Electronic Supplementary Material (ESI) for Nanoscale.

Supporting Information

Asymmetric angular dependence for multicolor display based on plasmonic inclination nanopillar array

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Experimental Section/Methods

Fabrication: Polystyrene (PS, Mw=18,000 g/mol, from Sigma-Aldrich LLC.) was dissolved in anisole and spin-coated to get 600 nm-thick resist film. The EBL system (Raith Elphy Quantum) is integrated in SEM (Carl Zeiss SEM-Sigma 300), and the exposure holder is locked on the SEM stage, so the tilted angle for EBL can be accurately controlled with the 5-axis stage. EBL is carried out at 20 kV acceleration voltage, the stage is tilted with 0^o, 10^o, 20^o, respectively. After development with xylene for 5 min, 100 nm-thick Aluminum film is deposited on the facing-up side of the inclined pillar array through the lab-designed Glancing Angle Deposition (GLAD) system.

Characterization: The structure is characterized with SEM. The optical properties impacted with structural inclination is investigated with the angle-resolved spectrum system in microscope (ARS, Ideaoptics Inc.). For reflection spectra, the incident angle is oppositely selected as $\theta=0^o$, $\pm20^o$, $\pm30^o$, $\pm40^o$, and 0^o-, 10^o-, 20^o-inclined structures are measured, respectively. For diffraction mapping, the incident angle is oppositely fixed at $\theta=\pm20^o$, and the 0^{th}-, and $\pm1^{st}$-order of reflection intensities for 0^o-, 10^o-, 20^o-inclined structures with omnidirection are collected, respectively. For all the measurements, TM polarized incidence is selected which is coplanar with the inclination direction of the structures. The samples of color printing are observed with CCD integrated in dark-field microscope, with a tungsten-halogen lamp offering broadband wavelength range of 200 to 900 nm, and a ×5 objective lens with the numerical aperture (NA) of

0.12 as collimator. During observation, the samples are shifted from left side to right side with a step of 0.5 mm to characterize the asymmetric reflection color generation.

Simulation: The asymmetric optical properties are analyzed with the 3D Finite Difference Time Domain (FDTD) method. The Broadband Fixed Angle Source Technique (BFAST) method in FDTD calculation is utilized to simulate oblique illumination with a broadband source. The varied incident angle from -40^{o} to 40^{o} and a broadband source from 400 nm to 800 nm is introduced for the analysis of far-field intensity and electric near-field distribution. For all the simulations, the polarization direction of the source is coplanar with the inclination direction of the structures, corresponding to the actual measurement condition.

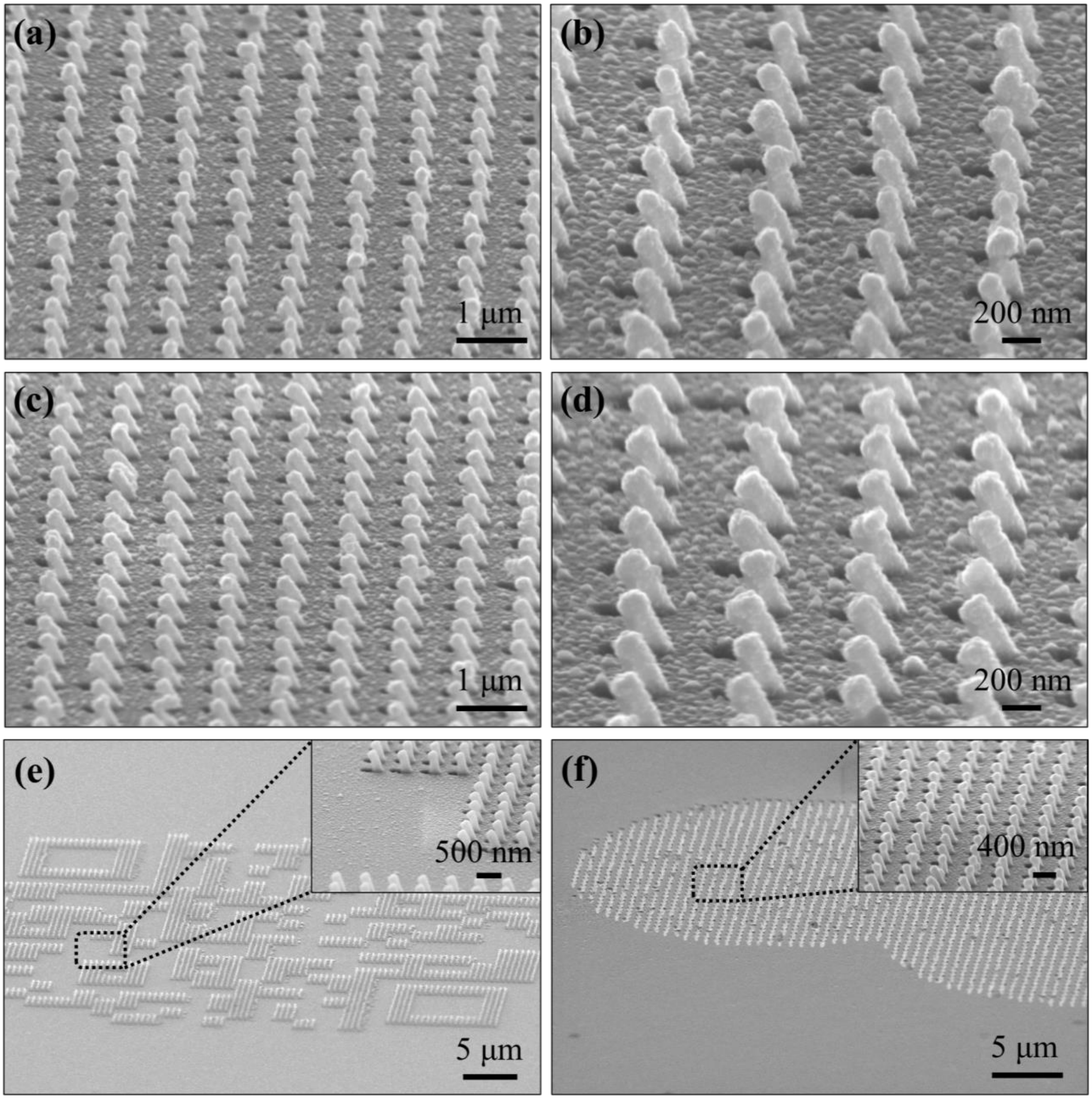

Fig. S1 85^{o}-tilted SEM images of a) large-area pattern of Al pillar arrays tilted with 10^{o} and b) its zoom-in image; c) large-area pattern of Al pillar arrays tilted with 20^{o} and d) its zoom-in image; e) "2D code" pattern containing Al pillar arrays tilted with 20^{o}; f) "flower" pattern containing Al pillar arrays tilted with 20^{o}.

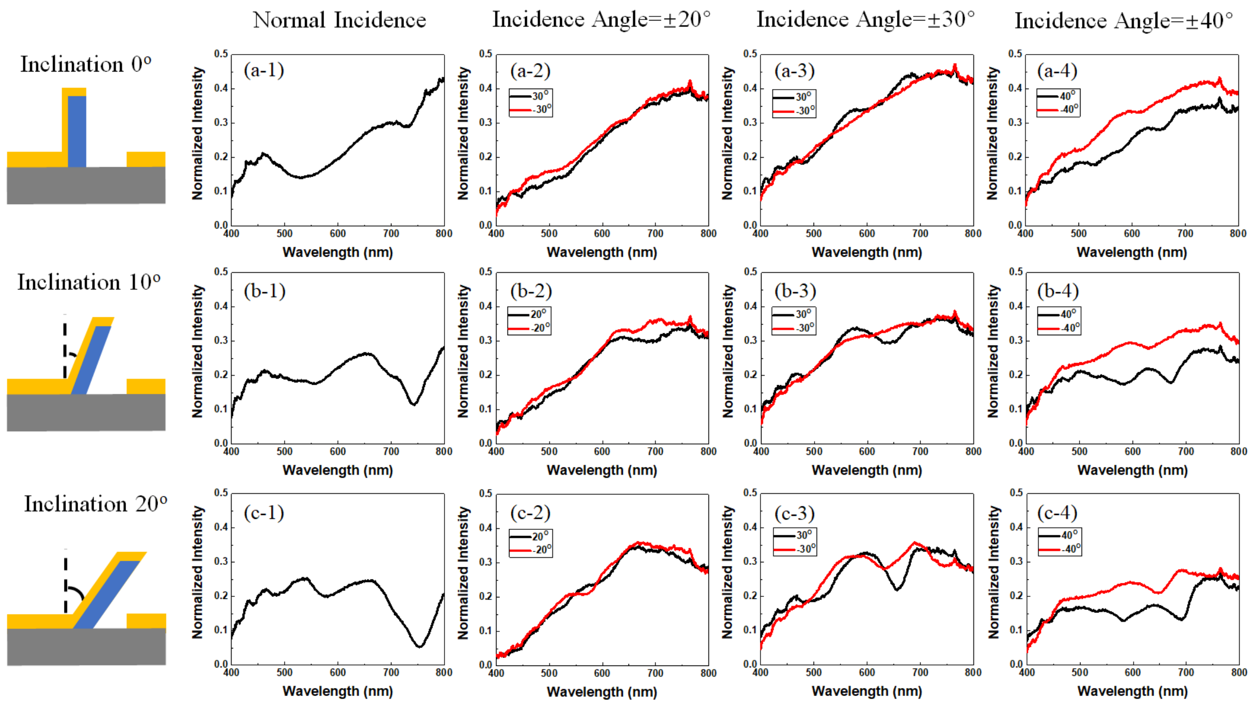

Fig. S2 Summary of reflection spectra of 600 nm-period nanopillar arrays inclined with a) 0°, b) 10°, c) 20° for the incident angles of 1) 0°, 2) ±20°, 3) ±30°, 4) ±40° with TM-polarization, as illustrated on the left side. The dark lines are for the positive incidence, and the red lines are for the positive incidence.

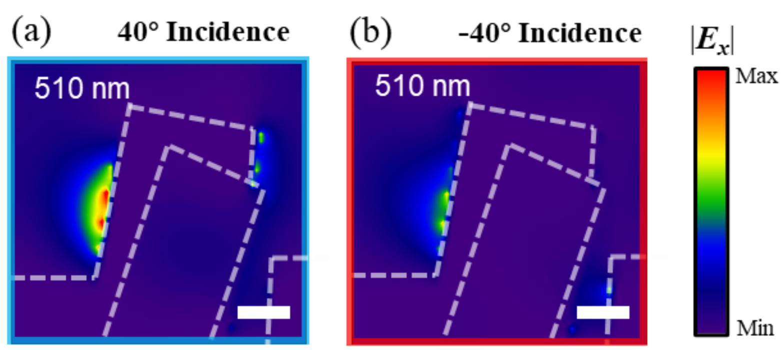

Fig. S3 Calculated |*E*| near-field distributions with 20°-inclined nanopillar array for a) 40° incidence and b) -40° incidence (bottom line) at the wavelength of 510 nm. The scale bar is 100 nm.

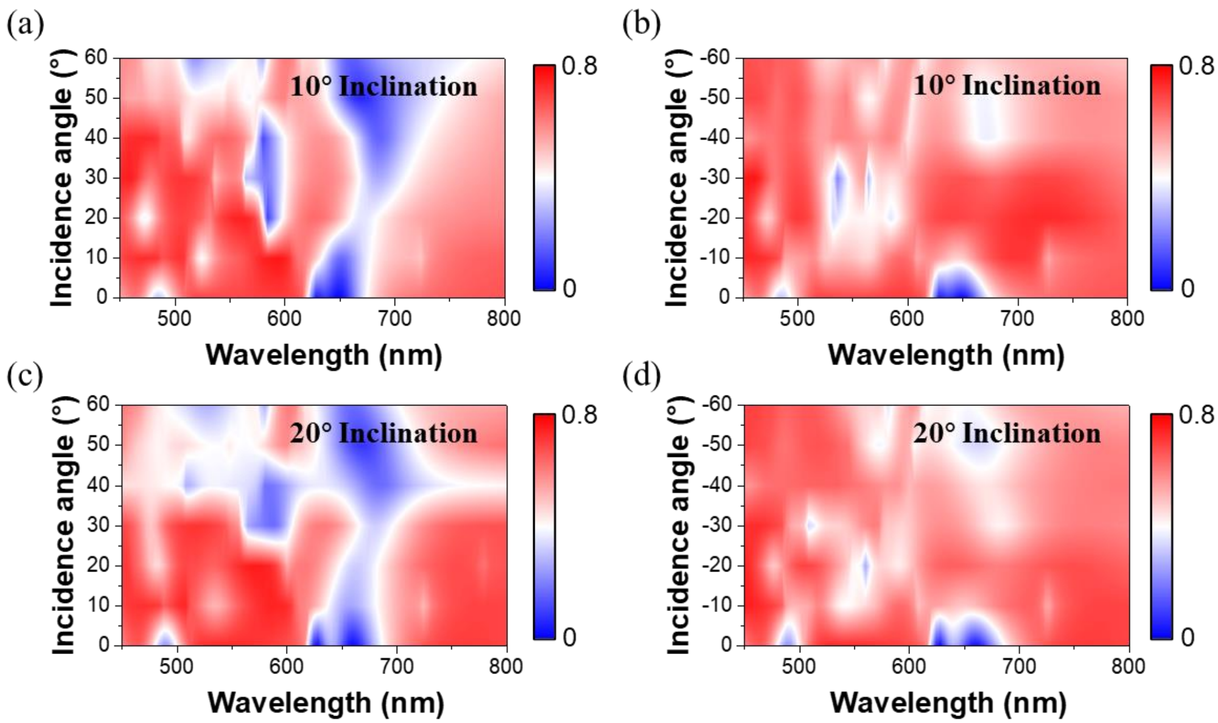

Fig. S4 Maps of simulated intensity difference for reflectance of 600 nm-period nanopillar arrays inclined with 10º and incident angles varied between a) 0º~60º, and b) 0º~-60º. Simulated maps of intensity difference for reflectance of 600 nm-period nanopillar arrays inclined with 20º and incident angles varied between c) 0º~60º, and d) 0º~-60º.

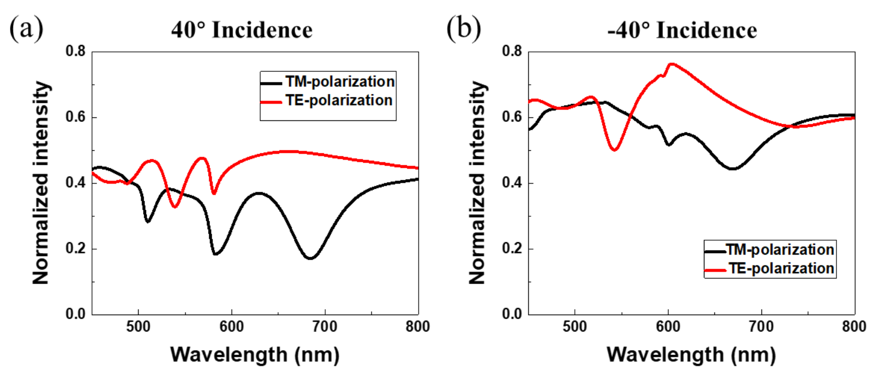

Fig. S5 Simulated reflection spectra of 20º-inclined nanopillar array for a) 40º incidence with TM-/TE-polarization, and b) -40º incidence with TM-/TE-polarization, respectively.

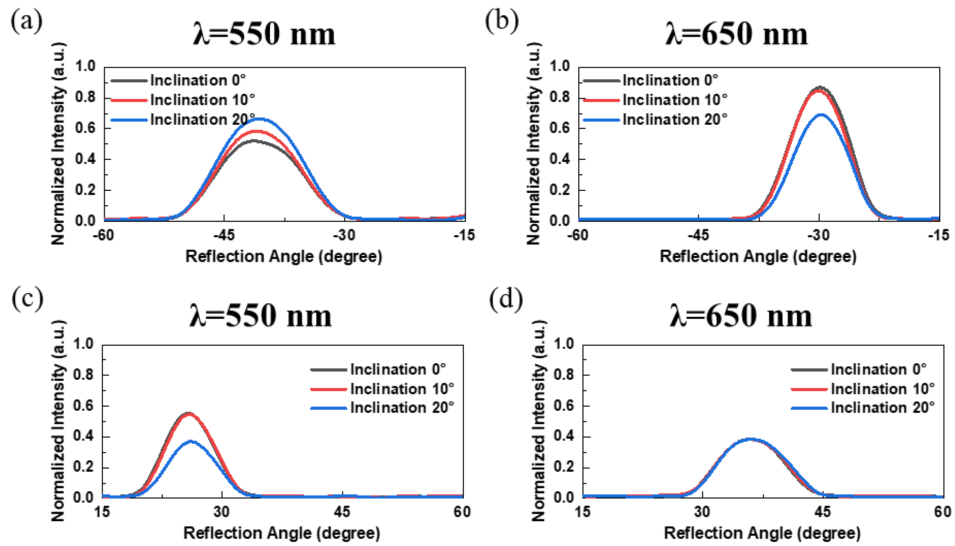

Fig. S6 Angular reflected power distribution at the wavelengths of 550 nm and 650 nm for varied structural inclination with a,b) -20º light incidence and c,d) 20º light incidence.

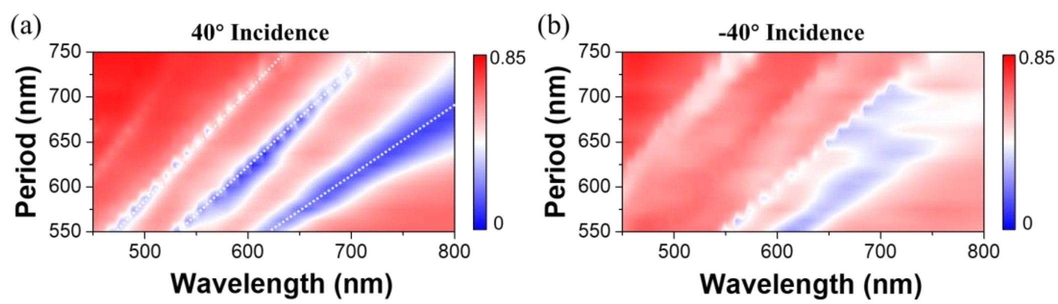

Fig. S7 Simulated normal reflection of the 20º-inclined pillar arrays with varied periods of 550~750 nm for a) 40º and b) -40º light incidence, respectively.

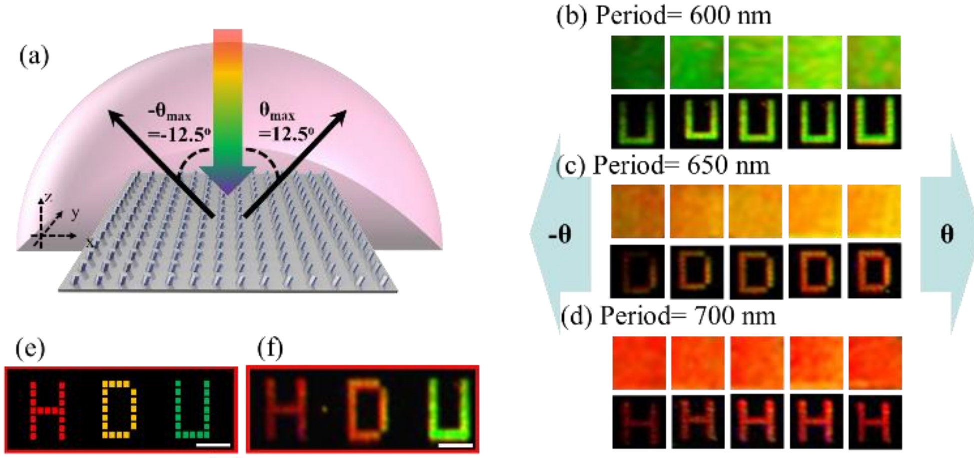

Fig. S8 a) Asymmetric multicolor display of 20°-inclined nanopillar array observed with varied opposite angles. The periods of the structures are b) 600 nm for green, c) 650 nm for yellow, d) 700 nm for red. e) The designed and f) the normal reflection color of "HDU" with varied periods for aimed color, which is the abbreviation of "Hangzhou Dianzi University". The scale bar in e,f) is 50 μm.