

# **Supporting Information**

S1. Scheme of the PL measurement setup

Fig. S1 Scheme of the PL measurement setup.

Fig. S1 is the scheme of PL measurement setup. The laser power is adjusted by an attenuator and monitored by a power detector, the polarization of the laser is controlled by using a group of Glan–Taylor prism and half-wave plate, and a band pass filter centered at 780 nm is used to clear the fluoresces from the laser cavity. The laser beam is focused onto the sample by a lens with 75 mm focal length, and the PL signal is recorded by using a CCD camera (Newton 970) through a monochromator (Andor, SP-970), in front of which a 785 nm notch filter is used to block the scattered laser. The exposure time of the spectrum is 0.2 s.

## S2. Calculation of peak power intensity from the incident power

For the incident power of 180 mW as an example, considering the incident angle of 65 degrees, the laser spot size of  $6.98 \times 10^{-10}$  m<sup>2</sup>, and the average power density of 25.8 kW/cm<sup>2</sup>, the peak power density is calculated to be 2.61 GW/cm<sup>2</sup> with the following formula,

$$P_{peak} = \frac{P_{average}}{(f \times L)}$$

where f=76 Mhz, L=130 fs.

Table S1, list of the average powers and their corresponding peak power intensities.

Paverage (mW)	100	102	140	170	180	
Average power density (kW/cm <sup>2</sup> )	14.4	14.6	20.1	24.4	25.8	
Peak power density (GW/cm <sup>2</sup> )	1.45	1.48	2.03	2.47	2.61	

#### **S3.** Dark noise background of the transient PL measurement.



Fig. S2 Dark noise background of the transient PL measurement with integration time of 60 s and 600 s, respectively.

Fig. S2 gives the dark noise background of the transient PL measurement by using the time-correlated single photon counting (TCSPC) system as shown in Fig. S1. The left and right panels of Fig. S2 present the dark background data obtained with the integration times of 60 s and 600 s, respectively. For the transient PL data given in Fig. 3 of the main context, the contribution of the background was deducted directly by subtracting the counts from background to that from the samples.

#### S4. Excitation polarization dependence of longitudinal SPR of Au-Al<sub>2</sub>O<sub>3</sub>-Al.



Fig. S3 Excitation polarization dependence of longitudinal SPR of Au-Al\_2O\_3-Al

Fig. S3 gives excitation polarization dependence of longitudinal SPR of Au-Al<sub>2</sub>O<sub>3</sub>-Al. As the polarization angle increases from 0 to 80 degrees,  $f_E$  decreases from 29.4 to 0.9, which implies that the longitudinal SPR is hard to be excited by an *s*-polarized laser.



### S5. Cross-section SEM image of Au-Al<sub>2</sub>O<sub>3</sub>-Al and TEM image of Au nanorods

Fig. S4 Cross-section SEM image of Au-Al<sub>2</sub>O<sub>3</sub>-Al sample which has Au diameter of  $\sim$ 13 $\pm$ 1 nm. (a) Cross-section SEM image of AAO membrane. (b) TEM image of Au nanorods. The length and average diameter of Au nanorods are 34 $\pm$ 2 nm and 13 $\pm$ 1 nm, respectively, the corresponding aspect ratio is about 2.8.



## S6. Extinction and PL spectra of Au-Al<sub>2</sub>O<sub>3</sub>-Al under different excitation conditions.

Fig. S5 Extinction and PL spectra of Au-Al<sub>2</sub>O<sub>3</sub>-Al excited at different laser wavelength and laser status. (a) is the extinction spectrum and the corresponding PL spectrum of Au-Al<sub>2</sub>O<sub>3</sub>-Al under the  $P_{exc}$  of 2.61 GW/cm<sup>2</sup>. The length and diameter of Au nanorod is about  $34\pm2$  nm and  $13\pm1$  nm, respectively. (b) is the PL spectra of Au-Al<sub>2</sub>O<sub>3</sub>-Al excited by both pulsed and continuous wave (CW) laser beams. (c) and (d) are PL spectra of Au-Al<sub>2</sub>O<sub>3</sub>-Al excited by a pulsed laser at 730 nm and 830 nm, respectively. The excitation laser here was *p*-polarized.

Fig. S5 (a) presents the extinction and PL spectra of Au-Al<sub>2</sub>O<sub>3</sub>-Al excited by a 782 nm laser under the  $P_{exc}$  of 2.61 GW/cm<sup>2</sup>. The periods of the oscillations appear in PL spectrum agree well with that of the extinction spectrum of Au-Al<sub>2</sub>O<sub>3</sub>-Al, and this oscillations originate from the interference between the light reflected from the top and bottom surfaces of the AAO membranes.<sup>1</sup> Fig. S5 (b) gives the PL spectra of Au-Al<sub>2</sub>O<sub>3</sub>-Al excited by both pulsed and CW laser beams, the results show that the broadband SC only emerges under the excitation of pulsed laser which has stronger peak intensity (2.61 GW/cm<sup>2</sup>) than that of the CW one (25.8 KW/cm<sup>2</sup>). Fig. S5 (c) and (d) show the SC spectra under the  $P_{exc}$  of 2.61 and 1.48 GW/cm<sup>2</sup>, respectively. The results demonstrate that the SC could be excited in a wide excitation wavelength range.

### S7. Fabrication details of Au-Al<sub>2</sub>O<sub>3</sub>-Al samples

The treated Al sheets (99.99%) were exposed to a  $H_2SO_4$  acid (0.3 M) solution under a constant voltage of 19 V in an electrochemical cell at a temperature about 4 °C. The aluminum oxide layer produced by the first anodization step was removed by wet-chemical etching in a mixture of phosphoric acid and chromic acid. The thickness of the barrier layer between the end of the pores and the Al substrate was about 6 nm which was controlled by decreasing the thinning voltage to 5 V step by step during the thinning process. The lengths of the Au nanorods in different samples were controlled by changing the deposition time. The underlying Al substrate was dissolved by a  $CuCl_2(0.44M)$  and HCl(1.2M) solutions.

S8. Transmission spectra of band-pass (BP) filters and Notch Filters (NF).



Fig. S6 Transmission spectra of band-pass (BP) filters and notch filters (NF) used in the experiments. (a)-(e) are filters of BP780, NF785, BP730, SW650, BP830 and NF830, respectively.

#### references

1 Y.-L. Wang, Z.-J. Yang, Z.-S. Zhang, X.-N. Peng, L. Zhou, Z.-H. Hao and Q.-Q. Wang, *The Journal of Physical Chemistry C*, 2014, **118**, 16060-16066.