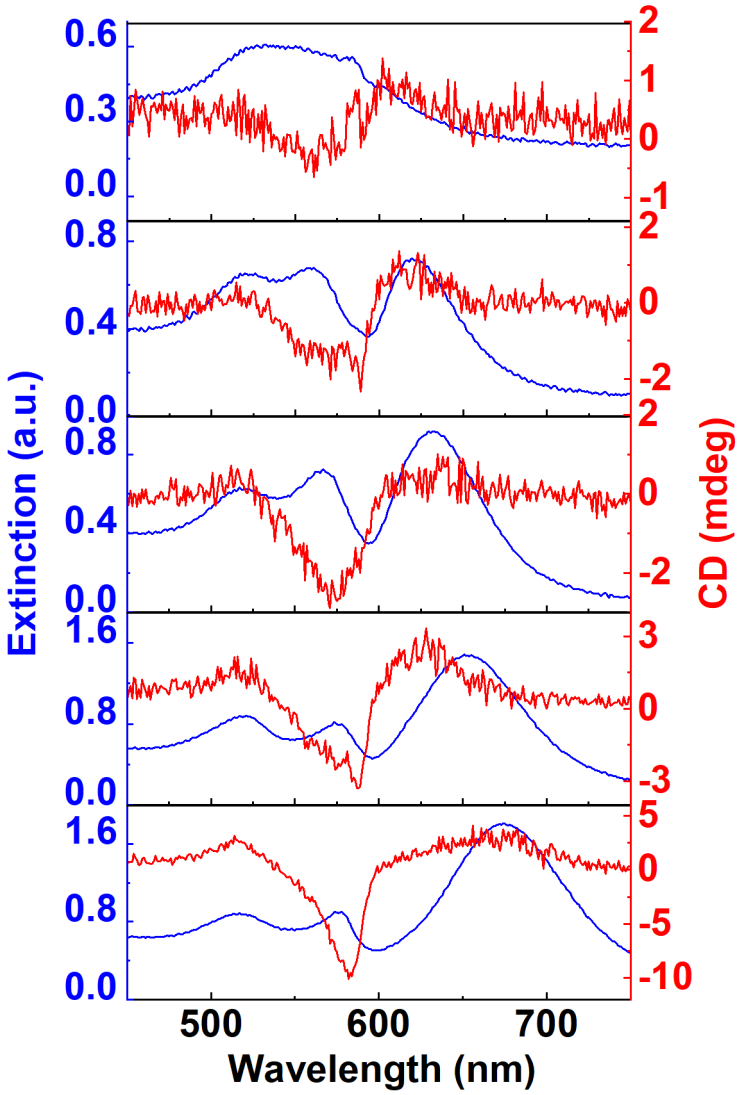

Figure S6. Measured extinction (blue) and CD (red) spectra of Au@TDBC NRs. The ensemble LSPR peaks of bare Au NR samples are centered at 570, 596, 616, 635, and 670 nm, from top to bottom, respectively.

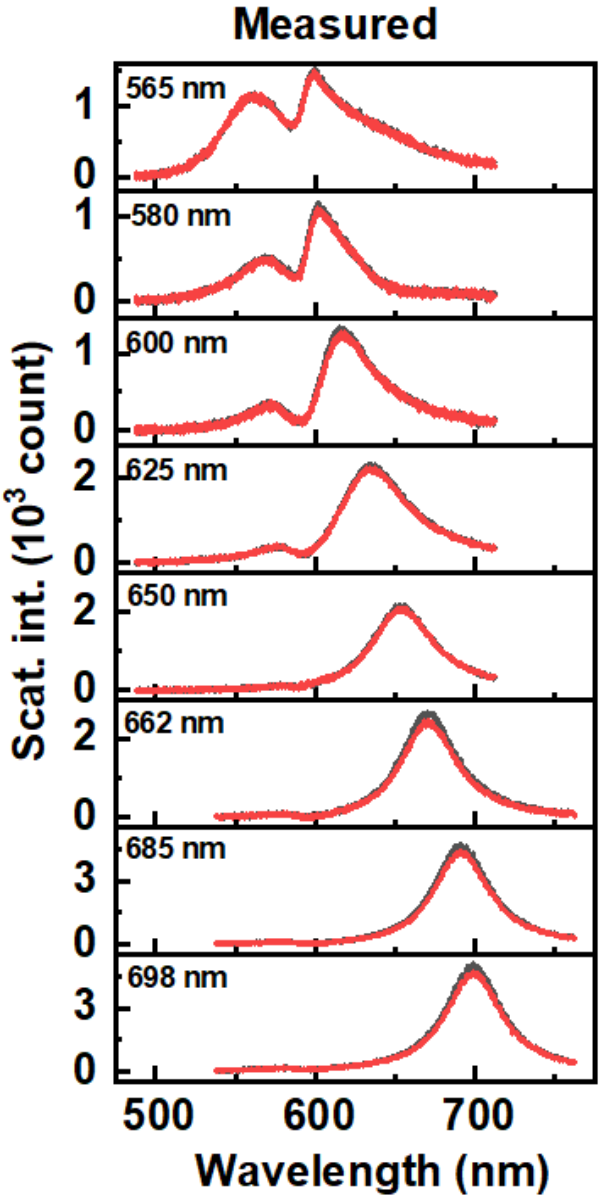

Figure S7. Measured DFM scattering spectra of the Au@TDBC NRs under the excitation of CPL with LCP (black) and RCP (red).

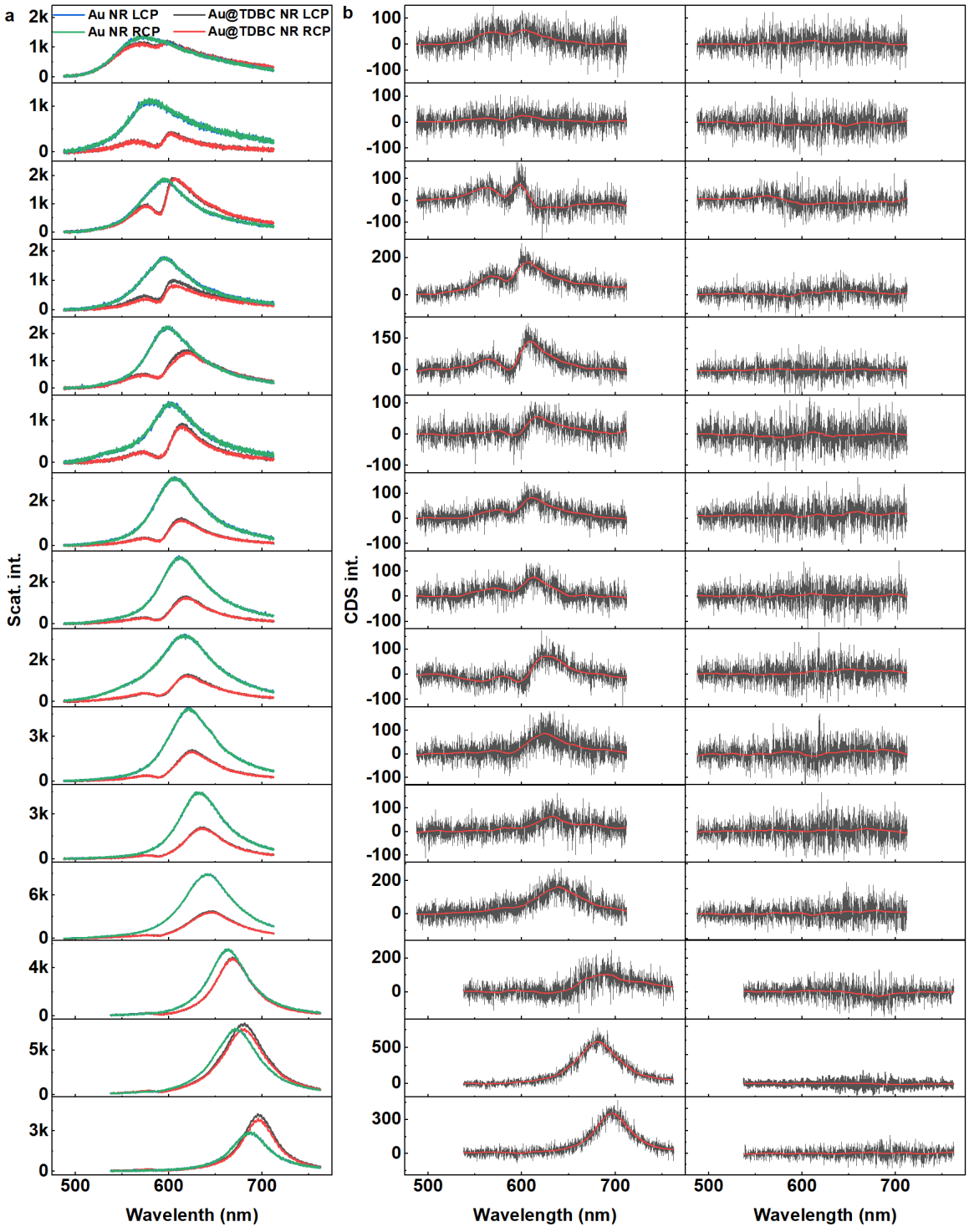

Figure S8. (a) Measured DFM scattering spectra of the Au NRs with and without the adsorption of TDBC under the excitation of CPL with LCP and RCP. (b) Single particle CDS spectra of Au NRs with (left panel) and without (right panel) the adsorption of TDBC molecules.

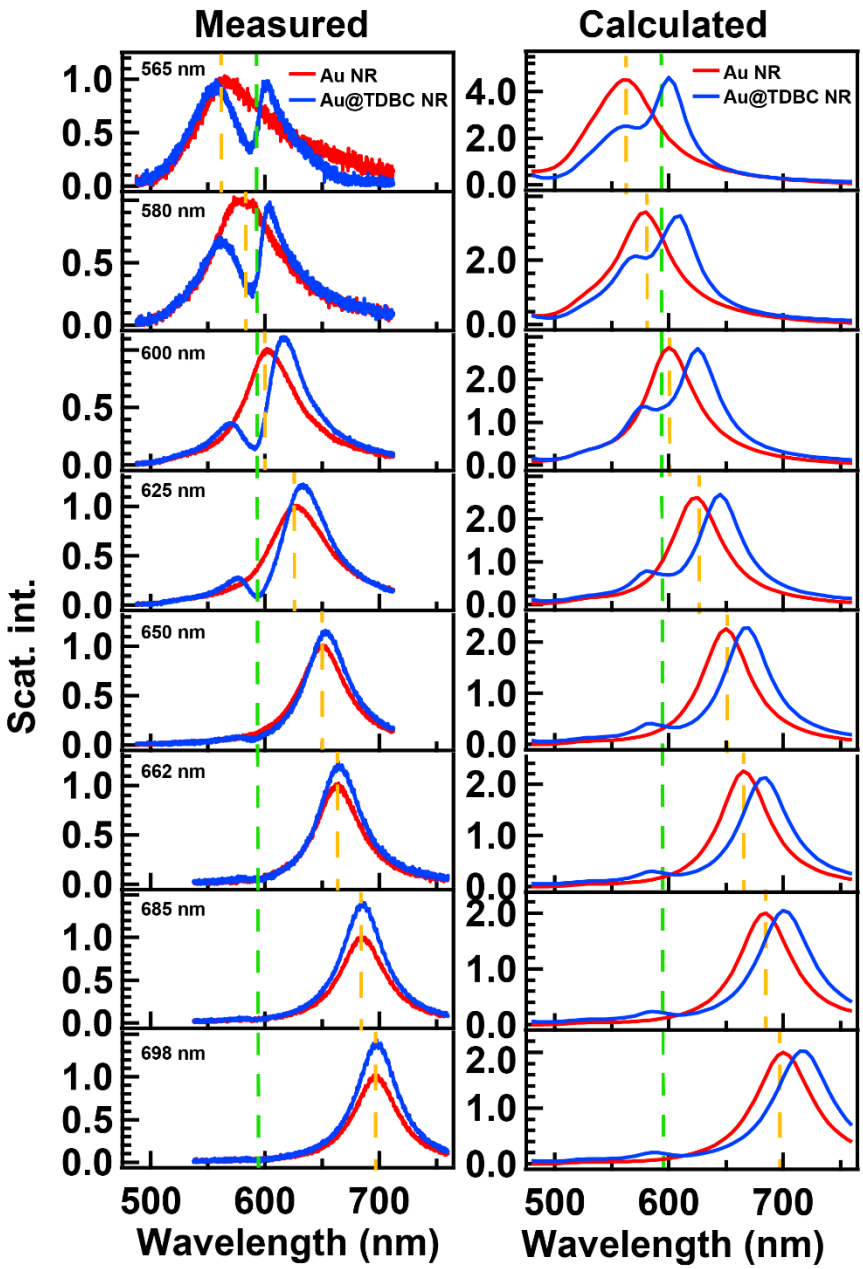

Figure S9. Single particle scattering spectra of Au NRs adsorbed with TDBC molecules. Left panel: measured scattering spectra of Au NRs with (blue) and without (red) the adsorption of TDBC molecules. The bare Au NRs possess increasing plasmonic resonance wavelengths from top to bottom, being indicated as orange dashed lines. The excitonic band of the TDBC J-aggregates at 590 nm is indicated as the green dashed line for comparison. Exposure time is 40 s. Right panel: calculated spectra. The calculated spectra of the bare Au NRs are normalized to those of the Au NRs adsorbed with TDBC by multiplying proper factors. The factor is indicated by the y axis.

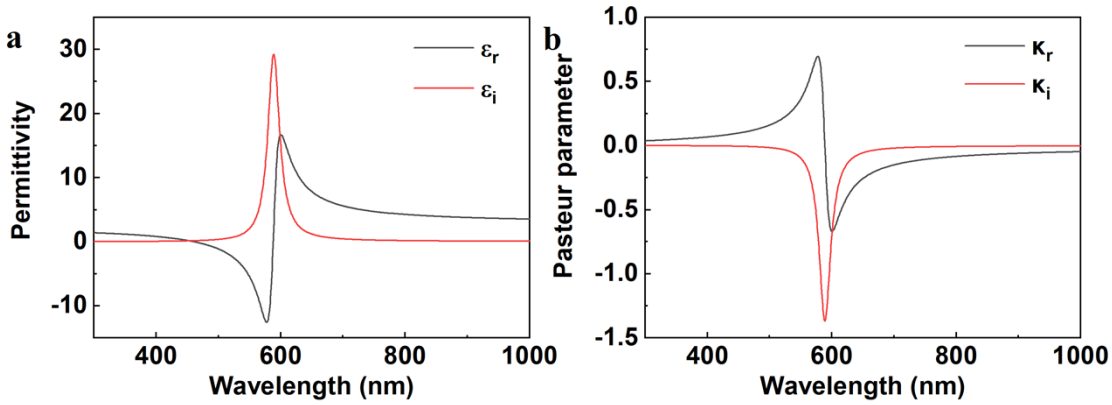

Figure S10. Dispersion of permittivity ε (a) and Pasteur parameter κ (b) of the TDBC J-aggregates.

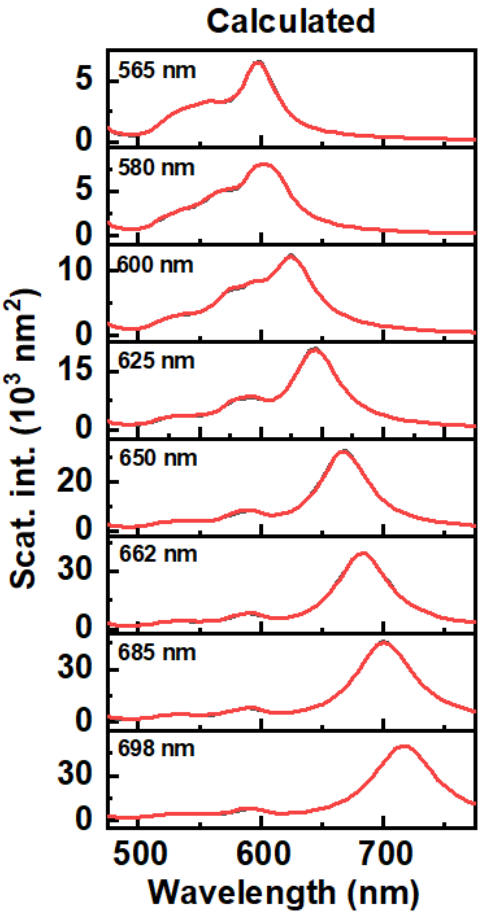

Figure S11. Calculated scattering spectra of the Au@TDBC NRs under the excitation of CPL with LCP (black) and RCP (red).

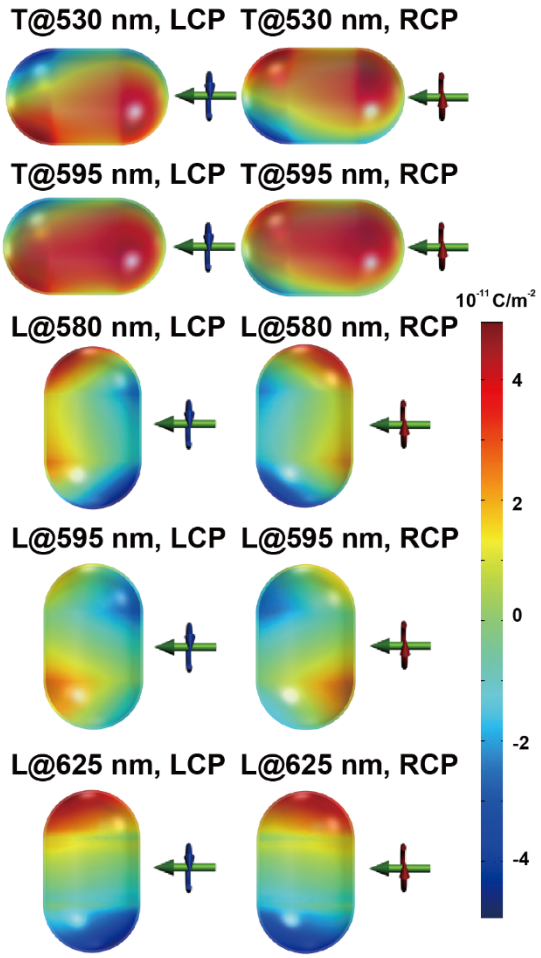

Figure S12. Charge density profiles of the nanostructures at the resonance wavelengths of the T and L modes.

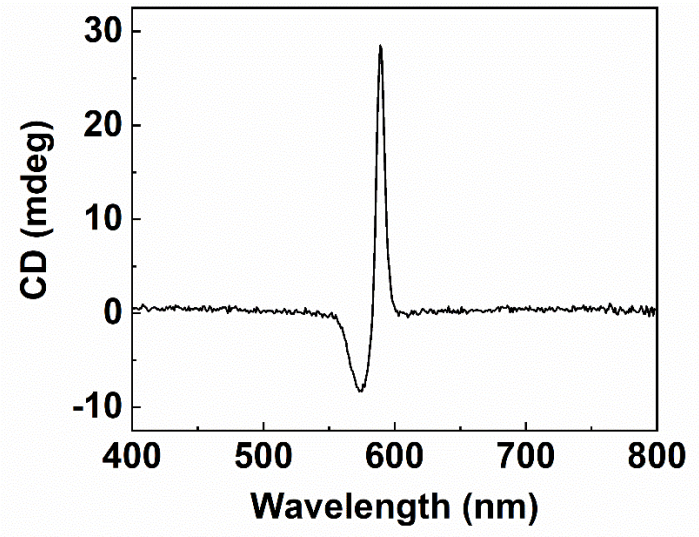

Figure S13. Measured CD spectra of TDBC using Chirascan-plus Circular dichroism spectrometer from Applied Photophysics. 30 μM TDBC is dispersed in 0.5 mM CTAB solution. The optical path is 10 mm, and the spot size is about 2 mm.