Supporting Information

Super Asymmetric Cross Antenna Structure with Tunable Dual-Frequency Resonance

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S1. Parameter Optimization

The tip of nanorod is set as a taper and the sharp angle of taper is called θ in the super asymmetric cross antenna structure (SACA). The angle θ and the width of nanorod (W) are taken into account in order to better understand the influence of the structural parameters of the SACA on the dual-frequency resonance. In order to study the effect of single variable, W is adjusted by changing the height of taper (h) to ensure an unchanged θ. Similarly, θ is adjusted by changing h to ensure an unchanged W. **Figure S1** shows the effect of θ and W on the location of the dual-frequency resonance. In **Figure S1a**, at W = 0.18 μm, when θ increases, the location of the low-frequency mode remains basically unchanged at 1644 cm^{-1}, whereas the high-frequency mode is red-shifted from 3277 cm^{-1} to 3166 cm^{-1}. Throughout the entire range of θ, in order to achieve a better electric field enhancement (see Section 3.1.1 for details) and desired frequency range (~3300 cm^{-1}), the value of θ should be set to 62°. As shown in **Figure S1b**, at θ = 62°, the locations of two resonances remain roughly unchanged as W increases. However, by considering the electric field enhancement of the dual-frequency resonance (see Section 3.1.1) and the effect of W on the actual machining difficulty, the preferred value of W is 0.18 μm. In summary, W and θ are optimized to be 0.18 μm and 62° respectively.

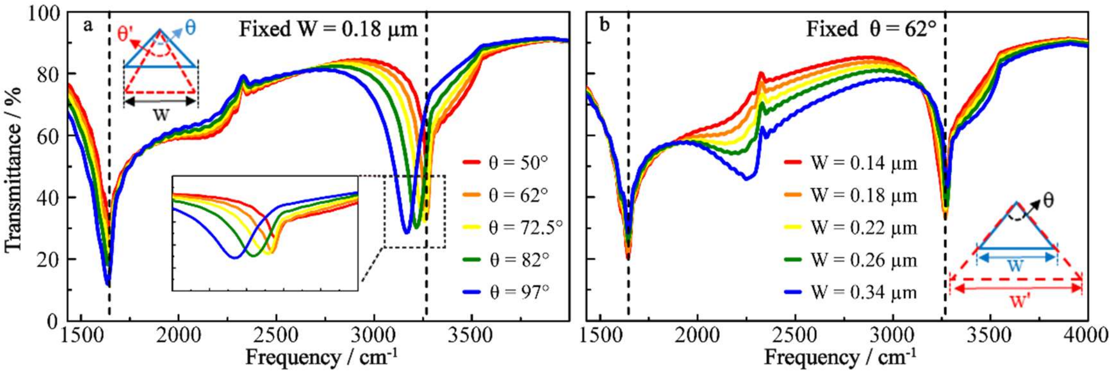

Figure S1. The spectral response of the SACA obtained by adjusting the sharp angle of taper (θ) and the width of nanorod (W). **(a)** Transmission spectra by regulating θ and at fixed W. Considering the actual machining difficulty, the preferred value of W is set to 0.18 μm. **(b)** Transmission spectra by regulating W and at fixed θ. Black dashed frame is a drawing of partial enlargement to show the change of the high-frequency mode.

S2. The Effect of L1-L3 dimer

The transmission spectra of a single L_1, a single L_3 and an L_1-L_3 dimer are shown in **Figure S2a**, in which the polarized electric field of incident light is along the y-axis, and the structural parameters of L_1, L_3 dimer and L_1-L_3 dimer are the same as those of the super asymmetric cross antenna structure (SACA). It is found that the single-frequency mode of L_1 is located at υ_1 = 1644 $cm^{-1}$and that of L_3 is at υ_4 = 3501 cm^{-1}. A dual-frequency resonance is generated as L_1 and L_3 are approaching and coupled to each other to form an L_1-L_3 dimer, which are located at υ_2 = 1677 $cm^{-1}$and υ_3 = 3466 cm^{-1} respectively. Compared with the case of a single nanorod, the location of the L_1-L_3 dimer is shifted due to the interaction of L_1 and L_3. The electric field distribution of the L_1-L_3 dimer is shown in **Figure S2b** and **S2c**. For the low-frequency mode (1644 cm^{-1}), the electric field of the L_1-L_3 dimer is mainly distributed at the tip of L_1, and a small amount of that exists at the tip of L_3. For the high-frequency mode (3466 cm^{-1}), the situation is opposite.

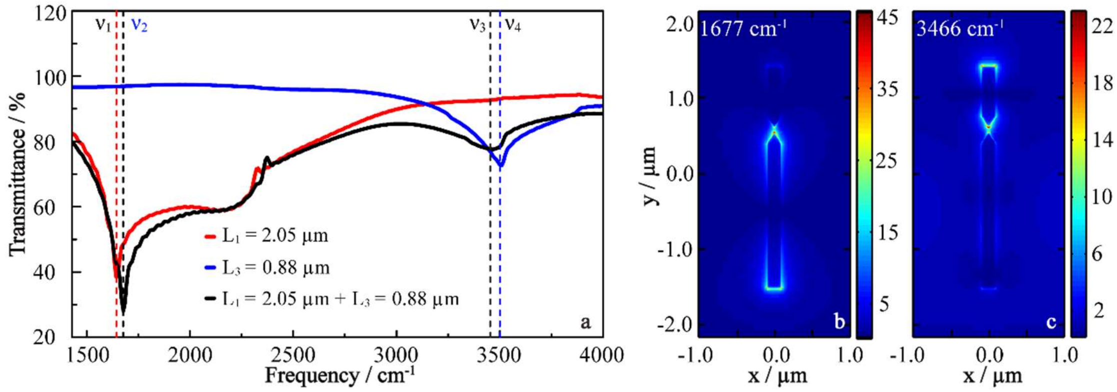

Figure S2. Calculated transmission spectra and electric field distribution for three types of nanorods. **(a)** Transmission spectra of a single L_1, a single L_3 and an L_1-L_3 dimer with υ_1 = 1644 cm^{-1}, υ_2 = 1677 cm^{-1}, υ_3 = 3466 cm^{-1} and υ_4 = 3501 cm^{-1}. **(b-c)** The electric field distribution of the L_1-L_3 dimer.

S3. The effect of L2-L2 dimer

Figure S3 shows the influence of the orthogonal gap spacing of the L_2-L_2 dimer (g) on the microcavity structure of the SACA. As shown in **Figure S3a**, the location of the low-frequency mode remains unchanged and that of the high-frequency mode is shifted from υ_2 = 3269 cm^{-1} to υ_3 = 3363 cm^{-1} as g increases. As shown in **Figure S3b-e** and also in **Figure 6e-f**, the electric field distribution of the low-frequency mode is mainly concentrated on the tip of L_1 with a weak contribution showing at the edge of the L_2-L_2 dimer.

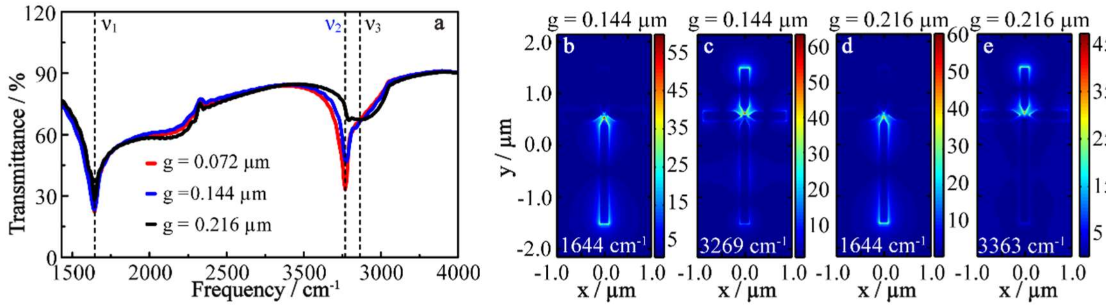

Figure S3. The influence of the orthogonal gap spacing of the L_2-L_2 dimer (g) on the microcavity structure of the SACA. **(a)** Transmission spectra of three different structural parameters with υ_1 = 1644 cm^{-1}, υ_2 = 3269 cm^{-1} and υ_3 = 3363 cm^{-1}. **(b-c)** The electric field distribution of two different structural parameters.