

Multiband tunable exciton-induced transparencies: Exploiting both strong and intermediate coupling in a nanocube-hexagonal-nanoplate heterodimer J-aggregates hybrid: supplement

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Multiband tunable exciton-induced transparencies: Exploiting both strong and intermediate coupling in a nanocube-hexagonal-nanoplate heterodimer J-aggregates hybrid

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S1. Charge distributions of several higher order modes of the heterodimer in the less-coupled region.

The charge distributions of higher order modes residing in the less-coupled region of Fig. 1d in the main text are presented in Fig. S1. Figure S1 shows that there is nearly no coupling between nanocube (NC) and hexagonal nanoplate (NH), and the resonances are solely determined by the single NC (see individual NC modes in main text Fig. 1c and b1). Due to the less-coupled property, the wavelengths of the resonance peaks are the nearly same as the NC with one-to-one correspondence.

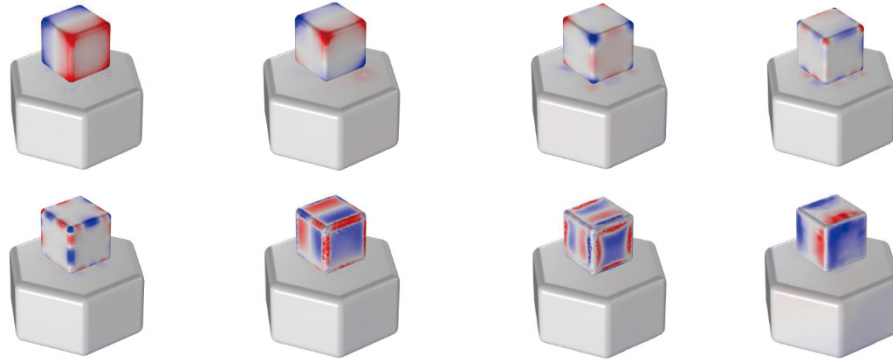


Fig. S1. The charges distribution of modes of AgNC-AuNH heterodimer in the less-couple region.

S2. Fano dips as a result of coupling between higher-order dark mode and bright $BD_c^1 D_h^1$ mode.

Figure S2 shows the scattering and absorption of the heterodimers with several nanogap thicknesses, where three other higher-order modes are demonstrated (see markers and corresponding charge distributions on right). Taking gap = 1 nm. for instance, we could observe two other minor Fano dips (green and purple markers) in scattering (Fig. S2a), which

corresponds to the two evident absorption peaks in Fig. S2b. And if we turn to the lower-order mode (blue markers), which has a bonding quadrupole-quadrupole ($BQ_c^1 Q_h^1$) distribution, we would find that though it has evident resonances in absorption (blue markers in Fig. S2b) the corresponding scattering shows no dips at all. It is due to the orthogonality between quadrupole and dipole. That's to say, $BQ_c^1 Q_h^1$ will have no coupling between $BD_c^1 D_h^1$, which leads to no interference dip in the scattering.

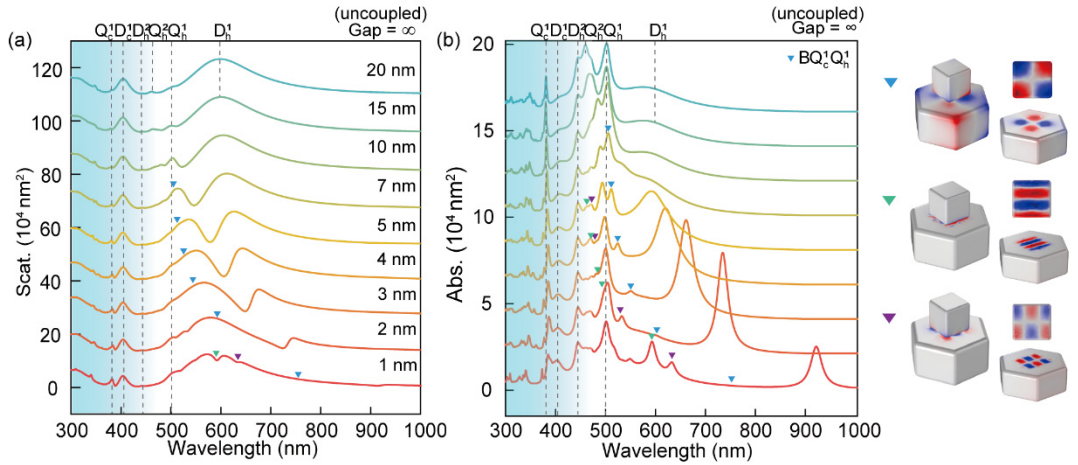


Fig. S2. Scattering (a) and absorption (b) cross-sections of a AgNC on AuNH heterodimer with different gap distances. The charges distribution of three higher-order modes of heterodimer on the right.

S3. Fitting of the scattering spectra.

Obviously, there are extra modes near the EIT window of the scattering (e.g., ~ 2.4 eV in Fig. S3, and ~ 2.1 eV in Fig. S4), which deviate the solution from a temporal coupled mode theory (TCMT) which accounts for the two oscillators. In order to better fit the EIT window, after weighing out the computation accuracy and complexity, we use the combination formula of TCMT[1] and a single Lorentz function, as follows:

$$Q_{\text{sca}} = A_1 \left[|i(\omega - \omega_1) + \Gamma_1/2 + \frac{g^2}{i(\omega - \omega_2) + \Gamma_2/2}|^2 \right] + A_2 \left[\frac{1}{\pi} \left(\frac{\Gamma_0/2}{(\omega - \omega_0)^2 + (\Gamma_0/2)^2} \right) \right] + C \quad (\text{S1})$$

The first term of the formula is from the TCMT, the second term accounts for the Lorentzian term. ω_1, ω_2 are resonance frequencies of two modes, and Γ_1, Γ_2 are their linewidths. Despite we decompose the scattering into coupled lineshape (via TCMT) with an extra Lorentzian, the scattering spectrum is still hard to be perfectly fitted. It is the extremely large radiative damping harbored by the $BD_c^1 D_h^1$ mode that makes it deviate from the dipolar lineshape, which means the $BD_c^1 D_h^1$ itself is not a rigorously Lorentzian. Thus, the TCMT or coupled oscillator model[2] based on Lorentzian oscillations could hardly perfectly match and fit the spectra. That being said, we still could obtain high R-squares.

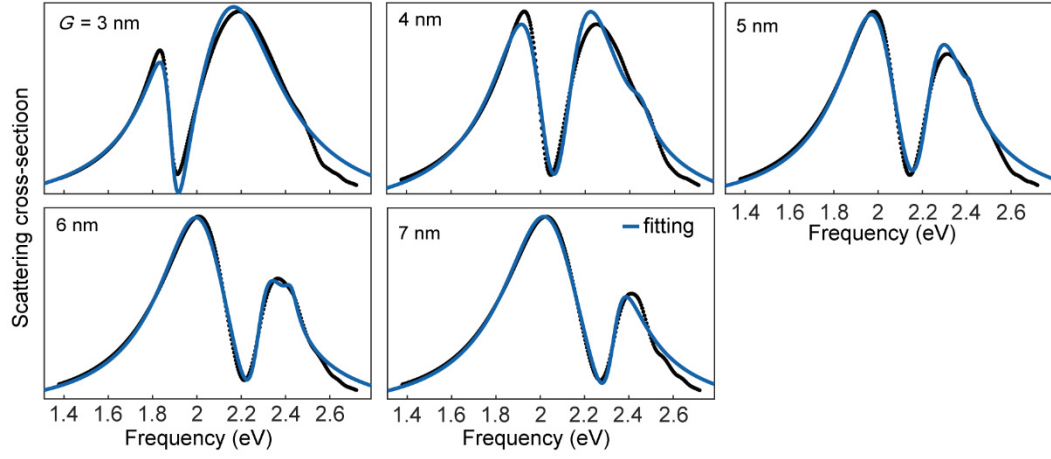


Fig. S3. The scattering cross-sections (black) and fitted results (blue) of the heterodimer with different gap distances.

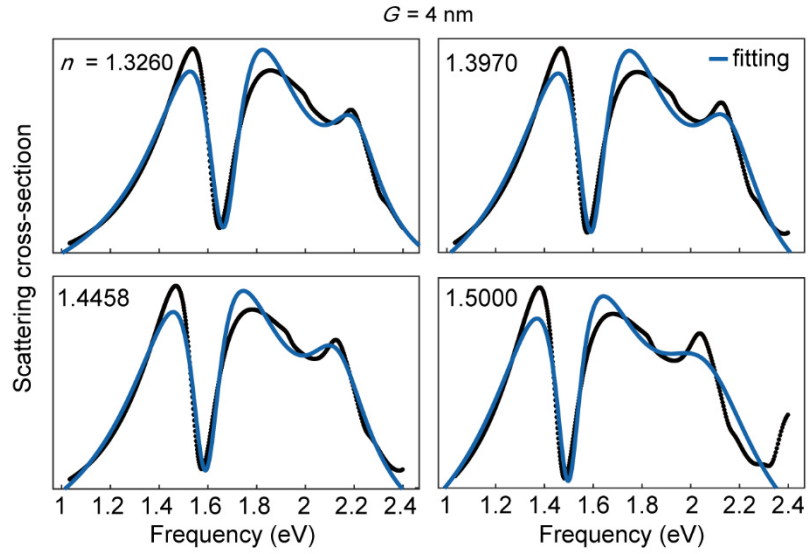


Fig. S4. The scattering cross-section (black) and fitting results (blue) of the heterodimer with different refractive indices in $G = 4$ nm.

	g (meV)	$\Gamma_1/2$ (meV)	$\Gamma_2/2$ (meV)	R-square (Coefficient of determination)	of
$G = 7$ nm	121.1	259.5	39.42	0.9927	

$G = 6 \text{ nm}$	138.4	228.9	55.47	0.9940
$G = 5 \text{ nm}$	132.4	253.5	47.16	0.9886
$G = 4 \text{ nm}$	125.6	286.2	41.08	0.9805
$G = 3 \text{ nm}$	116.9	300.9	20.09	0.9788
$G = 4 \text{ nm}$ $n = 1.3260$	111.6	332.6	40.96	0.9732
$G = 4 \text{ nm}$ $n = 1.3970$	106.7	346.4	41.17	0.9651
$G = 4 \text{ nm}$ $n = 1.4458$	105	350	39.01	0.9674
$G = 4 \text{ nm}$ $n = 1.5000$	98	350	34	0.9199

Table S1 The fitting parameter of scattering cross-sections of the heterodimer with different gaps and refraction indices in Fig. S3, 4.

S4. FOM of the NC-NH heterodimers with different materials and gaps.

We investigated the gap and material dependence of the FOM, by linearly fitting the resonance-refractive index data. It shows the FOM is material dependent, and the silver has the most outstanding property, i.e., FOM: AgNC-AgNH > AgNC-AuNH > AuNC-AuNH. And with the gap shrinking down (from 4 nm to 2 nm, for instance), the FOM tends to increase.

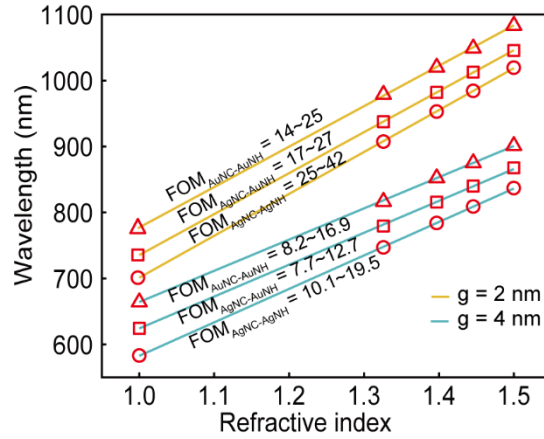


Fig. S5. The FOM of the NC-NH heterodimer $BD_1^1Q_1^1$ modes with different materials. Markers represent the resonance peaks of each mode with different refractive indexes.

S5. The scattering cross-section of the heterodimer with different thicknesses and radius of rounding curvature.

Figure S6a shows the scattering cross-section of the heterodimer as a function of AuNH rounding curvature. The curvatures radius range from 2 nm to 40 nm, and the spectra basically don't change. The scattering cross-section of the heterodimer as a function of AuNH thickness. The linewidth of the EIT window gradually narrows with the increase of thickness, while the dip position and spectral intensity change slightly.

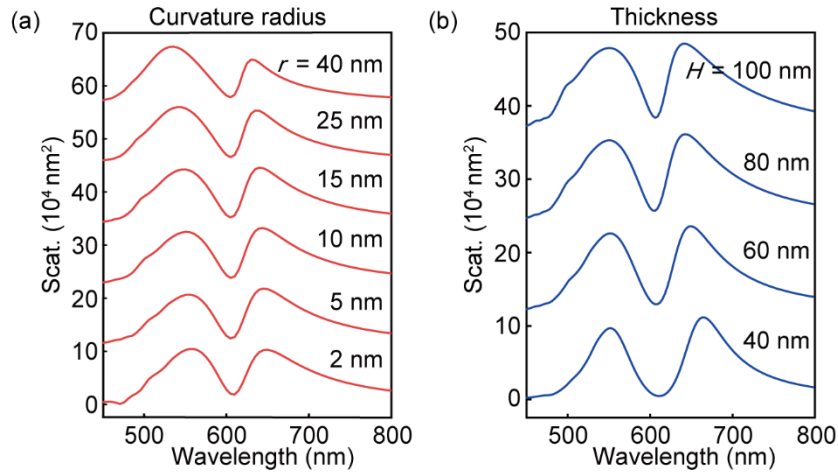


Fig. S6. The scattering cross-section of the heterodimer as a function of AuNH rounding curvature (a) and AuNH thickness (b). Other parameters of the AgNC and AuNH are as same as that in Fig. 1.

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