Electronic Supplementary Material (ESI) for Nanoscale.

Supplementary Information

High-Q, Low-Mode-Volume and Multiresonant Plasmonic Nanoslit Cavities Fabricated by Helium Ion Milling

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S1. An SEM image of a 10 nm nanoslit cavity

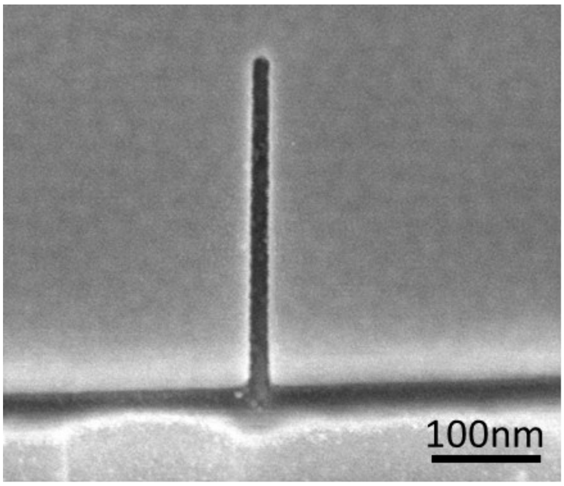

Figure S1. SEM image of a 10×300 nm nanoslit cavity.

S2. Dispersion curves of nanoslit cavities

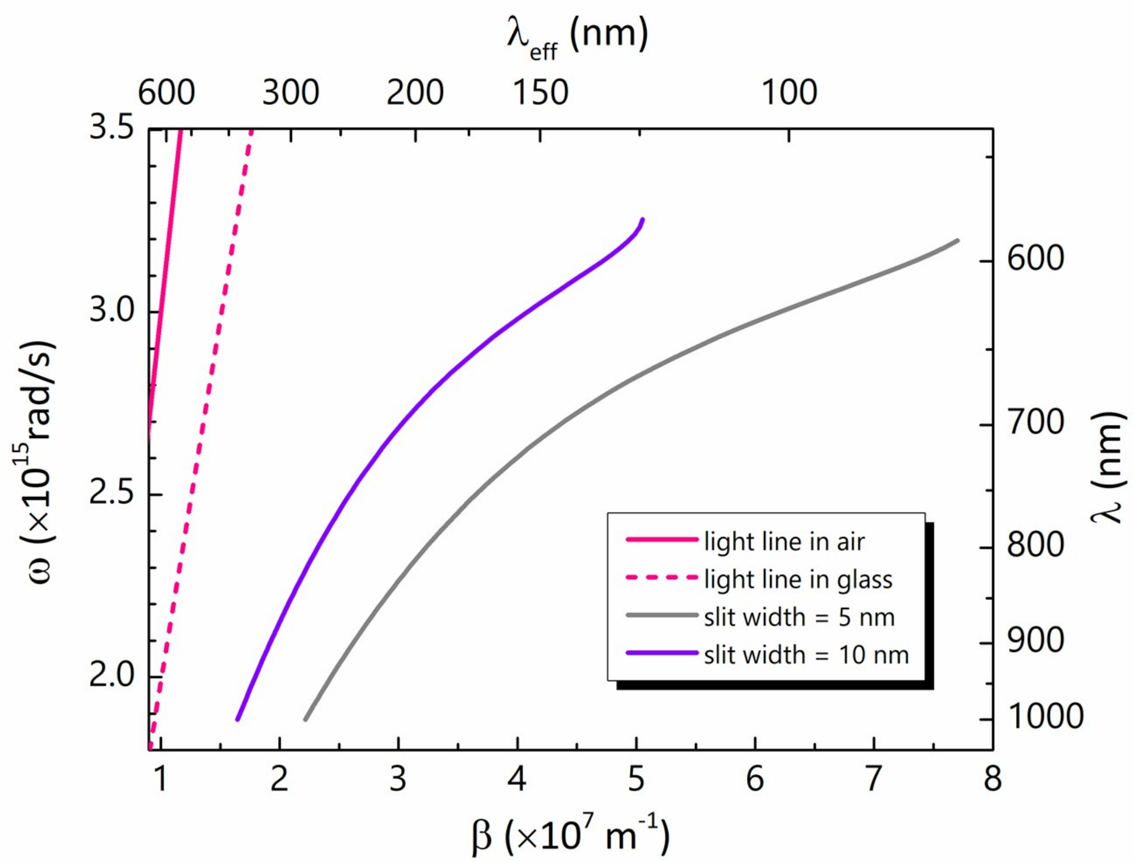

Figure S2. Mode dispersion for both 5 and 10 nm nanoslit cavities. 5 nm cavities exhibit smaller effective wavelength than 10 nm cavities. The curves were obtained using software FDTD MODE Solutions and no higher-order modes were detected in the simulations.

S3. Illustration of optical setup

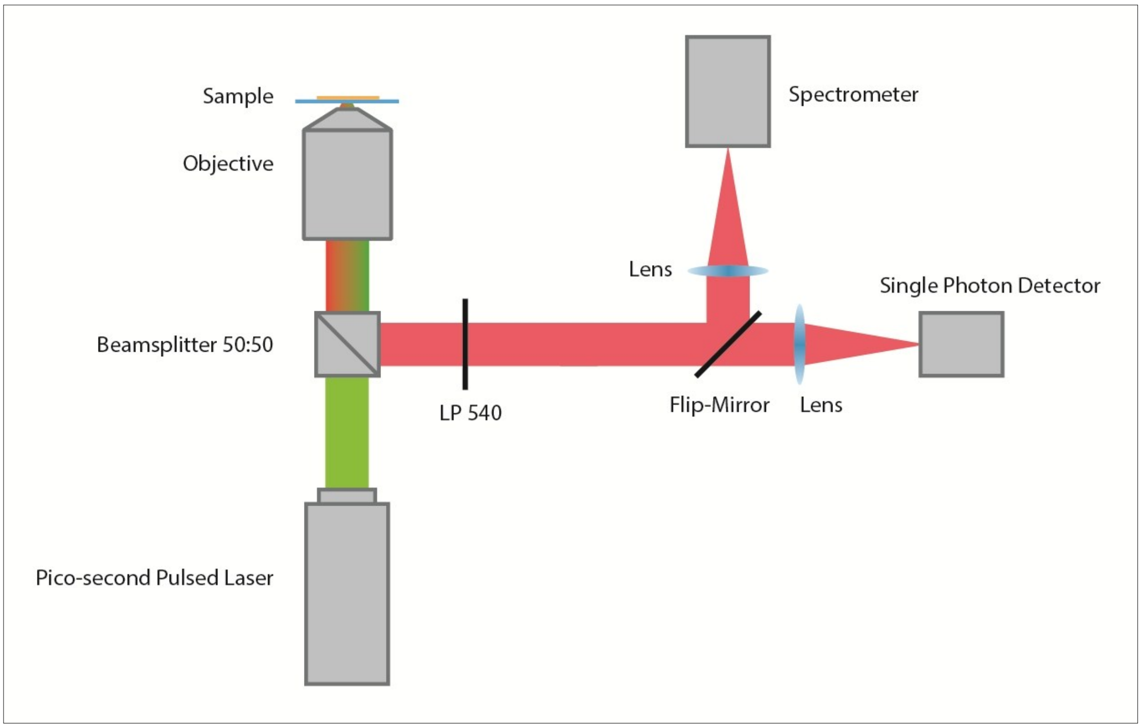

Figure S3. Sketch of the optical setup. The light of the pulsed laser source passes a 50:50 beamsplitter before entering the objective. The focused light excites the nanoslit cavities and the emitted PL is collected via the same objective. The PL light is then reflected off the beamsplitter and any residual excitation light is blocked by a longpass filter. Afterwards, the remaining light is detected via a single photon detector or, by using a flip mirror, analyzed with a spectrometer.

S4. 1PL Spectra processing

For each nanoslit cavity, the original raw PL signal was collected near its open end and a background signal was collected from a nearby point on the strip edge. Figure S4 shows an example of how we processed the PL spectrum of a 10 nm wide and 180 nm long nanoslit. The background signal (Figure S4a, red curve, PL signal from the edge) shows a very broad band centering around 640 nm without any features. By contrast, the raw PL from the nanoslit (Figure S4a, black curve) shows characteristic plasmon peaks on top of the broad background PL. We subtracted the background PL from the raw PL of the nanoslit and the processed spectrum is shown in Figure S4b. The 1st and 2nd order resonance peaks of the nanoslit are clearly displayed. The spectra shown in Figure 2 are the results of 10-point FFT smoothing performed with Originlab Pro software. It is noted that the processed spectrum shown in Figure S4b still shows a small background because it is not possible to completely remove the contribution from the edge and Au strip. Therefore, each processed PL spectrum still exhibits variable background.

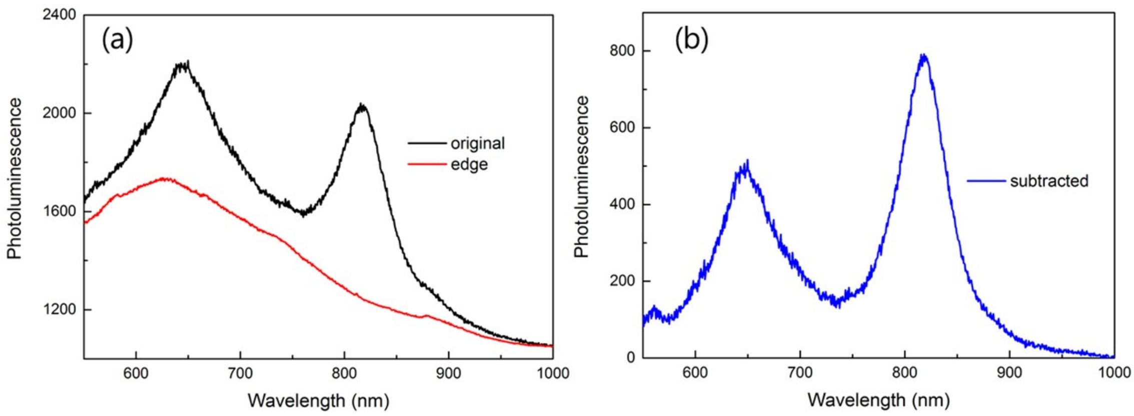

Figure S4. (a) Raw PL spectrum (black) from a 10nm wide, 180 nm long nanoslit and the spectrum from nearby edges (red). (b) Processed spectrum of the nanoslit by subtracting the edge spectrum from the raw nanoslit spectrum.

S5. Extraction of the Q-factor for each spectrum

To extract the Q-factors of the plasmon resonances, we fit the resonance peaks with multiple Lorentz functions in the frequency domain and calculated the Q-factor as the ratio between the resonance frequency and the peak's FWHM. As shown in Figure S4, the processed spectrum still exhibits a small variable background. Therefore, the processed spectra underwent further baseline corrections as shown in the following two examples.

Figure S5 shows the peak fitting to the PL spectrum of the 10 nm wide, 180 nm long nanoslit cavity using OriginPro software. A small baseline was manually subtracted from the original spectrum as shown in Figure S4b. The black line represents the processed PL spectrum while the blue and green lines represent the Lorentz fitting peaks to the two resonant modes. The red line displays the sum of the two fitted peaks showing a good resemblance to the original spectrum (black line). From the fitting parameters, we derived that the Q factors for the 1st (green curve) and 2nd order (blue curve) modes are estimated at 19 and 8, respectively.

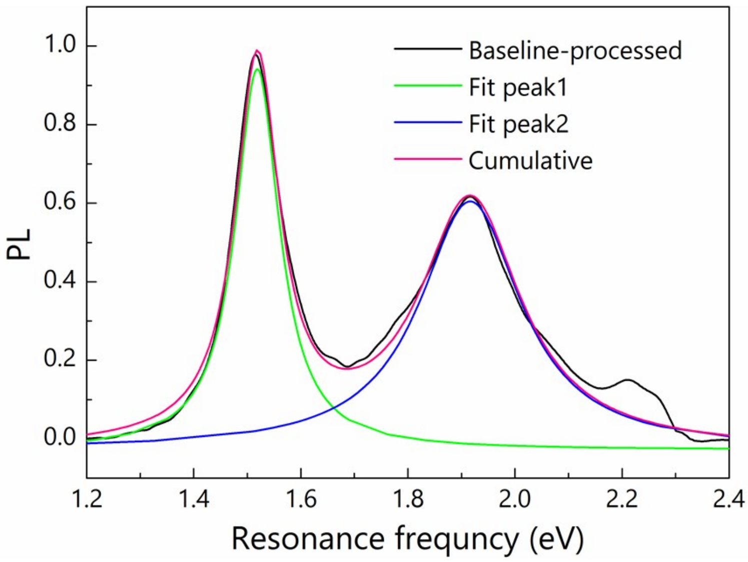

Figure S5. Lorentz fitting to the processed PL spectrum in the frequency domain for a 10 nm wide, 180 nm long nanoslit cavity. The blue and green curves represent the Lorentz fitting to each individual resonance peak and the red line is the sum of the two fitting curves.

As we mentioned earlier, the PL spectrum from each nanoslit cavity still shows variable background even after the processing demonstrated in **Supplementary**

Information S4. The 180 nm long nanoslit shows a very small background after processing as shown in Figure S4b. For other nanoslits, the background can be relatively large. Therefore, a baseline correction is applied in order to extract the Q-factors of the plasmon resonances, which is illustrated in Figure S6 for the 10 nm wide, 300 nm long nanoslit cavity. The baseline correction and the subsequent Q-factor extraction were both done in OriginPro software. The baseline, as shown in Figure S6a, was created by choosing several anchor points on the spectra in the range away from the resonances. After the baseline subtraction, the spectrum was fitted with Lorentz functions and the Q-factors were extracted: 22 and 13 for the two resonance peaks (2nd and 3rd order modes) in Figure S6b.

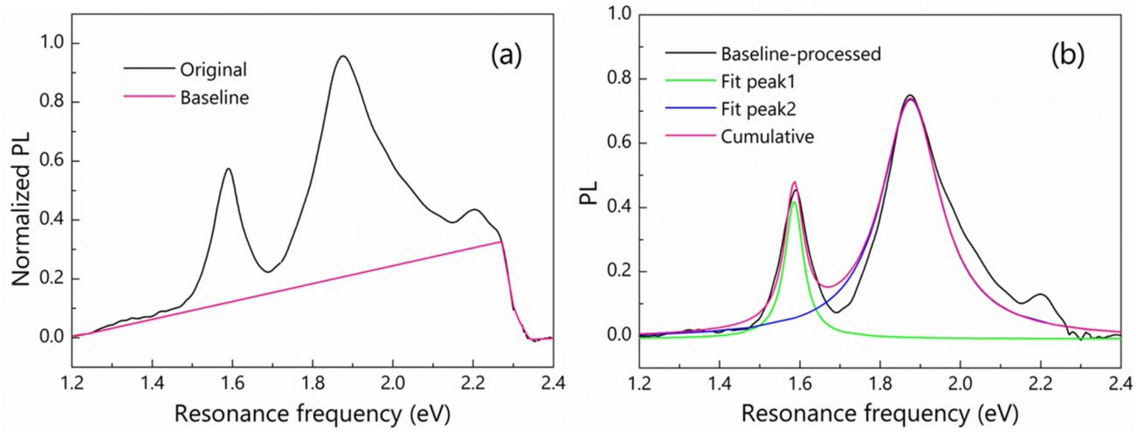

Figure S6. Extraction of the Q-factors for the 10 nm wide, 300 nm long nanoslit cavity. (a) The original PL spectrum and the baseline to be subtracted. (b) Lorentz fitting to the two resonance peaks. The blue and green curves are the fitting peaks for the two resonance peaks and the red curve is the cumulative sum of the two fitting peaks.

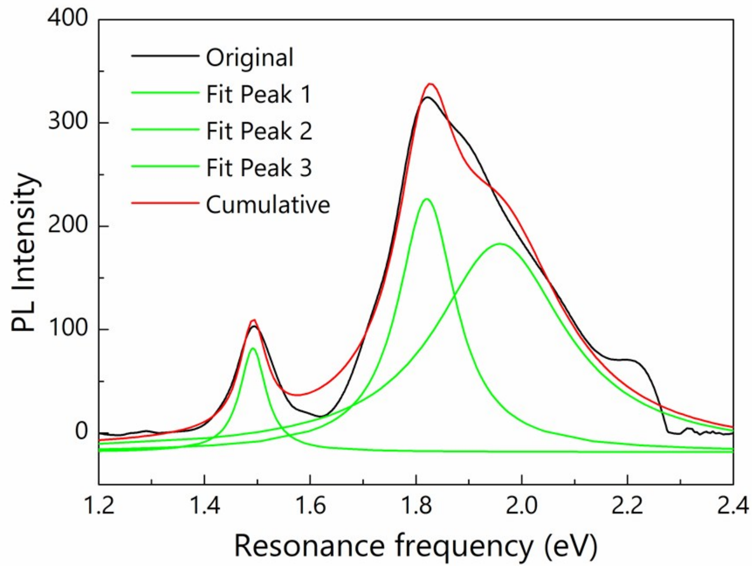

Fig. S7. Lorentz fitting to the spectrum from a nanoslit cavity: 10 nm in width and 320 nm in length. A Q-factor of 24 is obtained for the resonance peak ~1.49 eV.

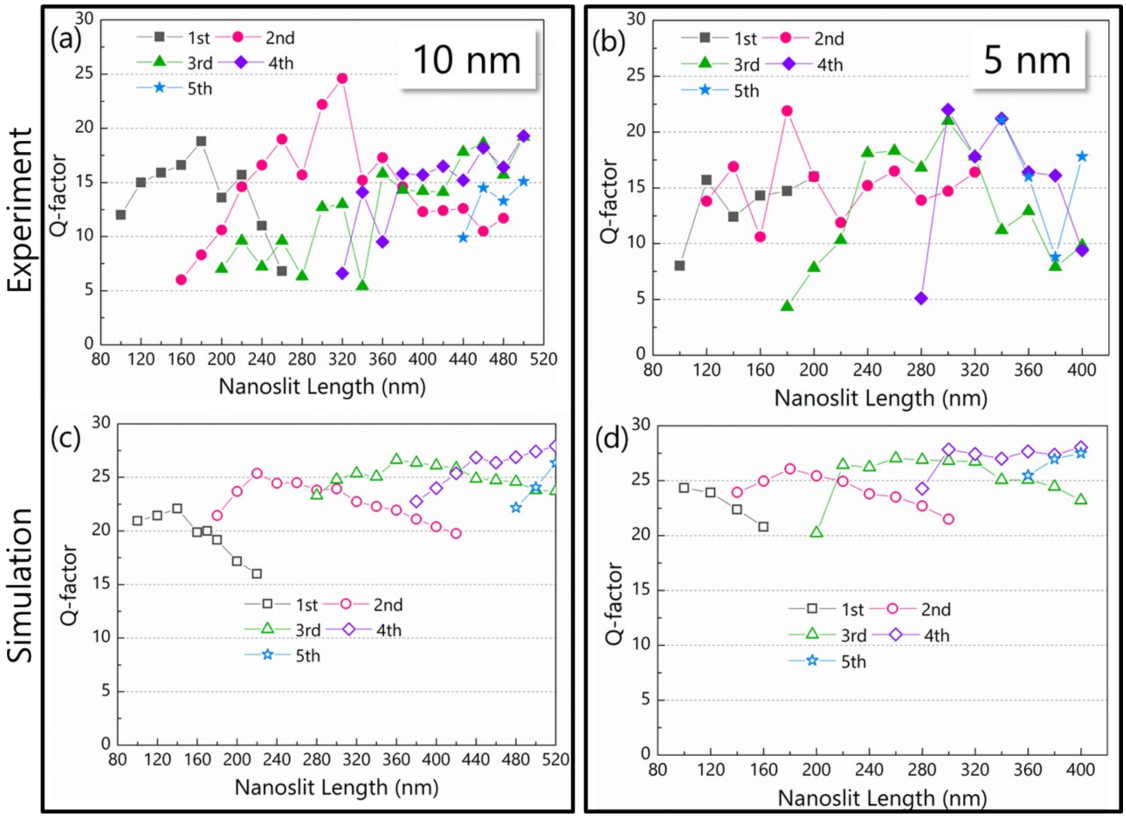

Figure S8. The Q-factors for 10 nm nanoslit cavities (left column) and 5 nm nanoslit cavities (right column). The top row shows the Q-factors obtained from the PL spectra and the bottom row shows the data obtained from simulations.

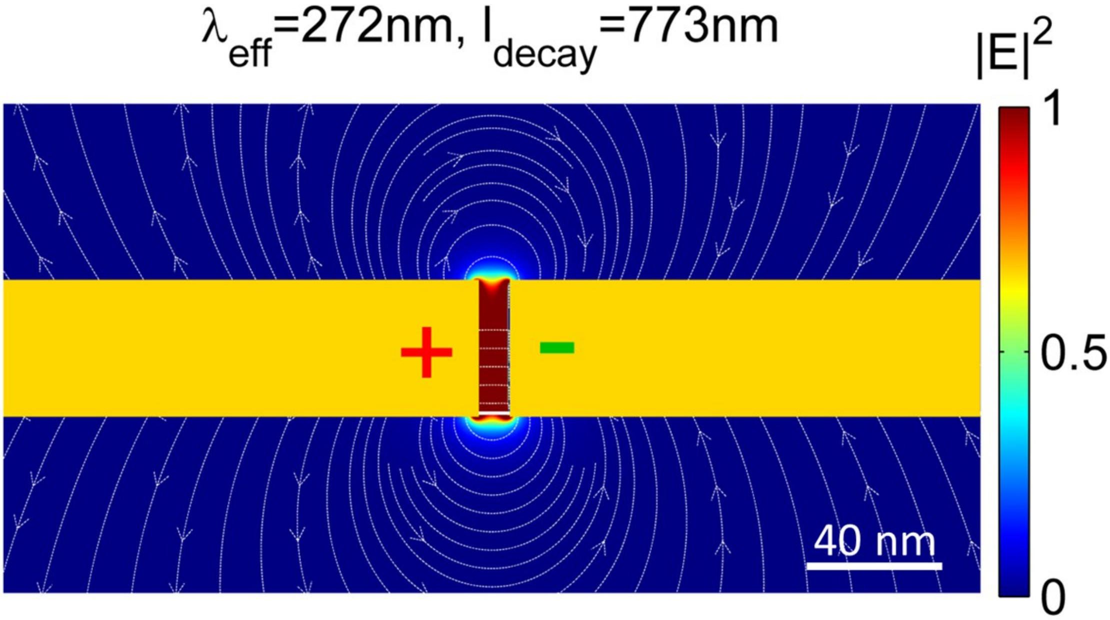

Figure S9. Modal pattern of a 10 nm wide nanoslit cavity with resonance wavelength at 800 nm. The electric field lines are almost perpendicular to the gold/air interface resulting in lower losses that help to enhance the resonant mode quality.