Research article

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Near-zero reflection of all-dielectric structural coloration enabling polarization-sensitive optical encryption with enhanced switchability

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Abstract: Structural coloration using metasurfaces has been steadily researched to overcome the limitations of conventional color printing using pigments by improving the resolution, lowering the toxicity, and increasing the durability. Many metasurfaces have been demonstrated for dynamic structural coloration to convert images at the visible spectrum. However, the previous works cannot reach near-zero scattering when colors are turned-off, preventing it from being cryptographic applications. Herein, we propose a completely on/off switchable structural coloration with polarization-sensitive metasurfaces, enabling full-colored images to be displayed and hidden through the control of the polarization of incident light. It is confirmed that the nanostructure exhibits the polarization-

dependent magnetic field distributions, and near-zero scattering is realized when the polarization of incident light is perpendicular to the long axis of the nanofins. Also, the metasurfaces are made up of triple-nanofin structures whose lengths affect locations of resonance peaks, resulting in full-color spectrum coverages. With such advantages, a QR code image, a two-color object image, and an overlapped dual-portrait image are obtained with the metasurfaces. Such demonstrations will provide potential applications in the fields of high-security information encryption, security tag, multichannel imaging, and dynamic displays.

Keywords: anticounterfeiting; cryptography; dynamic metasurface; full-color display; structural color; switchable metasurface.

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1 Introduction

Structural coloration, the generation of colors through the use of micro- or nanostructures, has been steadily studied due to advantages such as long-term durability, high resolution, and lower toxicity compared to conventional pigment-based colorations [1–4]. Photonic crystals have been utilized to express structural colors [5], however, since the structures are larger than the working wavelength, the resolution of images produced by photonic crystals is limited [6]. Structural coloration from metasurfaces achieves super-resolution imaging beyond the diffraction limit [7, 8] and opens the door to multifunctional displays using periodic subwavelength nanostructures [9]. Phenomena that arise from light-matter interactions such as localized surface plasmon resonances (LSPRs) [10–13] and Mie-resonances [14, 15] allow for the production of structural colors from metasurfaces. Plasmonic metasurfaces based on LSPRs induce collective electron oscillations that enable the absorption and scattering of light [16, 17]. The location of the resonant

wavelength depends on the geometry of the constituent meta-atoms that make up the plasmonic metasurface. In contrast, all-dielectric metasurfaces can support Mie-like resonance [18]. Low optical loss of dielectric materials suggests the possibility of expansion of the achievable color gamut and the expression of highly saturated spectral colors. Structural coloration through all-dielectric metasurfaces has been demonstrated with various materials including amorphous silicon (a-Si) [19–23], silicon nitride (SiN) [24], gallium nitride (GaN) [25], and titanium dioxide (TiO₂) [26, 27].

Polarization-sensitive coloration is fascinating in the sense that the color can be immediately switched without changing the geometry or material of the metasurface [20, 28]. Polarization-sensitive coloration have been reported to achieve this, such as cross-shaped [29, 30], rectangular [19, 31], elliptical [32, 33] nanostructures and 1D gratings [21, 34]. An interesting application of polarization-sensitive coloration is in optical cryptography and image encryption devices [19], where the color can be switched “on” or “off” depending on the polarization of the incident light. However, the resonances of previously polarization-sensitive metasurfaces are not sufficiently diminished in the “off” state [21], but dim colors are generated instead [19, 21, 29, 35]. The residual colors at the turned-off condition degrade the performance of optical cryptography and image encryption. Although some efforts using phase change materials to change structural colors on metasurfaces have also been made, performances of color switching and

active control are not enough for practical usages [36–38]. In this sense, metasurfaces that produce full structural colors in the “on” state and near-zero reflectance in the “off” state have been rarely reported.

In this work, we present full-color metasurfaces with switchable reflection properties by the incident polarization angle (θ) where the color can be turned-off completely. The all-dielectric metasurfaces consist of triple-nanofin structures composed of hydrogenated amorphous silicon (a-Si:H). Mie-resonances are induced in the visible region under x-linearly polarized light to produce the desired color in reflection, while under y-linearly polarized light, no color is observed. In other words, colors are expressed when the polarization of the incident light is parallel to the long axis of the nanostructures, and suppressed when it is perpendicular. The simulated and experimented color palettes are compared using the CIE 1931 and HSV color spaces. The experimentally fabricated and measured metasurfaces confirm the capability of producing red, green, and blue colors that switch off to black when the polarization of the incident light is changed. This is also confirmed through the HSV color space as the brightness of the colors drops to almost zero. As demonstrations of encryption applications of this design, a dynamic QR code image, a switchable two-color object image, and an overlapped dual-portrait image are realized. These triple-nanofin metasurfaces (TNMs) could be used for applications such as ultrahigh-resolution images, full-color displays, optical storage media, security tags, and encryption technologies.

Figure 1: Schematic illustration of switchable metasurfaces. Colored images are switched on and off by the polarization angle (θ). The colors appear under (A) horizontally polarized incident light ($\theta = 0^\circ$) and concealed under (B) vertically polarized light ($\theta = 90^\circ$). (C) Schematic of the subpixel and superpixel with the periodicity (P). A single subpixel consists of a triple-nanofin structure with the same height (h) and width (w), and distance (d) with varied lengths (l_1 , l_2 , and l_3). One superpixel consists of four subpixels. An oblique angle SEM image of a fabricated sample is shown in the top right image.

2 Results and discussion

A schematic of the TNMs that are able to turn their color on and off depending on the incident polarization angle is shown in Figure 1. Under horizontally polarized incident light ($\theta = 0^\circ$; parallel to the long axis of nanostructures), the TNMs display an image (Figure 1A), while in contrast, the image is concealed when the light is vertically polarized ($\theta = 90^\circ$; perpendicular to the long axis of nanostructures) (Figure 1B). Each nanofin structure has a high aspect ratio between the width and length (Figure 1C). The TNMs consist of an array of periodic superpixels with periodicity (P). Each superpixel is made up of four subpixels arranged in a square lattice. The nanofins are composed of hydrogenated amorphous silicon (a-Si:H) that exhibits a high

refractive index in the visible range (approximately 3.5). TNMs which have the same height (h) and width (w) with variable lengths (l_1 , l_2 , and l_3) provide wide coverage of the color space compared to dual-nanofin metasurfaces (Supplementary Notes 1). The w must be smaller than 50 nm to maximize the turn-off property (Supplementary Notes 2). Each nanofin is spaced apart from adjacent nanofins with the same distance (d).

Numerically calculated and experimentally measured color palettes of the TNMs under $\theta = 0$ and 90° illumination are shown in Figure 2. The lengths of the nanofins, l_1 , l_2 , and l_3 , vary from 30 to 270 nm in steps of 60 nm to obtain a wide color coverage under conditions of $P = 900$ nm, $h = 250$ nm, $w = 50$ nm, and $d = 50$ nm. In both the simulations and experiments, the metasurfaces express full-color gamuts at

Figure 2: Numerically simulated and experimentally measured colors where $P = 900$ nm, $h = 250$ nm, $w = 50$ nm, and $d = 50$ nm. (A) Numerically calculated color palettes at $\theta = 0^\circ$ (top) and $\theta = 90^\circ$ (bottom). (B) Experimentally measured color palettes at $\theta = 0^\circ$ (top) and $\theta = 90^\circ$ (bottom). (C) Simulated (green dashed line), and experimental results (blue dashed line) plotted on the CIE 1931 chromaticity diagram at $\theta = 0^\circ$. The colors are represented in the HSV color space for (D) simulated and (E) experimental results at $\theta = 0^\circ$ and $\theta = 90^\circ$.

$\theta = 0^\circ$ with the colors suppressed at $\theta = 90^\circ$ (Figures 2A and B). The simulated (green dash line) and measured (blue dash line) color palettes are plotted on the CIE 1931 chromaticity diagram to identify the color coverage under $\theta = 0^\circ$ (Figure 2C). To investigate the switching off capabilities, the producible colors are plotted in the HSV color space, where the azimuth angle, a ratio of radial distance from the center, and height of the point corresponds to hue, saturation, and value, respectively (Figures 2D and E). At $\theta = 0^\circ$, the colors cover the HSV cone, while sinking down to the lower area at $\theta = 90^\circ$. This relates to the saturation and value tending towards zero, which corresponds to black. Minor differences between the calculated and measured color palettes can be attributed to fabrication defects including secondary electron scattering in the electron-beam lithography process and the side-wall effect during reactive ion-beam etching despite a reflection of unavoidable process errors in the simulation model (Supplementary Note 3) [39–42].

The intensities of the peaks of the red, green, and blue color spectra decrease when the incident polarization angle is changed from $\theta = 0^\circ$ to 90° , and near-zero reflection is achieved (Figure 3A). The reflectance spectra ($R(\lambda, \theta)$) are quantified by a function of wavelength (λ) and the polarization angle (θ) by the following equation [17, 19],

$$R(\lambda, \theta) = R_H(\lambda) \cos^2 \theta, \quad (1)$$

where $R_H(\lambda)$ is the reflectance spectrum of designed color pixels under horizontally polarized incident light $\theta = 0^\circ$. The geometry of triple-nanofins with a high refractive index of dielectric materials supports different Mie-resonant spectra depending on polarization angle. The reflected spectrum from the green metasurface shows the resonance peak around 515 and 660 nm. The magnetic field distribution of the TNMs with $l_1 = l_2 = l_3 = 270$ nm is calculated to identify the strongest peak (Figure 3B). The magnetic fields are confined in the triple-nanofins at $\theta = 0^\circ$ and the intensity gradually diminishes as the polarization changes to $\theta = 90^\circ$.

A geometric study is conducted to investigate the interactions between the nanostructures and lattice resonance of the TNMs (Figure 4). The coupling between adjacent nanofins is analyzed for two designs, by varying d from 0 to 55 nm with $l_1 = 30$ nm, $l_2 = 270$ nm, and $l_3 = 30$ nm and $l_1 = l_2 = l_3 = 270$ nm (Figure 4A) at $\theta = 0^\circ$. In both cases, the resonant wavelengths are blue-shifted as d increases. The shift of the resonant wavelengths is distinctly affected by the size of outer nanofins. The resonant wavelengths of the TNMs with shorter outside nanofins are almost independent to d ; however, the resonant wavelength of the longer nanofins is rapidly blue-shifted as d increases. The reflectance spectra of the TNMs with $l_1 = l_2 = l_3 = 270$ nm have been

Figure 3: (A) Polarization angle dependency of red, green, and blue metasurfaces where the polarization angle increases from 0° to 90° in steps of 10° . The intensity of reflectance spectra decreases gradually as the polarization is rotated from 0° to 90° . The reflectance spectra of the three colors are expressed in the different scales in order to clarify the spectral peaks. Since the green structures have a larger volume of hydrogenated amorphous silicon, it has the strongest resonant peak compared to red and blue ones. (B) The magnetic field distribution at $\lambda = 670$ nm varies for different polarization angles. $l_1 = l_2 = l_3 = 270$ nm and the height of cross-section of field distribution is $h/2$. The intensity of the magnetic field is normalized with respect to the maximum of 0° .

calculated while varying P from 600 to 1000 nm (Figure 4B). The resonant wavelength is steadily red-shifted from 580 to 680 nm as P increases. This linear relationship between the resonance wavelength and P indicates that the peak is due to lattice resonances [43].

Applications of cryptography are experimentally demonstrated with a QR code image, a two-color object image, and an overlapped dual-portrait image (Figure 5). The QR code is encoded with a green-colored metasurface (Figure 5A). Since the bare glass exhibits higher reflectance ($\sim 8\%$) than the TNMs at $\theta = 90^\circ$, the background space is filled with rectangular-shaped nanopillars to match the reflectance of TNMs. For incident light with polarization of $\theta = 0^\circ$, the green QR code is visible, while it disappears

Figure 4: Effect of geometrical parameters on the resonance peaks.

(A) The reflectance spectra of TNMs with (i) $l_1 = 30$ nm, $l_2 = 270$ nm, and $l_3 = 30$ nm and (ii) $l_1 = l_2 = l_3 = 270$ nm when d is varied from 0 to 55 nm. The reflectance intensities are normalized to themselves for clarity. (B) The reflectance spectra of TNMs ($l_1 = l_2 = l_3 = 270$ nm) when the periodicity of the superpixel is varied from 600 to 1000 nm.

under $\theta = 90^\circ$. This functionality can also be used to encode two different pictures into the same metasurface that can be displayed or hidden depending on the polarization direction. Images of a tree and a bird with different colors are alternated between for $\theta = 0^\circ$ and $\theta = 90^\circ$, with both images visible at $\theta = 45^\circ$ (Figure 5B). This switchability is achieved by designing TNMs composed of horizontally aligned green structures and vertically aligned blue structures. Furthermore, an overlapped image is encoded into a metasurface with the portraits of *Einstein* and *Boltzmann* (Figures 5C and D). The images are gradually converted when the polarization angle is rotated from 0° to 90° . The differences in the colors between the intended and fabricated images are due to an insufficient source intensity and a slightly mistuned white balance of the optical microscope.

3 Conclusions

We present polarization-sensitive TNMs with color palettes and imaging cryptographic applications that can be switched on and off depending on the polarization of the incident light. Full-color coverage is achieved at turn-on and near-zero reflectance is achieved at turned-off for both numerically calculated and experimental results. The reflection spectra and the magnetic field distribution depending on the incident polarization angles have been calculated to clarify gradual color change. Geometrical studies about d and P have been conducted to investigate the reflectance spectrum. Although the results have a narrower viewing angle (Supplementary Note 4) than other structural color devices [44, 45], the performance which is displayed with the CIE and HSV color diagrams

Figure 5: Cryptographic applications of the polarization-sensitive TNMs.

(A) A green QR code image is turned-on and -off when θ changes from 0° to 90° . (B) Images of a tree and a bird using green and blue TNMs. The blue bird image is turned-off at $\theta = 0^\circ$, while the green tree is turned-on, while the opposite is true at $\theta = 90^\circ$. An intermediate image is displayed at $\theta = 45^\circ$. Two polarization-selective overlapped images are demonstrated with the portraits of *Einstein* and *Boltzmann*. (C) Intended images of the TNMs. (D) Captured images using an optical microscope from the fabricated TNM for $\theta = 0^\circ$, 45° , and 90° .

shows these color filter designs significantly reduce residual colors when turned-off. As a validation of the completely switchable encryption, practical applications of QR code encryption, two-color image switching, and overlapped images resolving are demonstrated. Our polarization-sensitive metasurfaces could be integrated and used as high-security information encryption, security tag, multichannel imaging, and dynamic display [46–48].

4 Experimental section

4.1 Numerical modeling

Reflectance spectra were calculated using the commercially available finite-difference time-domain (FDTD) simulation software, Lumerical Solutions. Periodic boundary conditions were used for the x - and y -axis and perfectly matched layers were used for the z -axis. The refractive index of the SiO_2 substrate was set to 1.462. The refractive index of a-Si:H was obtained through ellipsometry. Each simulation result was calculated with periodic superpixel structures. The colors were calculated with a daylight (D50) illumination source.

4.2 Sample fabrication

The metasurfaces were fabricated by electron-beam lithography and patterning transfer processes with dry-etching. Firstly, on cleaned fused silica substrates, a 250 nm layer of a-Si:H was deposited by plasma-enhanced chemical vapor deposition (BMR technology, HiDep-SC). A 75 nm layer of photoresist (Microchem, PMMA 495 A2) was spin-coated and baked at 180°C for 5 min. Conductive polymer (Espacer 300Z) was spin-coated on the a-Si:H thin film at 2000 rpm for 1 min to reduce the charging effect. The nanopatterns were exposed using electron-beam lithography (ELIONIX, ELS-7800, 80 kV, 100 pA), then the exposed patterns were immersed with a developer (MIB-K:IPA = 1:3, Microchem) for 12 min at 0°C . A 25 nm chromium (Cr) layer was deposited using the electron-beam evaporator. The residual photoresist layer was removed using 60°C hot acetone for 12 h. The leftover Cr was transferred to the a-Si:H patterns using inductively coupled plasma reactive ion etching. Finally, the Cr mask was removed using Cr etchant for 2 min.

4.3 Optical measurement

The reflectance spectra of the metasurfaces were measured using an optical setup using a Xenon-lamp (Newport) light source. The intensity of the reflectance was measured with a spectrometer (HORIBA, iHR320). The QR code and tree-bird images were captured with the optical setup above while the portrait images were captured with a conventional optical microscope.

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Author contribution: C.J. and Y.Y. contributed equally to this work. J.R., C.J. and Y.Y. conceived the idea and initiated the work. C.J. and Y.Y. designed the structural color filter and simulated the reflectance spectra. Y.Y. fabricated the metasurfaces with electron-beam lithography. Y.Y., C.J. and T.L. obtained images using the Fourier-transform infrared spectroscopy and optical setup. J.J., J.M. and S.M. provided conceptual advice and theoretical analysis for structural color images. C.J., Y.Y. and J.R. mainly wrote the manuscript, T.B. and J.J. are partially involved in writing. J.R. guided the entire project. All the authors have accepted responsibility for the entire content of this submitted manuscript and approved submission.

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