Electronic Supplementary Material (ESI) for Nanoscale

Electronic Supplementary Information

Three-tiered Au nano-disk array for interacting broadband light

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Experimental details

Fabrication of the three-tiered Au disk:

To make a bilayer resist stack, 100-nm-thick polymethylglutarimide (PMGI, MicroChem) was spin-coated onto a Si substrate and baked at 180°C for 3 min. Then, a 50-nm-thick ZEP (ZEP520A, Nippon Zeon Co.) resist was spin-coated onto the PMGI and baked at 180°C for 3 min. The electron beam exposures were conducted with a beam accelerating voltage of 50 KeV. After development of the ZEP resist with a commercial developer (ZED-N50, Nippon Zeon Co.), the sample was immersed in an alkaline aqueous solution (NMD-3, JSR Micro) for 5 s to form an undercut profile in the PMGI resist. Next, a multilayer with a structure of 10 nm Cr (top)/(20 nm Au/2 nm Cr/20 nm SiO_2/2 nm Cr)×2/20 nm Au /2 nm Cr (bottom) was deposited on the patterned polymer template by magnetron sputtering with a base pressure of less than 10^{-7} Pa. After lift-off in acetone and removal of PMGI in NMD-3, metal residues near the Au disks were etched by Ar plasma. Finally, the Cr and SiO_2 layers were chemically removed by Cr etching solution (mixture of $HClO_4$ and $(NH_4)_2[Ce(NO_3)_6]$) and diluted hydrofluoric acid (1% HF aqueous solution). The prepared Au disk arrays on Si substrates were examined by scanning electron microscopy (SU-8000, Hitachi) and atomic force microscopy (L-traceII, SII NanoTechnology Inc.) in a tapping mode.

Electromagnetic simulation:

Electromagnetic simulations were performed using three-dimensional finite-difference time-domain software (Lumerical FDTD solution 6.5). For the calculation of extinction, absorption, and scattering cross sections in Figure 1, simulation mesh was set to a 1.0-nm cubic grid, and perfectly matched layers (PML) were used as a boundary condition. The isolated nanostructures were placed in a uniform medium of air and illuminated from the top with a total-field scattered-field (TFSF) source. TFSF sources are used to divide the computation volume into total field (incident plus scattered field) and scattered field only regions. The two three-dimensional monitor boxes were used to calculate the scattered and the absorbed power.

For the calculation of the scattering properties from the array of Au nanostructures on the Si substrate (Figure 3b), periodic boundary conditions were used for the lateral direction to model the arrayed structure, and PML conditions were used for the vertical direction. The periodicity of the nanostructures was set to 1 μm. The arrayed nanostructures were illuminated from the top with a plane wave source. The two two-dimensional monitors to get the reflected power into the air and the transmitted power into the Si were placed at 200-nm and 0-nm above the Si surface, respectively.

The light absorption in a thin Si substrate in Figure S2 was calculated with the two two-dimensional monitors placed at the Si surface and 750-nm below the Si surface. Except for the positions of these monitors, other conditions are identical to those for Figure 3b. The absorption in a thin Si was estimated from the intensity difference of the light passed though the two monitors.

Table S1. Scattering peak positions and maximum absorption cross section of three-tiered Au disks. The thickness of the intermediate SiO_2 layer was varied from 15 to 60 nm, without changing the dimensions of Au disks (diameter = 80 (top), 140 (middle), 200 (bottom) nm, thickness = 20 nm for all).

SiO_2 thickness	Scattering peak #1 (nm)	Scattering peak #2 (nm)	Scattering Peak #3 (nm)	Max. absorption cross section (a.u.)
15 nm	600	700	900	4.0 @ 890 nm
20 nm	580	680	840	3.8 @ 820 nm
30 nm	570	670	780	3.0 @ 770 nm
40 nm	560	660	780	2.5 @ 760 nm
60 nm	560	660	760	2.1 @ 740 nm
Isolated single disks	560 (disk dia. 80 nm)	650 (disk dia. 140 nm)	730 (disk dia. 200 nm)	1.8 @ 640 nm (disk dia. 140 nm)

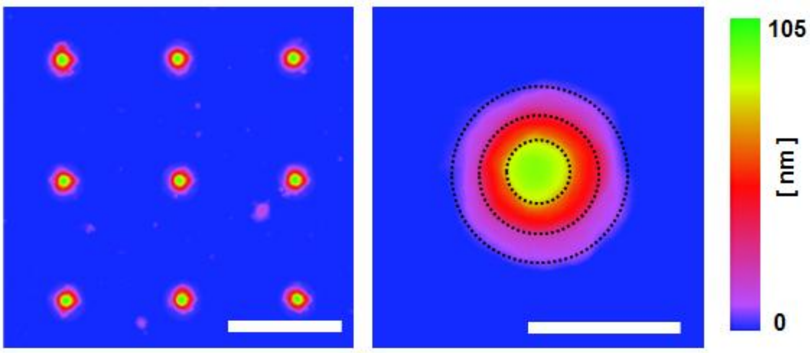

Figure S1. Topographic images of three-tiered Au disks obtained from tapping mode atomic force microscopy (AFM). Scale bars in AFM images are 1 μm (left) and 200 nm (right).

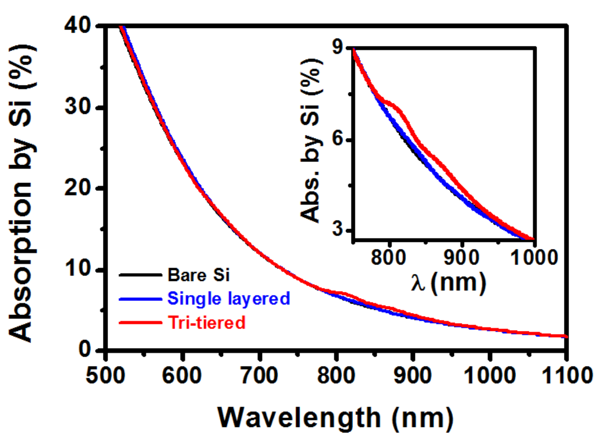

Figure S2. The simulated fraction of the light absorption in a 750-nm-thick Si without nanostructures (black line), with the array of single nanodisks (blue line), and with the array of tri-tiered nanodisks (red line). The inset graph is an enlarged view of the wavelength range of 700 ~ 100 nm. Simulation conditions are described at the experimental details in this Electronic Supplementary Information.