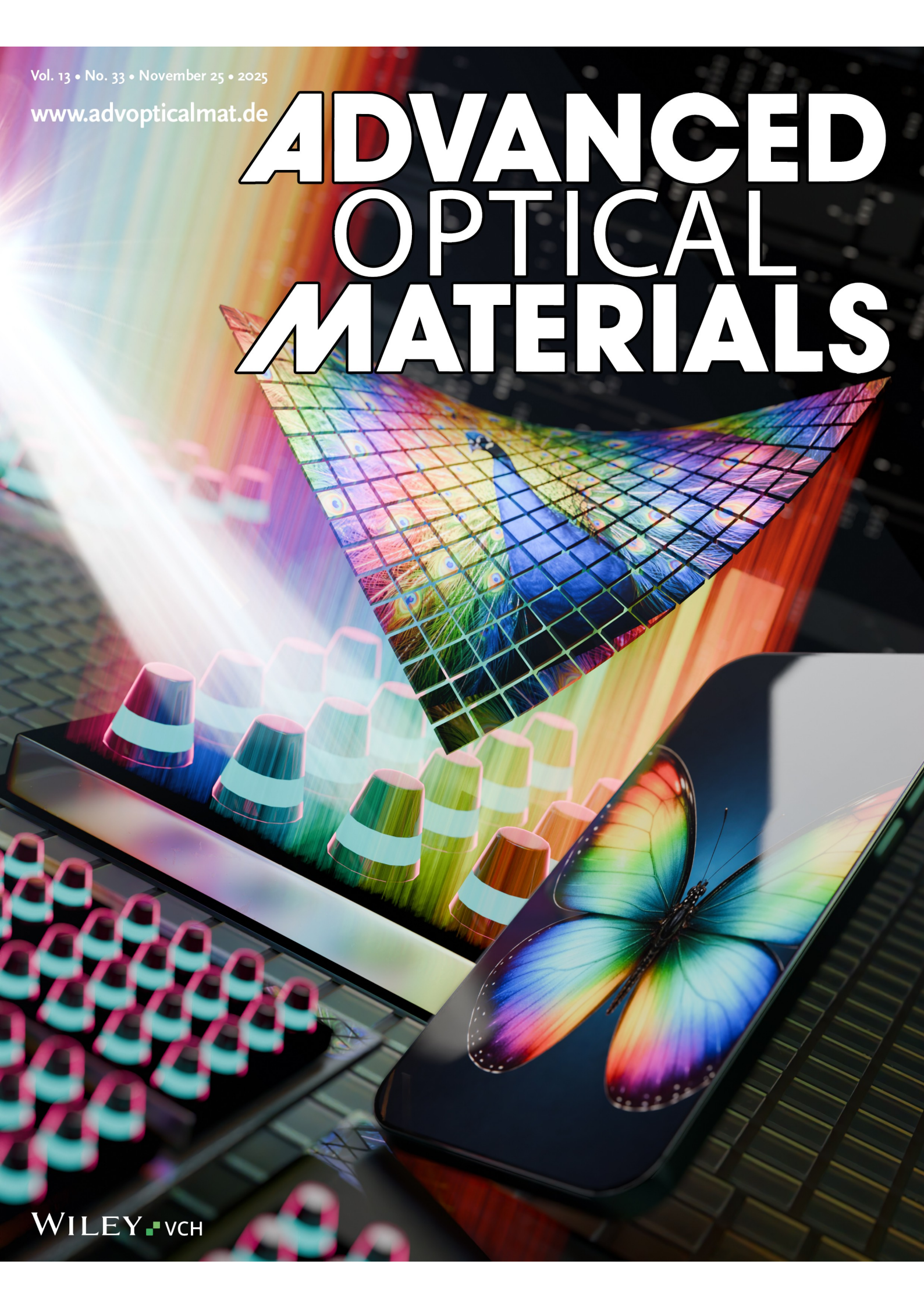


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ADVANCED OPTICAL MATERIALS

The background of the cover is a complex, vibrant illustration. It features a large, curved, grid-like structure that resembles a lens or a diffraction grating, displaying a full rainbow spectrum. Below this structure, there are several cylindrical optical components, possibly waveguides or resonators, arranged in a row. In the foreground, a smartphone is shown with a colorful butterfly on its screen. The entire scene is set against a dark, textured background, possibly representing a microchip or a complex optical device.

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RESEARCH ARTICLE **OPEN ACCESS**

Multi-Dielectric Metasurfaces for Ultrabright, Tunable Structural Color and Reconfigurable Optical Filtering with Extraordinarily Large Color Span

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ABSTRACT

Color is a fundamental element of human perception, impacting emotional and behavioural responses, and color space serves as an essential metric in advanced optical imaging and display technologies. Here, we propose and experimentally demonstrate a multi-dielectric metasurface architecture that enables bright, ultra-saturated, superior color generation with unprecedentedly high reflectivity up to 93%. The proposed metastructure consists of multi-layered truncated cones of gallium phosphide (GaP) sputtered between a pair of titanium dioxide (TiO₂) layers carefully constructed to induce both magnetic dipole (MD) and magnetic toroidal dipole (MTD) modes. The engineered design achieves a record experimental CIE 1931 color gamut of 222.1% in sRGB, 164.5% in Adobe RGB, and 117.4% in Rec. 2020. The pronounced dependence of the fabricated metastructures on their dielectric environment allows for exceptional color tunability across the visible spectrum via their thin film coating with consistently high reflectivity. Finally, as proof-of-concept demonstrations, we show reconfigurable, high-performance filters and reflectors employing the same design framework by precisely tailoring the geometry of the metasurface. The proposed architecture paves a promising route toward next-generation unexceptionally bright digital displays, printing technologies, and advanced photonic applications.

1 | Introduction

Light absorption-based dyes and pigments are the conventional generators of color; however, they suffer from chemical instability at elevated temperatures and exhibit a typically wide-band reflection spectrum, thus allowing only for a narrow color gamut [1]. Although the atomic-scale quantum emitters generate pure and vivid colors, their controlled manufacturing and continuous reliance on external power remain as major barriers to their widespread applications [2, 3]. With the advancement of nanofabrication, there has been growing interest in structural

color generation by exploiting subwavelength-scale nanodisks [4–6], nanogaps [7, 8], nanoparticles [9, 10], and gratings [11, 12]. Continued advancement expanded applications of these structures to digital display and imaging [13], color printing [14], information storage [15, 16], security [17], sensing [18], and colored photovoltaics [19].

Plasmonic metal nanostructures can produce a wide range of colors with a high resolution; however, due to their inherent Ohmic losses, their resulting colors are not bright [20, 21]. Similarly, the high extinction coefficient at shorter wavelengths

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hinders the most widely used electronic/optoelectronic material, silicon, from generating the vivid blue and violet hue [22, 23]. Even though the transparent dielectric materials such as TiO_2 , Si_3N_4 , and their combinations can overcome this limitation, their low refractive index prevents them from achieving a wide color gamut [24–28]. Although notable progress has been reported in structural color generation, a substantial gap remains in achieving commercial-level colors for advanced display applications [13, 15]. To date, no study has reported experimental demonstration of 100% Rec. 2020 of CIE 1931 chromaticity diagram coverage, the ultimate target for next-generation displays. Furthermore, the reflectivity reported in previous studies has not been radiant enough compared to other color-generation technologies [19, 20, 22, 25]. In addition, limited progress has been made in spectral tunability, where intensity and saturation often degrade, and tunability is restricted to an invariant structural framework [29–31]. Also, the splitting of resonances limits the conversion of a narrow-band color filter into a wide-band filter and reflector [24, 30]. Highly selective, efficient, polarization-independent, broad spectral linewidth filters and reflectors are essential for optical instrumentation [32], optical cavity construction [33], optical microscopy [34], spectroscopy [35], and astronomy [36]. Multi-layer thin films make highly reflective Bragg reflectors bulky and complex [37, 38], and the metal-based reflectors suffer from significant Ohmic loss [39, 40]. Therefore, to date, a pressing demand continues for a simple, widely reconfigurable, efficient optical filter and reflector in the visible spectrum.

In this work, we conceptually propose and experimentally demonstrate a class of multi-layer dielectric metasurfaces made of truncated cones (TCs), achieving 70% to 93% reflection efficiency (average 83%) across the visible spectrum. These engineered TC-shaped layers of gallium phosphide (GaP) and titanium dioxide (TiO_2) deliver ultra-pure colors experimentally with full-width-at-half-maximum (FWHM) reflection of 21.78 nm on average by suppressing unwanted resonances. Notably, the measured CIE 1931 color gamut areas expand to 222.1% of sRGB, 164.5% of Adobe RGB, and 117.4% of Rec. 2020 – representing the highest experimentally reported values of color space to date, to the best of our knowledge. Additionally, using the dielectric environment, such as PMMA, polystyrene, and PVK coatings, enables vivid color tuning, where PVK achieves the highest average wavelength tuning of 90 nm with an average reflectivity of 90%. To gain deeper insight into the bright colors along with large color space achieved by the proposed TC-shaped dielectric multi-layers, we further identified the underlying resonance mechanism for all observed outcomes by analysing the electric and magnetic field distributions as well as the multipolar decomposition. In addition, we account for the secondary colors of pink and magenta for the first time from a dielectric metasurface by employing a linearly polarized source. Finally, we show a highly selective, polarization-invariant tunable filter and broadband reflector by tailoring the diameter and constituent layers of the meta-atom using our metasurface architecture with a measured maximum top-flat linewidth of 70 nm. We present microscopic printed color images of the proposed design as a proof-of-concept demonstration.

We employ a hybrid meta-atom composed of TiO_2 and GaP tri-layer to maximize reflection efficiency and structural stability while minimizing the absorption loss. Even though GaP is a

promising material in nanophotonics, offering a higher refractive index and relatively lower absorption loss in the visible window, it is still unexplored for structural color generation. The well-developed GaP metastructure fabrication procedure has enabled the production of a wide range of shapes and structures [41–43]. The molecular beam epitaxy (MBE) and metal-organic chemical vapor deposition (MOCVD) are typically used to deposit single-crystal GaP film [44, 45]; however, expensive installation and selective substrate limit their widespread use. In comparison, RF sputtering is a simple, cost-effective method to deposit amorphous GaP film suitable for photonic applications [46, 47]. An all-GaP metasurface with a reasonable scattering volume might not achieve high reflection efficiency due to its extinction coefficient at shorter wavelengths; similar limitations are seen in silicon-based metasurfaces. Thereupon, we sandwich a thin GaP layer between two layers of transparent dielectric material of TiO_2 to ensure a high scattering volume while minimizing the absorption loss.

A multi-layer design with a gradient refractive index is chosen to suppress the Fabry-Pérot cavity mode resonances between the top and bottom surfaces [27]. Instead of a conventional cylindrical structure, we choose a truncated cone shape (TCS) to eliminate the electrical dipole (ED) resonance, where the base diameter is 120 nm larger than the top. We carefully thinned the GaP layer to 90 nm to support the target magnetic dipole (MD) with a minimum absorption loss. Figure 1a schematically depicts the array of multi-layer metasurface with a magnified view of the unit cell, and Figure 1b presents the top-view SEM image. The rational placement of GaP in the lower half allows a wider diameter to support a magnetic toroidal dipole (MTD) for broadband filtering. The top layer of TiO_2 is chosen at 140 nm; however, due to the TCS, the diameter is not large enough to support a Mie resonance. The bottom TiO_2 layer is set to a very thin 50 nm to eliminate unwanted resonances. Therefore, the overall thickness of metasurfaces becomes 280 nm, providing ample scattering volume and structural stability to obtain bright and vivid colors. The period of the meta-atoms ranges from 296 to 456 nm in 20 nm steps (9 different periods), with the corresponding base diameter variation from 176 to 272 nm in 12 nm increments. Thin films of GaP and TiO_2 were deposited on a glass substrate using RF sputtering, followed by electron beam lithography (EBL) patterning and inductively coupled plasma (ICP) dry etching. The measured refractive index and top SEM images of nine periods are presented in Sections S1 and S2. The detailed fabrication and angle measurement procedure of the meta-atom is provided in the Methods section and Section S3.

We tuned the base diameter from 144 to 344 nm for a period of 376 nm to capture the full range of resonant modes. All numerical analyses of the resonance modes within the meta-atoms were performed using the commercially available finite-difference time-domain (FDTD) software Lumerical. Figure 1c shows that the Mie resonances are progressively excited with the diameter increment, where more modes emerge at larger diameters. We select a base diameter of 270 nm from Figure 1c to investigate the mode decomposition, where two strong resonant modes appear at 605 and 558 nm. Figure 1d,e depicts an example of the field analysis using the horizontal and vertical cross-sectional electric field ($|E|$) and magnetic field ($|H|$) maps, as well as electric field vector distribution profiles. Figure 1d confirms that the resonant

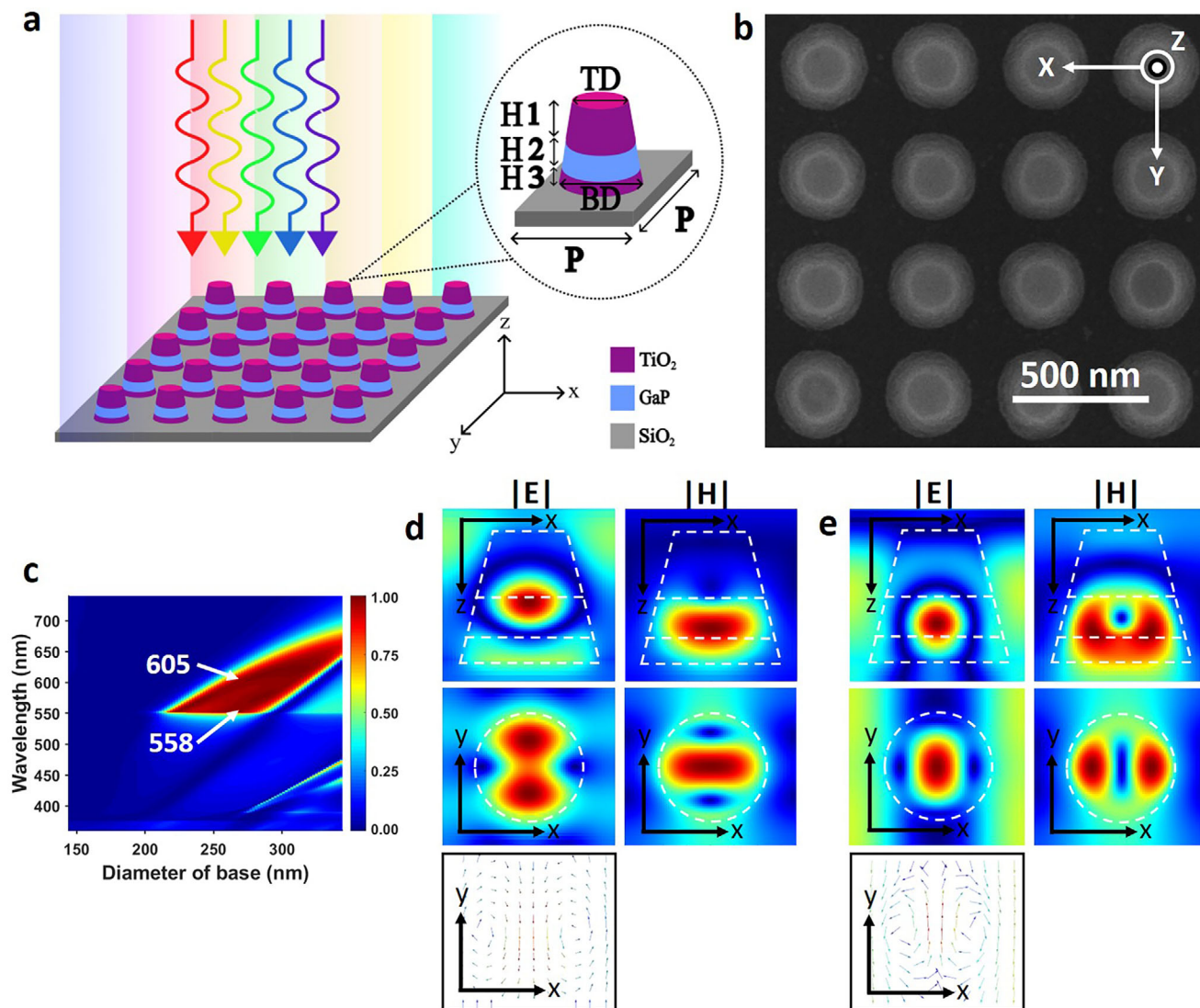


FIGURE 1 | Structural design, image, and mode decomposition of the proposed truncated cone-shaped (TCS) multi-layer metasurface. (a) Schematic illustration of the hybrid metasurface array under a normally applied source. The unit cell (inset) shows TCS geometry with TiO_2 (H1 140 nm, H3 50 nm) and GaP (H2 90 nm) layers; TD and BD are top diameter and base diameter, respectively. (b) Scanning electron microscope (SEM) image of the fabricated metasurface array from the period, p 436 nm, with the TD and BD of 140 and 260 nm, respectively. (c) Numerical reflectance intensity spectra for the increasing base diameter, BD, versus wavelength, for a fixed period of 376 nm. (d, e) Electric field ($|E|$) and magnetic field ($|H|$) maps and electric field vector distribution profiles at resonant wavelengths of 605 and 580 nm, representing the magnetic dipole (MD) and magnetic toroidal dipole (MTD) resonances for the base diameter of 270 nm.

mode at a longer wavelength is the well-known MD mode, while Figure 1e is characterized as the MTD mode. An MTD mode is excited within a high refractive index material disk, where the two opposite circular displacement currents form horizontally, creating a torus of magnetic field that forms vertically [48, 49]. Here, we chose a base diameter of 224 nm for the period 376 nm to generate color, where only one MD exists, enabling all energy to be concentrated in this MD mode.

The reflection performance and color purity were assessed in Figure 2a,b with the spectra of nine samples for the periods ranging from 296 to 456 nm. A homemade optical setup based on the back-focal plane (BFP) imaging was built to investigate the experimental results (see Method and Figure S4). For the period 456 nm, the reflection peak appears at the wavelength of

670 nm (numerical) and 673 nm (experimental), and the reflection efficiency is 96.3% (numerical) and 70% (experimental); also, the FWHM is 15.0 nm (numerical) and 23.4 nm (experimental). Similarly, for the period 436 nm, the corresponding numerical and experimental values are 642 and 645 nm, 99.4% and 73%, and FWHM of 17.0 and 23.1 nm, respectively. The numerical and experimental reflection peaks make an average 29 nm blue shift with each period and base diameter decrement. The overall experimental efficiency of our structure ranges from 70% to 93% (average 83%), with around 90% efficiency in the blue-violet region, outperforming all previous structural color generation reports. For instance, a silicon cylindrical metasurface in DMSO environment has an efficiency range of 15% to 74% [22], while a TiO_2 square-shaped metasurface shows a reflection efficiency of around 60% in the blue-violet region and an overall

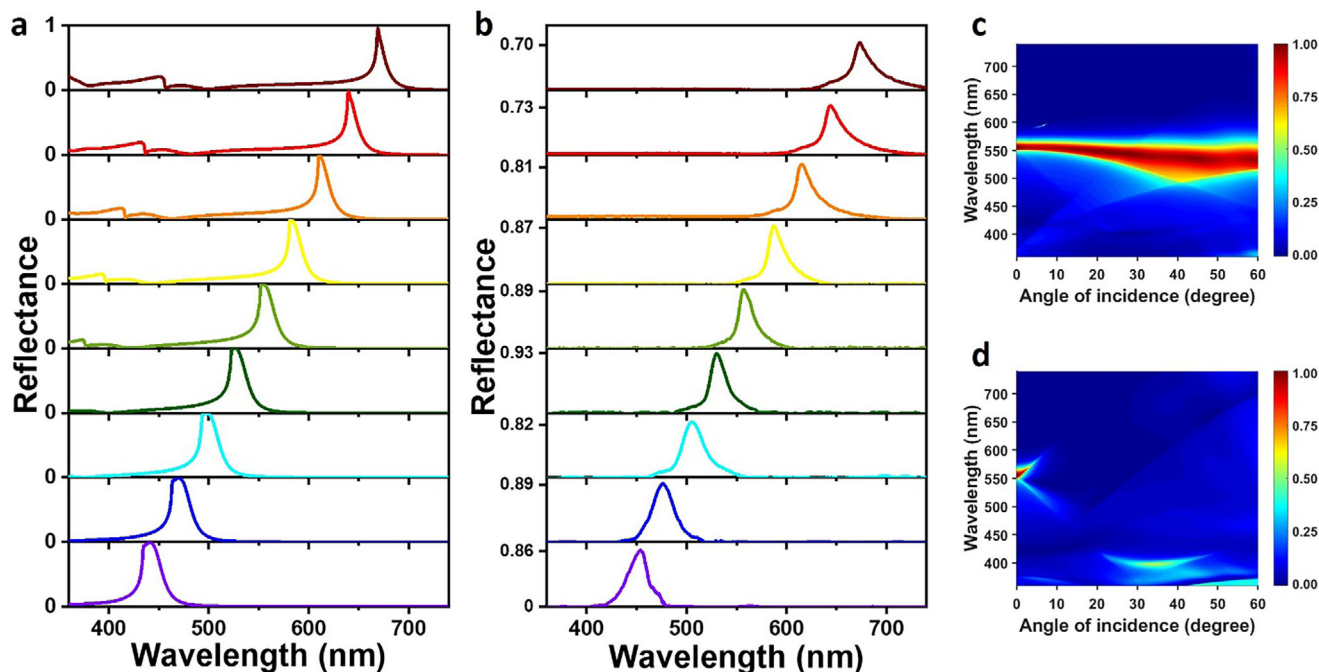


FIGURE 2 | Reflectance spectra and angular dependence on the applied source exhibiting high-performance reflectivity. (a, b) Numerical and experimental reflection spectra from the arrays of multi-layered TCS metasurface where the period ranges from 296 to 456 nm (bottom to top) in steps of 20 nm with the base diameter varying from 176 to 272 nm in increments of 12 nm. (c, d) Numerical angle-dependent reflectance for the period 376 nm for the s- and p-polarizations of incident light, showing strong angular stability for the s-polarization and rapid degradation for the p-polarization.

experimental efficiency of less than 70% [24]. Besides, a multi-dielectric $\text{SiO}_2\text{-TiO}_2\text{-Si}_3\text{N}_4$ metasurface has a relatively higher efficiency (60% to 89%), but still does not exceed 90% [27]. There is a weak resonance due to magnetic quadrupole on the numerical reflectance at a shorter wavelength in Figure 2a for the period 376 to 456 nm, which does not appear in the experimental reflectance in Figure 2b, attributed to the significant intensity difference between the simulated and experimental results as well as the limitations of the experimental setup. Additionally, the average numerical and experimental FWHM are 16.93 and 21.78 nm, confirming the ultra-saturated colors.

Figure 2c shows the angular independence for the s-polarization, while the p-polarization exhibits (Figure 2d) strong angular dependency for the period of 376 nm, where the base and top diameters are 224 and 104 nm. The MD mode is aligned with the electric field oscillation direction of the light source, which remains unchanged for the s-polarization, causing minimal effect on the MD mode. Figure 2c shows that the resonance remains narrow and intense up to 20°; after which, the spectral linewidth broadens gradually, adversely affecting color saturation. In contrast, the abrupt change of electric field oscillation for the p-polarization disrupts MD inside the meta-atom after a few degrees, causing the sudden disappearance of resonance (Figure 2d). We also compare our truncated cone shape (TCS) with the conventional cylindrical shape while keeping the height, period, and layer thicknesses the same for both. The numerical reflectance analysis in Section S5 demonstrates that the TCS effectively eliminates the unwanted ED mode, enhancing the intensity and purity of the target MD mode. Besides, the mode decomposition and location of ED in the meta-atom are presented in Section S6. To further evaluate the performance, we numer-

ically compare the reflectance with the all- TiO_2 and all-GaP metasurfaces in Section S7, where the height of all metasurfaces is fixed at 280 nm.

Since the photographed color quality is influenced by the camera sensors, illumination intensity, and the quality of the objective lens, we developed three custom microscope setups with three cameras – a Samsung Galaxy A31, a Canon DS126631 digital camera, and an Olympus DSX 1000 Opto-digital microscope (see Method Section). The photographed colors of violet, blue, cyan, green, chartreuse, yellow, orange, red, and Indian red (wine red in the Canon camera) for nine periods varying from 296 to 456 nm (bottom to top rows) are presented in Figure 3a–c. The purpose of varying sample areas is to compare the quality of structural colors with conventional pigments and dye-based printed colors, which are typically in the order of 25 μm . The color quality remains consistent for the side length of 50 to 10 μm for all systems, underpinning the high resolution, while the blurred images for the order of 5 μm are attributed to the limitation of the imaging system rather than the metasurface itself. Previously, several reports have reported color generation using a single-pixel plasmonic or 2×2 dielectric nanodisks metasurface [20, 22], which has generally been insufficient for practical applications due to their weak resonance intensity. We placed 42 experimental sample spectra (9 samples in Figure 2b, and 33 samples in Figure S9a) onto the CIE 1931 chromaticity diagram encompassing a broad chromaticity space (black stars in Figure 3d) to evaluate the color performance further. The rigorous measurement methodology (see Section S11) quantifies the exceptional area spanning 222.1% of sRGB (Standard Red, Green, and Blue), 164.5% of Adobe RGB, and 117.4% of Rec. 2020. To the best of our knowledge, these three measurements

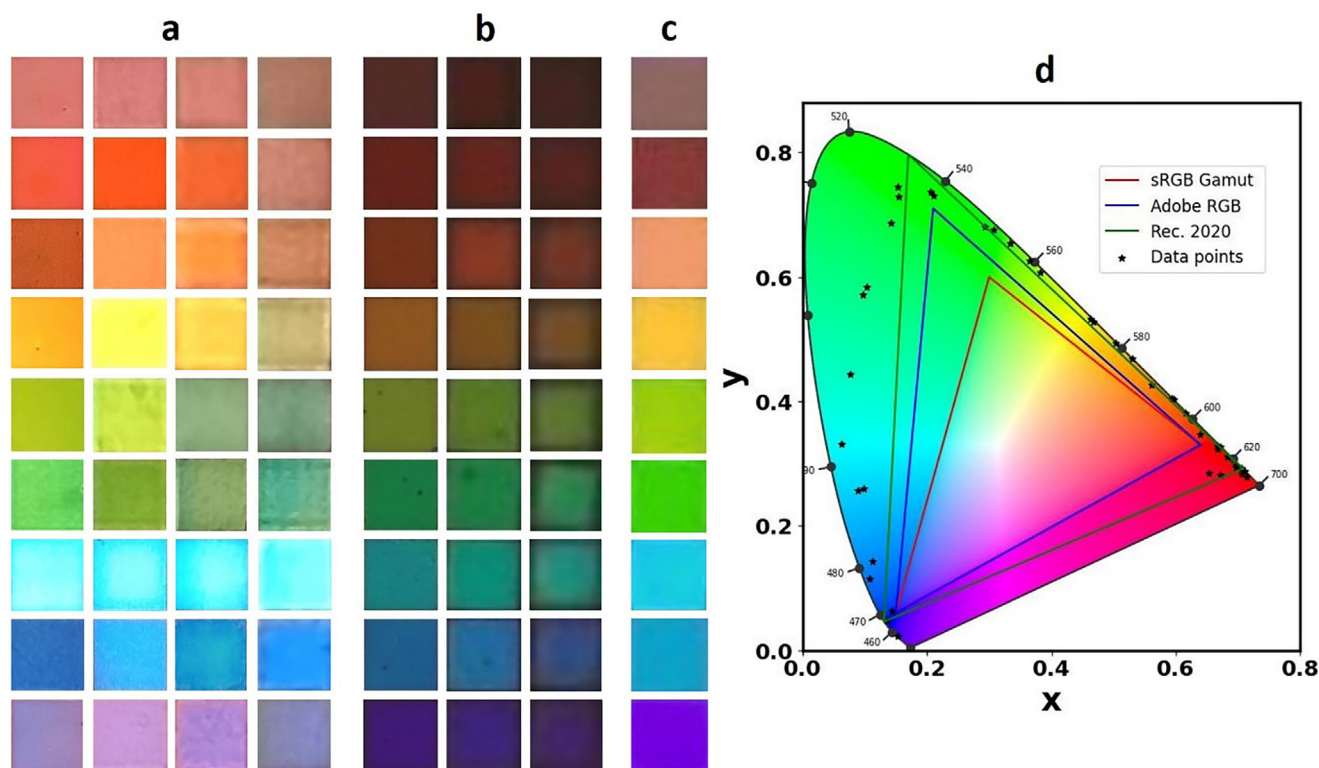


FIGURE 3 | Structural color demonstration and chromaticity diagram analysis. (a) Experimental color palettes photographed by smartphone camera (Samsung Galaxy A31) through a 10× objective of microscope lens from square metasurfaces with side lengths of 50, 20, 10, and 5 μm (left to right). (b) Corresponding color palettes photographed by a Canon DSL26631 digital camera through a 10× objective from the square metasurfaces of side lengths of 50, 20, and 10 μm (left to right). (c) Experimental color palette photographed by an Olympus DSX 1000 Opto-digital microscope from the square metasurface of side length 50 μm. (d) Experimental color palette on the CIE 1931 chromaticity diagram for the 42 experimental samples (black stars) of metasurface side 50 μm, covering an exceptionally wide color gamut.

are the largest-ever demonstrated gamut areas and, notably, the first-ever reported instance that exceeded 100% of Rec. 2020. In comparison, several remarkable reports include a maximum of 109% of Adobe RGB from the plasmonic material [21], 97.2% of Rec. 2020, and 68% of Rec. 2020 using the dielectric metasurfaces [22, 27], which are the previously reported best in their classes.

We studied numerical refractive index modulation of the dielectric environment from 1.0 to 2.0 for a fixed period of 396 nm, as demonstrated in Figure 4a. The pronounced resonance maintains high reflection intensity with minimal mode splitting or spectral distortion up to the refractive index of approximately 1.70. We select three transparent polymers with distinct refractive indices: polymethyl methacrylate (PMMA, $n = 1.49$), polystyrene ($n = 1.589$), and poly(9-vinyl carbazole) (PVK, $n = 1.683$) to realize the color variation experimentally. The control numerical demonstration of the wavelength shift of 54, 70, and 96 nm based on MD mode transition upon coverage of PMMA, polystyrene, and PVK, respectively, relative to the air covering is shown in Figure 4b. Experimental results are confirmed this trend with a high reflectance level of 92% for PMMA and polystyrene and 94% for PVK, while the reflectance is 87% in air (Figure 4c). The corresponding average numerical wavelength tuning is 51, 67, and 90 nm, with the respective experimental reflectance values are 88%, 88%, and 90% for PMMA, polystyrene, and PVK across nine different periods, presented in Section S12. To develop a better understanding of the resonant mechanism

behind the spectral tuning, we examine the electric field map across the halfway cross-sections of the GaP layer in three media (Figure 4d), using the identical geometry of Figure 1d,e. The increasingly delocalized field of MD and MTD into the surrounding dielectric medium enables the larger effective mode volume, resulting in a resonance red shift. More details of the mode decomposition when covered by polystyrene are provided in Section S13.

Figure 4e depicts the photographs of the adaptive color transition across the period ranging from 296 to 416 nm with three dielectric coatings. The violet hue in air transits to the denim blue, the rifle green, and the lion yellow for the period 296 nm; similarly, the blue hue in air shifts to the aquamarine, the celadon, and the sunglow yellow for the period 316 nm when covered with PMMA, polystyrene, and PVK. All periods exhibit a gradient and vibrant color evolution with a robust intensity. Furthermore, chartreuse and yellow colors emerge at shorter periods (336 and 356 nm) with PVK coverage, compared to their visual appearance at longer periods (376 and 396 nm) in the air environment, supporting the substantial wavelength tunability. The real-time color tuning is also achievable by integrating microfluidic channels containing transparent oils and liquids beside the solid polymer coatings. Additionally, the pronounced environmental sensitivity indicates the capability for sensing applications. The adaptive and rigorous color transitions for varying dielectric environments validate the design's structural versatility and fidelity.

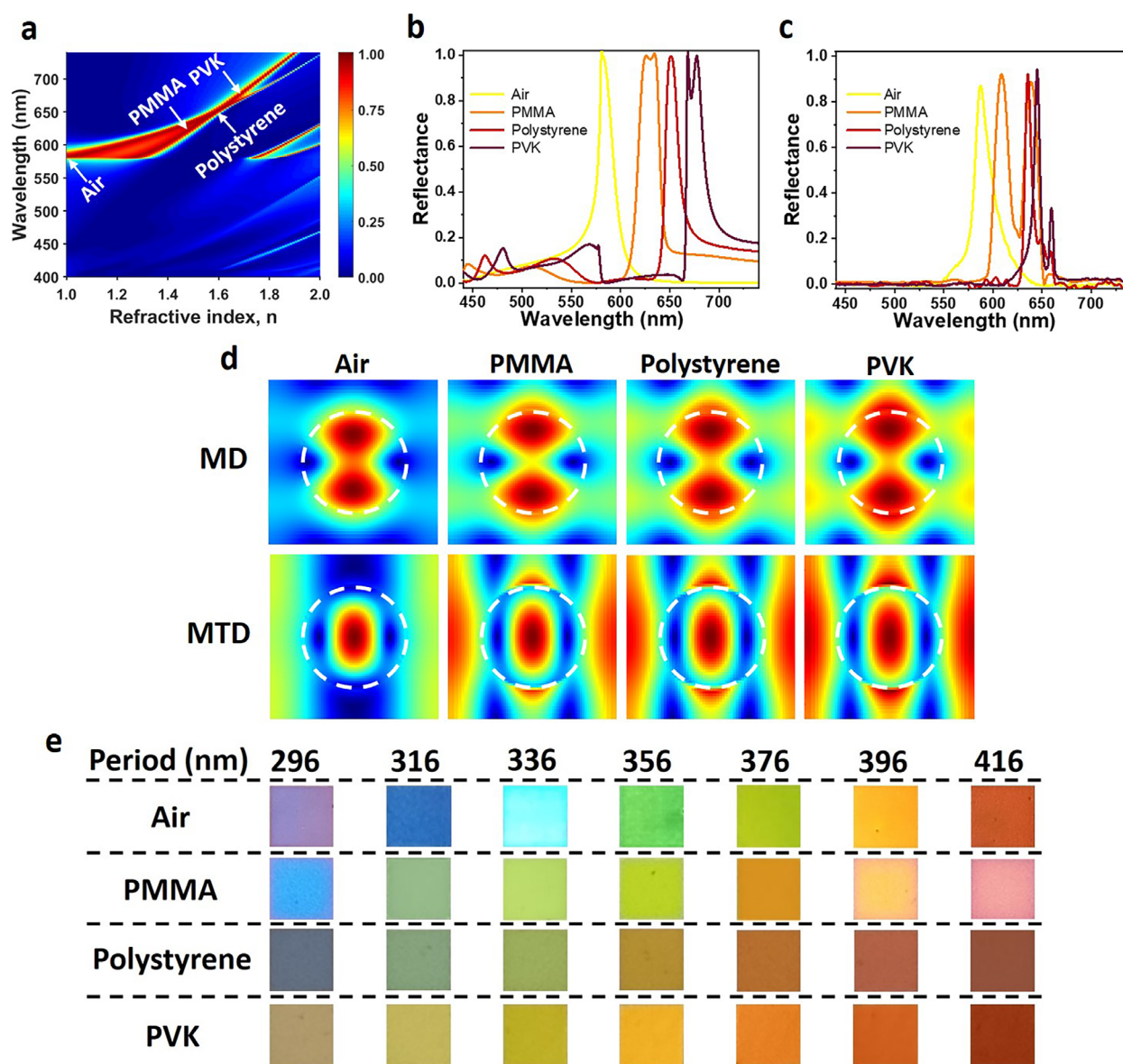


FIGURE 4 | Color tuning using dielectric environment, mode decomposition, and experimental color response. (a) Numerical wavelength modulation for a fixed period of 396 nm as a function of the environmental refractive index from 1.0 to 2.0. (b, c) Numerical and experimental reflectance for the environment of air ($n = 1.0$), PMMA ($n = 1.49$), polystyrene ($n = 1.589$), and PVK ($n = 1.683$). (d) Cross-sectional numerical electric field map for the different transparent environments affecting the mode volume of MD and MTD by a period of 376 nm through the mid-plane of the GaP layer. (e) Experimental photographed color palettes taken by a smartphone camera through a 10x microscope objective showing the response of the varying environments.

The coupling of MD and MTD manifests as a progressive broadening of the spectral linewidth by increasing the diameter of the TCS unit cell. Figure 5a,d exhibits the monotonic increment of spectral linewidth where the top spectral linewidth (90% reflection) broadens to 59 nm (numerical) and 34 nm (experimental) for the diameter 320 nm with a high reflectivity of 100% (numerical) and 87% (experimental). Besides this top wide-spectral linewidth, both have an incredible selectivity (changes of spectral position from 10% to 90% reflection, average of both sides) value of 39.7 nm (numerical) and 25 nm (experimental). This spectacular experimental selectivity is crucial in optical communication to select the smaller transition band, minimizing bandwidth loss. The numerical (Figure 5b) and experimental (Figure 5e) tunability of filters demonstrates high reflectivity,

while the selectivity decreases gradually in smaller periods. To realize the largest spectral linewidth, we numerically increase the thickness of the GaP layer as represented in Figure 5c. The spectral linewidths broaden gradually following the thickness increment without affecting the top-flat linewidth and selectivity. To realize this experimentally, we fabricated a structure of GaP with a thickness of 130 nm, which demonstrates the maximum 70 nm top-spectral linewidth along with a reflectivity level of 90% (Figure 5f), corresponding to a 53 nm red shift, compared to the numerical linewidth of 88 nm and 100% reflectivity (Figure 5c). This red shift is likely due to the fabrication-induced variations. Additional numerical broadband reflection tuning is provided in Section S15. Importantly, all measurements regarding the filters and reflectors confirm the

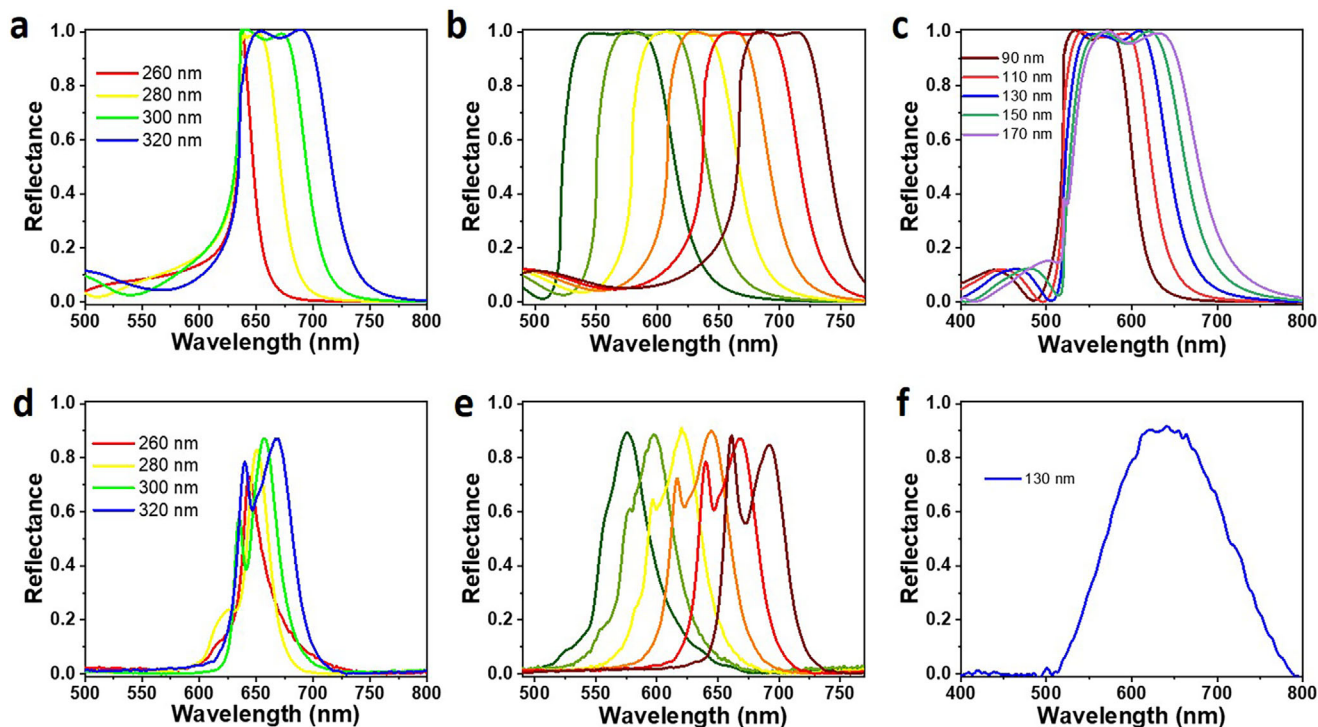


FIGURE 5 | Spectral tuning and broadband reflectance for the applications of highly selective filters and wide-spectrum reflectors. (a, d) Numerical and experimental reflectance spectra for varying base diameter from 260 to 320 nm and a fixed period of 436 nm, while GaP thickness is 90 nm. (b, e) Numerical and experimental reflectance tuning for varying period from 356 to 456 nm featuring a base diameter 60 nm larger than those of the corresponding color generation structures. (c) Numerical reflectance spectra for varied GaP thickness from 90 to 170 nm in 20 nm steps, while the base and top diameters are 260 and 140 nm with a fixed period of 356 nm. (f) Experimental reflectance for GaP thickness of 130 nm, including top and bottom TiO_2 layers of 120 and 30 nm, while the period is fixed at 356 nm.

independence of the polarization state of the applied light source.

Finally, as a proof-of-concept demonstration, we fabricated four microscopic “UNAM” patterns with identical printing dimensions, but distinct periods for each. Figure 6a represents the top view of the SEM pattern with the gradual changing of periods from the largest to the smallest. Figure 6b demonstrates the corresponding printing colors for Figure 6a, where unpolarized and linearly polarized light sources illuminate the top and bottom rows. The excitation of pink and magenta colors (Figure 6b, bottom row) from the period 416 to 456 nm is exceptional in the dielectric-based structural color metasurface. As the secondary colors, they required the simultaneous excitation and effective mixing of red and blue hues. The polarization-sensitive excitation of multiple resonant modes arises from the period 416 to 456 nm, where the same pattern excites red in Figure 6c (top row) and magenta (bottom row) colors depending on the influence of source polarization states. A more detailed demonstration of the polarization dependency on the source light is provided in Section S17. Figure 6d,e further illustrates the influence of the dielectric environment by presenting the adaptive and vivid color modulation. Additionally, the transparent coating effectively suppresses the reflectance from the fused silica wafer, leading to a visually smoother unpatterned area (Figure 6d,e). Further experimental color printings and discussion on the impact of dielectric environment are provided in Section S18.

2 | Conclusion

In summary, we have proposed and experimentally demonstrated a dielectric metasurface architecture of multi-layered truncated cones with outstanding performance of ultra-pure and bright color generation. Through strategic design and careful implementation, the proposed metastructure effectively suppresses the unwanted resonances and achieves the CIE 1931 color chart to the highest ever coverage area, to the best of our knowledge, which is the largest span ever reported to date. Furthermore, excellent color tuning is demonstrated by exploring the sensitivity to the dielectric environment of PMMA, polystyrene, and PVK. A detailed analysis based on multipole decomposition explains the fascinating underlying resonant mechanism, revealing magnetic dipole and toroidal modes. Additionally, the tunable spectral linewidth establishes the platforms of high-performance optical filters and reflectors. Finally, a vibrant printing color display with large spectral linewidth tunability represents a compelling alternative to conventional technologies for advanced display and photonic applications.

3 | Methods

3.1 | Numerical Calculations

All numerical reflectance spectra and mode decompositions were calculated using commercially available software Lumerical

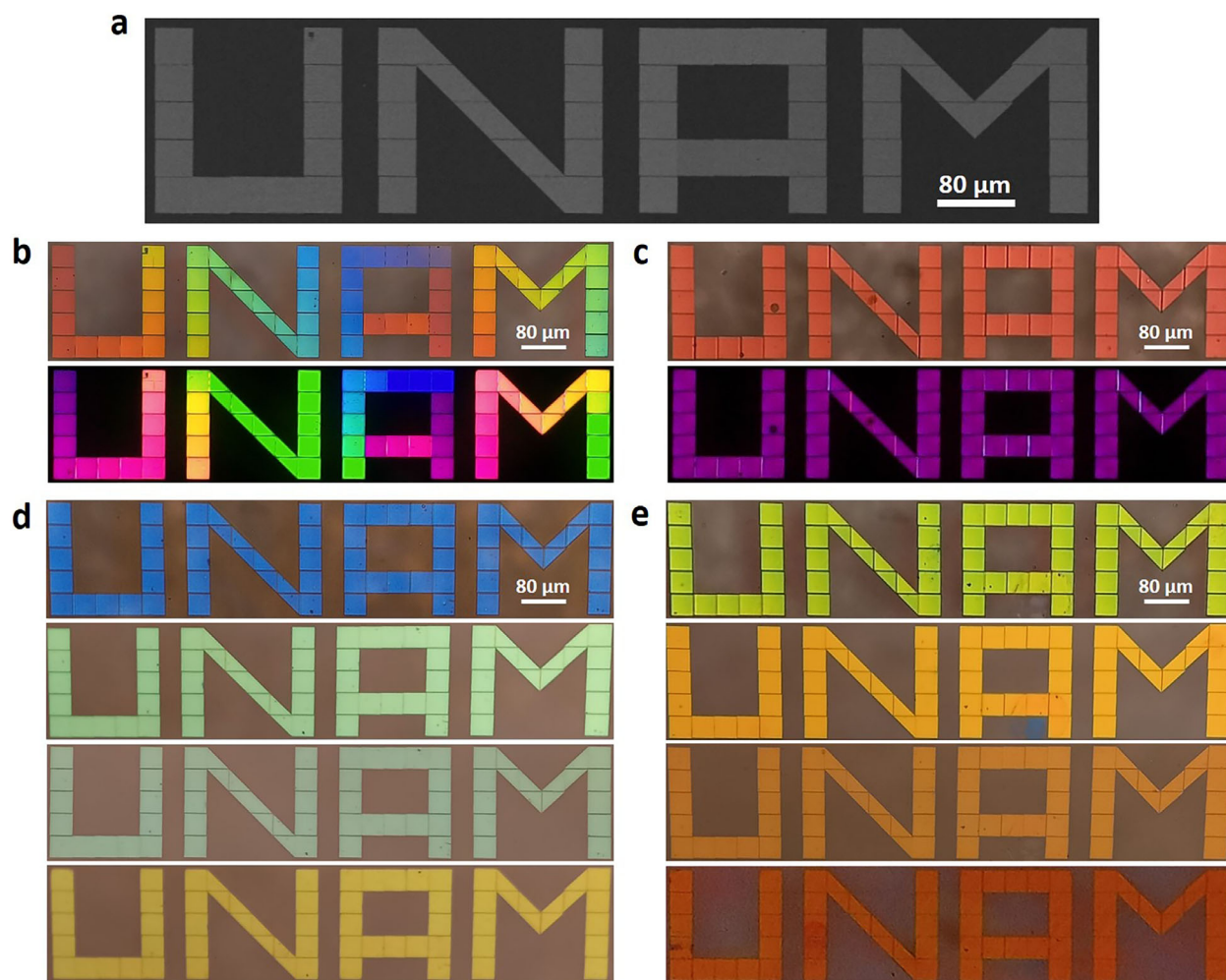


FIGURE 6 | SEM image, microscopic color printing, and sensitivity to polarization state and dielectric environment. (a) Top-view SEM image of the fabricated UNAM (Ulusal Nanoteknoloji Araştırma Merkezi) pattern, where dimensions are 1.0 mm by 0.2 mm with a 40 μm small block. (b) Unpolarized bright field illuminated color transition from wine red to violet (top row), and corresponding polarization-sensitive hues (bottom row), photographed by ZESIS and Olympus microscope, respectively. (c) Unpolarized bright field illuminated red printing (top row) changes to magenta printing (bottom row) under the influence of a linearly polarized source. (d, e) Post-fabrication printed color tuning of blue (d) and chartreuse (e) hues to aquamarine, celadon, sunglow yellow (d), and amber, carrot orange, cardinal (e), confirming the sensitivity to PMMA (second row), polystyrene (third row), and PVK (last row).

based on the finite-difference time-domain (FDTD) method. The periodic boundary condition was applied horizontally to match the periodic structure, while the perfectly matched layer (PML) was used vertically to absorb the reflected and transmitted light. The plane wave source of the Bloch/periodic wave type was employed as a normally applied source. In contrast, the Broadband Fixed Angle Source Technique (BFAST) wave type is applied for the inclined illumination. The refractive indices of PMMA, polystyrene, and PVK was chosen to be 1.49, 1.589, and 1.683 for the numerical simulations. Here, we chose SiO₂ (Glass) – Palik material from the software material database as the substrate.

3.2 | Fabrication

Fabrication was performed using a top-down process flow with electron beam lithography (EBL) and inductively coupled plasma etching (ICP). First, we cleaned the fused silica wafers using

Hellmanex and DI water solution in the sonicator, followed by acetone and isopropanol. A standard Piranha solution (H₂SO₄: H₂O₂ = 3:1) was used to make wafers ultraclean. After rinsing with water and drying using the N₂ gas flow, the wafers were placed on the hotplate to remove water molecules. Then, the wafers were transferred to the chamber of the RF sputter. We deposited TiO₂ and GaP thin films in high-vacuum conditions. After that, the electron beam resist (PMMA) was deposited by a spinner and baked for 3 min on the hot plate. The deposited materials, fused silica wafers, and PMMA are electrically non-conductors that can lead to charge accumulation during the EBL process. To avoid this, a conducting polymer E-Spacer 300Z (SHOWA DENKO) was spin-coated (7 nm) on top of PMMA. Then, the EBL was performed to make the cylindrical pattern on PMMA, followed by the development with MIBK: IPA (1:3), IPA, and DI water. A 50 nm thick chromium (Cr) layer was deposited on the patterned wafers by the e-beam evaporator (MIDAS e-beam Evaporator). Then, the lift-off process was carried out with acetone, removing the PMMA and unwanted Cr to obtain the Cr hard mask. After

drying the wafers with N₂ gas flow, the ICP (STS Multiplex ICP) performed dry etching (ICP power 400 W, bias power 350 W, gas flow: CHF₃ 20 sccm, SF₆ 20 sccm, and O₂ 4 sccm) to achieve the target geometry. The sidewall angle can be adjusted by tuning the ICP bias power. Finally, the remaining Cr was removed using the chrome etchant. We prepared polystyrene and PVK solutions by adding polystyrene powder (Sigma-Aldrich) and PVK powder (Aldrich) into toluene. A spinner was used to deposit PMMA, polystyrene, and PVK for the optical characterization. Similarly, toluene was also used to remove the polystyrene and PVK thin film.

3.3 | Optical Characterization

The reflectance measurement was performed by a back-focal plane (BFP) imaging setup connecting with a slit spectrometer and a CCD camera. This arrangement includes a 4f lens assembly with a halogen lamp for illumination, mirrors, a Mitutoyo Plan Apo 10× objective (NA 0.28), a spatial filter, and a polarizer (see Part S4). Two custom optical setups photographed the images with different optical microscopes and cameras. A smartphone camera (Samsung Galaxy A31) with a ZESIS (Axio Scope AI, 10× objective, NA 0.2) microscope took one set of bright field microscopy images. A Canon DSI26631 camera with a CASCADE MICROTECH (IRIX, 10× objective, NA 0.25) microscope took another set of bright field microscopy images. In addition, an Olympus DSX 1000 Opto-digital microscope was employed for the unpolarized and linearly polarized illumination and associated image acquisition.

Author Contributions

Md Rumon Miah conceived the idea, performed all numerical simulations, fabricated and characterized all samples, and wrote the first manuscript. Yavuz Selim Aydın prepared the chromaticity diagrams and wrote the relevant part. Hilmi Volkan Demir supervised the project at all steps and wrote the final manuscript.

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Conflicts of Interest

None of the authors have a conflict of interest to disclose.

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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Supporting Information

Additional supporting information can be found online in the Supporting Information section.

Supporting File: adom71173-sup-0001-SuppMat.docx.