Nano-optic features such as nanopillars, subwavelength resonators and Mie voids are all emerging as new tools for creating realistic, durable and environmentally sound alternatives to traditional pigments.
Structured Colors with Dielectric Nanoresonators

Joel Yang, Hao Wang, John Chan You En, Mario Hentschel and Yuri Kivshar
Visual perception of colors plays a vital role in our daily lives. Human efforts to create colors have evolved from collecting natural dyes and pigments to the predominant use of industrial synthetic dyes in modern society, driven by the need to reduce costs and meet specific requirements. However, colors generated from dyes and pigments degrade due to radiation damage, photobleaching or heat. These substances might also contain toxic compounds that pose risks to human health and the environment. And the relatively large size of pigment particles—commonly on the order of tens of micrometers—severely limits their applications.

Dye-free structural colors present a compelling alternative, offering unprecedented fade resistance, print resolution, scalable manufacturability and manipulation of viewing-angle dependence. These colors result from the physical interaction of light with nanostructured materials, a phenomenon that has naturally evolved predominantly in some insects, birds and plants. It is believed that nanostructural colors serve various functions such as warning, camouflage, physiological constraints (for example, color can be related to UV protection) and mate selection.

Inspired by nature, researchers have explored the use of various materials, including metals, semiconductors and dielectrics, for artificial structural-color generation. While metals have been extensively investigated for color applications through the mechanism of surface plasmon resonance, they suffer from intrinsic loss, oxidation and high cost. Dielectric systems have thus gained attention, due to their lower loss, greater flexibility in tuning resonances and fabrication strategies that align with industrial standards.

Dielectric nanostructures from high-refractive-index (HRI) materials such as silicon, titanium oxide, silicon nitride, diamond and gallium nitride exhibit strong optical responses, and their behavior can be understood through Mie theory, providing another available platform for nano-optics and leading to the emergence of Mie-tronics. With multipolar interferences and bound states in the continuum (BICs), high values of the quality factor ($Q$) can be achieved, promising vibrant color generation.

In contrast to color generation via plasmonics and Mie scattering, low-refractive-index (LRI) materials ($n < 1.7$) typically require larger dimensions for color generation, which limits control and resolution. Examples include periodic structures like photonic crystals, total internal reflection with microdroplets, and thin-film interference. Transparent and cost-effective LRI materials, such as glass, plastics and polymers, are widely available. Identifying new mechanisms for generating colors with subwavelength dimensions using these materials enables the design of dye-free, colorful devices with unique properties and applications. Further, the use of LRI structures could leverage biodegradable polymers (such as chitosan and cellulose) for sustainable development.

This feature focuses on recent discoveries, including 3D-printed LRI nanopillars for a 3D color palette; air voids on the surface of HRI dielectrics (Mie voids), which exhibit Mie-like scattering for individual color control; and BICs for highly saturated color generation. Simply controlling nanostructural morphology and geometry enables vivid full colors and varying levels of gray, providing a nanoscale-resolution “magic brush” for painting. Further, we believe these novel concepts and methods enable unprecedented control over light-matter interaction and underpin the development of next-generation optical devices for security, communications, displays, ultrafast computation, compact biosensing and quantum technology.

**Nanopillars**

To achieve a full range of colors from a single LRI material through simple geometric modulation in
Inspired by nature, researchers have explored the use of various materials, including metals, semiconductors and dielectrics, for artificial structural-color generation.

A single patterning step, nanoscale additive manufacturing—specifically, two-photon polymerization lithography (TPL)—can overcome the constraints of conventional binary lithography. TPL offers advantages such as rapid prototyping, customized design, low cost and minimal waste, enabling the fabrication of structures with hundred-nanometer length scales.

In recent work, a group led by several of the authors achieved the first full color palette in a 3D color space at the single-nanopillar level, by tuning the three printing parameters of the nanopillar—height (H), exposure time (T) and period (P).

In our work, we observed that individual LRI nanopillars (roughly 300–500 nm in diameter) that are shorter than 0.7 μm exhibit multiple shades of gray, which are challenging to achieve with resonant nanostructures made of HRI materials. As the nanopillars increase in height, they begin to behave like leaky waveguides, which results in wavelength-selective effects. The hues of single nanopillars vary dramatically across the gamut of colors with the height of the nanopillars. Saturation can be further tuned by varying the periodicity—a larger spacing “dilutes” the color; a smaller one increases the saturation. Notably, the variation of periodicity negligibly changes the hue, a pattern distinct from that of the more familiar diffractive effect. Converting all experimentally obtained spectra into colors achieves a wide coverage of the full hue–saturation–brightness (HSB) color space.

This full color palette allows arbitrary colorful and grayscale images to be mapped into HTP channels, analogous to RGB channels in image-processing software. Using a 3.2-μm pixel size (to fully utilize the available colors) enabled nanopillar printing of a colorful, 300×240-pixel painting of apples. A 240×300-pixel grayscale image of a cat, meanwhile, was created with nanopillars at a fixed 0.8-μm period, while heights and exposure times varied freely. As a result, a single nanopillar represents each pixel, enabling 18 grayscale levels in the print. Despite the tiled appearance caused by variations across different write fields and equipment limitations, the details of the original images are preserved.

As nanopillars are directional scatterers, different visual effects can be realized under different illumination conditions. A microscopic, 2000×658-pixel replica of the famous Chinese calligraphy artwork *Preface to the Poems Composed at the Orchid Pavilion* was 3D-printed with a fixed 0.8-μm pixel size. Under brightfield transmissive illumination, grayscale Chinese ink details and red seals are preserved on a white background.

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3D printing with two-photon polymerization lithography (left) enables the creation of full-color and grayscale images using low-refractive-index nanopillars. The printed image is formed by nanopillars designed with different heights, periodicities and diameters (right).

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reproducing traditional Chinese cocoon-paper calligraphy. Under glancing-angle illumination (darkfield), grayscale inversion and color desaturation are caused by strong broadband scattering, replicating the other primary artistic form of Chinese calligraphy, a stone rubbing on a black background. These results underscore the potential for realizing a 3D color design space using a simple geometry and a single patterning step by TPL.

**BICs and Mie resonators**

Another approach, BICs, can enable highly saturated structural colors. In optics, BIC refers to the perfect confinement of light with specific frequencies in a structure, though these specific frequencies belong to a continuous range of frequencies in the incident light spectrum. Conventional wisdom holds that light with those specific frequencies will couple with the continuous spectrum and radiate out of the structure, resulting in leaky resonance modes. Yet BICs defy this conventional wisdom—remaining localized with no radiation despite being part of the continuous spectrum (see “Engineering with Bound States in the Continuum,” OPN, January 2020, p. 38).

BICs were proposed in 1929 by von Neumann and Wigner, who mathematically constructed an artificial quantum potential that produces a discrete state with positive energy and zero linewidth inside the continuous spectrum. Due to its zero linewidth, a pure BIC has an infinitely large $Q$ factor—achievable only if a material were to have infinite size or zero or infinite permittivity. In practice, quasi-BICs can be achieved only with finite (albeit large) $Q$ factors given finite sample sizes, material absorption and structural imperfections.
In a recent example, bound states in the continuum were used to experimentally demonstrate a highly saturated red pixel not found in nature.

Nonetheless, quasi-BICs are still useful for applications that demand narrowband spectral filters.

In a recent example, several of us used BICs to experimentally demonstrate a highly saturated red pixel not found in nature. This red pixel approximates a reflectance spectrum with unity reflectance at red wavelengths (beyond 600 nm) and zero reflectance elsewhere, characteristic of Schrödinger’s “ideal” color pixel. Saturated red had been challenging in structural color, as typical nanoresonators supporting fundamental modes at red wavelengths also support additional higher-order modes at blue wavelengths (380 nm to 480 nm), and as the resonances of typical nanoresonators are much broader than that of Schrödinger’s ideal color pixel.

To overcome these challenges, the group fabricated amorphous silicon (a-Si) nano-antennas on a quartz substrate to support two partially overlapping quasi-BIC modes at red wavelengths and total suppression of higher-order modes at blue wavelengths. The a-Si nano-antennas are coated above with hydrogen silsesquioxane (HSQ) resist, whereas the quartz substrate is coated below with black dye. The HSQ resist suppresses the reflectance at blue wavelengths, whereas the black dye reduces background signals in the reflectance spectrum. Because of the sharp spectral transitions and narrow bandwidths enabled by quasi-BICs, the a-Si nano-antennas produce red pixels that, remarkably, show saturation in both simulation and experiment that exceeds the saturation of commercially available cadmium red pigment.

**Mie voids**

Interestingly, voids in HRI materials can also resonantly confine radiation. Practically speaking, spherical voids in a homogeneous medium are very hard to create; to realize such Mie void resonances experimentally, one has to bring the voids to the surface. Using focused ion beam (FIB) milling or mask-based dry etching, one can create cylindrical voids in a solid silicon wafer.

In recent work in our labs, voids of different depth and diameter, milled into a silicon wafer, enabled creation of distinct colors originating from single voids. Owing to the refractive index of silicon, Mie voids are about a factor of four larger than solid features such as those already discussed, and thus Mie voids show very large resonant cross-sections, resulting in intense scattering. This larger size as well as the reduced confinement at the inverted air–silicon interface mean that the Q factors of Mie void resonances are smaller, which results in colors close to natural colors based on light absorption.
In order to visualize the full tunability, one can vary the two defining parameters—void depth and diameter—in a controlled fashion over a large parameter space; the large parameter sweep enables a variety of different colors and even white and black hues. The large available color range enabled a microscopic replica of a portion of the painting Improvisation No. 9 by Wassily Kandinsky using Mie voids in silicon, with a period (that is, pixel size) of 900 nm. Depth and diameter are varied to reproduce the colors correctly.

Outlook

By manipulating the response to incident light fields, dielectric nanoresonators offer a full color palette for nanoscale pixel-based painting. This advance significantly enhances data storage capacity, aids in anti-counterfeiting measures and improves optical security. For instance, randomly positioned nanoresonators and voids can create unique physical unclonable functions for each product, acting as independent colorfu units without substantial correlated effects. Elliptical or rectangular resonators can encrypt polarization-dependent multiple information objects in a single device, while the strong wavelength-dependent
Innovations in structural colors from dielectric resonators will, we believe, impact various fields, thereby boosting the development of next-generation metadevices.

responses of nanoparticles can be utilized for multicolor selection, introducing new channels for white-light orbital angular momentum and holograms, thereby increasing communication capacity.

Achieving full color generation with simple geometrical parameter-tuning of nanopillars made of cost-effective LRI materials extends the material range beyond lossy metals and brittle HRI materials. 3D-printed nanostructures such as those described in this feature can be easily transferred to other polymer and glass materials with moulding methods, offering great potential for flexible optoelectronics. Biocompatible materials further expand applications to complex bio-inspired optical functions, medical and health monitoring and other uses. Additionally, TPL-based 3D-printing enables the fabrication of almost arbitrarily complex 3D photonic crystals with spectral selectivity in the visible band, finding applications in complex 3D integrated optical devices and topological photonics.

The strong light confinement in HRI Mie resonators, characterized by high-Q resonances with narrower spectra, facilitates the generation of very saturated structural colors, benefiting sensing applications. Mie voids in particular are well-suited for optical sensing and trapping experiments. They can also be integrated into hybrid systems with quantum emitters, up-conversion nanoparticles, fluorescent molecules, excitons in gallium arsenide or defect centers (color centers) in diamond or silicon carbide.

The concept of superscattering, emerging from the physics of BICs, allows the pushing of the scattering cross-section of a multipole resonance beyond the established limit, opening new possibilities for ultracompact sensing applications based on structural colors. Moreover, the fabrication of Mie resonators and Mie voids with HRI materials such as silicon and gallium nitride is compatible with industrial CMOS processes. This compatibility enables the development of multifunctional color filters, color routers and spectrometers with promising applications in on-chip optoelectronics, integrated photonic circuits, optical imaging and computing.

Envisaging future developments, the integration of design methods like inverse design and machine-learning-based optimization strategies could achieve a larger color gamut with novel nanoresonators and structure designs, stepping toward a universal nanophotonic color pixel. Such a pixel would consist of two Schrödinger subpixels that could be used to separately tune hue and luminosity for display applications. With the continuous evolution of Mie-ronics, materials and fabrication technologies, innovations in structural colors from dielectric resonators will, we believe, impact various fields—from colorful photovoltaics to nanolasers and detectors to augmented reality to Li-Fi and more—thereby boosting the development of next-generation metadevices.

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References and Resources