Tunable Multimode Plasmon-Exciton Coupling for Absorption-Induced Transparency and Strong Coupling

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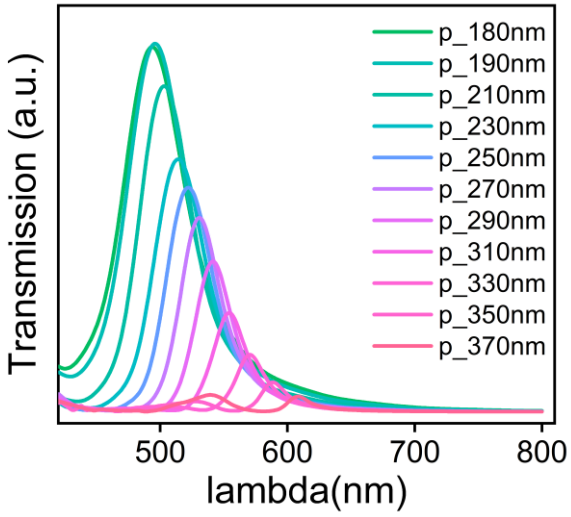

Figure S1. Bare SPP modes with respect to period.

II Hybrid Polariton Energy Calculated with Two-oscillator Coupled Model

The interaction between single plasmonic mode and molecular excitons based on a two-oscillator coupled model is $\varepsilon_{\pm} = \frac{E_{\mathrm{SPP}} + E_{\mathrm{dye}}}{2} \pm \sqrt{\frac{G^2 + \left(E_{\mathrm{SPP}} - E_{\mathrm{dye}}\right)^2}{4}}$, showing great theoretical model and parameters G for different polaritonic bands. Results can be seen that SPP(1,0) and (1,1) modes play dominant roles in UP and UMP bands, respectively, while both of them are partly responsible for LP band.

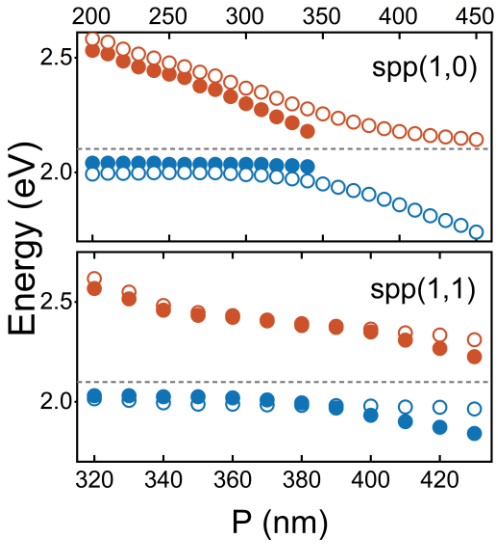

Figure S2. The interaction between single SPP(1,0) (top panel) or SPP(1,1) (bottom panel) mode and molecular excitons based on two-oscillator model for period 320-430 nm.

III Transmission spectra of different molecular absorbance

Figure S3 shows the Rabi splitting as a function of the normalized absorbance of the molecules (0, 0.3, 0.6, 1, 2.5) for the 350 nm period array, as shown in Figure 4a. The variation between the LP and UMP band with the increasing molecular absorbance gets explicitly in order to exhibit the square root dependence. Notably, the LP band with low absorbance shows the Fano dip which originate from the subtle interference with SPP(1,0) and the Fano dip disappears with high collective molecular dipoles.

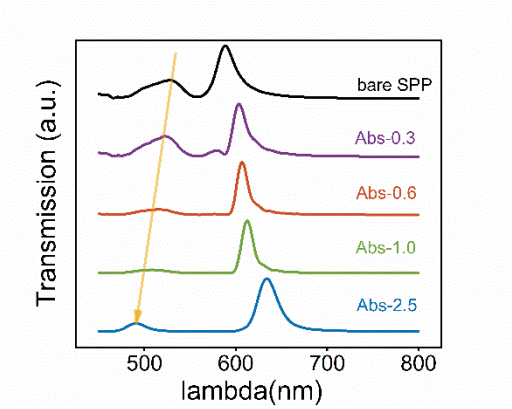

Figure S3. Transmission intensity with respect to molecular absorbance.

IV Scattering Intensity Calculated with Scatter Theory[1]

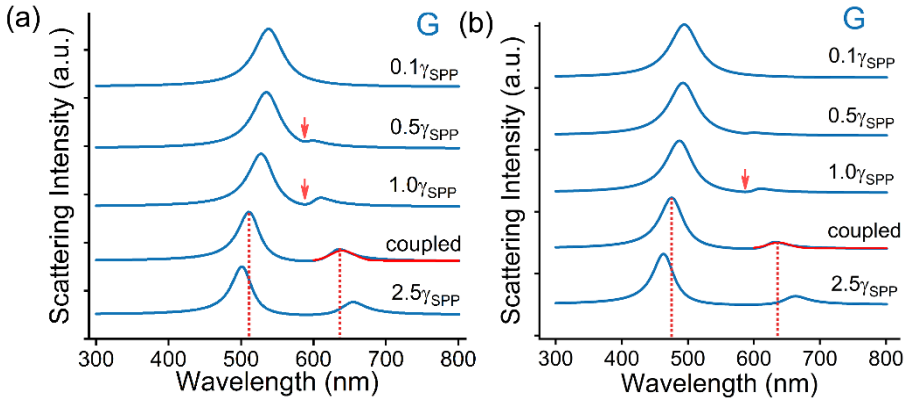

Figure S4. Comparison of calculated scattering intensity for the (a) 200 nm and (b) 350 nm period array. The resonance frequency and FWHM of SPP mode are $\omega_{SPP} = 2.5\text{eV}$, $\gamma_{SPP} = 0.27\text{eV}$ for 200 nm, a $\omega_{SPP} = 2.3\text{eV}$, $\gamma_{SPP} = 0.21\text{eV}$ for 350 nm. The molecular transition frequency and width are $\omega_{SPP} = 2.1\text{eV}$ and $\gamma_{em} = 0.08\text{eV}$.

In Figure 5 of the main text, transmission spectra and angle dispersion of P = 200 nm and 350 nm array show that the characteristic nature of AIT effect is non-dispersive which stand for short and long period. The scattering intensity with respect to the coupled strength G and SPP decay γ_{SPP} are shown in Figure S4. When G □ γ_{SPP}, bare SPP mode is dominant of hybrid states in both period. Fano dip gradually shows at the energy of molecular absorption maximum (590 nm) and obviously antisymmetry of right peak can be seen. Physical mechanism of Fano interference originates from interaction between discrete (sharp) and continuum (wide) energy. For the given

γ_{SPP} in our regime, the low coupled strength means that huge inconsistent exists in molecular exciton and SPP modes. Moreover, for long period array, Fano dip is more obvious and right peak is more symmetric. The Gaussian fitting is plotted in Figure S4 (red line) which the coefficient of determination R^2 of (a) and (b) are 0.949 and 0.934.

Reference

(1) Pelton, M.; Storm, S. D.; Leng, H. Strong Coupling of Emitters to Single Plasmonic Nanoparticles: Exciton-Induced Transparency and Rabi Splitting. *Nanoscale* **2019**, *11*, 14540–14552.