Supplementary Information for Fabrication of Plasmonic Arrays of Nanodisks and Nanotriangles by Nanotip Indentation Lithography and their Optical Properties

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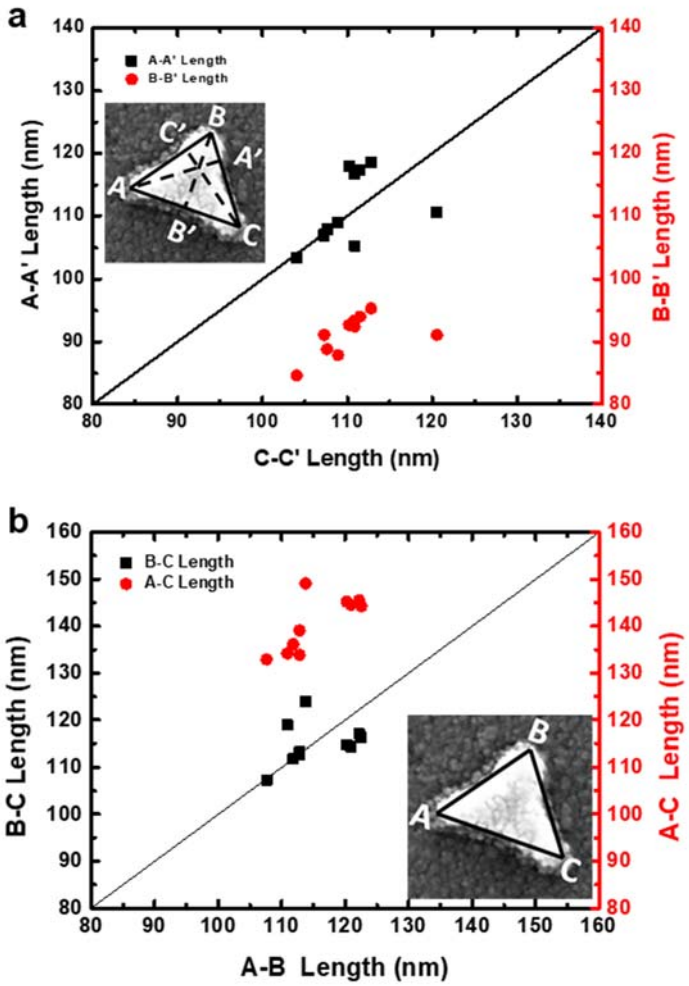

Figure S1. SEM analysis of the perpendicular line and edge lengths of the Au nanotriangle. a) Lengths of lines perpendicular to the baseline along A-A', B-B', and C-C' (inset, black dashed line) are measured and plotted as a function of the length of C-C'. Two lengths of A-A' and C-C' (black square) show a good agreement with the linear curve with a slope of one, indicating that A-A' = C-C'. The length B-B' (red circle) is approximately 20 nm shorter than the length C-C'. b) Lengths of the edge lines of the triangle along A-B, B-C, and A-C (inset, black line) are measured and plotted as a function of length A-B. Black line is an eye guide with a slope of one. Two lengths along A-B and B-C are similar (~120 nm), whereas the length along A-C is approximately 20 nm longer than the others.

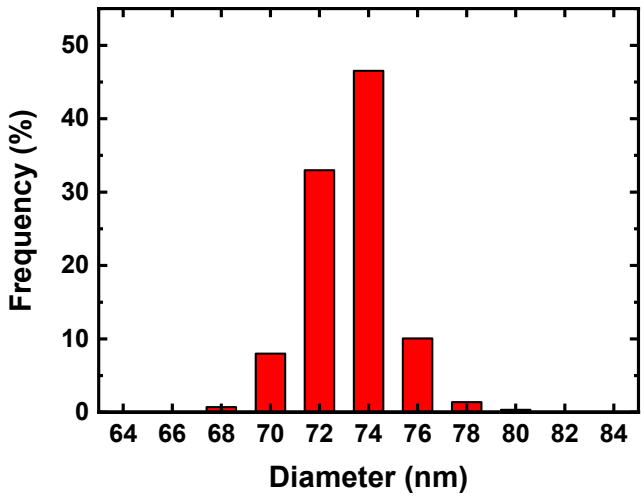

Figure S2. Histogram of diameter of Au nanodisks fabricated by NTIL using single AFM tip measured by scanning electron microscope (SEM).

Table S1. Statistics of the diameter of Au nanodisks fabricated by NTIL using single tip array by array

	Array 1 (180/900 NDs)	**Array 2** (180/900 NDs)	**Array 3** (180/900 NDs)	**Array 4** (180/900 NDs)	**Array 5** (180/900 NDs)	**Array 6** (180/900 NDs)	**Array 7** (180/900 NDs)	**Array 8** (180/900 NDs)	**Overall** (1440/7200 NDs)
Average (nm)	73.9	74.6	73.3	73.4	72.2	73.4	72.3	73.4	73.3
Standard deviation (nm)	2.2	1.3	1.1	1.2	1.2	1.4	1.6	1.4	1.6
Coefficient of variation (CV, %)	3.0	1.7	1.5	1.6	1.6	1.9	2.2	2.0	2.2
Maximum (nm)	79.0	78.0	75.7	76.1	72.3	75.4	75.1	77.4	79.0
Minimum (nm)	68.3	72.6	71.0	70.3	70.0	70.2	69.6	70.9	68.3

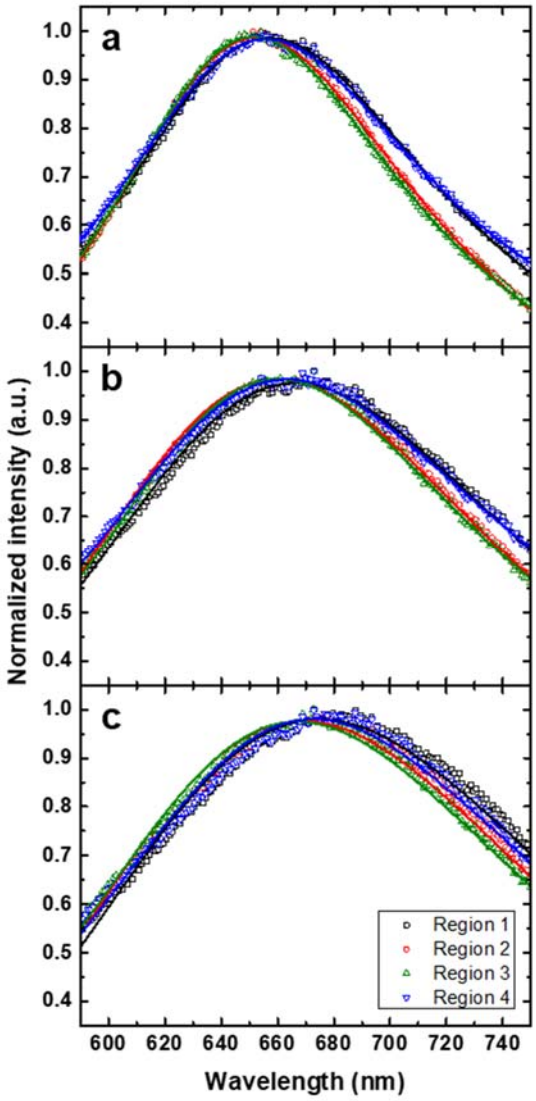

Figure S3. Spatial variation of LSPR spectra in a nanodisk array. a-c) Normalized LSPR spectra of the four regions of each gold nanodisk array fabricated with different indentation forces of 10.3 (a), 11.3 (b), and 12.4 μN (c). The solid lines represent the Lorentzian fitted curves.

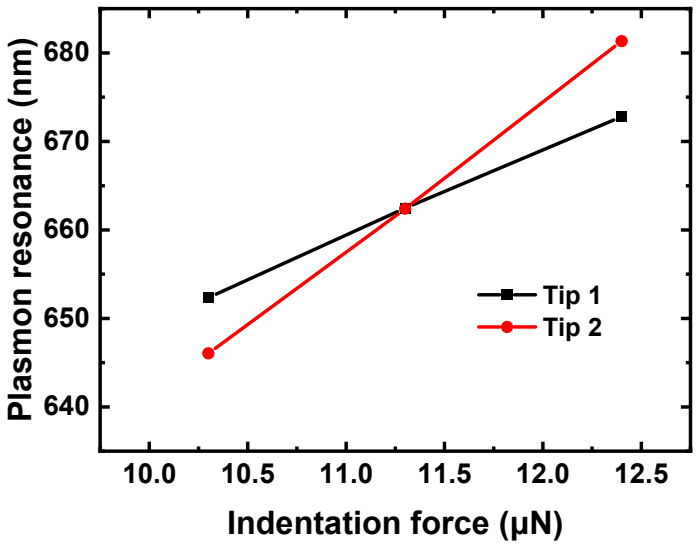

Figure S4. Localized surface plasmon resonance (LSPR) spectral peak positions of Au nanodisk arrays fabricated by NTIL as a function of indentation force. Two different AFM tips (AD-40-AS, Adama) were used for the patterning.

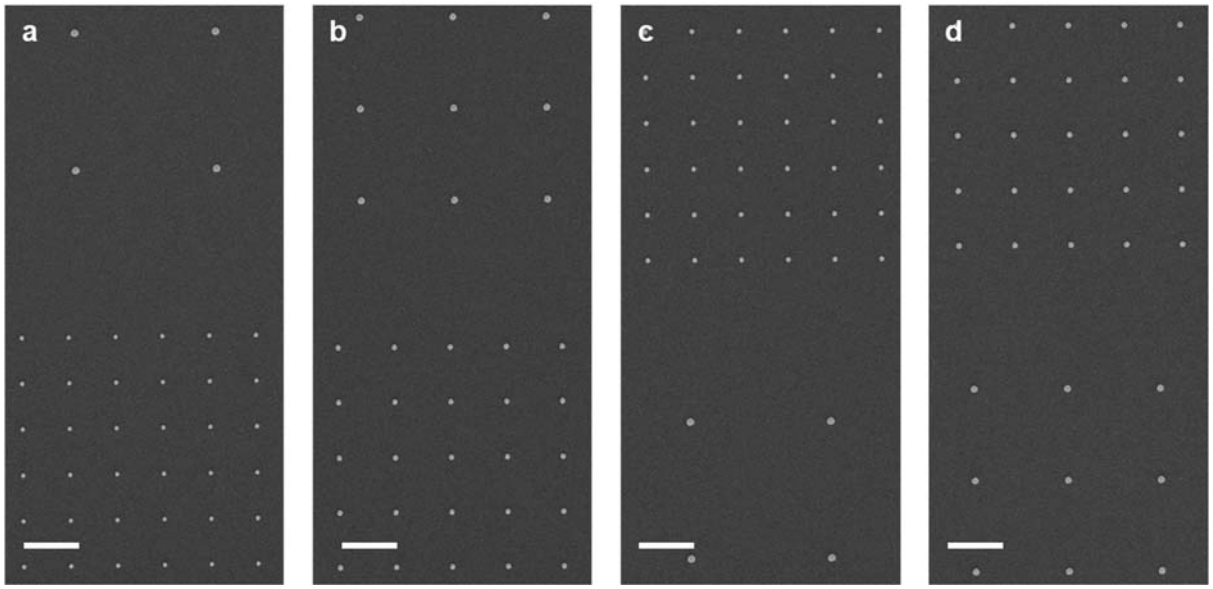

Figure S5. Scanning electron microscope (SEM) images of nanodisks arrays fabricated by NTIL with various array configurations. a-d) SEM images acquired from the selected areas of A to D in Figure 3d, respectively. Diameter sizes of nanodisks are 94 nm, 79 nm, 62 nm, and 54 nm for 2x2, 3x3, 5x5, and 6x6 array configurations, respectively. Scale bar is 2 µm.

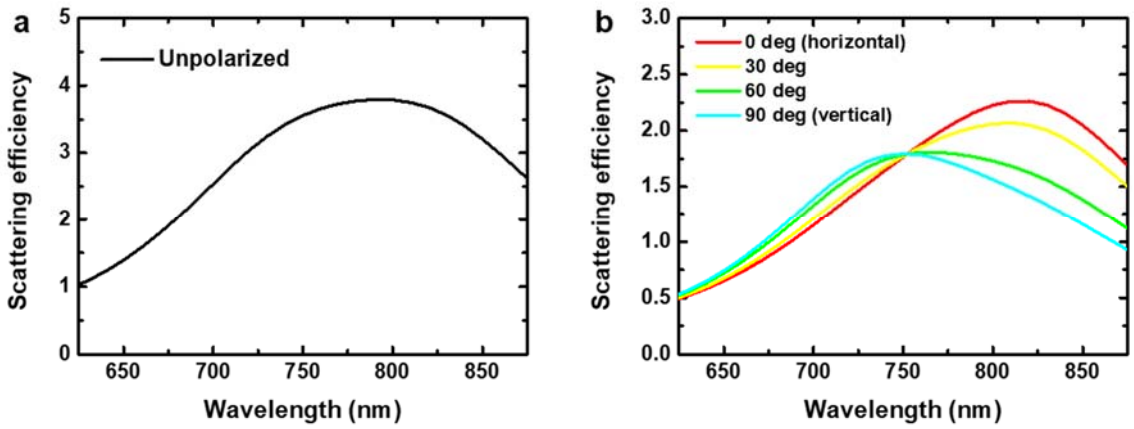

Figure S6. Calculated scattering efficiency spectra of a triangular nanostructure. a-b) Spectra with a ring-shaped unpolarized dark-field illumination (a) and with a ring-shaped polarized dark-field illumination (b). The length of the base and altitude of the triangular nanostructure were 160 and 110 nm, respectively. The incident angle was 60° with respect to the *z*-axis, and the incident plane was rotated around the center of the nanostructure with the step of 10°. With the rotation of the polarizer from 0° to 90° in b), the peak around 820 nm decreases, whereas the peak around 740 nm increases. This result shows good agreement with the experiments in Figure 4a.

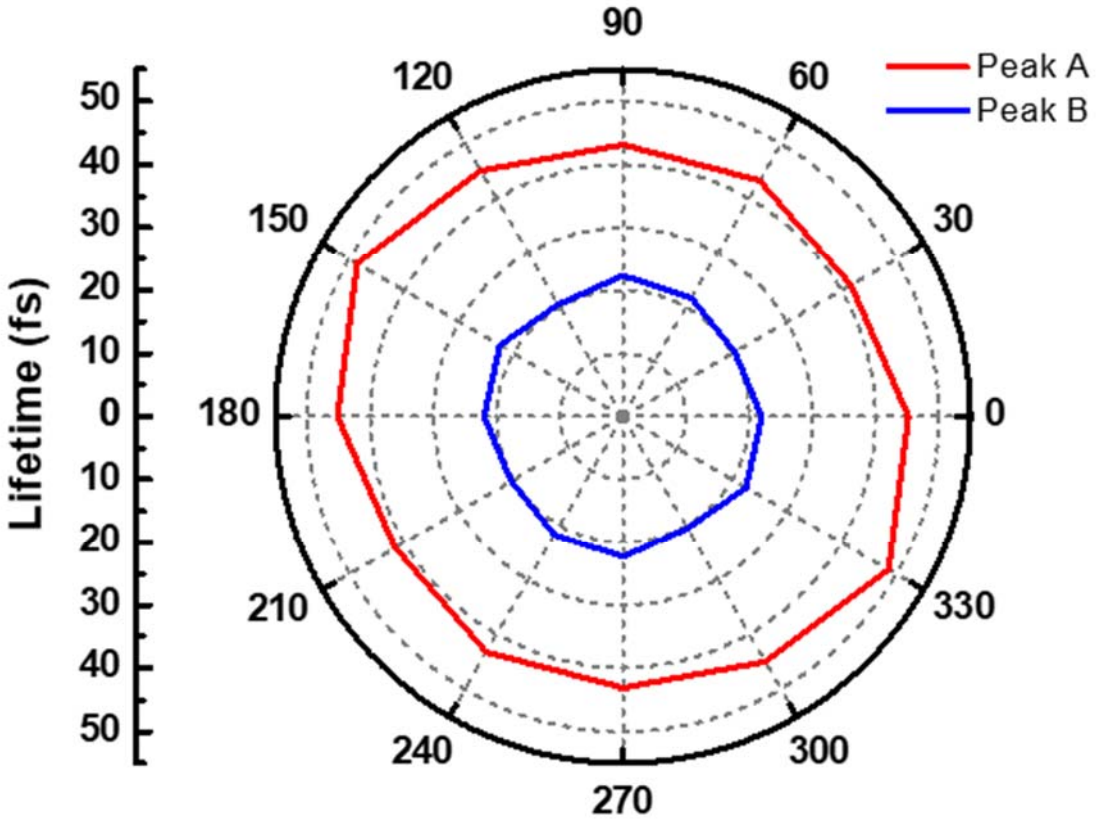

Figure S7. Polar plot of the LSPR lifetime in the Au nanotriangle dependent on the incident polarization. LSPR lifetime was extracted from the peak width in the dark-field optical scattering measurement. Spectral positions of Peak A and Peak B correspond to 820 and 730 nm, respectively, as shown in Figure 4a.

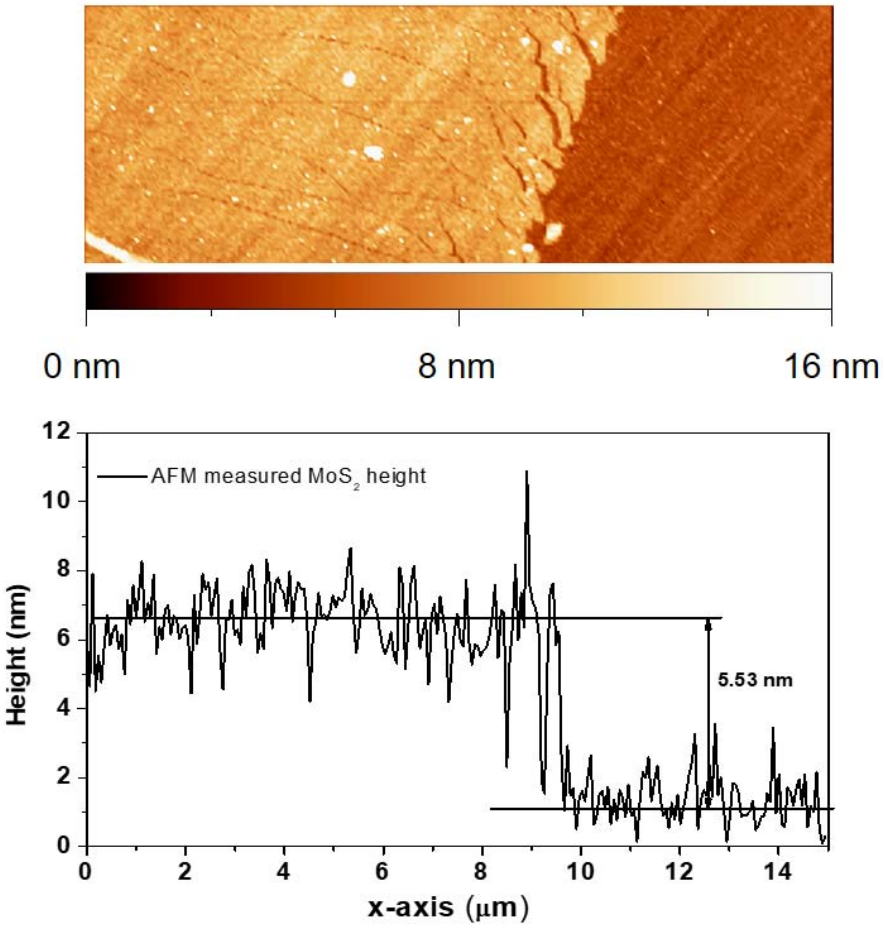

Figure S8. AFM thickness measurement of a few-layer MoS_2 film transferred onto the substrate with NTIL-fabricated nanostructures. (top) AFM morphology of the MoS_2 film. The scan range was 15 x 5 μm. The color bar indicating the measured thickness is shown below. (bottom) Cross-section profile of the AFM morphology along the *x*-axis.

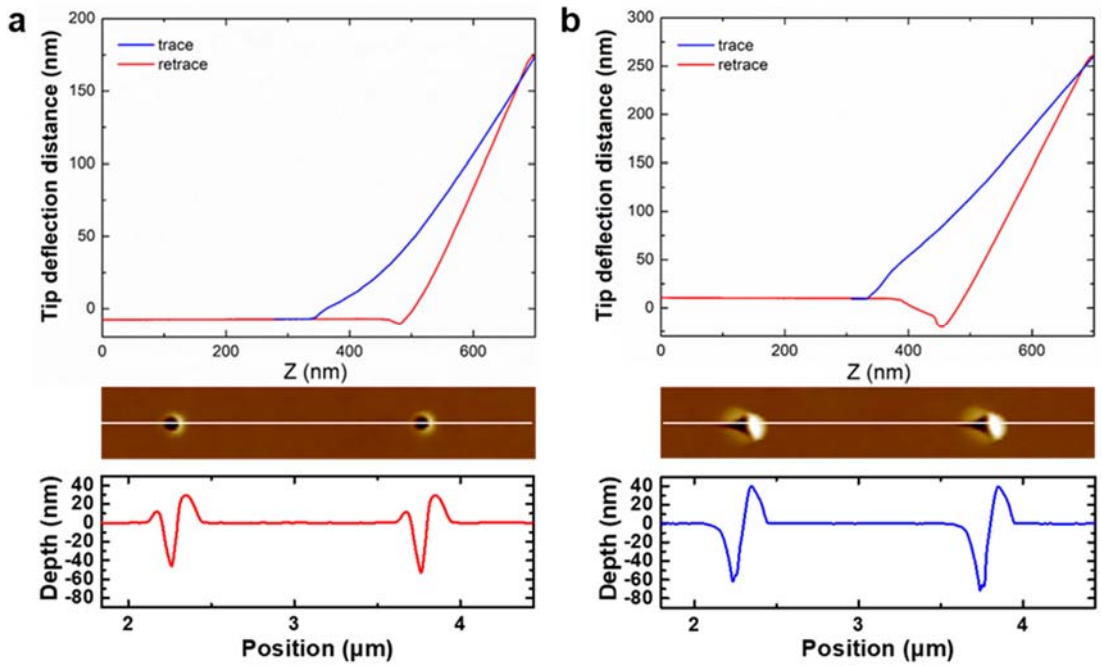

Figure S9. Force–distance curve, indented AFM image, and its cross-section profile. a) Circular shape in a mask layer with a force of approximately 7.2 µN (tip deflection distance 180 nm, spring constant 40 N m^{-1}, AD-40-AS). b) Triangular shape in a mask layer with a force of approximately 10.5 µN (tip deflection distance 250 nm, spring constant 42 N m^{-1}, PPP-NCHR).

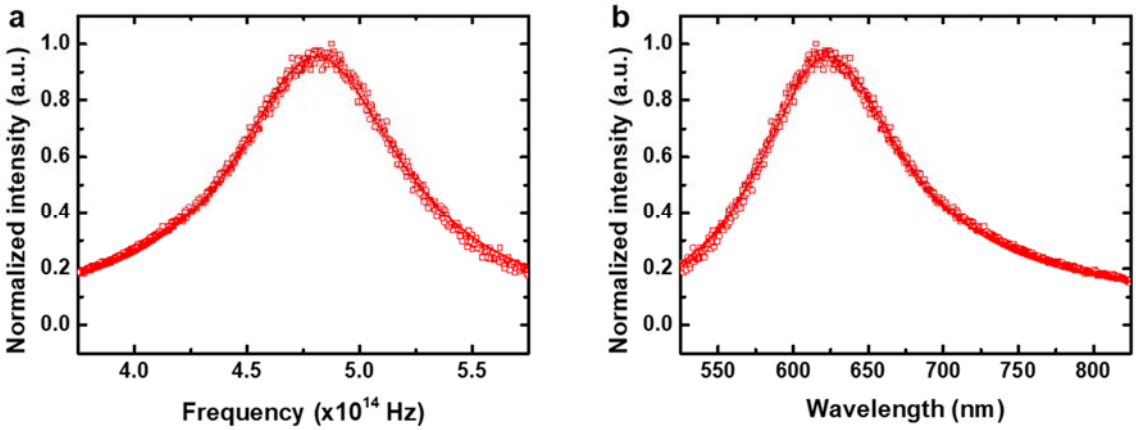

Figure S10. Representative optical scattering spectra of a gold nanodisk array. a-b) The spectra of the frequency domain (a) and its corresponding wavelength domain (b). The solid lines represent the Lorentzian fitted curves in the frequency domain.