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Grayscale Photomasks Enable Noncontact Optical Patterning of Hydroxypropyl Cellulose Gels for Dynamic Art

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ABSTRACT

Hydroxypropyl cellulose (HPC) forms cholesteric photonic gels with dynamic structural color, but most HPC systems are tuned globally or patterned through serial fabrication. Here we report a mask-encoded parallel photonic manufacturing strategy that locally regulates the reflected wavelength of acrylamide-containing HPC (AM-HPC) photonic gels. UV-induced photopolymerization produces a dose-dependent blueshift consistent with cholesteric-pitch compression, yielding a 204 nm (662–458 nm) tuning window in 68 wt.% AM-HPC under time-controlled exposure and spatial features down to 50 μm . To translate this response into programmable manufacturing, we combine Gaussian-process active-learning calibration with an experimentally measured mask-opacity-to-UV transfer function. This workflow converts two-dimensional target wavelength fields into grayscale optical masks, enabling spatially varying UV doses to be delivered simultaneously across a continuous, unsegmented gel. A single 10 s exposure produced multilevel structural-color images with an 85 μm mask-pixel pitch (13 840 pixels cm^{-2}) in a continuous gel. The patterned gels retain thermally responsive structural color, allowing dynamic modulation of encoded images. These results establish a proof-of-concept digital-to-material workflow for parallel, composition-specific programming of apparent structural color in continuous HPC photonic gels.

1 | Introduction

Structural color encodes optical response in physical architecture rather than chemical pigments, enabling vivid and potentially durable colors from periodic nanoscale organization. Research is now advancing beyond static coloration toward dynamic, spatially programmable patterns [1–5]. This transition remains difficult in sustainable soft materials, where the molecular mobility that enables dynamic responsiveness often produces uniform, sample-level optical changes rather than independently programmable local colors. Hydroxypropyl cellulose (HPC), a water-

soluble cellulose derivative, is an attractive platform because concentrated HPC self-assembles into cholesteric photonic phases with visible structural color, while retaining renewability, processability, and responsiveness to concentration, temperature, hydration, and mechanical deformation [6–9]. However, these stimuli typically tune the entire material or broad regions of it. As a result, HPC gels have been widely explored as responsive coatings, sensors, and photonic skins, but have not yet been readily transformed into high-resolution, image-like photonic media in which each location carries a prescribed structural color [10].

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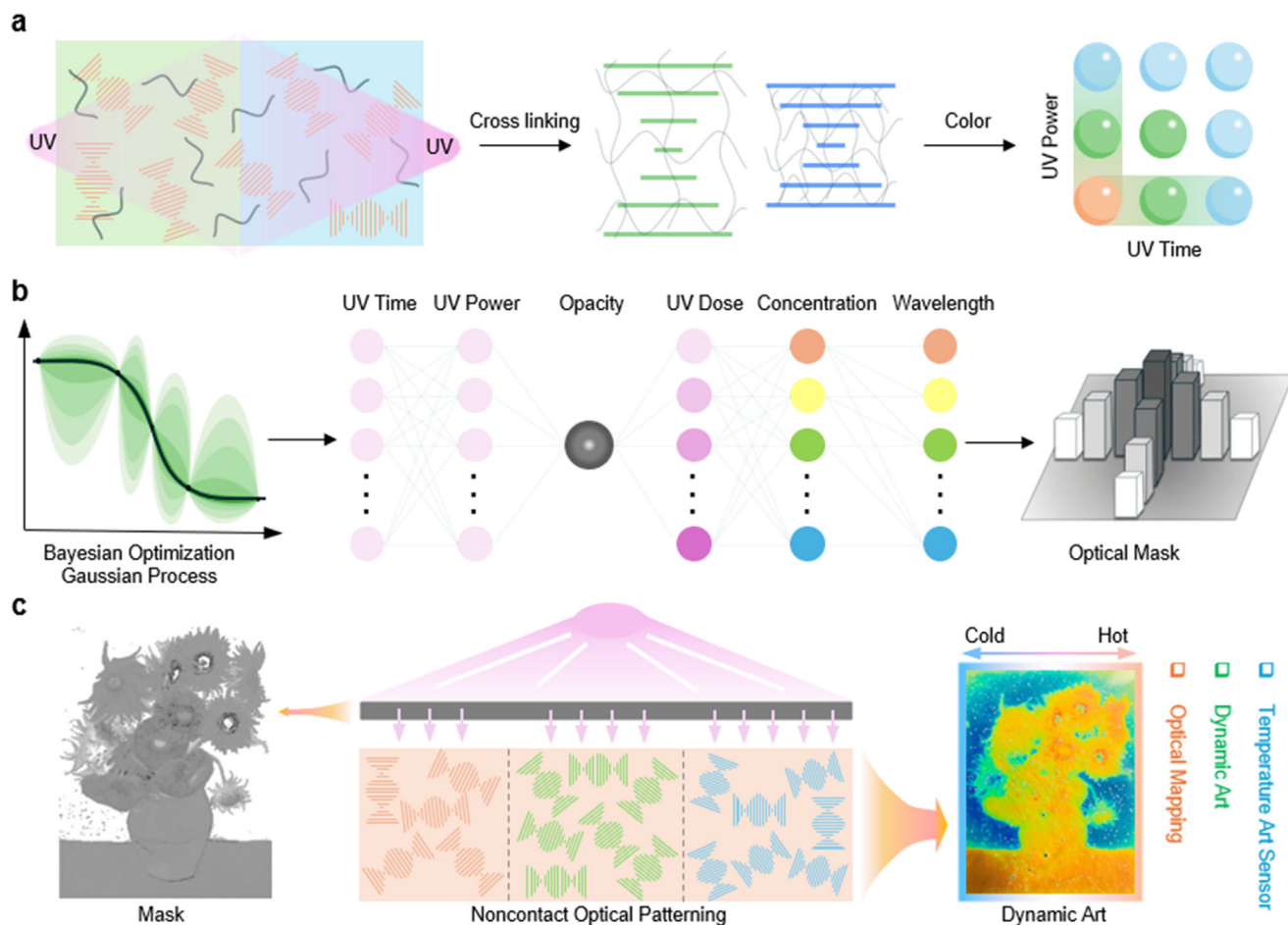


FIGURE 1 | Contactless, light-guided, and programmable spatial control strategies for photonic gels. (a) UV tuning of structural color. (b) Bayesian optimization (BO) driven digital mask encoding. (c) Optical manufacturing for pixel-level structural color programming.

Recent advances in chemical modification, photopolymerization and three-dimensional printing have expanded the design space of HPC-based photonic materials [11, 12]. Crosslinking strategies have been employed to preserve the cholesteric helical structure and regulate the color, scattering characteristics, angular dependence, and polarization response of solid HPC films, thereby further expanding the range of material formats available for HPC-based photonic systems [13–15]. Mechanically responsive HPC materials have also progressed from roll-to-roll manufactured photonic laminates and self-supporting hydrogels that change color under deformation to arrays driven by fluid pressure [9, 16], in which discrete regions of a continuous HPC film function as independently controlled display pixels. In parallel, direct ink writing, three-dimensional printing, extrusion, and fiber forming methods have enabled structurally colored filaments, objects, and continuous fibers with increasingly complex macroscopic geometries [17–19]. Photochemical strategies can stabilize or modulate cholesteric organization [14, 20, 21], while additive manufacturing enables geometric patterning of structural color inks [22, 23]. Yet these approaches do not fully solve the central digital-to-material translation problem: how to convert a desired color map directly into a local cholesteric-pitch map inside one continuous HPC gel. Most existing methods either tune the pitch globally, rely on serial deposition or printing, or pattern external geometry rather than quantitatively writing the internal helical pitch at arbitrary positions. Achieving programmable

structural color therefore requires a manufacturing rule that links a controllable external input to a predictable wavelength output at every pixel. Light is an especially powerful input for this purpose because its dose, intensity, and spatial distribution can be patterned rapidly, remotely, and without contact.

Here we report a mask-encoded optical manufacturing strategy that uses ultraviolet light to write structural color directly into photopolymerizable acrylamide-modified HPC (AM-HPC) photonic gels. UV-induced reorganization of the cholesteric mesophase produces coupled changes in the helical pitch, average refractive index, and orientation of the cholesteric domains relative to the incident light, thereby converting local UV dose into a predictable blueshift of the reflected wavelength. In 68 wt.% AM-HPC, this process provides a 204 nm wavelength tuning window and spatial features down to 50 μm . To make this response programmable, we combine Gaussian-process active-learning calibration with a grayscale-mask transfer function that links target wavelength, UV dose, and local mask opacity. A digital image can then be converted into a single photomask and written in one 10 s exposure, producing multicolor structural images at 13 840 pixels/cm² with 85 μm pixels while retaining the intrinsic thermal responsiveness of HPC. Together, this approach transforms HPC from a globally responsive cholesteric gel into a digitally addressable photonic material by bridging digital color information and continuous cholesteric pitch patterning through

an integrated workflow of image decoding, dose calibration, and mask-mediated photopolymerization.

2 | Results and Discussion

2.1 | Design Framework for Mask-Guided Optical Manufacturing

A data-driven optical manufacturing framework was established to achieve spatially programmable structural color in acrylamide-modified HPC (AM-HPC) photonic gels, as illustrated in Figure 1. The core design principle relies on light-guided regulation of the cholesteric mesophase within a continuous soft photonic matrix. In AM-HPC gels, localized UV exposure induces photopolymerization-driven network formation and subsequent reorganization of the cholesteric mesophase. This reorganization is associated with coupled changes in the effective helical pitch, average refractive index, and orientation of the cholesteric domains relative to the incident light, collectively shifting the reflected wavelength toward shorter wavelengths. The dose-dependent optical response therefore enables predictable modulation of the reflected wavelength and corresponding structural color. As illustrated in Figure 1a, spatially patterned UV exposure can therefore write different structural color states into the same gel. In this strategy, light functions not only as an external stimulus, but also as a contactless optical manufacturing tool for writing internal photonic structures with spatial precision.

To convert this physical response into a programmable fabrication route, a quantitative digital-to-material translation framework was introduced. The optical response of the AM-HPC system was parameterized by correlating UV irradiation time, UV power density, mask opacity, HPC concentration, and the resulting structural color wavelength. Bayesian optimization (BO) was employed to efficiently construct this nonlinear mapping, providing the predictive basis for translating desired optical outputs into fabrication parameters (Figure 1b).

Based on this quantitative relationship, digital image information can be converted into a manufacturable optical mask. Specifically, the target image is decomposed into pixel-level optical information, which is subsequently transformed into the corresponding wavelength distribution and further encoded into opacity values through the calibrated UV response model. During optical manufacturing, the mask opacity spatially modulates the incident UV field, enabling continuous and pixel-level programming of structural color across a two-dimensional plane in a single exposure step (Figure 1c). This design framework provides the conceptual and quantitative basis for the following sections, where UV-responsive optical behavior, predictive modeling, mask design, and high-resolution structural color image reconstruction are discussed in detail.

2.2 | UV-Tunable Structural Color in AM-HPC Photonic Gels

AM-HPC gels with concentrations ranging from 68 to 78 wt.% were prepared to examine their UV-programmable optical response (Figure 2a). The combined influence of concentration

and UV irradiation time on structural color is summarized in Figure 2b, with the corresponding original color images provided in Figure S1. Before UV irradiation, increasing AM-HPC concentration shifts the initial color from red to green, cyan, and blue, indicating a concentration-dependent reduction of the cholesteric pitch and a corresponding shift toward shorter reflected wavelengths. Upon UV exposure, all formulations exhibit a time-dependent blueshift, confirming that UV irradiation provides an effective handle for regulating the cholesteric organization of the AM-HPC matrix. The magnitude of this UV-induced color modulation is strongly concentration-dependent. Lower concentration gels, particularly 68 wt.% AM-HPC, display a broad red-to-blue transition, whereas higher-concentration gels show a narrower tuning range because their initial cholesteric pitch is intrinsically shorter. These results establish the concentration- and irradiation time-dependent optical response of AM-HPC gels and provide the basis for subsequent quantitative wavelength analysis and programmable optical manufacturing. Pixel-resolved image processing was further used to convert the visual color evolution into quantitative wavelength information (Figure 2c; Figure S2 and Tables S1–S6). A region of 1.6 cm × 1.6 cm was selected from each AM-HPC image, and the average RGB values were extracted at a sampling density of 3600 pixels/cm². The extracted RGB values were converted into reflected wavelengths, providing a quantitative description of the structural color response under different concentrations and UV irradiation times. As shown in Figure S2, the processed color blocks preserve the concentration- and time-dependent color gradients observed in the original images, while Tables S1–S6 list the corresponding RGB values and wavelength assignments. This image-based quantification converts spatial color information into comparable wavelength datasets, enabling reliable analysis of UV-induced pitch modulation.

The wavelength evolution extracted from the pixel-resolved analysis is shown in Figure 2d. All AM-HPC samples exhibit a progressive decrease in the camera-derived apparent wavelength with increasing UV irradiation time, confirming UV-induced blue shifting of the cholesteric photonic response. The observed wavelength window becomes narrower with increasing AM-HPC concentration. Because the tunable window is derived from camera-based wavelength extraction, the reported range represents the visible wavelength range that can be resolved within the limitations of camera acquisition, RGB readout, and subsequent wavelength conversion. The 68 wt.% AM-HPC gel shows the broadest modulation, with the reflected wavelength decreasing from 662 to 458 nm, corresponding to a 204 nm tuning window. The tuning range then decreases to 166 nm for 70 wt.% AM-HPC, 122 nm for 72 wt.% AM-HPC, 84 nm for 74 wt.% AM-HPC, 36 nm for 76 wt.% AM-HPC, and only 12 nm for 78 wt.% AM-HPC. This trend suggests that lower concentration gels allow more pronounced UV-induced pitch modulation, whereas higher concentration gels start from intrinsically shorter cholesteric pitches and thus provide a narrower window for further blue shifting. Accordingly, the 68 wt.% AM-HPC composition was selected for subsequent dose-wavelength calibration and pixel-level structural color programming because it provided the broadest accessible red-to-blue tuning range among the investigated formulations.

Beyond spectral tunability, the AM-HPC system enables localized structural color programming within a continuous gel plane. As

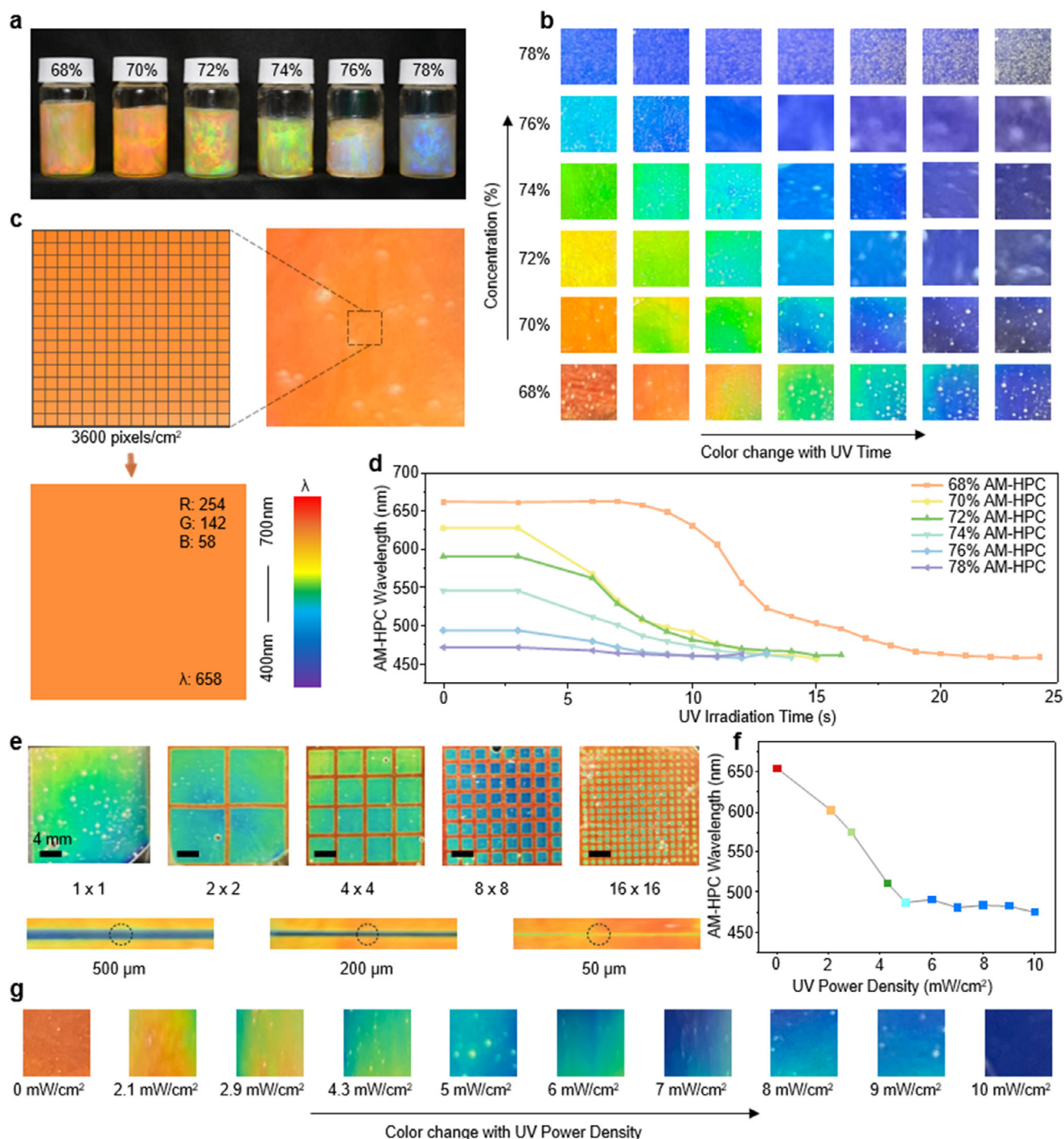


FIGURE 2 | UV-tunable structural color of AM-HPC photonic gels. (a) Photographs of AM-HPC gels ranging from 68 to 78 wt.%. (b) Structural color evolution of AM-HPC as a function of concentration and UV irradiation time (10 mW/cm²). (c) Quantitative extraction of wavelength. (d) Wavelength of photonic gels from 68 to 78 wt.% under UV irradiation. (e) Spatially resolved optical manufacturing. (f) Wavelength evolution of 68 wt.% AM-HPC under different UV power densities at a fixed exposure time of 10 s. (g) Structural color evolution of 68 wt.% AM-HPC under different UV power densities.

shown in Figure 2e, predefined regions of a single AM-HPC gel were selectively exposed to UV light, generating spatially confined color changes while the surrounding unexposed areas retained their original optical state. This result confirms that UV-induced reorganization of the cholesteric mesophase can be restricted to targeted locations, allowing different regions of the same gel to carry distinct reflected wavelengths. Such localized control demonstrates that structural color can be written into

the internal cholesteric architecture without physical separation, serial ink deposition, or multilayer assembly. By further reducing the irradiation area, spatial features down to 50 μm were achieved, highlighting the high-resolution capability of this optical manufacturing strategy. In addition to exposure time, UV power density provides another effective handle for structural color programming. For 68 wt.% AM-HPC, increasing UV power density at a fixed exposure time produces an overall blueshift of

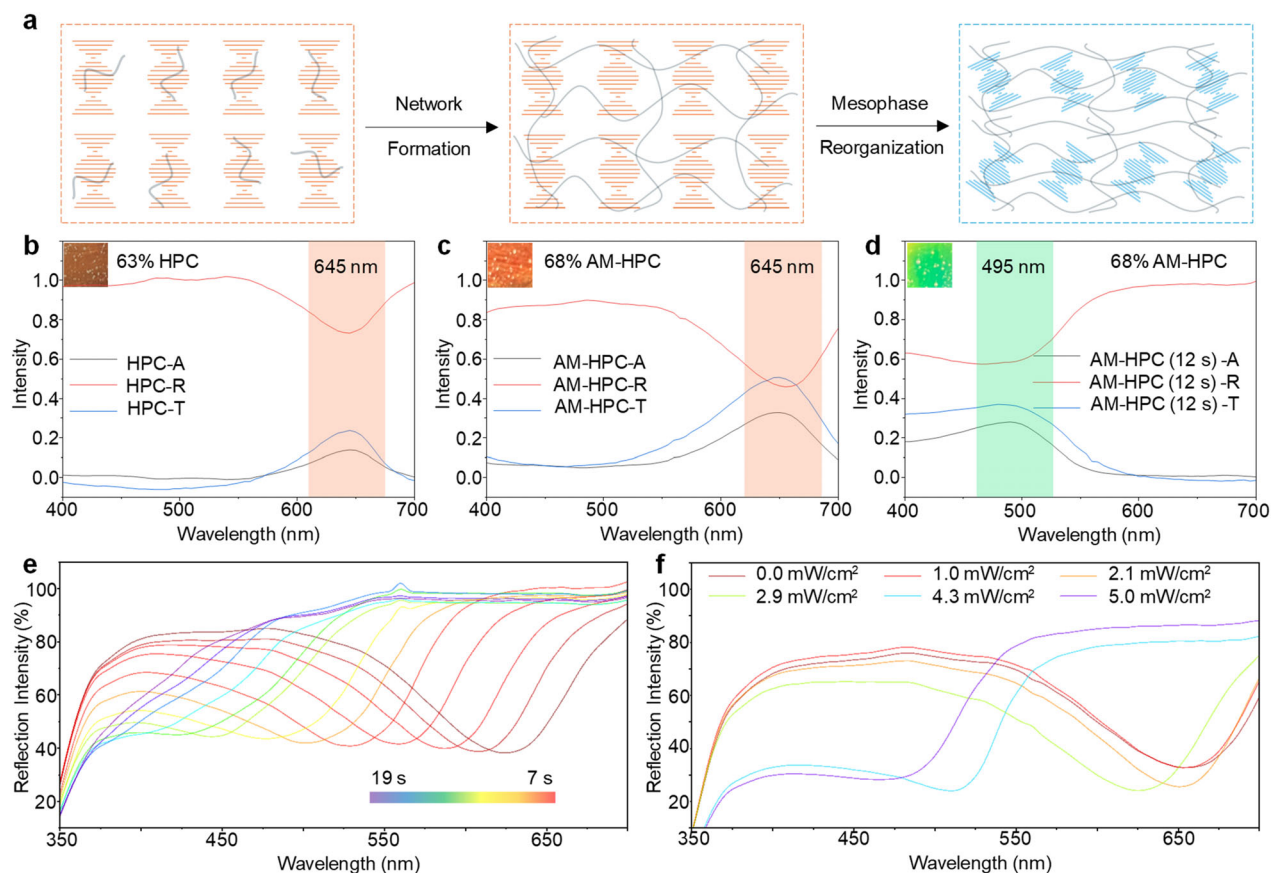


FIGURE 3 | Optical evidence for UV-induced pitch modulation. (a) Schematic illustration of the UV-induced crosslinking process. (b) Transmission (T), reflection (R), and absorption (A) spectra of 63 wt.% HPC. (c) 68 wt.% AM-HPC before UV irradiation, and (d) 68 wt.% AM-HPC after UV irradiation (10 mW/cm²). (e) Evolution of reflection spectra of 68 wt.% AM-HPC under varying UV irradiation times (from 7 to 19 s in 1 s increments) at a power density of 10 mW/cm², and (f) 68 wt.% AM-HPC at the UV power densities indicated in the panel, with a fixed irradiation time of 10 s.

the reflected wavelength from 655 nm at 0 mW/cm² to 474 nm at 10 mW/cm² (Figure 2f; Figure S3 and Table S7). At a UV power density of 5 mW/cm², the AM-HPC gel already achieves a wavelength shift spanning from red to blue. The corresponding mean color images show a clear transition from red to green, cyan, and blue (Figure 2g; Figure S3), consistent with the RGB-wavelength values listed in Table S7. Under the standardized imaging conditions, the camera-derived apparent wavelength ranged from a maximum of 662 nm in the red region to a minimum of 458 nm in the blue region. Together, these results establish a quantitative and spatially controllable platform for UV-directed structural color programming in AM-HPC photonic networks. The ability to correlate concentration, UV irradiation time, UV power density, and wavelength provides the essential basis for subsequent digital encoding and image reconstruction.

2.3 | Optical Spectral Evolution During UV-Guided Structural Modulation

To elucidate the mechanism underlying UV-regulated structural color evolution, the optical responses of HPC and AM-HPC systems were systematically investigated. As illustrated in Figure 3a, UV irradiation induces a two-stage structural evolution in AM-HPC, involving initial polymer-network formation followed by reorganization of the cholesteric mesophase. This structural

evolution may lead to changes in the cholesteric pitch, average refractive index, and orientation of the cholesteric domains relative to the incident light. These coupled changes produce the pronounced blueshift. To ensure comparison at a comparable optical state, 63 wt.% HPC and 68 wt.% AM-HPC were selected because they exhibit similar reflection wavelengths prior to UV irradiation (645 nm). The baseline optical properties of the system were first examined using 63 wt.% HPC. As shown in Figure 3b, the transmission, reflection, and absorption spectra of 63 wt.% HPC exhibit typical cholesteric photonic characteristics, where a distinct reflection band corresponds to selective Bragg reflection, accompanied by complementary features in transmission and absorption. These results confirm that the observed coloration originates from the periodic helicoidal structure rather than molecular absorption. Upon introducing the photo crosslinking agent, the optical response of the system is modified. The 68 wt.% AM-HPC sample before UV irradiation (Figure 3c) retains spectral features broadly comparable to those of 63 wt.% HPC, but with subtle variations in transmission and absorption, indicating additional light-matter interactions introduced by the photo crosslinking agent.

After UV irradiation, the optical spectra of AM-HPC evolve significantly (Figure 3d). The reflection band shifts toward shorter wavelengths, accompanied by corresponding changes in the reflection spectrum, indicating that UV-triggered photopoly-

merization alters the cholesteric organization of the AM-HPC matrix. For cholesteric liquid crystals, the reflected wavelength is described by $\lambda \approx \bar{n}P \cos \theta$, where P is the cholesteric pitch, $\bar{n}$ is the average refractive index, and θ is the angle between the helical axis and the incident light. Accordingly, the observed blueshift is primarily attributed to photopolymerization-induced reorganization of the cholesteric phase, which leads to changes in the cholesteric pitch, average refractive index, and orientation of the cholesteric domains relative to the incident light. The coupled effects of these structural changes collectively shift the reflection band toward shorter wavelengths [20]. At the same time, the absorption increases, reflecting the optical response of the photo crosslinking components during the photopolymerization process. The dynamic optical response during photopolymerization was further analyzed by monitoring the reflection spectra under controlled UV conditions. At a fixed UV intensity of 10 mW/cm², increasing irradiation time leads to a continuous and systematic shift in the reflection minima (Figure 3e), reflecting the gradual evolution of the photonic structure. Notably, a small spectral peak emerges during this process, indicating additional absorption contributions beyond the intrinsic cholesteric response. Independent characterization of the photo-crosslinking agent system (Figure S4) shows distinct absorption bands overlapping with the AM-HPC spectral region, confirming that this peak originates from the photo crosslinking agent rather than the cholesteric structure itself. Importantly, this characteristic absorption feature is significantly diminished after UV-induced crosslinking. The disappearance of this peak indicates the consumption or transformation of the photo-crosslinking agent during the photopolymerization process. To decouple the effects of UV power density and time, the reflection spectra were also recorded under varying UV power densities at a fixed irradiation time of 10 s (Figure 3f). Similar spectral trends are observed, confirming that the optical response is governed by the cumulative UV dose rather than a single parameter. Higher UV intensities accelerate the crosslinking process, leading to more pronounced spectral shifts within the same irradiation duration. As shown in Figure 3e,f, continuous UV irradiation produces a larger wavelength shift within a shorter effective time than intermittent exposure. This behavior indicates that uninterrupted irradiation sustains a more efficient photochemical process, whereas dark intervals during intermittent exposure partially interrupt the evolution of the cholesteric pitch [24]. The corresponding reflection-spectrum-derived wavelengths of the 68 wt.% AM-HPC gel as a function of UV irradiation time are provided in Figure S5 for independent spectroscopic comparison. During the initial stage (0–7 s), both the camera-derived apparent wavelength and the spectroscopically extracted UV–vis wavelength remain relatively stable. The camera-derived apparent wavelength is approximately 662 nm, whereas the UV–vis spectra-extracted wavelength is approximately 625 nm. During the subsequent blue-shifting stage (7–19 s), the camera-derived wavelength exhibits a pronounced nonlinear evolution, while the wavelength extracted directly from the UV–vis spectra shows a more nearly linear decrease. This difference is mainly influenced by the built-in characteristics and image processing pipeline of the camera, as the analysis involves image to RGB and RGB to wavelength conversion. Therefore, the camera-derived wavelength should be regarded as an apparent colorimetric wavelength rather than a direct spectroscopic measurement.

The reflected wavelength of a cholesteric photonic gel is governed by the Bragg relationship $\lambda \approx \bar{n} P \cos \theta$ (Figure S6), where $\bar{n}$ is the average refractive index, and P is the cholesteric pitch [25]. In AM-HPC, UV exposure initiates acrylamide crosslinking within the HPC-rich matrix. After the initial network formation, continued photopolymerization further induces reorganization of the cholesteric phase, leads to changes in the cholesteric pitch, average refractive index, and orientation of the cholesteric domains relative to the incident light, and thereby shifts the structural color toward blue. The corresponding shift of the reflection and transmission bands, together with the attenuation of the photoinitiator-related absorption feature after UV exposure, supports a photopolymerization-driven structural evolution rather than a purely optical absorption effect. Because similar spectral shifts are obtained by increasing either irradiation time or UV power density, the color evolution is best understood as a UV dose-regulated reorganization of the cholesteric phase. As evident from the results, the camera-derived apparent wavelengths were generally higher than those determined by UV–vis spectroscopy. This discrepancy is mainly attributed to the limited color discrimination of the smartphone imaging system. Accordingly, the camera-derived wavelengths were used for image-based color encoding and mask calibration, whereas UV–vis spectroscopy was used to characterize the actual spectral evolution.

2.4 | Bayesian Optimization Guided UV-Wavelength Relationships

A data-driven modeling framework based on Bayesian optimization (BO) was employed to establish a quantitative relationship between UV irradiation parameters and structural color evolution. This approach enables efficient exploration of the correlation between UV exposure conditions and the camera-derived apparent wavelength. To further verify the reliability of the BO-derived relationships, the same datasets were independently fitted using a conventional four-parameter logistic sigmoidal model, and the resulting trends and prediction errors were compared with those obtained from the BO model. The evolution of the UV time-wavelength relationship is shown in Figure 4a–c, where Gaussian process regression is iteratively applied to refine the predictive model. Starting from a minimal dataset of three sampling points, the model captures only the overall trend with relatively large uncertainty. Among these three points, two are boundary points that define the curve interval, while the third point helps estimate the confidence interval. As additional data points are incorporated (five and eight points), the prediction accuracy improves significantly, accompanied by a substantial reduction in the uncertainty range. This result indicates that the BO framework effectively guides data acquisition toward regions with higher information gain. As quantified in Figure 4d–f, the mean absolute error (MAE) and root mean square error (RMSE) decrease markedly from 21.41 and 27.11 to 1.09 and 2.18 nm, respectively, with increasing training data.

A similar strategy was applied to model the relationship between UV power density and reflection wavelength. As shown in Figure 4g–i, increasing the number of sampled data points from three to eight leads to a significant improvement in prediction accuracy. Correspondingly, the MAE and RMSE are reduced to

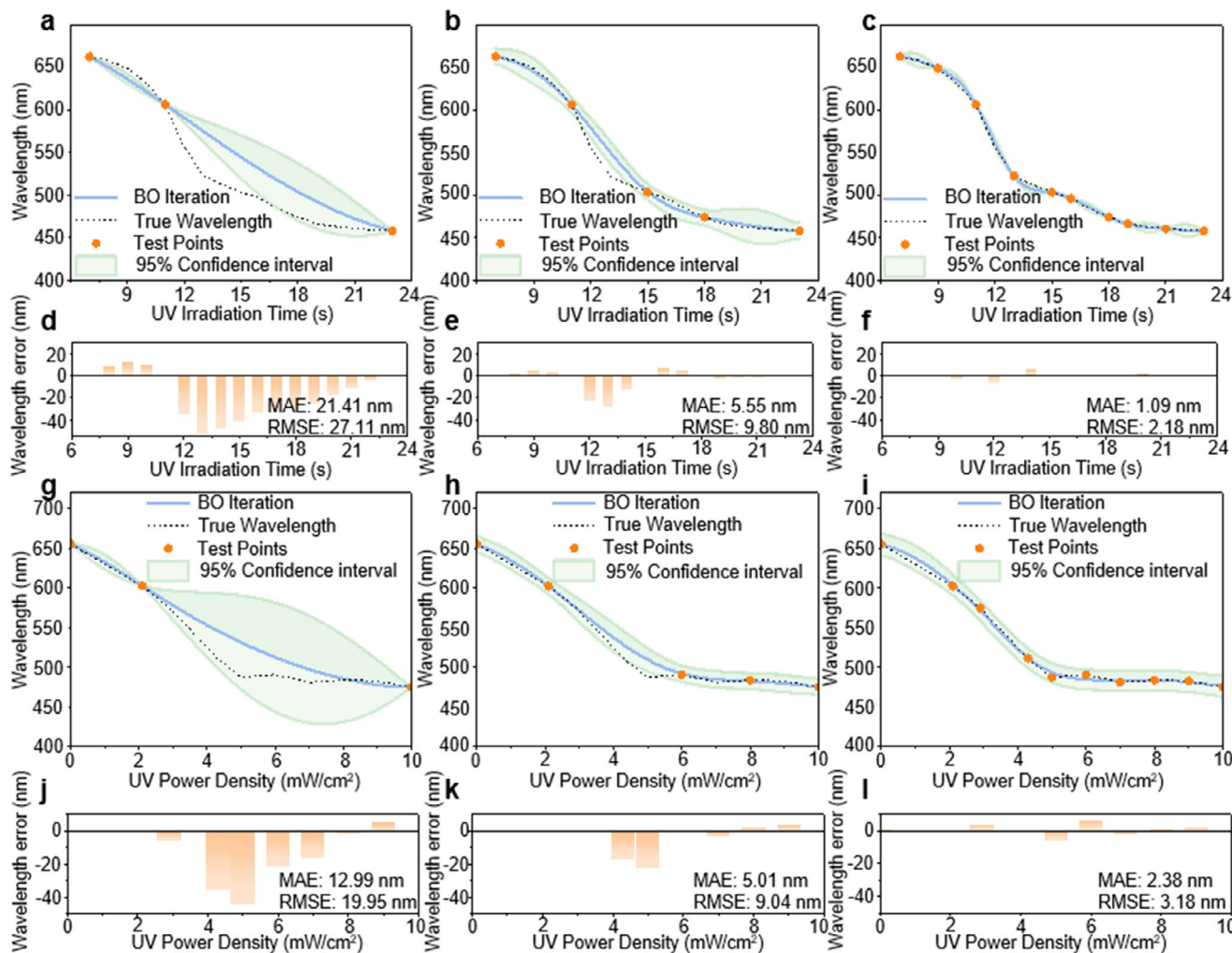


FIGURE 4 | Bayesian optimization-assisted modeling of UV-regulated structural color in photonic gels. (a–c) The fitting process for wavelength and UV irradiation time; (d–f) Mean absolute error (MAE) and root mean square error (RMSE) of the fitting process for wavelength and UV irradiation time; (g–i) The fitting process for wavelength and UV power density; (j–l) MAE and RMSE of the fitting process for wavelength and UV power density.

2.38 and 3.18 nm, respectively (Figure 4j–l). For comparison, the UV irradiation time and wavelength and UV power density and wavelength relationships were also fitted using a conventional four-parameter logistic model (Figure S7). The model reproduced the overall monotonic trends, but yielded MAE/RMSE values of 5.63/6.69 nm for UV irradiation time (Figure S7a,b) and 4.50/5.79 nm for UV power density (Figure S7c,d). The different performances of the two models can be understood from their mathematical formulations. The four-parameter logistic model assumes a predefined monotonic sigmoidal response characterized by two wavelength asymptotes, a transition midpoint, and a Hill coefficient. This compact formulation provides a physically intuitive description of the overall wavelength shift but has limited flexibility in representing local variations in the experimental response. In contrast, the Gaussian process surrogate used in the BO framework does not impose a fixed global functional form. Its posterior prediction is determined by the covariance between measured processing conditions and provides both a wavelength estimate and a pointwise uncertainty. This uncertainty enables subsequent calibration conditions to be selected adaptively from regions in which the model is least confident. The associated error analysis further confirms the effectiveness of the BO-

guided strategy, as both MAE and RMSE decrease consistently during the iterative fitting process. Notably, accurate prediction of the UV-wavelength relationship can be achieved with only a limited number of experimental data points, highlighting the efficiency of this approach. After establishing the quantitative UV-wavelength relationship, desired structural colors can be precisely accessed by selecting suitable UV irradiation times or UV power densities within the calibrated parameter space. The BO-guided model therefore converts wavelength design targets into experimentally actionable UV exposure parameters, allowing color control through either exposure duration modulation or incident power modulation. This relationship curve is essential for optical manufacturing because it enables target reflected wavelengths to be translated into controllable local UV doses for subsequent grayscale mask encoding and pixel-level structural color patterning. In this work, under appropriate UV power densities, 10 s is sufficient to cover the full wavelength tuning window from red to blue. Therefore, 10 s was chosen as the standard exposure time for the following studies. Overall, this data-driven framework establishes a robust mapping between UV exposure, including irradiation time and power density, and structural color. This capability not only provides a critical

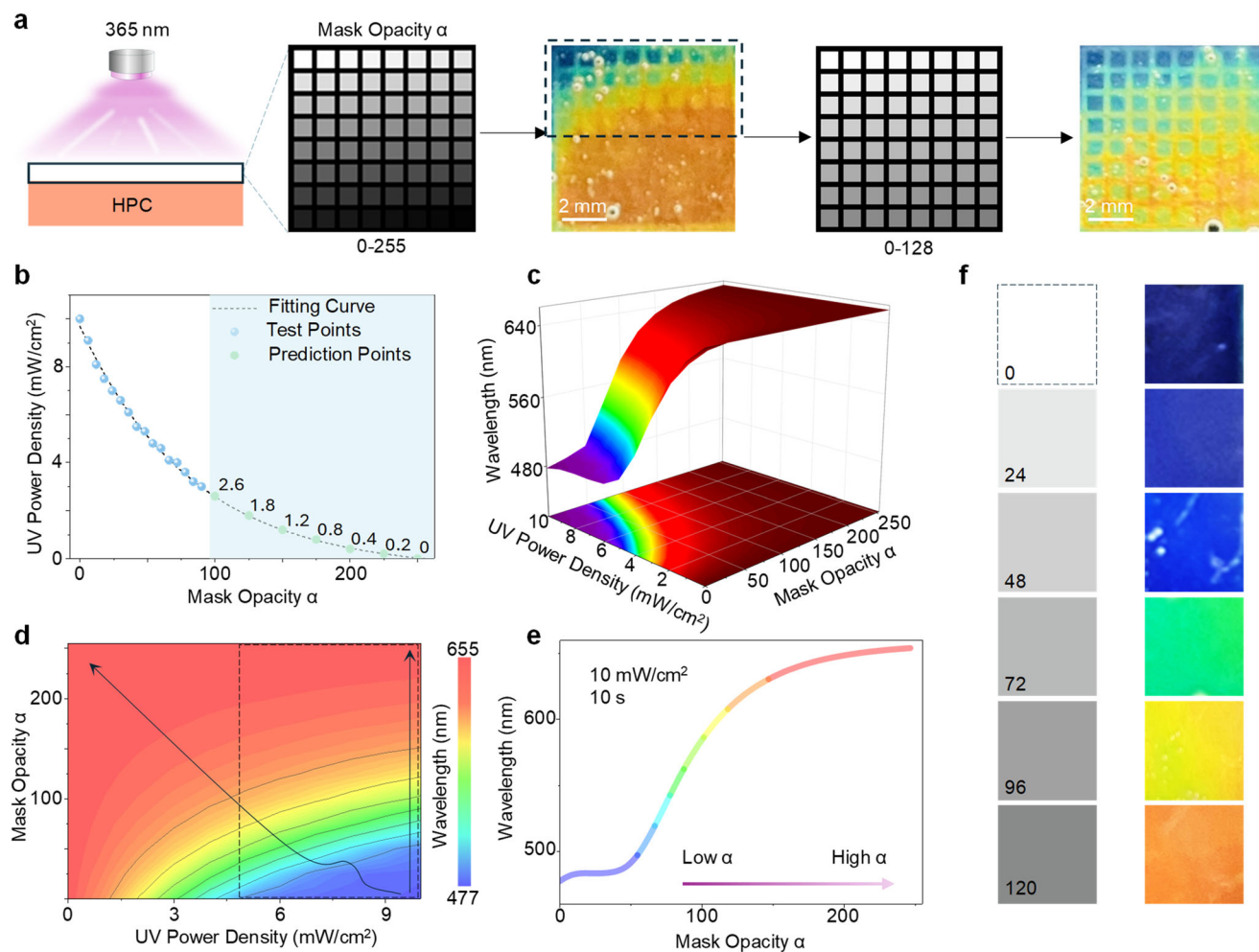


FIGURE 5 | Opacity-encoded optical masks. (a) Selection of the effective opacity range. (b) Monotonic fitting of the relationship between mask opacity α and filtered UV power density. (c) Three-dimensional response surface obtained by filtered UV power density-wavelength-mask opacity α . (d) Two-dimensional projection of the response surface. (e) Relationship between mask opacity α and wavelength under 10 mW/cm² UV irradiation for 10 s. (f) Structural color variation under different opacity values under 10 mW/cm² UV irradiation for 10 s.

foundation for digitally programmable, pixel-level photonic patterning via controlled UV modulation, but also offers general design guidelines for coupling UV regulation with structural color and other tunable material parameters.

2.5 | Opacity-Encoded Optical Masks for Quantitative UV Dose Translation

An opacity-encoded optical mask was developed to quantitatively translate digital grayscale information into spatially controlled UV dosage. The incident UV power density for mask-based optical manufacturing was selected based on the power-density response established in Figure 2f. Under fully transparent exposure conditions, 68 wt.% AM-HPC undergoes a red-to-blue structural color transition within 10 s when the UV power density reaches approximately 5 mW/cm². Because an opacity mask attenuates the incident UV field, the applied UV power density must be higher than this threshold to ensure that sufficient local UV dose reaches the gel after passing through the mask. Therefore, 10 mW/cm² was chosen as the standard incident power density for mask calibration. As shown

in Figure 5a, the full opacity range from 0 to 255 was then evaluated, and an effective working window of 0 to 128 was identified for continuous structural color tuning from red to blue. This opacity window enables grayscale mask values to be converted into controlled local UV doses for subsequent pixel-level color programming. The influence of mask opacity α on the transmitted UV power density was subsequently investigated. A monotonic fitting function was used to correlate mask opacity α with the transmitted UV power density (Figure 5b). The fitted curve shows that the effective UV intensity continuously decreases with increasing mask opacity α , confirming that mask opacity can serve as a reliable control parameter for local UV modulation. This result provides the basis for converting digital image information into optical manufacturing instructions. The fitting accuracy, evaluated by $R^2 = 0.9952$ and RMSE = 0.143 mW/cm² (Figure S8), further confirms that the mask opacity α -dependent UV attenuation can be reliably predicted. Because the reflected wavelength of AM-HPC is governed by UV dose-dependent pitch contraction, this calibrated relationship provides the quantitative basis for converting target structural colors into grayscale mask instructions for spatially programmed optical manufacturing.

By combining the fitted mask opacity α -filtered UV power density relationship with the previously established UV power density-wavelength relationship, a three-dimensional response surface was constructed (Figure 5c). The constructed response surface captures the coupled relationship among mask opacity, filtered UV power density, and wavelength. Increasing mask opacity reduces the filtered UV power density reaching the AM-HPC gel, whereas increasing filtered UV power density drives the reflected wavelength toward shorter values through stronger UV-induced reorganization of the cholesteric mesophase. Consequently, the reflected wavelength shows a positive dependence on mask opacity and a negative dependence on filtered UV power density. This trend indicates that low opacity regions generate blue-shifted colors by transmitting higher UV doses, while high opacity regions retain longer wavelength colors by suppressing local UV exposure. Under a fixed irradiation time of 10 s, this response surface directly links mask opacity, filtered UV power density, and the resulting structural color wavelength, thereby providing a quantitative basis for direct writing optical manufacturing. To define a practical fabrication window, the three-dimensional response surface was further projected into a two-dimensional parameter map (Figure 5d). This projection allows the operable region for full color programming to be visualized more intuitively. A pronounced red-to-blue transition is already visible at an incident filtered UV power density of approximately 7 mW/cm², showing that broad structural color modulation can be initiated under this condition. However, the deepest blue region is observed only when the incident filtered UV power density exceeds approximately 9 mW/cm². This region is located in the lower right triangular domain of the process map, where low mask opacity permits a high filtered UV power density to reach the gel. Therefore, within the selected opacity range of 0–128, an incident filtered UV power density above 9 mW/cm² is required to achieve the complete red-to-blue tuning window. Accordingly, 10 mW/cm² was selected for subsequent optical manufacturing to ensure sufficient filtered UV dose across the designed mask opacity range and to enable complete structural color programming. This result defines the practical operating regime for mask-guided optical manufacturing and ensures that the encoded mask opacity α values can be faithfully translated into predictable structural colors.

Based on this mapping, the relationship between mask opacity α and wavelength was further extracted at a fixed UV power density of 10 mW/cm² and UV irradiation time of 10 s, as shown in Figure 5e. This result indicates that structural color can be continuously programmed through opacity modulation. The corresponding structural color changes under different mask opacity α values are shown in Figure 5f, confirming that the encoded opacity values can be accurately translated into predictable optical outputs. These results establish a quantitative bridge from mask opacity α to UV dose and ultimately to structural color, thereby forming the key translation layer between digital image information and pixel-level optical manufacturing. Under the selected conditions in this work, namely 10 mW/cm² UV irradiation for 10 s, the mask opacity α -wavelength curve provides the direct calibration function for pixel-level color programming. By assigning an appropriate mask opacity, each AM-HPC pixel can be tuned across the red-to-blue structural color range under this fixed exposure condition. In principle, by combining the UV irradiation time-wavelength, UV power density-wavelength, and

mask opacity α -filtered UV power density relationships established above, the target structural color can also be modulated through flexible combinations of exposure time, incident UV power density, and mask opacity α . This integrated calibration framework enables digital color information to be translated into spatially resolved UV exposure fields for programmable photonic manufacturing.

2.6 | Pixel-Level Optical Manufacturing of Structural Color Images

The optical manufacturing strategy was demonstrated through spatially resolved structural color control using masks with different opacity distributions. As illustrated in Figure 6a, point, line, and area-based masks with spatially varying opacity were employed to regulate local UV dosage across a two-dimensional plane, thereby enabling deterministic tuning of the cholesteric mesophase at predefined positions. Representative examples are shown in Figure 6b, where discrete features, continuous line patterns, and area-confined geometries were directly written into the AM-HPC surface. These results demonstrate that the proposed approach enables not only regional optical manufacturing, but also programmable control over structural color with high geometric freedom.

Building on this spatial control capability, a digital workflow was developed for structural color image reconstruction. As shown in Figure 6c, the target image is first converted into a wavelength map and subsequently translated into an opacity-encoded optical mask. In this workflow, color information in the digital image is no longer treated as a surface pigment signal but is converted into a spatial distribution of target reflected wavelengths. Each wavelength value is then mapped to a corresponding mask opacity according to the calibrated opacity-wavelength relationship established above. In this way, digital image information is transformed into spatially resolved UV exposure instructions for mask-guided optical manufacturing.

The quantitative reconstruction process is detailed in Figure 6d. The target image was first sampled at a resolution of 13 840 pixels/cm², corresponding to a pixel size of 85 μ m $\times$ 85 μ m. The RGB value of each pixel was extracted and converted into the corresponding wavelength. Based on the previously established UV power density-wavelength relationship at a fixed UV irradiation time of 10 s, the target wavelength of each pixel was first converted into the required UV power density and then further translated into the corresponding mask opacity α value. Through this stepwise mapping, each digital pixel is assigned a specific opacity value that determines the local UV dose delivered to the AM-HPC gel. The resulting optical mask is therefore composed of spatially connected opacity units, each encoding the exposure condition required to generate a prescribed structural color. This pixel-wise conversion establishes a quantitative bridge between digital image input and spatially programmable photonic output, enabling image-like structural color patterns to be manufactured in a single UV exposure step.

Using this optical manufacturing strategy, a high-resolution structural color reconstruction of Van Gogh's *Sunflowers* was achieved in the AM-HPC (Figure 6e). The resulting image

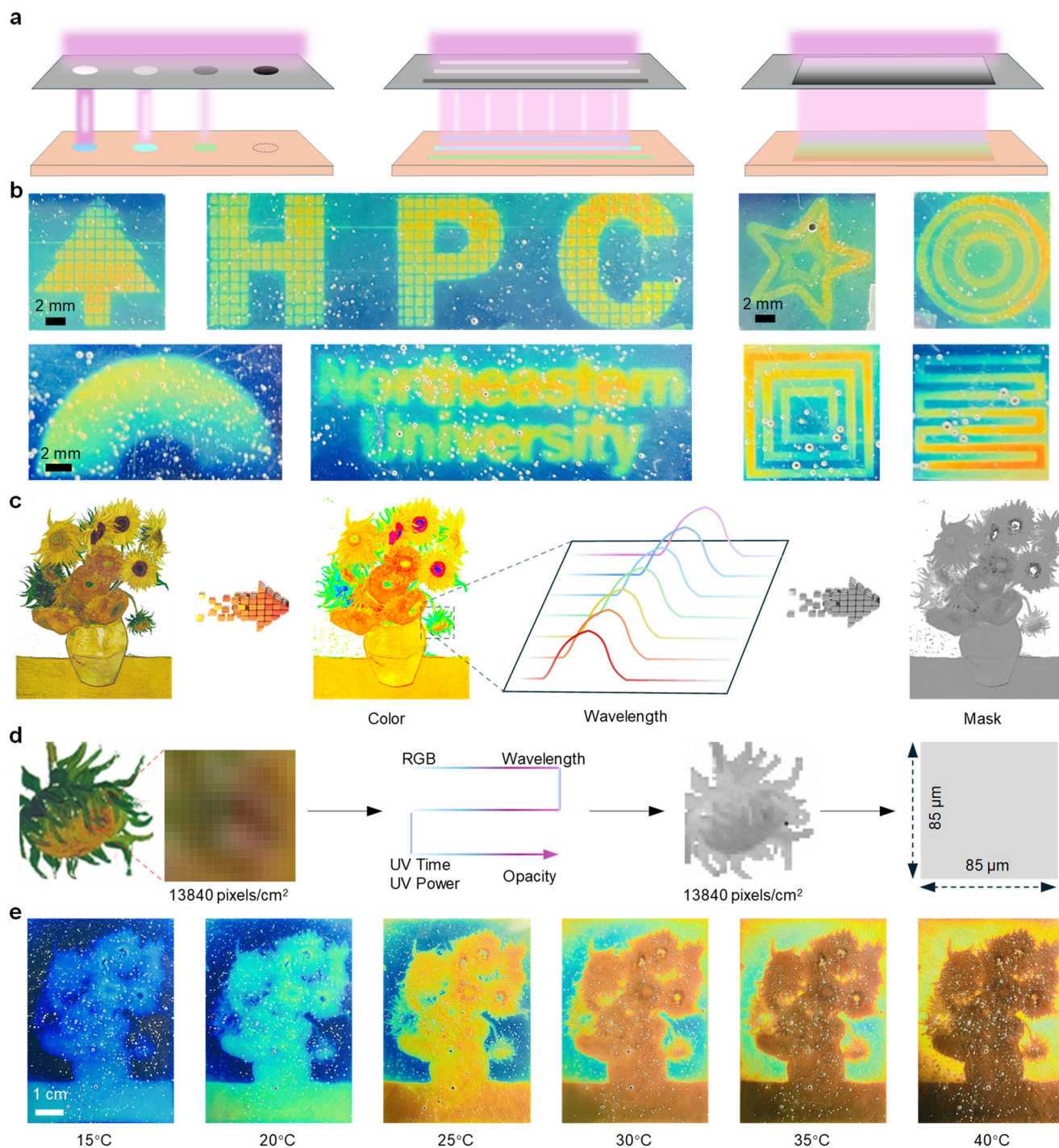


FIGURE 6 | Spatially programmable photonic patterning and high-resolution image reconstruction. (a) Schematic illustration of spatial control of structural color. (b) Point, line, and area-based structural color patterning. (c) Workflow for mask generation from a target image. (d) Parameter mapping for optical mask reconstruction. (e) Structural color image: Van Gogh's *Sunflowers*.

exhibits continuous color variation, clear local features, and faithful spatial correspondence with the target artwork. Moreover, the reconstructed image retains the intrinsic temperature responsiveness of the HPC system, showing dynamic structural color evolution under thermal variation. The generality of this strategy is further demonstrated by the structural color reconstruction of *Woman with a Watch* by Pablo Picasso (Figure S9), which also exhibits continuous image encoding and dynamic thermoresponsive optical evolution. The successful reproduction

of distinct artistic styles confirms that the present framework is not limited to a single target pattern but is broadly applicable to complex multicolor image reconstruction.

3 | Conclusion

In summary, this work establishes a contactless, light-guided strategy for achieving two-dimensional spatial programming of

cholesteric self-assembly in HPC-based photonic gels. By incorporating photo-crosslinking components into the HPC matrix, UV irradiation drives polymer-network formation and reorganization of the cholesteric mesophase. The resulting blueshift reflects a decrease in the optical periodicity, and may involve coupled changes in the effective helical pitch, average refractive index, and orientation of the cholesteric domains relative to the incident light. In the 68 wt.% AM-HPC system, this strategy achieves a broad wavelength-tuning window of 204 nm and spatial features down to 50 μm . A quantitative framework integrating Bayesian optimization with optical mask design was developed to capture the nonlinear relationships among UV irradiation time, UV power density, mask opacity, concentration, and wavelength, achieving accurate prediction with only 8 test points. Under a UV power density of 10 mW/cm^2 and an irradiation time of 10 s, light-directed assembly combined with Bayesian optimization-guided optical manufacturing enables single-step, pixel-level (reconstruction at 13 840 pixels/ cm^2 , 85 μm pixel pitch) structural color programming of complex images with high spatial fidelity, while preserving the intrinsic thermoresponsive behavior of the HPC system. This work provides a generalizable framework for coupling UV-regulated self-assembly with digitally addressable optical manufacturing. This work establishes a scalable route for translating digital information into spatially encoded photonic architectures, thereby expanding the design space of structural color materials toward more complex and functional optical systems. This strategy potentially provides a basis for future applications in adaptive displays, optical encryption, and responsive photonic interfaces.

4 | Experimental Sections

4.1 | Materials

Hydroxypropyl cellulose (HPC, $M_w \approx 4 \times 10^4$) was obtained from Nippon Soda Co. Acrylamide (AM), N, N'-Methylenebisacrylamide (MBA, crosslinker), and 2-Hydroxy-4'-(2-hydroxyethoxy)-2-methylpropiophenone were purchased from Fisher Scientific and used without further purification. Deionized (DI) water was employed for the preparation of all aqueous formulations. For UV curing experiments, a 365 nm ultraviolet light source was used (a commercial LED UV lamp purchased from Amazon). Transparent paper and carbon black were purchased from Amazon. Unless otherwise specified, all chemicals were utilized as received.

4.2 | Preparation of AM-HPC Gels

AM-HPC gels with solid contents of 68, 70, 72, 74, 76, and 78 wt.% were prepared. In all formulations, hydroxypropyl cellulose (HPC, 7 g per batch) served as the cholesteric matrix. Acrylamide was introduced as the polymerizable monomer at 10 wt.% relative to the mass of HPC, while N, N'-methylenebisacrylamide and 2-Hydroxy-4'-(2-hydroxyethoxy)-2-methylpropiophenone were each added at 1 wt.% relative to HPC (Table S8).

The photopolymerizable components were first dissolved in the calculated amount of deionized water to ensure homogeneous dispersion. Subsequently, HPC was gradually incorporated into

the precursor solution and mixed using a planetary centrifugal mixer at 3500 rpm for 5 min to obtain a uniform viscous cholesteric mixture. The resulting AM-HPC formulation was then sandwiched between two glass slides to form thin films of controlled thickness, which were subsequently subjected to ultraviolet irradiation for photopolymerization.

4.3 | Preparation of Mask

The grayscale photomask was fabricated by printing carbon black onto transparent paper to achieve spatially tunable ultraviolet (UV) attenuation. Mask opacity α is an 8-bit grayscale value from 0 to 255, where $\alpha = 0$ gives maximum UV transmission, and increasing α reduces transmitted UV intensity.

The mask opacity α was calculated from the transparency T (%) value according to

$$\alpha = 255 \left(1 - \frac{T(\%)}{100} \right)$$

To quantitatively calibrate the relationship between grayscale value and transmitted UV intensity, the printed masks were characterized under a fixed incident UV intensity of 10 mW/cm^2 (365 nm). The transmitted UV power through different grayscale regions was measured, and the corresponding attenuation behavior was evaluated.

4.4 | Reflected Color and Wavelength Determination of Structural Color

Structural color responses of the AM-HPC gels were quantified under standardized illumination conditions. Each sample was placed on a non-reflective black background to minimize stray reflection and ambient light interference. A daylight-equivalent white light source with a correlated color temperature of 6500 K was directed normal to the sample surface. Images were acquired vertically using an Apple iPhone 13. The illumination geometry, camera-to-sample distance, viewing angle, sample position, exposure, focus, and other acquisition settings were maintained consistently throughout all measurements to ensure reproducibility.

Image analysis was conducted in Python. Mean RGB values were extracted from predefined regions of 1.6 cm x 1.6 cm and subsequently converted into CIE 1931 chromaticity coordinates. The dominant wavelength (λ) corresponding to each structural color was calculated using established colorimetric transformation protocols. All measurements were carried out under identical optical conditions to ensure consistency. Because this conversion is affected by the spectral response and color discrimination of the smartphone imaging system, the resulting values are defined as camera-derived apparent wavelengths rather than absolute spectroscopic peak wavelengths. These values were used for image-based color analysis and mask calibration. No post-acquisition gamma adjustment, histogram equalization, histogram stretching, contrast enhancement, or color correction was applied to the images used for RGB analysis.

The resulting RGB- λ dataset was further utilized to train a Bayesian optimization model for establishing the quantitative relationship between UV irradiation parameters and the evolution of structural color.

4.5 | Computational Image Processing for Patterning

Input Dataset 1: wavelength paired with intensity:

$$\{(\lambda_i, I_i)\}_{i=1}^n$$

Input Dataset 2: intensity paired with opacity α :

$$\{(I_j, \alpha_j)\}_{j=1}^m$$

We seek a function $\alpha(\lambda)$ defined by two successive interpolations.

Step 1. Find k such that $\lambda_k \leq \lambda < \lambda_{k+1}$, then interpolate Dataset 1 to get intensity:

$$I(\lambda) = I_k + \frac{I_{k+1} - I_k}{\lambda_{k+1} - \lambda_k} (\lambda - \lambda_k)$$

where λ is the query wavelength, λ_k and λ_{k+1} are the two nearest measured wavelengths bracketing λ , and I_k and I_{k+1} are their corresponding measured intensities.

Step 2. Find j such that $I_j \leq I(\lambda) < I_{j+1}$, then interpolate Dataset 2 to get opacity α :

$$\alpha(\lambda) = \alpha_j + \frac{\alpha_{j+1} - \alpha_j}{I_{j+1} - I_j} (I(\lambda) - I_j)$$

where $I(\lambda)$ is the intensity obtained from Step 1, I_j and I_{j+1} are the two nearest measured intensities bracketing $I(\lambda)$, and α_j and α_{j+1} are their corresponding opacity α values.

4.6 | AM-HPC Gel Transmission, Reflection, and Absorption Light Measurement

The optical properties of the AM-HPC gels were evaluated using a UV-vis spectrophotometer (Shimadzu UV-1900, Japan). Transmission and reflection spectra were recorded over the visible wavelength range under normal incidence conditions. For transmission measurements, the samples were positioned perpendicular to the incident light path. Reflection and absorption spectra were obtained using the corresponding reflection and absorption modes of the instrument. For the UV-vis reflection spectra, the wavelength corresponding to the principal reflection minimum was taken as the characteristic wavelength of the cholesteric photonic band.

4.7 | Bayesian Optimization and Gaussian Process of UV Time/Power-Wavelength Curves

To establish the relationship between UV curing conditions (irradiation time and power) and the resulting structural color of the

AM-HPC, we model the calibration curve from UV parameters to dominant reflected wavelength λ using Gaussian Process (GP) regression with a Matérn ($\nu = 5/2$) kernel. Unlike fixed parametric models, the GP provides both a continuous mean prediction and a pointwise uncertainty estimate across the entire parameter range.

To establish the relationships between UV irradiation time or UV power density and the resulting wavelength of the AM-HPC gel, Gaussian process regression was employed as a surrogate model. The wavelength response was represented as

$$\lambda(x) \sim GP[m(x), k(x, x')]$$

where x denotes either UV irradiation time or UV power density, $m(x)$ is the prior mean function, and $k(x, x')$ is the covariance kernel. A constant-scaled Matérn 5/2 kernel combined with a white-noise kernel was used:

$$k(x, x') = \sigma_f^2 \left(1 + \frac{\sqrt{5}r}{l} + \frac{5r^2}{3l^2} \right) \exp\left(-\frac{\sqrt{5}r}{l}\right) + \sigma_n^2 \delta_{x, x'}$$

where $r = |x - x'|$, l is the characteristic length scale, σ_f^2 is the signal variance, and σ_n^2 is the observational-noise variance. The output wavelength values were normalized during model fitting, and the kernel hyperparameters were optimized with 15 optimizer restarts.

The calibration was performed through sequential uncertainty-guided sampling. Initially, the two boundary conditions at the endpoints of the investigated UV operating range were used as the training set. After fitting the GP surrogate model, the next experimental condition was selected from the unsampled candidates according to

$$x_{t+1} = \operatorname{argmax}_t \sigma_t(x)$$

where $\sigma_t(x)$ is the posterior predictive standard deviation at iteration t . The selected condition was experimentally measured and added to the training set, after which the GP model was refitted, and the uncertainty distribution was updated. The 95% predictive confidence interval was calculated as

$$CI_{95\%}(x) = \mu_t(x) \pm 1.96\sigma_t(x)$$

where $\mu_t(x)$ and $\sigma_t(x)$ are the posterior predictive mean and standard deviation, respectively. The procedure was repeated until the predictive uncertainty and validation errors were reduced to acceptable levels.

4.8 | Four-Parameter Logistic Fitting of UV Time/Power-Wavelength Curves

The relationships between wavelength and UV irradiation time or UV power density were fitted independently using the same four-parameter logistic (4PL) sigmoidal model:

$$\lambda(x) = \lambda_{\text{bottom}} + (\lambda_{\text{top}} - \lambda_{\text{bottom}}) / \left[1 + (x/EC_{50})^H \right]$$

where x represents either the UV irradiation time (s) or UV power density (mW/cm^2); $\lambda(x)$ is the predicted wavelength at a given x ; λ_{top} and λ_{bottom} are the upper and lower asymptotic wavelengths, respectively; EC_{50} is the UV irradiation time or UV power density corresponding to the midpoint of the wavelength transition; and H is the Hill coefficient describing the steepness of the transition. The irradiation time and power-density datasets were fitted separately using the same functional form but independent parameter sets.

Author Contributions

H. Z. conceived the concept and supervised the work. M. Y. prepared materials and apparatus, performed testing, organized data, and drafted the manuscript. Y. Z., S. S. K. S., and A.R.A. implemented the machine-learning workflow and developed the mask. Y. W. and R. B. assisted with material preparation and processing. N. P. and S. M. assisted with mask implementation. H. Z. guided the project and revised the manuscript.

Conflicts of Interest

The authors declare no conflicts of interest.

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 1: adfm78709-sup-0001-SuppMat.pdf.

Supporting File 2: adfm78709-sup-0002-VideoS1.mp4.