Hierarchical meta-assemblies for multiple length-scale synergetic structural coloration

Sirawit Tidma,[1] Metin Sitti,[2,*] and Mingchao Zhang[1,*]

[1]Department of Materials Science and Engineering, College of Design and Engineering, National University of Singapore, Singapore 117575, Singapore

[2]School of Medicine and College of Engineering, Koç University, Istanbul 34450, Turkey

*Correspondence: msitti@ku.edu.tr (M.S.); zhangmc@nus.edu.sg (M.Z.)

Received: August 17, 2026; Accepted: October 14, 2026; Published Online: October 15, 2026; https://doi.org/10.59717/j.xinn-mater.2026.100248

© 2026 The Author(s). This is an open access article under the CC BY license (https://creativecommons.org/licenses/by/4.0/).

Citation: Tidma S., Sitti M. and Zhang M. (2026). Hierarchical meta-assemblies for multiple length-scale synergetic structural coloration. *The Innovation Materials* **4**:100248.

Structural color emerges when light interacts with micro- and nanoscale structures arranged periodically across multiple length scales, such as nanoscale patterns nested inside microscale architectures. Traditional pigment-based coloration presents notable drawbacks, including potential toxicity and susceptibility to fading over time. By contrast, structural-color systems manipulate light through interference, diffraction, scattering, or guided propagation, potentially providing durable optical responses without relying on conventional molecular pigments. Biological blueprints from nature, such as peacock feathers and jewel beetle elytra, exhibit clear biological precedents of this structural approach. However, the fabrication and replication of such hierarchical structures have been technical challenges. Most existing artificial structural-color platforms are generally fabricated for a single structural scale, which inherently constrains their optical complexity, design versatility, and manufacturing scalability. A strategy that integrates optical architectures from the nanoscale to the macroscale with a continuously scalable fabrication process could therefore represent an important advance in structural photonics.

Song and co-authors have now addressed these limitations by developing a printable hierarchical meta-assembly using continuous roll-to-roll additive nanoprinting (R2R-ANP). Whereas conventional photonic crystals primarily rely on photonic bandgaps (PBGs) arising from Bragg scattering, the authors incorporated periodically assembled polystyrene nanoparticles (PS NPs) into curved micro-concave interfaces embedded within a PDMS matrix, enabling optical coupling between PBG reflection and total-internal-reflection (TIR)-mediated optical propagation. Each microscopic structure, or pixel, contributes to synergetic coloration, with the macroscopically perceived color arising from the combined optical responses of the center and rim of the meta-assemblies. Notably, this approach preserves exceptional scalability from nano-building blocks to meter-scale printable films. This work demonstrates how color and other optical responses can be programmed through geometric and structural design across multiple interacting scales.

The core innovation of this work lies in the deliberate combining of distinct optical phenomena that operate at different length scales. At the nanoscale, hexagonally close-packed PS NPs self-assemble into domed colloidal photonic crystals (DCPCs), giving rise to a base structural color governed by the PBG. Liquid PDMS is then infiltrated into the interstitial voids between PS NPs, and subsequent thermal curing crosslinks the PDMS to permanently lock PS NPs within the polymer matrix (Figure 1A).[1] At the microscale, peeling the cured PDMS from the substrate and flipping the embedded structures transform the initially domed DCPC interfaces into concave meta-assemblies. At the macroscale, these units are organized into patterned arrays and meter-scale films. Numerical simulations indicate that, for wavelengths within the PBG, the center-localized field and the interface-guided field develop competing spatial distributions, producing interference cancellation in their overlap region and suppressing rim propagation. Outside the PBG, TIR confinement and multibounce interference enhance wavelength-dependent fields near the concave interface. As a result, the meta-assembly microstructure exhibits two simultaneous colorations, which include a comparatively angle-insensitive PBG-dominated color at the center alongside a dynamically changing color around its rim. Such dual optical response cannot be replicated using conventional single-scale structures or uniform photonic crystals.

This work establishes a library of synergetic coloration across the visible spectrum by exploiting two largely, although not completely, decoupled structural parameters. The total perceived color results from the combination of the PBG-dominated center and the TIR-dominated rim, and each can be modulated independently. Increasing the nanoparticle diameter produces a bathochromic (red) shift in the PBG reflection, substantially changing the center color while causing only slight modulation of the rim response. By contrast, reducing the meta-assembly diameter shortens the effective TIR-mediated optical path and shifts the rim color towards shorter wavelengths, while the center color remains comparatively stable. The nanoparticle diameter and meta-assembly diameter thus provide two quasi-independent design variables for programming the composite optical response. The observed coloration also depends on illumination geometry. Under single-beam oblique illumination, the PBG-dominated reflection and TIR-dominated retroreflection are spatially separated (Figure 1B). Light entering a meta-assembly can undergo repeated TIR and multibounce interference along its curved interface, generating a wavelength-dependent retroreflective response. Under dual-beam illumination, the reflection and retroreflection contributions can be recombined, producing a mixed macroscopic color. This illumination-dependent mixing provides an additional level of optical programmability beyond that supplied by the two structural parameters.

The most significant contribution is the manufacturing strategy that enables this sophisticated optical architecture from nanoscale (10^{-9}) to macroscale (10^{0}) for industrial scaled-up production (Figure 1C). Unlike nanoscale 3D printing, which provides high spatial resolution but is limited in throughput by voxel-by-voxel fabrication, conventional colloidal self-assembly enables large-area nanoscale structures but offers limited control over their geometry and spatial arrangement. R2R-ANP bridges these regimes by combining spontaneous nanoparticle self-assembly with digitally defined printing and continuous roll-to-roll processing. The platform integrates inkjet deposition, nanoparticle self-assembly, PDMS infiltration, thermal curing, and embedded transfer into one manufacturing workflow, achieving nearly 100% fabrication yield while preserving nanoscale ordering. The platform achieves pixel customization and meter-scale film production containing more than 40 million meta-assemblies assembled from over 7×10^{13} nanoparticles. This integration provides an unusual balance between fabrication simplicity, manufacturing scalability, and multiscale structural programmability, overcoming a trade-off that remains difficult to address using either high-resolution three-dimensional printing or self-assembly alone.

A related study from the same group used polymer microdot templates containing Rhodamine B to direct the self-assembly of nanoparticles into programmable 1D optical heterostructures.[2] That method serves a different optical purpose and incorporates a molecular chromophore. Its long-term photostability would therefore need to be evaluated independently before a direct durability comparison with pigment-free structural-color systems could be made. A practical advantage of the R2R-ANP meta-assembly films is that they retain their optical responses under the accelerated laboratory stress tests reported in the study. The films maintained relatively consistent optical intensity over a broad temperature range and after exposure to water, acidic and alkaline solutions, and selected organic liquids, demonstrating resistance to several thermal and chemical conditions. Repeated washing has little effect on color loss due to strong interfacial bonding between PS meta-assemblies and the PDMS matrix, while typical color films suffer from color fading from repeated washing cycles. The film also exhibits excellent mechanical robustness, as it can withstand up to 200% tensile strain and thousands of stretching cycles without a loss in optical performance. These results establish encouraging mechanical resilience and laboratory-scale environmental stability, although they do not yet demonstrate multiyear durability under realistic outdoor conditions.

Despite this state-of-the-art strategy, several operational, physical, and material limitations remain. From a theoretical standpoint, the proposed optical mechanism is supported primarily by numerical electromagnetic simulations and far-field optical measurements. Experimental mapping of the angle-, polarization-, and position-resolved fields—for example, through Fourier-plane spectroscopy, back-focal-plane imaging, phase-resolved interferometric microscopy, or near-field optical imaging—could provide a more direct test of the simulated coupling between PBG reflection and TIR-confined propagation. The system also remains a passive optical platform whose most

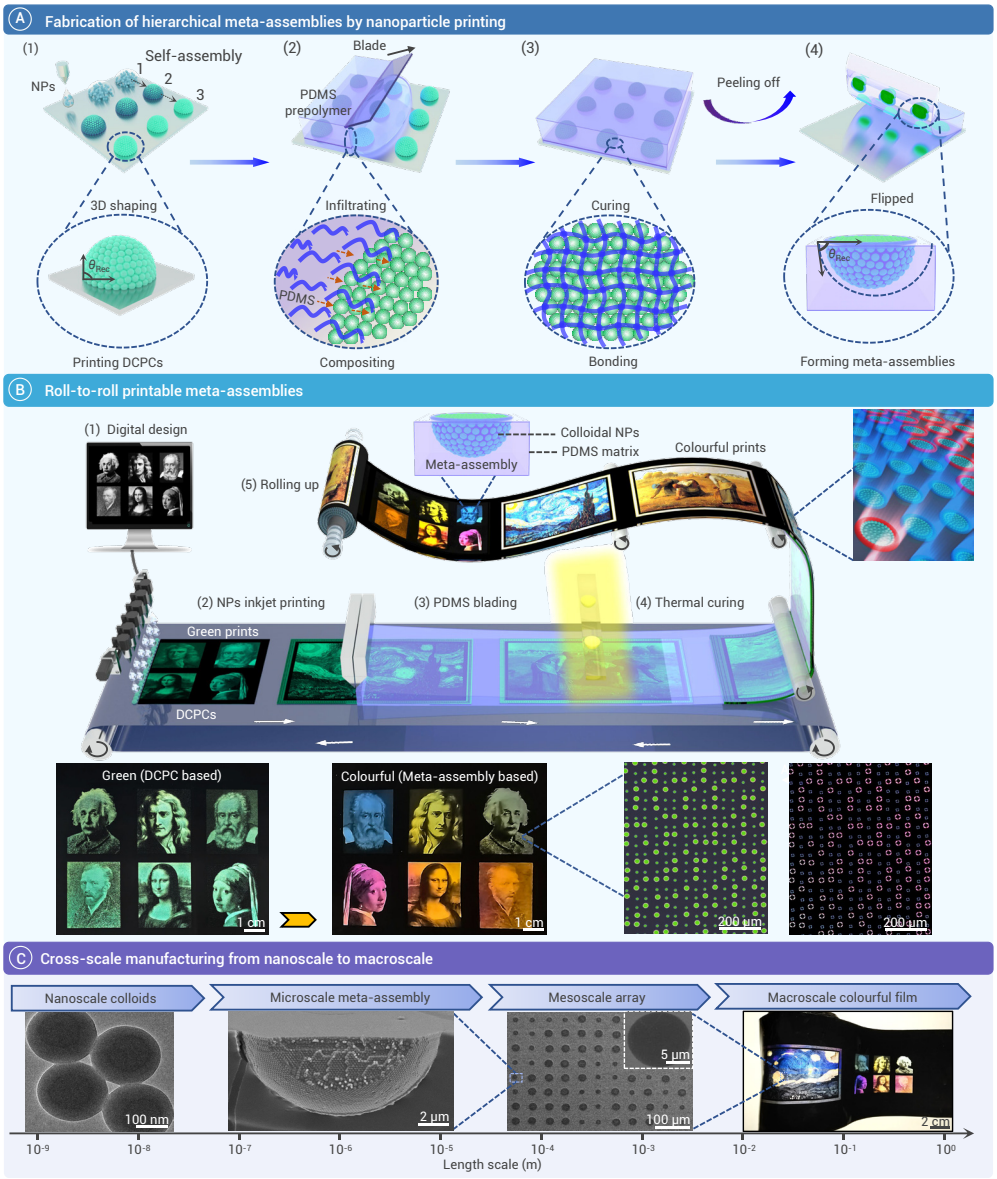


Figure 1. Multiscale fabrication of hierarchical colloidal photonic meta-assemblies (A) Schematic illustration of how domed colloidal photonic crystals are converted into hierarchical meta-assemblies embedded within PDMS. (B) Fabrication workflow of R2R-ANP. (C) Multi-scale manufacturing from nano- to macroscale.

distinctive macroscopic color mixing relies on controlled illumination geometry. Its performance under diffuse daylight, broadband indoor lighting, variable viewing distances, and uncontrolled observation angles should therefore be quantified before display or coating applications are considered.

Extending this work towards a dynamic reconfiguration system such as an adaptive smart coating or integrating with polymeric smart materials using thermal-responsive polymers, liquid crystals, or hydrogels could broaden functionality and color display.[3,4] Substituting nanoparticle types such as quantum dots, perovskites, or nanofibers could offer alternative routes to expanding the library.[5] Alternative dielectric colloids with higher refractive indices could broaden the accessible PBG range and enhance optical contrast, provided that particle monodispersity, printability, and self-assembly quality can be maintained. Incorporating emissive components such as quantum dots or perovskites might introduce additional luminescent functions, but these materials are not straightforward substitutes for PS NPs: they would create new challenges involving absorption, toxicity, photochemical stability, moisture sensitivity, and interfacial compatibility. Similarly, replacing spherical particles with nanofibers would alter the assembly topology and optical mechanism rather than simply expand the existing color library.

In terms of industrial viability, practical deployment will require qualitative evaluation of long-term outdoor durability, color stability under solar irradiation, humidity-temperature cycling, abrasion, contamination, color-coordinate stability, or techno-economic viability. If this approach is compatible with existing high-speed roll-to-roll lines while retaining structural precision and color uniformity, it could make the commercialization of photonic materials more practical. This meta-assembly concept nevertheless opens several exciting research directions. Combining this manufacturing strategy with AI-assisted inverse design may facilitate the rapid exploration of hierarchical architectures with tailored optical responses. More broadly, linking nanoscale self-assembly, microscale optical engineering, and continuous roll-to-roll manufacturing provides a foundation for constructing bio-inspired metamaterials that are simultaneously high-performing, customizable, and scalable at once.

In summary, Song and co-authors demonstrated more than just a new structural-color platform. Their printable meta-assembly strategy introduces a unified multiscale photonic architecture that reconciles optical sophistication with scalable manufacturing. The integration of PBG reflection and TIR within a single hierarchical building block creates optical functions that cannot be achieved by either mechanism alone. Although further evidence is needed regarding illumination-independent performance, long-term durability, process economics, and environmental impact, compatibility with continuous roll-to-roll processing brings hierarchical meta-assemblies an important step closer to practical photonic devices and coatings. These advances highlight how rational hierarchical design can transform optically simple, low-index polymers into sophisticated structural colors. This finding challenges the assumption in the optical materials field that high-refractive-index and costly constituents are necessary for high performance, opening opportunities for novel optical materials—demonstrating that even optically ordinary materials can produce exceptional optical functionalities.

REFERENCES

1. Li K., Chen J., Li H., et al. (2026). Printable meta-assemblies enable synergetic colouration. *Nature* **652**:1195–1203. DOI:10.1038/s41586-026-10408-8
2. Li K., Yu X., Yang D., et al. (2026). Printing-defined polymer microdot templates for deterministic 1D optical heterostructure patterning. *Polymer* **363**:130566. DOI:10.1016/j.polymer.2026.130566
3. Zhang M., Pal A., Zheng Z., et al. (2023). Hydrogel muscles powering reconfigurable micro-metastructures with wide-spectrum programmability. *Nat. Mater.* **22**:1243–1252. DOI:10.1038/s41563-023-01649-3
4. Zhang M., Pal A., Lyu X., et al. (2024). Artificial-goosebump-driven microactuation. *Nat. Mater.* **23**:560–569. DOI:10.1038/s41563-024-01810-6
5. Lyu X., Lei W., Gardi G., et al. (2026). Optofluidic three-dimensional microfabrication and nanofabrication. *Nature* **650**:613–620. DOI:10.1038/s41586-025-10033-x

FUNDING AND ACKNOWLEDGMENTS

FUNDING AND ACKNOWLEDGMENTS M.Z. acknowledges support and funding from the National University of Singapore Presidential Young Professorship Start-Up Grant (No. A-0010080-00-00). The funders had no role in study design, data collection and analysis, decision to publish, or preparation of the manuscript.

AUTHOR CONTRIBUTIONS

S. Tidma wrote the draft and arranged the figure. M. Sitti and M. Zhang supervised and revised the manuscript. All authors contributed to and approved the manuscript.

DECLARATION OF INTERESTS

The authors declare no competing interests.