SUPPLEMENATARY INFORMATION

Covalent bonding-assisted nanotransfer lithography for plasmonic nano-optical elements

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METHODS

Fabrication of 100-nm-pitch Si master

The 100-nm-pitch line-patterned Si master was fabricated by E-beam lithography.

Preparation of adhesive and spin-coating onto substrate

N-[3-(trimethoxysilyl)propyl]ethylenediamine,propane-1,3-diol, and di(propylene glycol) methyl ether, purchased from Sigma-Aldrich, were mixed with volume fractions of 1 %, 70 %, and 29 %, respectively, and mixed under ultrasonication for 30 s. The dispersed adhesive was coated by using a spin-coating system at 5000 rpm for 1 min on a substrate that had been plasma treated to enhance the hydrophilicity. Then, the substrate was baked on a hotplate at 150 °C for 1 min to evaporate the propane-1,3-diol and di(propylene glycol) methyl ether solvents.

Measuring thickness of SiO_2

SiO_2 of different thicknesses was deposited onto a bare Si wafer and the thickness of each sample was measured using a spectroscopic Ellipsometer (M2000D, Woollam, USA) at three locations, after which the obtained values were averaged.

X-ray photoelectron spectroscopy (XPS)

SiO_2 layers with thicknesses of 3.17 nm, 4.53 nm, and 9.41 nm were deposited on the non-patterned polymer stamp and then transferred to the adhesive coated substrate. Then, the atomic percentages of the fabricated samples were characterized by XPS (ThermoFisher Scientific, K-Alpha+, USA) as a function of the etch depth.

Meta-surface design

The meta-surface image was designed using PowerPoint (Microsoft, USA). Different diameters and periods of hole patterns were positioned based on RGB color and converted to an m-file using commercial MATLAB software (MathWorks, USA).

Fabrication of meta-surface Si master

The meta-surface m-file was converted to an AutoCAD (Autodesk, USA) file and used to fabricate the hole-patterned Si master using E-beam lithography.

Measuring permittivities of Ag and Al

Al and Ag with a thickness of 100 nm were deposited onto bare Si wafers, and the permittivites were then measured using a Spectroscopic Ellipsometer (M2000D, Woollam, USA).

Etching for fabrication of WGP

An etching machine (Plasmalab System 100, Oxford Instruments, Germany) was employed to etch SiO_2 anisotropically under CHF_3 and Ar gas at a 2:1 ratio. After the etching procedure, a SiO_2 nanopattern was formed with the same pattern as the stamp. The patterned SiO_2 acted as a hard mask for anisotropic Al etching under Cl_2 and BCl_3 gas at a 1:1 ratio.

Optical image characterization

High-resolution imaging of the meta-surface was obtained by optical microscopy (Eclipse LV 100, Nikon Instruments, Inc., USA).

Transmission spectrum characterization

The transmission spectra of the meta-surface and WGP were characterized using a spectrometer (QE Pro 6000, Ocean Optics, USA).

High-resolution surface and cross-sectional image analysis

The high-resolution surface and cross-sectional imaging was acquired by SEM (XL30SFEG, Philips Corporation, Netherlands, and JSM-7800F, JEOL, Japan), TEM (JEM-ARM200F, JEOL, Japan), FIB (Helios Nanolab, FEI, USA), AFM (XE-100, Park Systems, Korea) and EDS (Quantax-400, BRUKER, Germany).

Peel-off test experiment

A 90° peel test was performed to investigate the relative adhesion strength between the polymer stamp and substrate. After preparing the substrate, a polymer stamp of 20 mm in width was laminated onto the substrate using a roll-to-plate system. The substrate was firmly attached to a low-friction travelling stage moving horizontally as the polymer stamp was pulled vertically from the substrate at a constant speed of 50 mm/min. The polymer stamp was fixed on the grip installed at the end of the loadcell. As the polymer stamp was pulled upward, an interfacial crack between the polymer stamp and substrate was propagated and the vertical force was recorded as a function of time using the loadcell. The peel force was defined as the time-averaged vertical force during the interfacial crack propagation.

FDTD simulation

Metasurface: The reflection spectra of various diameters and holes arranged on a hexagonal lattice are calculated by using FDTD method. The dimensions are set to be the same as those of the fabricated samples. The unit cell of each sample is $a*\sqrt{3}a$ nm^2 where a is the lattice constant, and the boundary conditions of the four vertical sides, the upper and lower sides are used on the periodic, metal and perfectly matched layer boundary conditions, respectively. The plane wave source was perpendicularly injected to the nanostructured plane in the visible wavelength range of 380-830 nm.

WGP: The transmitted spectra of various gratings are simulated by FDTD method. The domain size of the calculation is $1000*1000*1700$ nm^3. The periodic boundary condition is used at four vertical sides, and the perfectly matched layer boundary condition is used for the upper and lower boundary conditions. The polarized plane wave sources pass through the WGP plane in the visible range of 380-830 nm, and the transmitted/incident intensity ratio is monitored according to the wavelength and polarization.

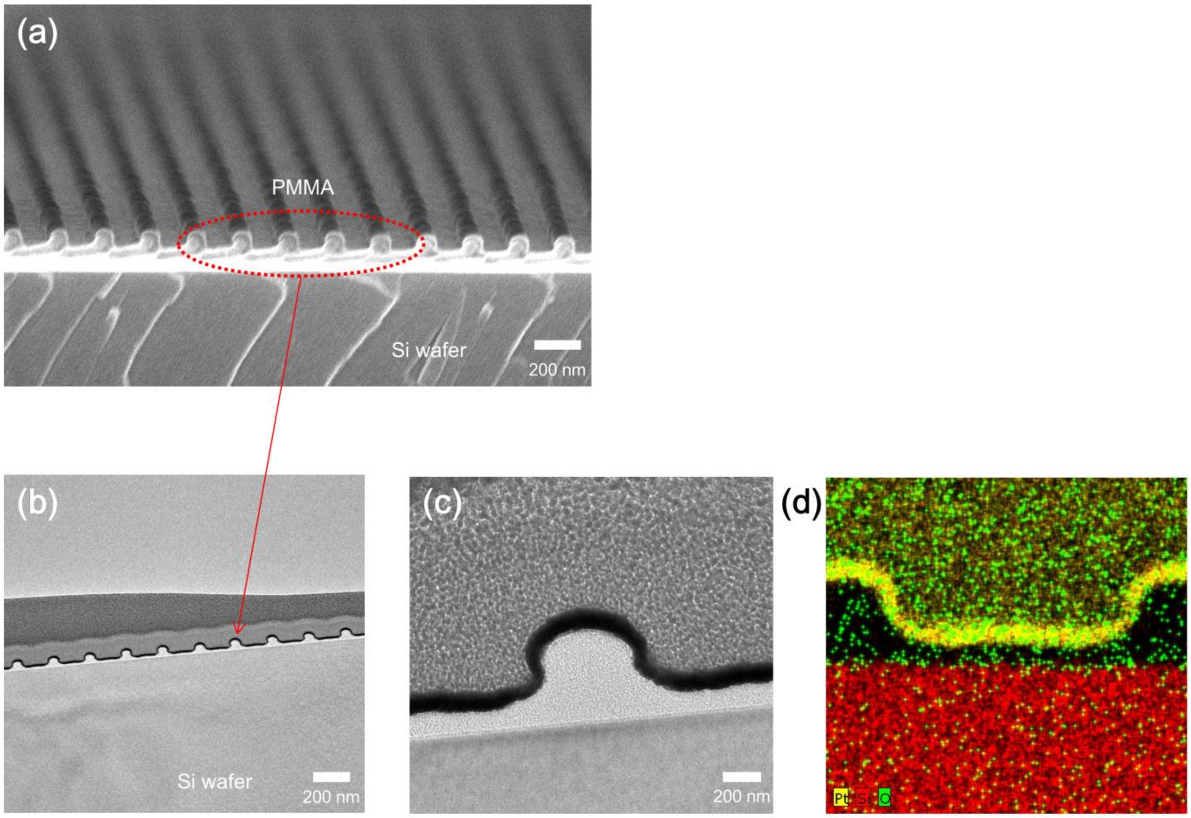

Fig. S1 Transfer printing using PMMA as adhesive. (a) SEM image of imprinted PMMA after transfer procedure, (b,c) TEM images of imprinted PMMA, (d) EDS image of imprinted PMMA.

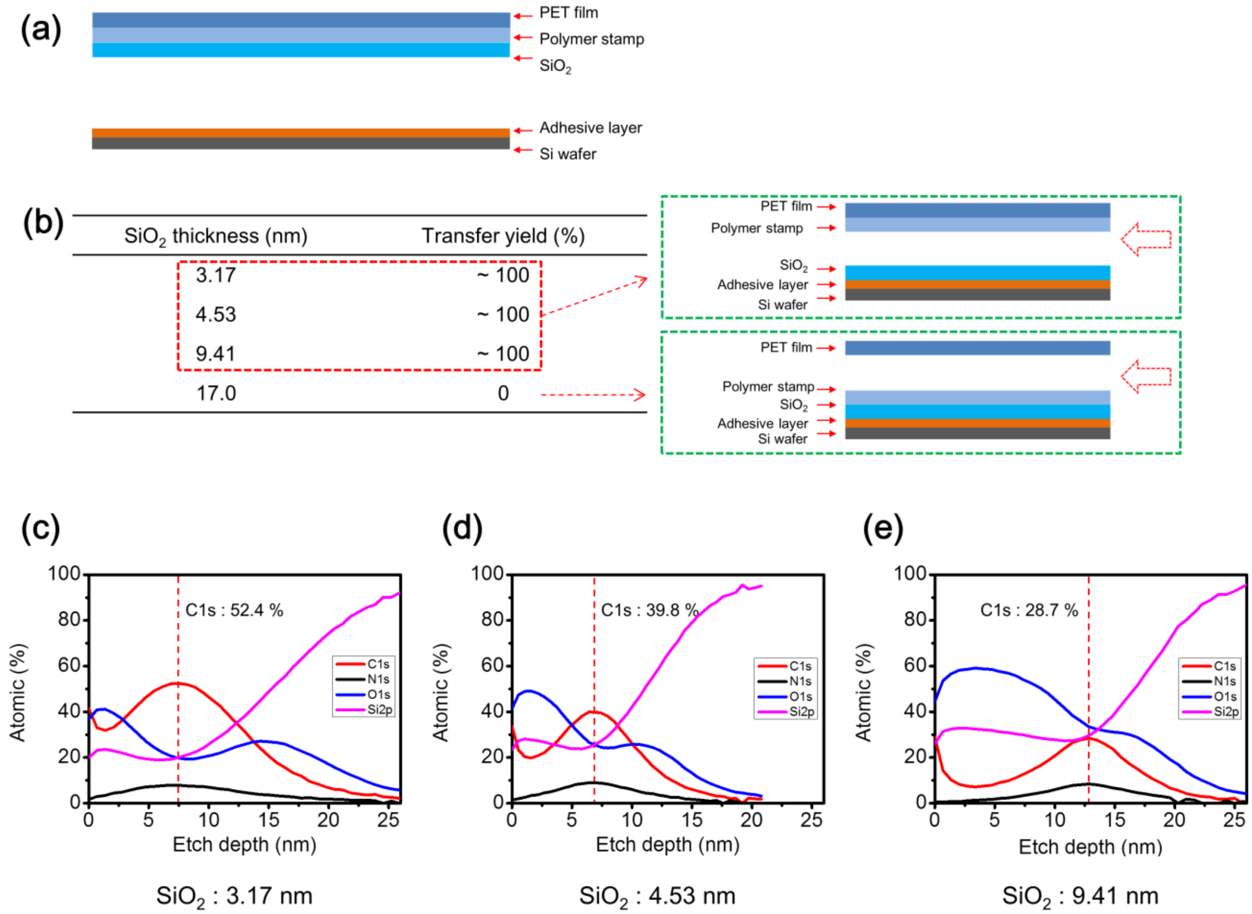

Fig. S2 Schematic diagrams of nanotransfer process depending on thicknesses of SiO_2 and XPS spectra (a) schematic diagram of prepared sample, (b) measured SiO_2 thickness, transfer yield and schematic diagrams after transfer procedure, based on the SiO_2 thickness (dotted arrow indicates the detached interface), (c, d, e) XPS spectra for different thicknesses of SiO_2.

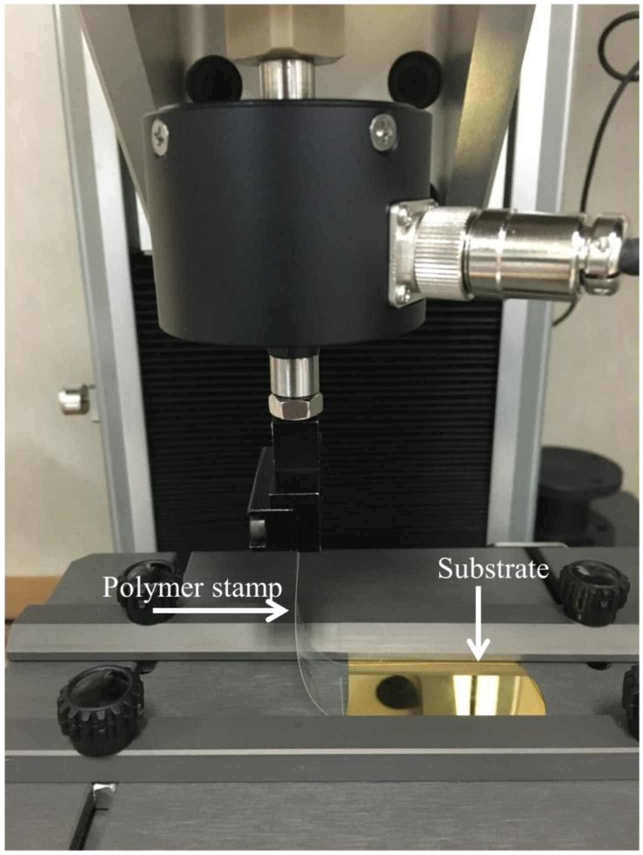

Fig. S3 A photograph of peel test set-up and peel force measurement.

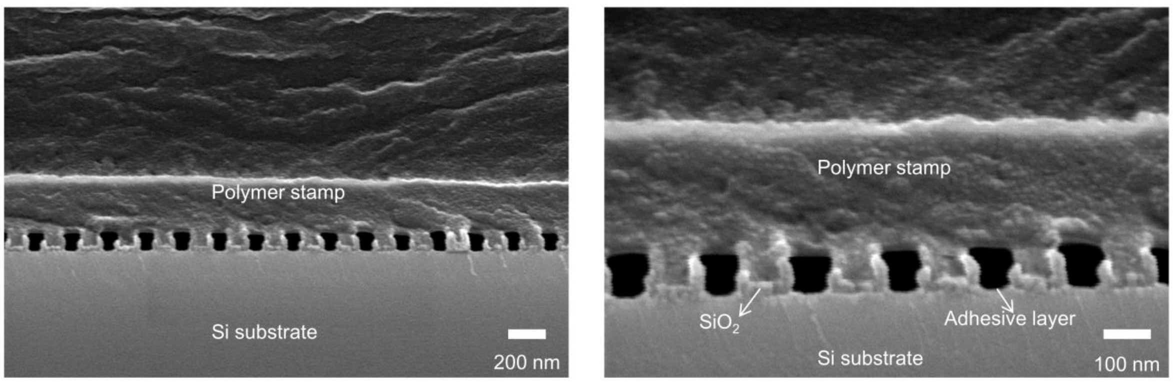

Fig. S4 SEM images of Si/adhesive/SiO_2 structure after peel-off test at different magnifications.

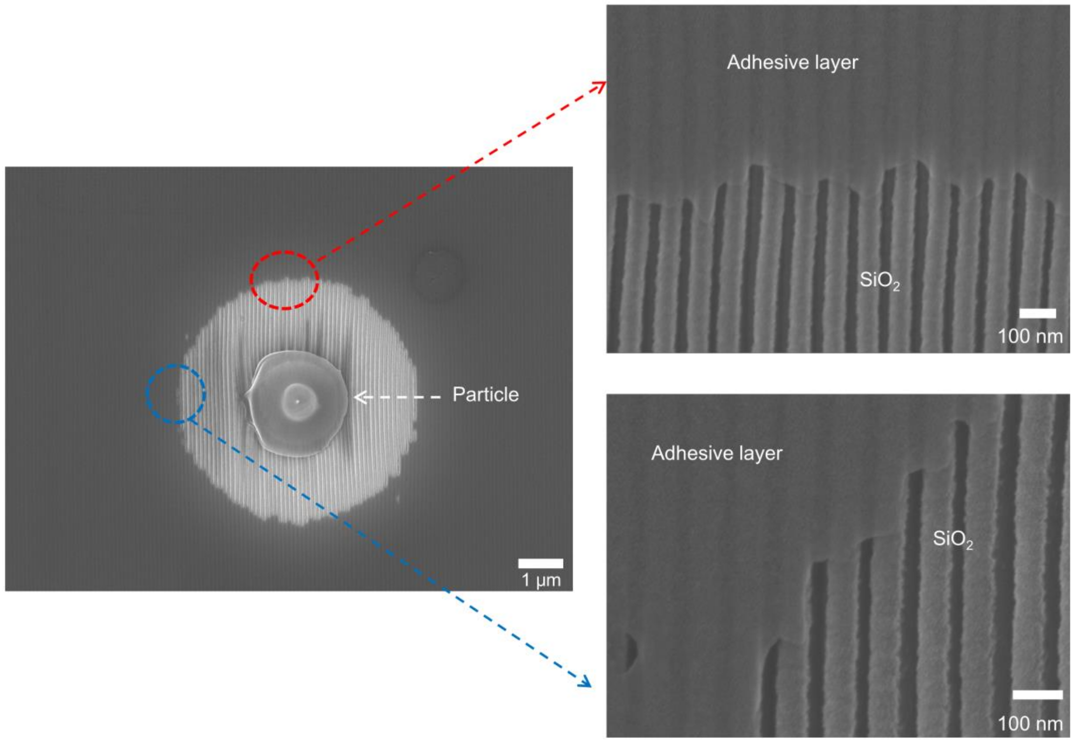

Fig. S5 SEM images of Au/adhesive/SiO_2 layers after peel-off test.

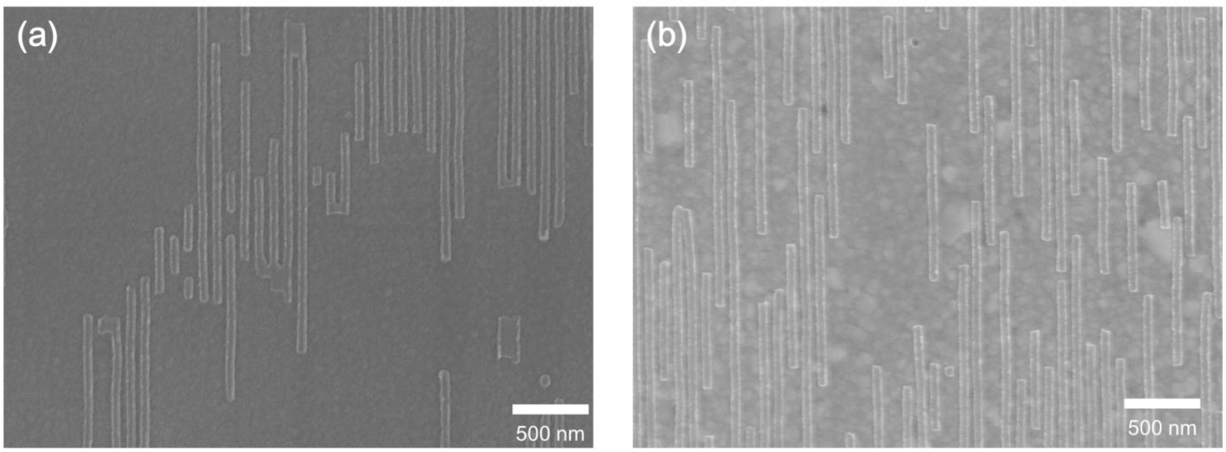

Fig. S6 SEM images of samples with transferred SiO_2 lines on the (a) Al substrate and (b) Ag substrate after peel test.

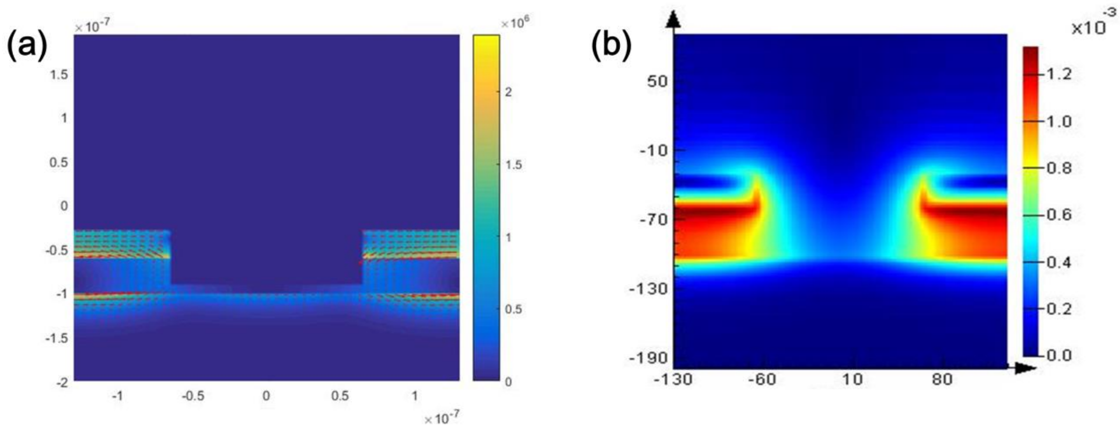

Fig. S7 Simulation results of electromagnetic field for meta-surface with period of 260 nm and diameter of 130 nm (a) current density vector at wavelength of 545 nm (b) magnetic field intensity at wavelength of 545 nm.

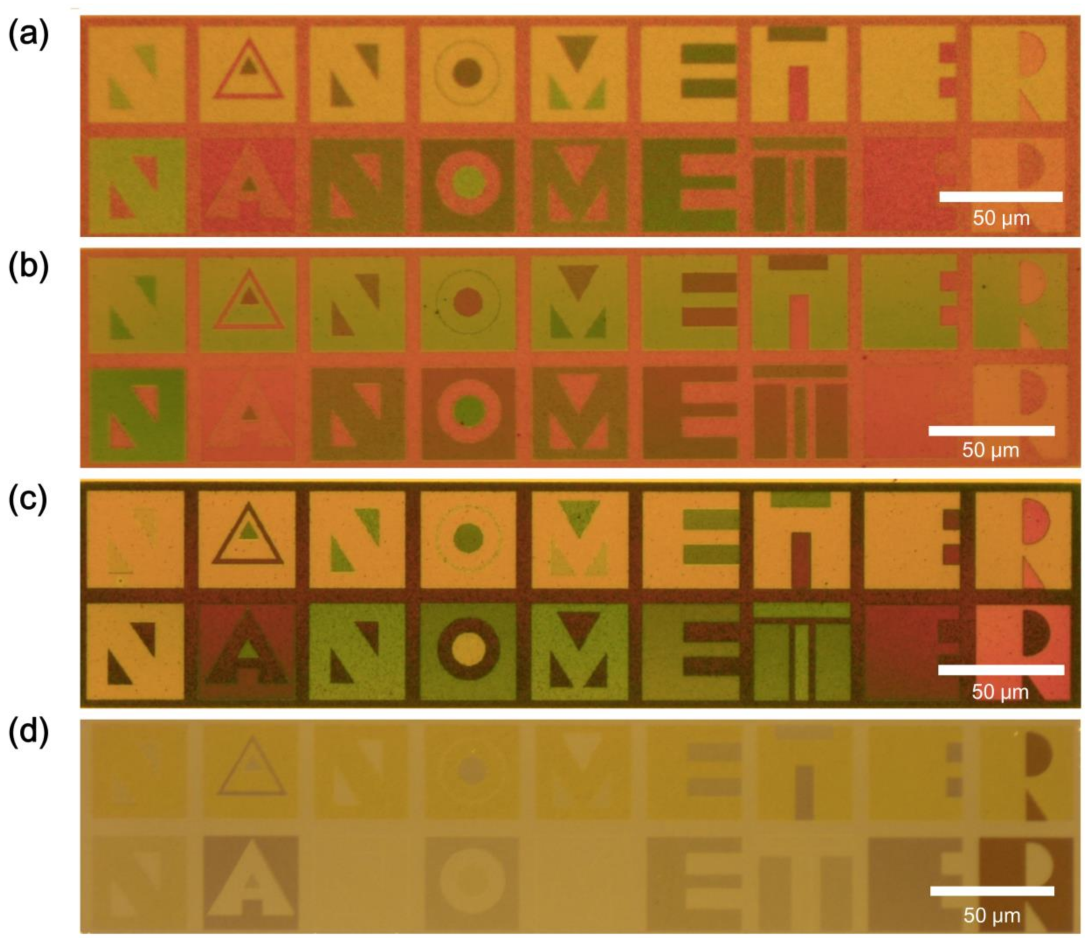

Fig. S8 Optical microscope images of meta-surfaces illuminated with halogen lamp. Fabricated meta-surface using (a) Ag, (b) Au, (c) Au and PMMA spin coating, and (d) Al.

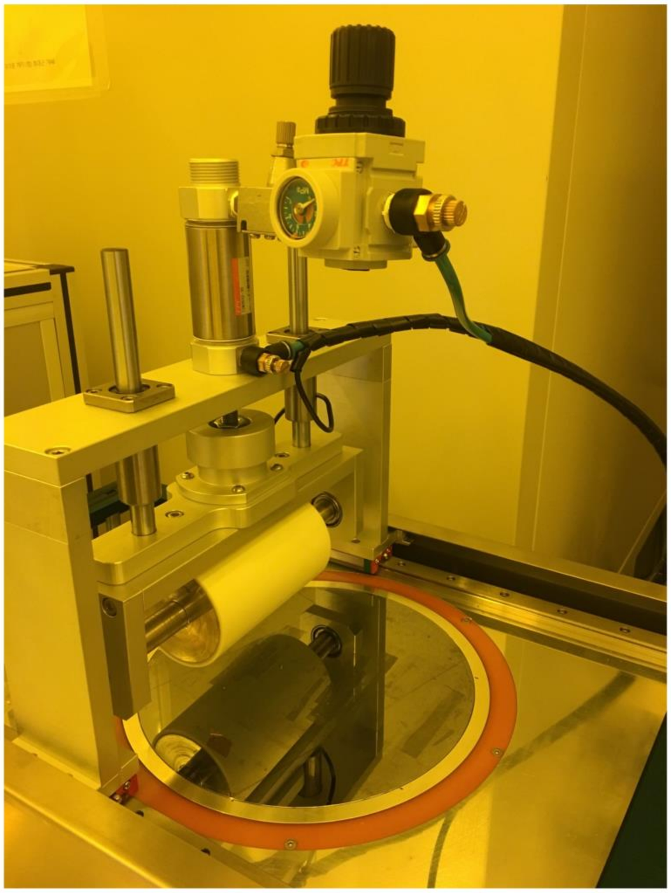

Fig. S9 Photograph of roll-to-plate transfer system.

Table S1. Measured surface roughness of substrates and transfer yield.

Samples	Min (nm)	Max (nm)	Mid (nm)	Mean (nm)	Rq (nm)	Transfer yield (%)
a	-0.647	1.028	0.191	0.000	0.161	~ 100
b	-13.167	8.087	-2.540	0.222	0.975	~ 100
c	-18.824	18.010	-0.407	0.622	3.823	~ 100
d	-33.061	78.458	22.698	0.603	7.922	~ 100
e	-48.000	177.844	64.922	3.826	18.770	0
f	-21.161	120.526	49.682	4.261	16.117	0

Table S2. Period, diameter, SiO_2 thickness, and CIE coordinate table for meta-surface

Period (nm)	Diameter (nm)	Thickness (nm)	x	y
200	70	30	0.3691	0.3590
260	110	30	0.3047	0.2895
260	130	30	0.3344	0.2620
260	160	30	0.3824	0.3228
280	130	30	0.2737	0.3154
300	130	30	0.2861	0.3815
300	150	30	0.2658	0.3570
320	150	30	0.3192	0.4019
360	180	30	0.3787	0.4433