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Revealing the plasmon coupling in gold nanochains directly from the near field

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Experimental and numerical simulation methods

Sample fabrication

The nanochain samples were fabricated on an Nb-doped TiO_2 (110) substrate by Electron Beam Lithography (EBL) followed by metal sputtering and lift-off techniques. The substrates were sequentially cleaned with acetone, methanol, and ultra-pure water in an ultrasonic bath. Then the EBL resist (ZEP520A) diluted with a ZEP-A thinner (1:1) was spin-coated onto the substrates at 1000 rpm for 10 s and at 4000 rpm for 90 s. After that, the substrates were prebaked on a hot plate for 2 min at 150 ℃. Patterns of nanochains were formed by using a high-resolution electron-beam lithography system (ELS-F125; Elionix) operating at 125 kV. After development in ZED-n50, a 2 nm-thick titanium adhesion layer was first deposited via sputtering (MPS-4000, ULVAC), followed by deposition of a 36 nm-thick gold film. Lift-off was done by successively immersing the samples in anisole, acetone, methanol, and ultra-pure water in an ultrasonic bath with 5 mins for each solution.

PEEM measurements

A PEEM system (PEEM-III, Elmitec) was employed to investigate the near-field properties of the Au nanochains. The main excitation source is a Ti:sapphire femtosecond laser oscillator (Tsunami, Spectra-Physics), which delivers 100-fs laser pulses with the central wavelength tunable from 700 nm to 930 nm at a repetition rate of 77 MHz. The laser beam was focused onto the sample surface at an incidence angle of 74° from the surface normal with a focal spot of approximately 40 μm × 150 μm using a lens (focal length f =150 mm). An additional Hg lamp was used to characterize the morphologies of the structures via a linear photoemission process and to provide the guidelines to determine the location of the plasmonic hot spots. The near-field spectral properties were obtained by integrating the photoemission yield over the whole field of view (FOV=10 μm) at different excitation wavelengths. The PEEM images under femtosecond laser excitation could be regarded as nonlinear mapping of the nanostructures.

Finite-difference time-domain (FDTD) simulations

Numerical simulations of the near-field distribution and the surface charge distribution of the gold nanochains were performed using the FDTD software package Lumerical. The Nb-doped TiO_2 substrate was assumed to have a constant refractive index of n= 2.6. The optical properties of gold were obtained using the data from Johnson and Christy. A plane wave light source was incident on the nanochains at an incidence angle of 74°. For the excitation of the L-modes, the in-plane wave vector and polarization are along the chain axes. A nonuniform mesh grid was used for discretization, but a 2-nm-resolution overlaid mesh was used over the whole nanochain. The other simulation conditions were chosen according to the experimental parameters.

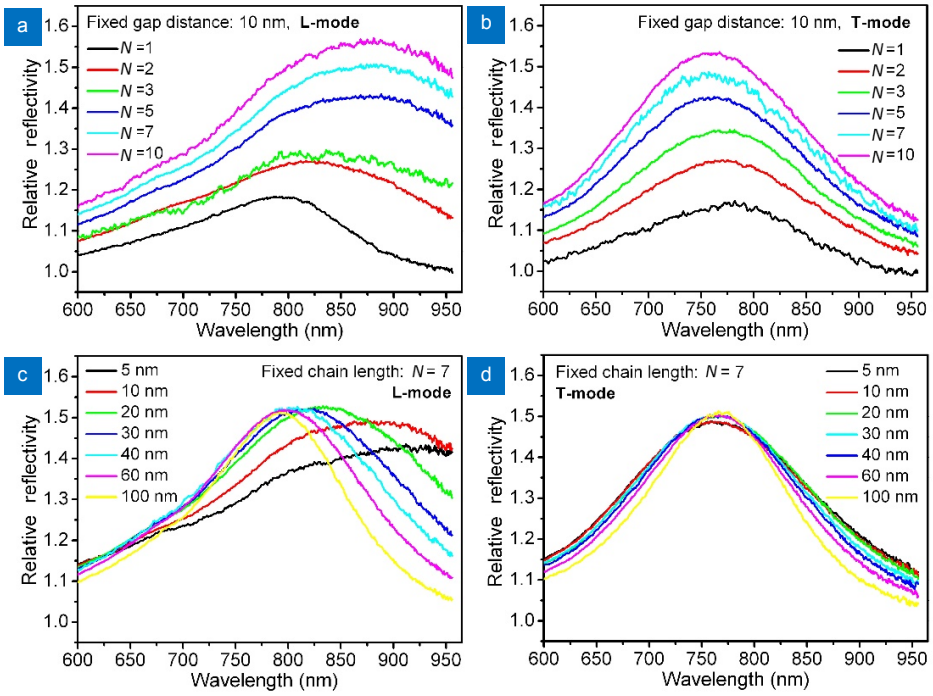

Fig. S1 | Far-field reflection spectra of the nanochains with different (a,b) chain lengths and (c,d) gap distance for both L-mode (a,c) and T-mode (b,d).

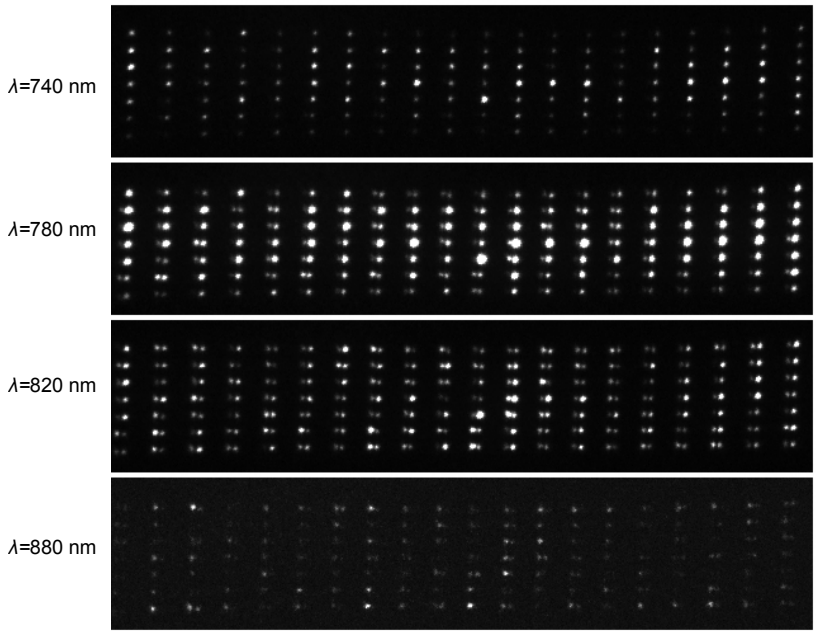

Fig. S2 | PEEM images of an array of nanochains (*N*=7, gap distance: 100 nm) under T-mode excitation at different wavelengths. Noting that in our experiments no observable photoemission with the excitation of s-polarized laser pulses, in order to excite the T-mode LSPR, we still use the p-polarized light but rotate the sample by 90°.