Supporting Information

Tunable Surface Plasmon Polaritons and Ultrafast Dynamics in 2D Nanohole Arrays

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Content:

S1. Sample fabrication: Nanoimprint lithography (NIL) utilized in this work mainly comprises two processes: thermal NIL and UV NIL. A transparent polymer stamp (from Obducat AB, Sweden) is used for duplicating the nanostructure from the nickel master mold by thermal NIL. Then, a 2-inch silicon wafer spin-coated by a UV curable resist (Tu2-170, from Obducat AB) of about 210 nm is covered by the polymer stamp and processed at 65°C with a pressure of 30 bar for 1 min under UV exposure. Afterwards, a 2 nm adhesive layer of chromium is deposited on the substrate prior to a gold layer of about 100 nm using electron-beam evaporation (see ref. [1] for fabrication details)

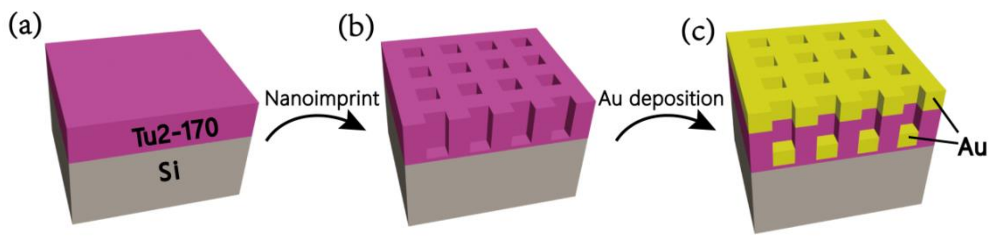


Figure S1. Sample fabrication processes

S2. Large-area SEM image: The image show that the 2D plasmonic arrays have uniform periodic nanostructures in a large area.

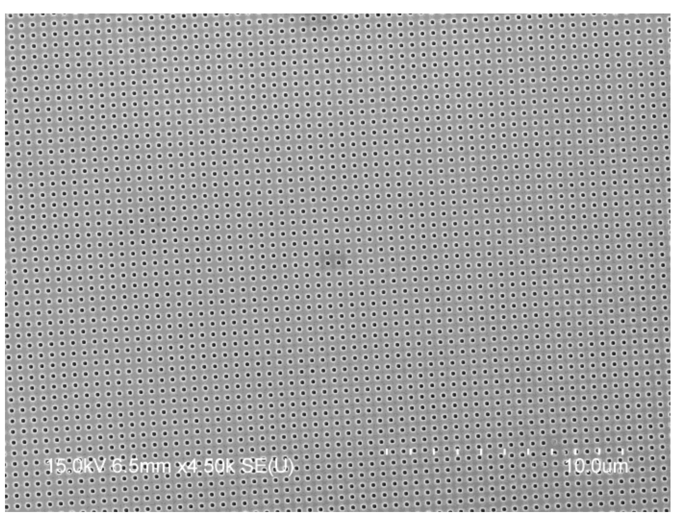


Figure S2. The SEM image in a large area.

S3. AFM image: The AFM image shows the depth profile. Extracting from the black dashed line after surface analysis, we can obtain the depth of the hole is ∼180 nm.

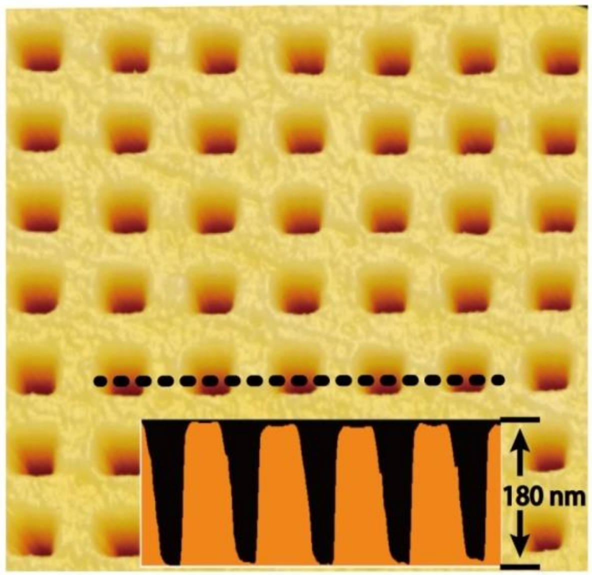


Figure S3. AFM image of the array.

S4. Simulations in the range of 350 ~ 450 nm

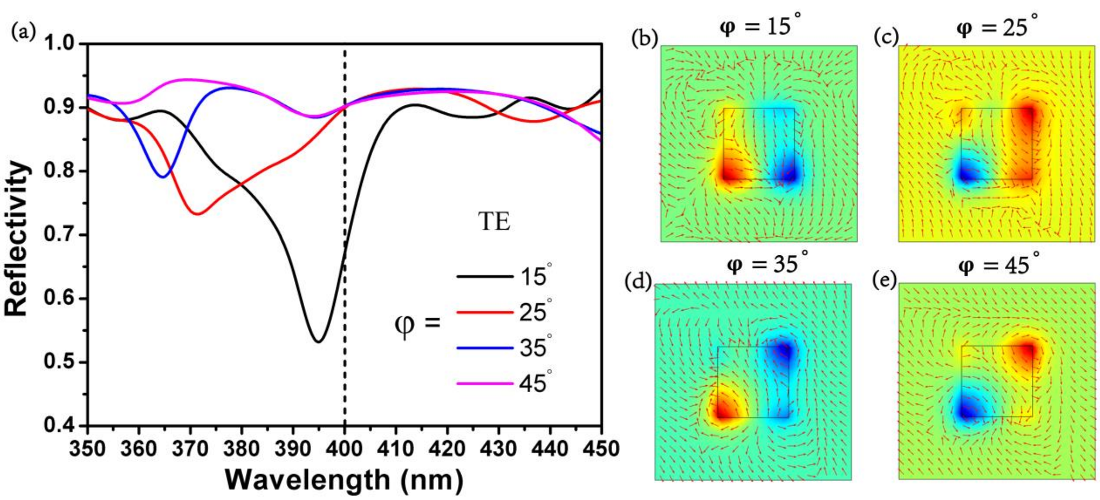
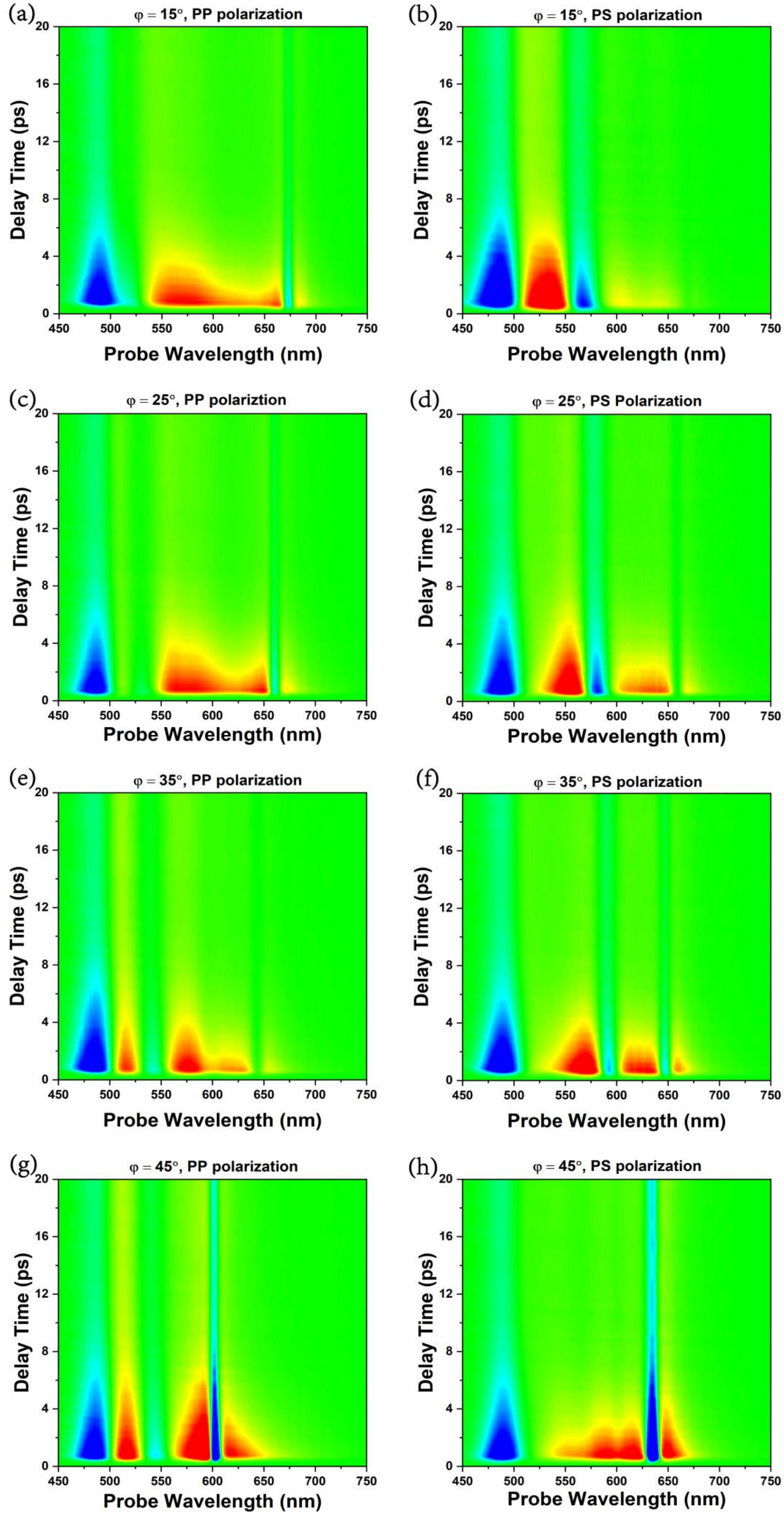


Figure S4. (a) The simulated reflectance spectra with azimuthal angles of 15 ° ~ 45 ° in the wavelength range of 350 ~ 450 nm under TE polarization, and (b-e) corresponding electromagnetic field profile of a single nanohole cell at the wavelength of 400 nm.

S5. Ultrafast maps of all azimuthal angles


(a)
φ = 15°, PP polarization
(b)
φ = 15°, PS polarization
(c)
φ = 25°, PP polariztion
(d)
φ = 25°, PS Polarization
(e)
φ = 35°, PP polarization
(f)
φ = 35°, PS polarization
(g)
φ = 45°, PP polarization
(h)
φ = 45°, PS polarization
Delay Time (ps)
Probe Wavelength (nm)
0
4
8
12
16
20
450
500
550
600
650
700
750

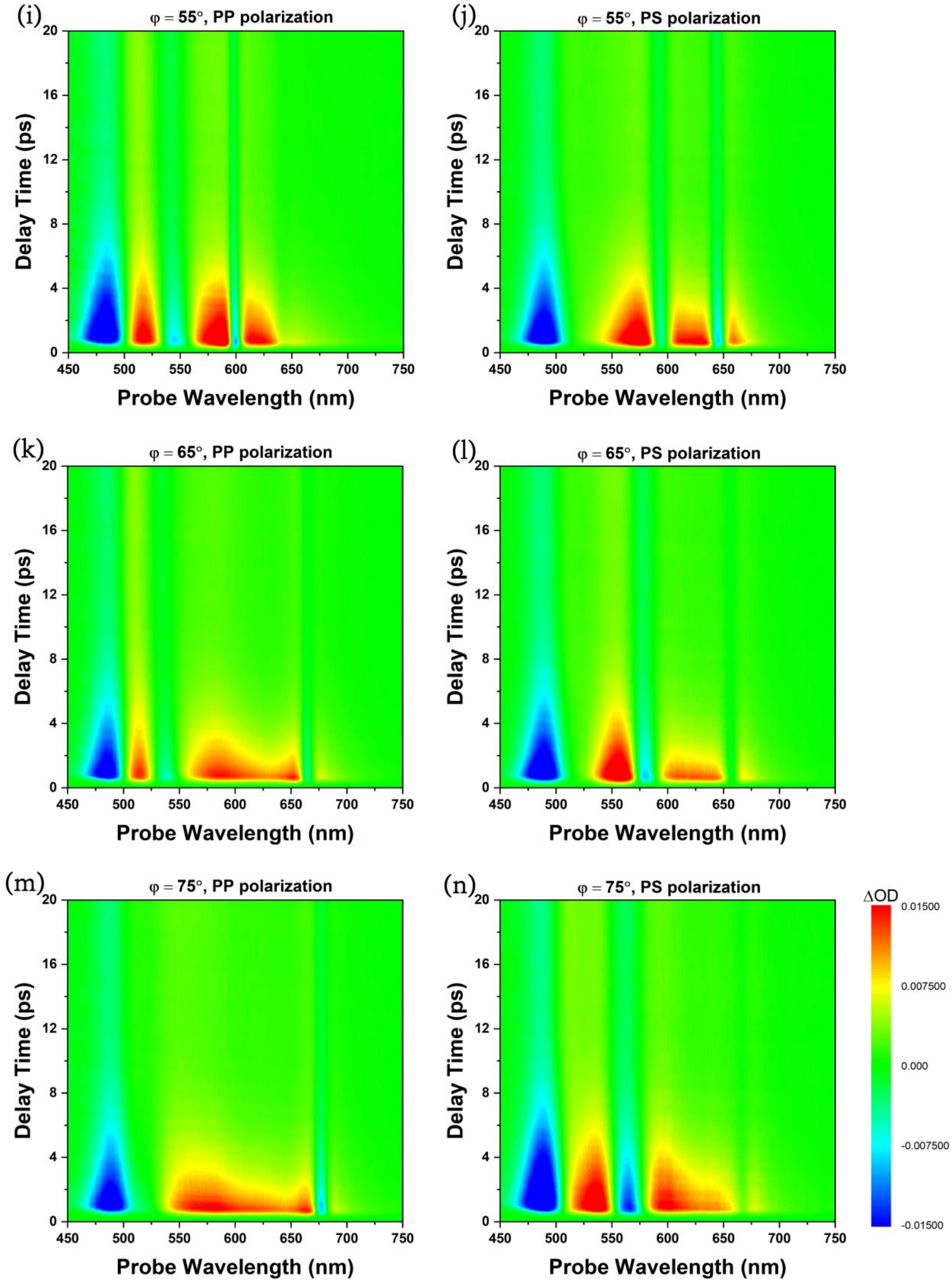


Figure S5. The ΔOD maps at all azimuthal angles

S6. Spectral shifts around plasmon dips

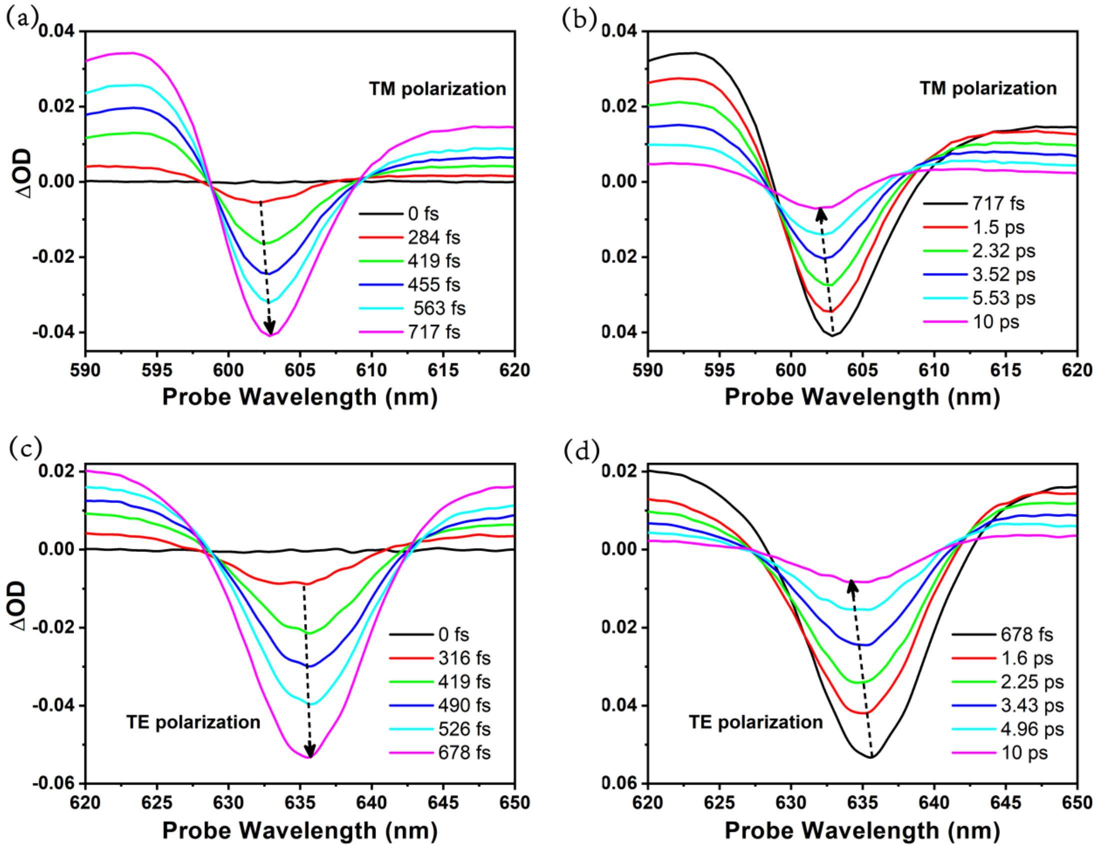


Figure S6. Enlarge Fig. 3 around the most intense plasmon dips. The black dashed lines guide the trends. Until ~ 700 fs, the spectra continue to broaden and redshift, and then to narrow and shift back.

S7. Fit of Figure 3 with dynamics

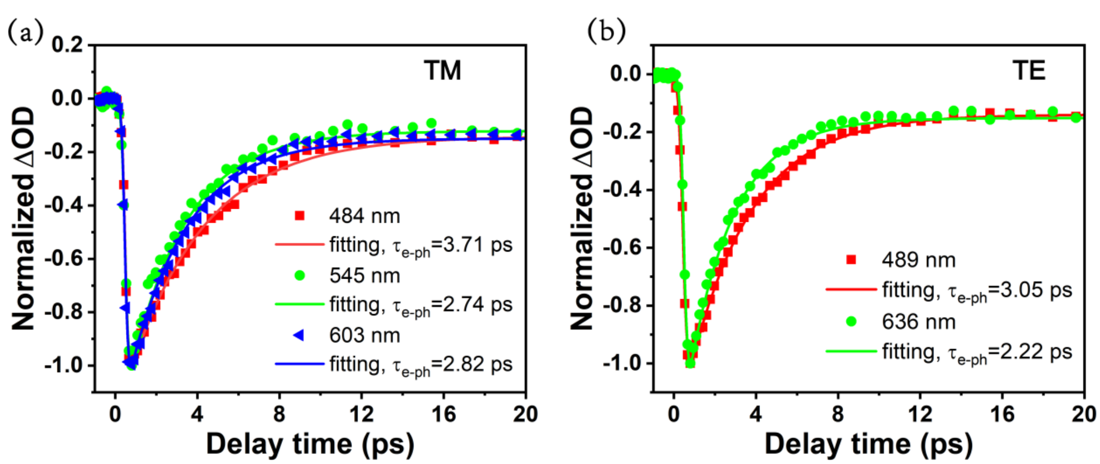


Figure S7. The corresponding time dynamics fitting of Figure 3 by a biexponetial decay functions

S8. Power-dependent transient ΔOD spectra

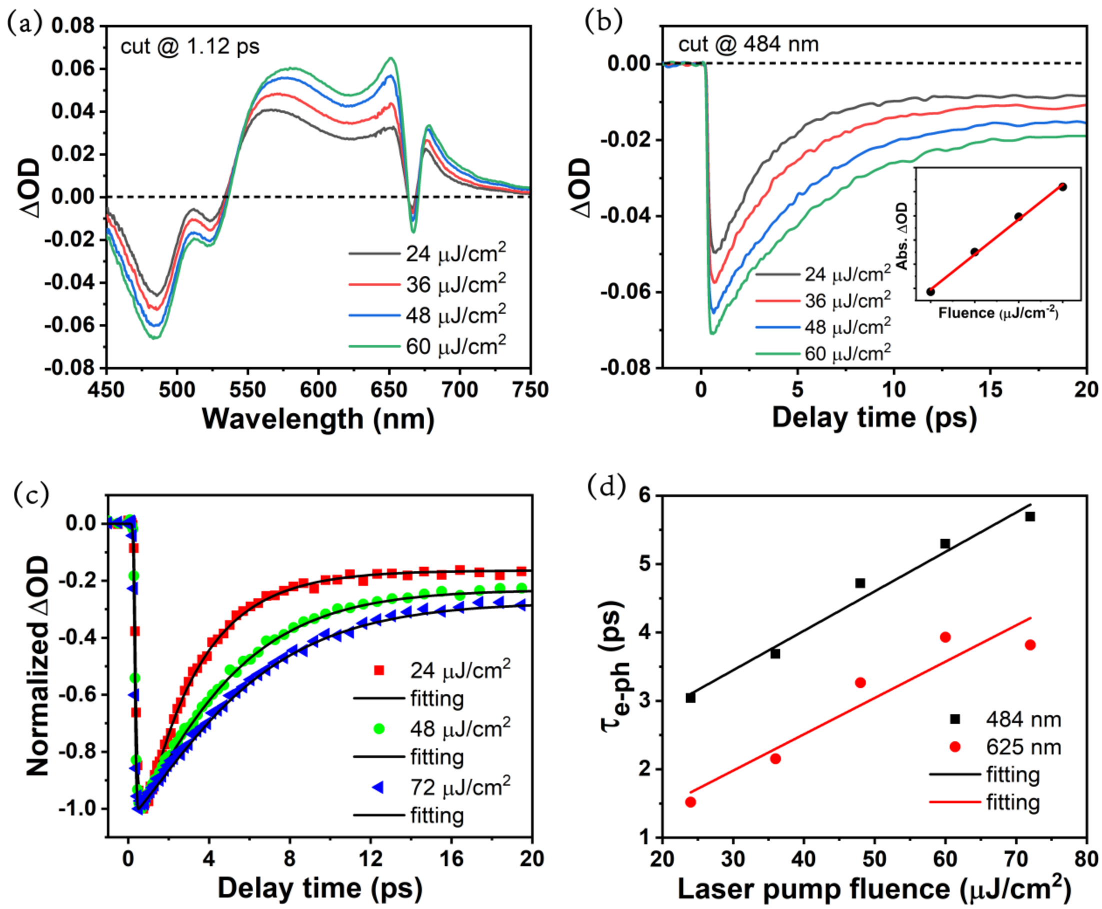


Figure S8. (a) Fluence-dependent transient ΔOD spectra around 20 ° azimuthal angle at the delay time of 1.12 ps. (b) Fluence-dependent ΔOD kinetics at the wavelength of 484 nm. Inset: Absolute ΔOD versus the pump fluence. (c) Fluence-dependent normalized ΔOD kinetics for three specific fluences of 24, 48, 72 μJ/cm^2. (d) A plot of the electron-phonon relaxation times against the pump fluence.

S9. Phase matching equation and band gap

The dispersion relation of surface plasmon at a dielectric-matal interface can be written as

$$K_{sp} = \frac{\omega}{c}\sqrt{\frac{\varepsilon_d \varepsilon_m}{\varepsilon_d + \varepsilon_m}}. \quad ①$$

The x-component of the wave vector can be described as

$$K_x = K sin\theta sin\varphi + m(2\pi/d), \quad ②$$

and the y-component of the wave vector can be described as

$$K_y = K sin\theta cos\varphi + n(2\pi/d), \quad ③$$

Where m and n denote the integer representing the diffraction order (m, n=±1, ±2, ..).

The conservation of energy and momentum can be achieved simultaneously if the surface plasmon wave vector in Eq. ① equals the wave vector of the incident light on the metal surface,

$$K_{sp} = \left(K_x^2 + K_y^2\right)^{1/2}. \quad ④$$

Combining the above relations, we can obtain the phase matching equation,

$$\frac{2\pi}{\lambda_{SPP}}\sqrt{\frac{\varepsilon_{Au}}{\varepsilon_{Au}+1}} = \sqrt{\left(\frac{2\pi}{\lambda_{SPP}} sin\theta sin\varphi + m\frac{2\pi}{P}\right)^2 + \left(\frac{2\pi}{\lambda_{SPP}} sin\theta cos\varphi + n\frac{2\pi}{P}\right)^2}.$$

Corresponding curves are shown below by using related MATLAB programs [2] (θ set to 23°).

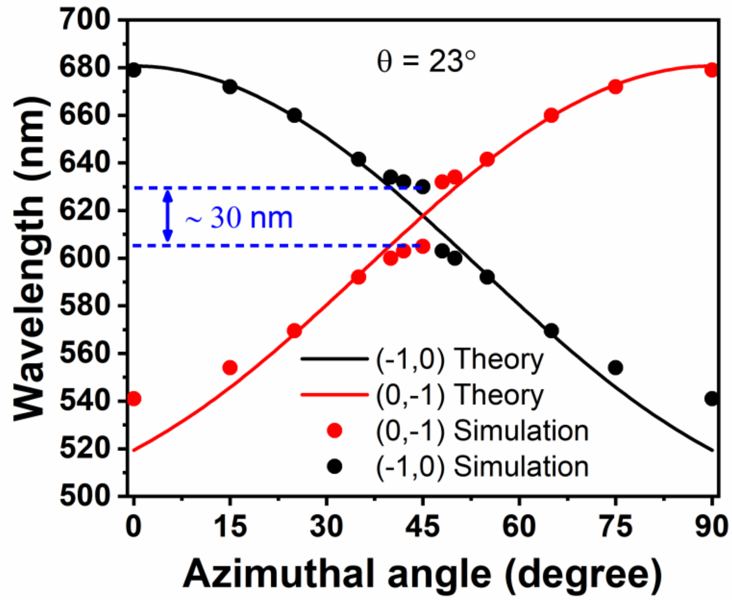


Figure S9. Theoretical curves of phase matching equation and the spectral positions of simulated SPP modes. The spectral positions of simulated (0,-1) mode (black solid circles) and (-1, 0) mode (red solid circles) as a function of the azimuthal angles The blue double arrow indicates the plasmonic band gap of ～30 nm.

S10. Contour map of Figure 4b

According to the phase-matching equation, two (0, -1) and (-1, 0) SPP modes shift monotonically from φ= 0° to φ= 90°. For example, (0, -1) SPP mode continues the redshift from φ= 0° to φ= 90° (Fig. S10a), instead of first proceeding with the redshift and then turning towards the blueshift after φ= 45° (Fig. S10b).

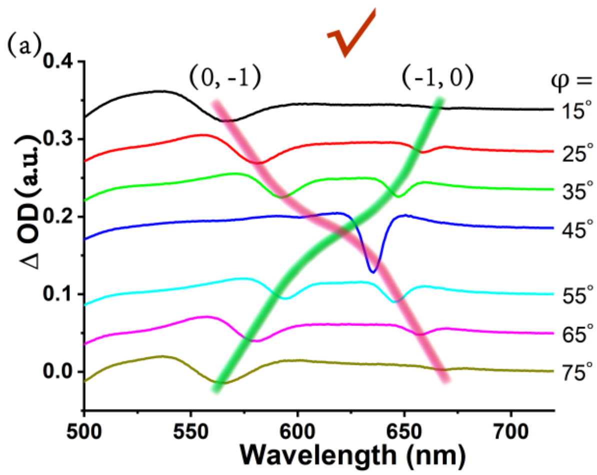


Figure S10. Illustration of modes evolution represented by contour map for Figure 4b.

S11. Field profiles under TM polarization

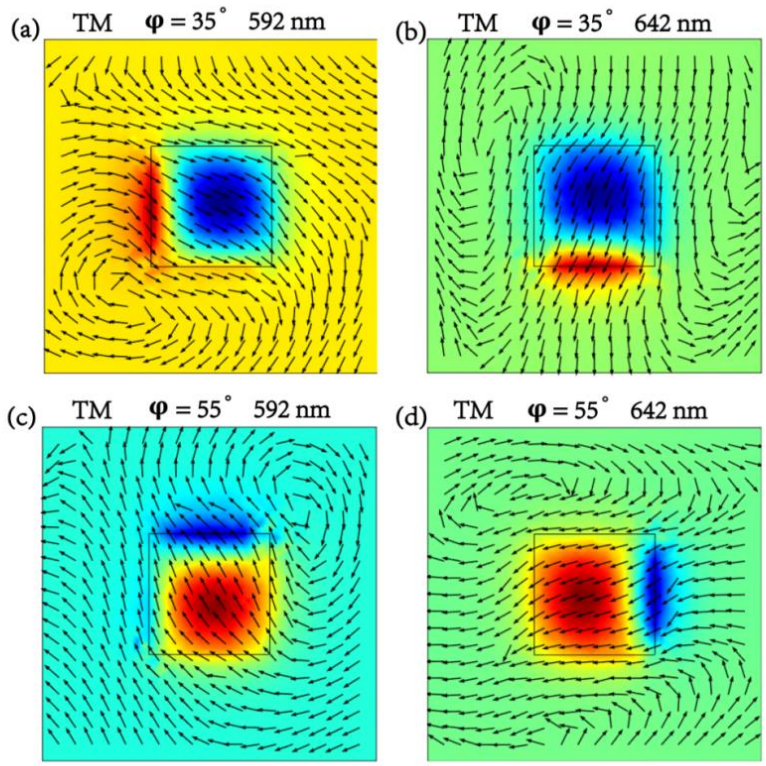


Figure S11. Electric field pattern of Ez and magnetic field vector distribution of Hx, Hy under TM polarization The azimuthal angles are 35° and 55° in (a, b) and (c, d), respectively. Note that: All intensities are normalized by the same scale bar.

REFERENCES

1. J. Zhu, L. Zhang, Y. Bai, H. Liu, N. Feng, J. Zhou, B. Zeng, T. Lin, and Q. H. Liu, "Simultaneous Fabrication of Two Kinds of Plasmonic Crystals by One Nanoimprint Mold," IEEE Photon. Technol. Lett. **29**, 504-506 (2017).
2. B. Ung and Y. L. Sheng, "Interference of surface waves in a metallic nanoslit," Opt. Express **15**, 1182-1190 (2007).