

Electronic Supplementary Information

for

Facile three-dimensional nanoarchitecturing of double-bent gold strips on Roll-to-Roll nanoimprinted transparent nanogratings for flexible and scalable plasmonic biosensors

1. Fabrication of double-bent gold strips

Double-bent gold (Au) strips (DAS) were fabricated by two main steps; the transparent nanograting structure was first imprinted on a PET substrate using a UV-curable polymeric resin, and then the Au layer was deposited over the nanograting at a specific angle. For nanoimprinting lithography (NIL), a custom-built 15"-wide roll-to-roll (R2R) NIL system (roll width ~ 400 mm, rolling speed up to 1 m/min,) was used. To prepare a flexible stamp (mold) that wraps around an imprinting roll, a desired pattern (nanograting) was transferred from a rigid master mold to a thin polydimethylsiloxane (PDMS) pad. Details on the rigid mold fabrication [1] and pattern transfer process [2] can be found elsewhere. A UV-curable polyurethane acrylate (PUA; MINS-311RM, Minuta Tech) resin was airbrushed on a clean PET; the resin thickness can be controlled by the PUA concentration (25-100 %) and airbrushing time (3-5 seconds). The PUA-coated PET substrate was then conveyed along the substrate roll. The PDMS mold-attached imprinting roll and the PUA-coated substrate roll make conformal contact to each other at a controlled pressure (~ 1 bar). As rolling proceeds, the PUA nanograting is continuously created on a PET substrate, which is instantly cured by the UV light (365 nm peak wavelength) at the outlet of a contact zone. To complete DAS, a 30-nm-thick Au layer was then thermally evaporated (UTE-01, Ultec Co., Ltd.) at three different angles (5°, 35°, and 50°) on the fully-cured PUA nanograting by using an angle-adjustable stage for sample mounting.

2. Electromagnetic simulation

Electromagnetic simulations were performed using two-dimensional (2D) finite-difference time-domain software (Lumerical FDTD solution 8.9). For the calculation of extinction, absorption, and scattering cross sections in Figure 2, simulation mesh was set to a 0.1-nm square grid. Perfectly matched layers and periodic boundaries were used as a boundary condition for vertical and lateral direction. The periodicity of the nanostructures was set to 200 nm. The direction of incident light is from top to bottom with the electric field polarized in the lateral direction. The two 2D monitors to get the reflected power into the air and the transmitted power into the backside of the nanostructure were placed at 250-nm above and 100-nm below the substrate surface, respectively. Figure S1 shows the schematic illustration of the FDTD geometry, the modeled nanostructures used for Figure 3 and the squared magnitude of the local electrical field amplitude around the double-bent gold stripes. Figure S2 shows the simulation geometry for modeling the nanostructure shown in Figure 2(a) and its calculated extinction cross section. The thicknesses of Au films in Figure S1(a) and Figure S2(a) were set to 20 nm and 30 nm, respectively, to consider the thickness change from the oblique-angle incidence of Au flux.

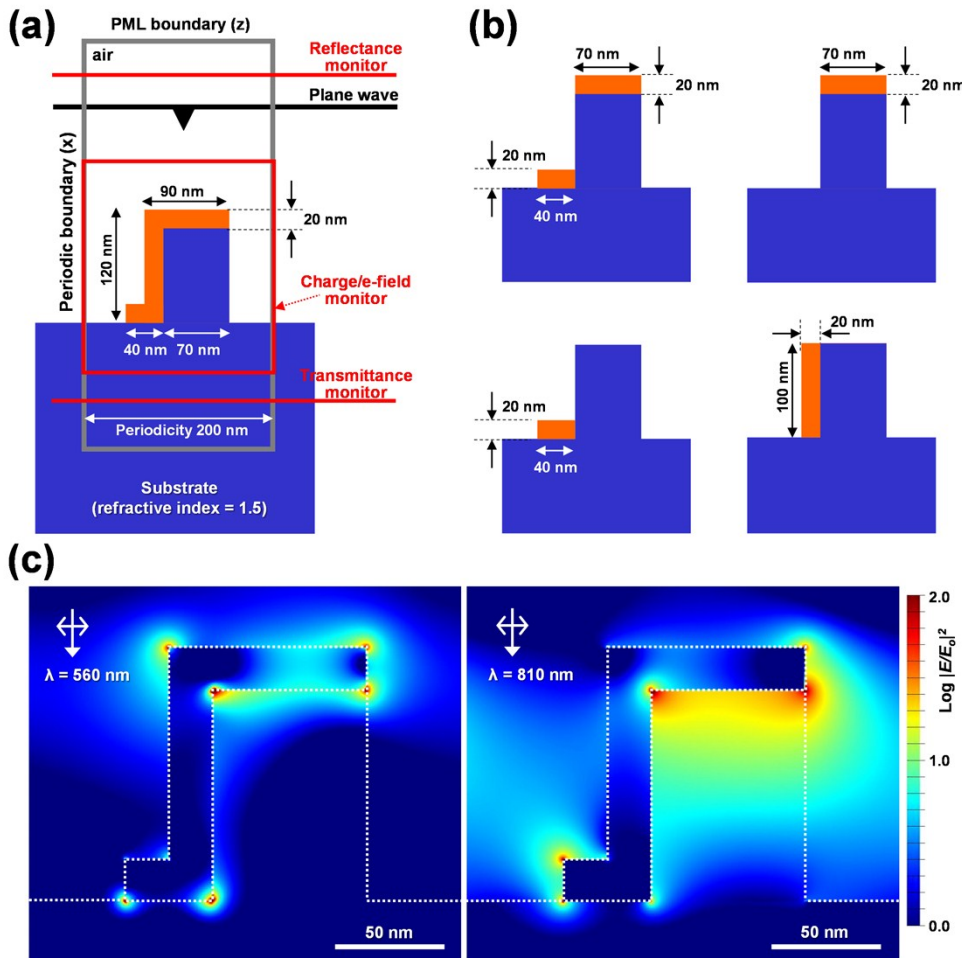


Figure S1. (a,b) Schematic illustration of (a) the FDTD simulation geometry and (b) the modeled nanostructures used to obtain the simulations results in Figure 3. Purple- and orange-colored regions are gold and polyurethane, respectively. (c) Squared magnitude of the local electrical field amplitude around the double-bent gold stripes. The wavelengths of the incident light were (left) 560 nm and (right) 810 nm.

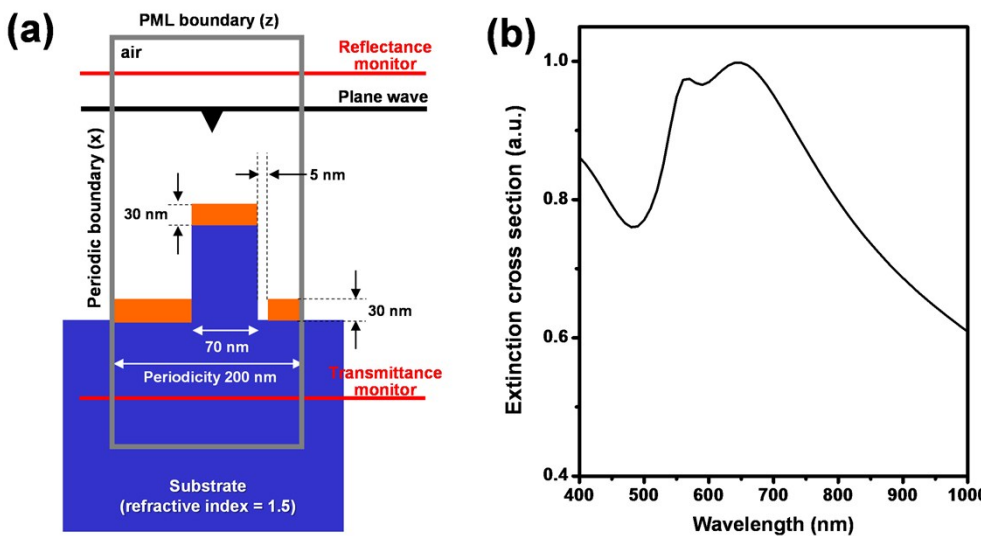


Figure S2. (a) Schematic illustration of the FDTD simulation geometry for modelling the nanostructure shown in Figure 2a and (b) its calculated extinction cross section.

3. Absorbance spectra of the Au strips

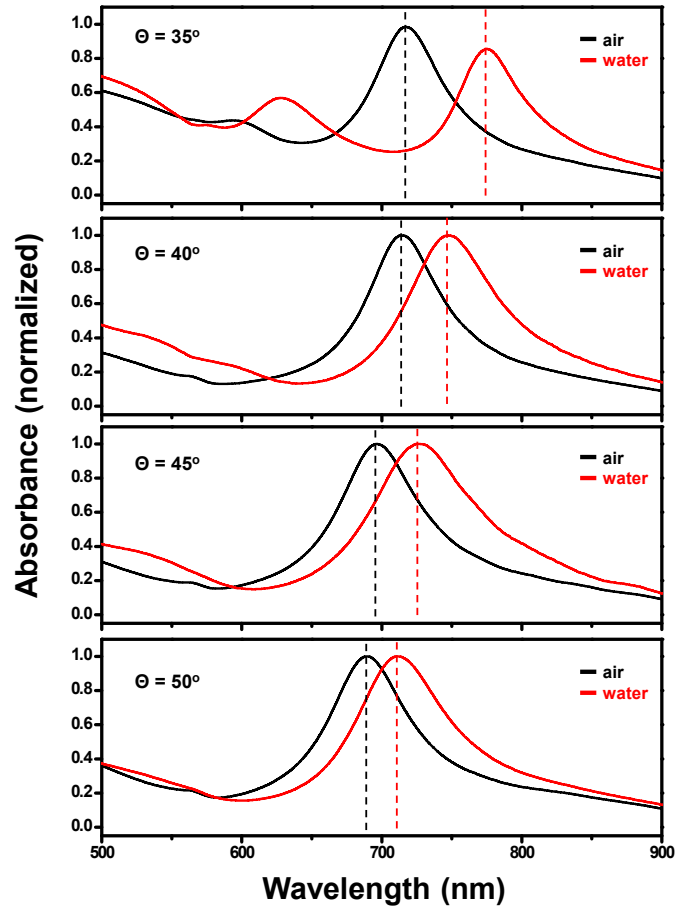


Figure S3. Measured absorbance curves of the Au strips on the 1D nanogratings. The deposition angle of Au was changed from 35° to 50°, and indicated in the graph. Black and red curves were measured from the samples in air and water, respectively. Dashed lines are for eye-guide.

References

- [1] M. G. Kang, M. S. Kim, J. S. Kim, and L. J. Guo, *Adv. Mater.*, 2008, **20**, 4408.
- [2] M. K. Kwak, H.-E. Jeong, and K. Y. Suh, *Adv. Mater.*, 2011, **23**, 3949