Plasmonic Optical Interference

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3D optical simulations

We employed a 3D FDTD simulation to identify the optical coupling characteristics between interference and surface plasmons excited on pores of a plasmonic gold thin layer. Based on an AAO structure with a hexagonal array of nanopores, we built a 3D simulation model (Figure S1) composed of an Al mirror substrate at the bottom, a porous alumina dielectric layer in the middle, and a plasmonic gold thin layer at the top. A simple unit model with air holes at the center and each vertex was expanded to whole space by periodic boundaries in the x and y direction. The PML layer was used as the z top boundary to eliminate unnecessary reflection, and the PEC boundary was used as the z bottom boundary. A slice of the xz plane is shown in Figure S1b. All geometric parameters of the model were based on real values. Gold thickness was 20 nm and the pore-to-pore distance was 100 nm. In our model, the thickness of the AAO layer and the corresponding depth of air pores varied from 100 to 300 nm. Plane wave pulses with an effective wavelength bandwidth from 380 to 780nm across the whole visible range, were propagated and reflected by our structure, only considering the vertical incidence. Reflection flux and spatial profiles of the electronic field in our noticeable domain were recorded and analyzed. We paid attention to the horizontal slice of the mouth of the gold pore where plasmons were excited and the vertical slice of the AAO hole where interference occurred.

Preparation of AAO templates by two-step anodization

AAO templates with hexagonally arrayed nanopores are typically fabricated by twostep anodization of aluminum. A pure aluminum (Al) sheet (99.999 %) of thickness 0.5 mm was electropolished in a mixture of perchloric acid and ethanol (HClO₄:C₂H₅OH = 1:4 volumetric ratio) to remove surface irregularities, at a constant voltage of 20 V and temperature of 7 °C. Next, 0.3 M oxalic acid solution was used to fabricate AAO structures for 12 hours (first anodization) at 40 V and 15°C. After that, the AAO layer was removed in a mixture of 1.8 wt % chromic acid and 6 wt % phosphoric acid for 6 hr at 65°C. The highly ordered AAO templates were obtained by a second anodization under the same conditions as the first anodization. The AAO depth was determined by the duration of the second anodization. The initial pore diameter was 32 nm without a widening process, and a periodic pore-to-pore distance (λ) of 100 nm. To increase the AAO pore size, the AAO templates were etched in 0.1 M phosphoric acid at 30°C.

Preparation of 3D plasmonic nanostructure.

We prepared an Al sheet of 5 cm x 6 cm for the anodization, to form a large-area AAO template of 5 cm x 5 cm. The thickness of AAO templates was controlled by the anodization time (Figure S2). Since AAO is porous alumina, the AAO layer itself becomes a dielectric layer, and the Al beneath the AAO naturally becomes a reflecting mirror layer. After preparing AAO templates, 20 nm-thick gold was deposited as a plasmonic layer on the templates by thermal evaporation. Thus, we could produce a 3D plasmonic structure with three components; a plasmonic layer at the top, a dielectric layer in the middle, and a mirror layer at the bottom.



Figure S1. Geometry of the 3D plasmonic nanostructure used for simulation. (a) SEM image of the 3D plasmonic nanostructure. (b) Geometrical model for simulation based on the SEM image of the 3D plasmonic nanostructure



Figure S2. AAO thickness (t_{AAO}) as a function of anodization time. AAO thickness increased linearly with anodization time.



Figure S3. Effect of AAO pore size on optical behavior of 3D plasmonic nanostructures. Au thickness on the top was 20 nm and AAO dielectric thickness was 200 nm. As the pore size increased, the SPR peak was blue-shifted due to the decreasing dielectric constant.

Theoretical design rule approach to determine the AAO thickness where the maximum absorption occurs.

- Effective refractive index for an AAO template

$$1.7 \approx 1.77 \frac{\left(100 \times 100\sqrt{3} - 2 \times \pi (16)^2\right)}{100 \times 100\sqrt{3}} + 1 \times 2 \times \pi (16)^2$$

- Relationship between AAO thickness and maximum absorption.

$$\left(t_{AAO} + 20 - \frac{\lambda - 400}{4}\right) \times 1.7 = \frac{\lambda}{2} \times \frac{m}{2}$$
(2)

where m>0 integer, and the absorption is maximum when m is odd.





Figure S4. Local field distribution for the strong absorption. (a) At 400 nm, the 3D plasmonic nanostructures with $t_{AAO} = 160$ nm and 280 nm show maximum plasmonic absorption. (b) At 605 nm, the 3D plasmonic nanostructures with $t_{AAO} = 120$ nm and 300 nm show the maximum plasmonic absorption. Based on Eq. (1), we could create a relationship between AAO thickness and a given wavelength for the maximum absorption. We needed the calibration factor (the third term in the parenthesis of Eq. (2)) as the wavelength increased from 400 nm to 680 nm in the full visible range.



Figure S5. Structural color of "KHU" capitals for "RGB" colors. (a) Fabrication process of "RGB" colored "KHU" capitals. First, we prepared an Al sheet covered by a polymer protector. We only opened the Al at the positions of KHU capitals. After dipping the Al sheet in the acid, we pulled out the sheet up to each capital according to the time. After finishing the anodization for each capital, we could not detect any color on the capitals. After evaporating gold on the sheet, we detected different brilliant colors on the capitals. The SEM images show the AAO thickness for each capital. (b) The corresponding reflectance spectra for "RGB" positions in "KHU" capitals.



Figure S6. Flexible 3D plasmonic nanostructure on a PET substrate. The size was about $3 \times 3 \text{ cm}^2$. The Au thickness was 20 nm, and the AAO thickness was 140 nm.