

# Pigment-free Color Printing Technology for Customized Structural-Color Graphics

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Structural color is reflection colors solely developed by periodic nanostructures in the absence of chemical pigments, providing unique visual impression and applications. Previously reported methods for colloidal crystallization to produce periodic nanostructures call for time-consuming multi-step processes and delicate conditions so that they are unable to produce structural color patterns. In this work, we newly formulate colloidal inks that simultaneously achieve the proper rheological conditions for direct writing and spontaneous formation of colloidal crystals with high crystallinity. With the inks, we can produce multi-colored graphics with customized designs on various substrates while controlling color hue, brightness, and saturation. Furthermore, angle-dependency and mechanical property are controllable and writing speed is significantly improved. This new printing technology produces optical devices, anti-counterfeiting patches, and aesthetic coatings tailored for applications, providing high potential for industrialization.

## 1. Background (objectives)

Structural colors are aesthetic colors developed by constructive interfaces of light diffracted from periodic nanostructures in the absence of chemical pigments. In nature, there are many examples of structural colors. Structural colors never fade and are biocompatible as long as a structure made of non-toxic materials persists. More importantly, structural colors are determined not by materials but by structures so that various colors are available even for a single set of materials. With these advantages, structural colors are promising for aesthetic coatings, anti-counterfeiting patches, colorimetric sensors, and various optical devices. To develop structural colors, colloidal particles have been used as a building block to construct crystalline lattices. However, conventional methods, based on evaporation of dispersion media, usually involve time-consuming and delicate processes for crystallization and patterning.

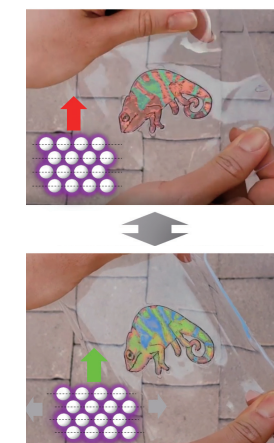
## 2. Contents

To overcome the limitations of conventional approaches, we optimally formulated colloidal inks for direct writing of colloidal crystals without a slow evaporation process. The inks were composed of silica particles dispersed in a photocurable resin. The particles were rendered to be repulsive to secure high ink stability and cause spontaneous crystallization in the resin medium. At the same time, the volume fraction of silica particles is set to be sufficiently high to satisfy the rheological property required for direct writing. The inks can be printed on various target surfaces including glasses, plastics, metals, papers, and even fabrics with high definition, which show iridescent structural colors. The colloidal crystals written on target surfaces have a face-centered cubic (fcc) lattice. Interestingly, the orientation of fcc crystals is dictated by the shear forces that the inks experience during the writing and spreading on a target surface. For the line segments, crystal orientation varies from the top to the bottom in the cross-section due to different directions of shear flow. By contrast, faces formed by the fusion of lines show uniform crystal orientation across the entire cross-section due to the shear flow during the fusion. The uniform orientation enables the photonic surfaces to have reflectivity at the resonant wavelength as high as 90%. The minimum width of lines is approximately

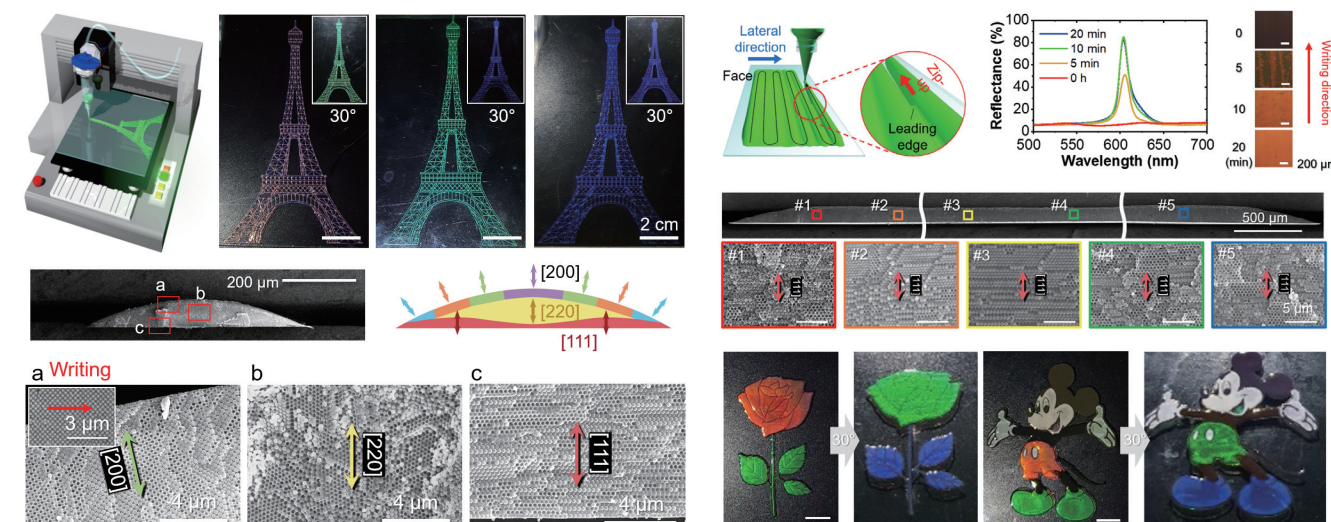
## 3. Expected effect

hair thick and the maximum writing speed is approximately 15 mm/s. As the inks are formulated to have high viscosity, there is no blurring at the boundaries between two inks printed on the same surfaces in the absence of photocuring. Therefore, any customized multicolor graphics are printable with multiple distinct inks. The mechanical property of printed graphics is determined by the choice of the ink resin. The resin that forms rigid polymer through photocuring is available for stable and invariant coloration, whereas the resin for the tough polymer is good for foldable graphics. The use of the resin for elastic polymer enables the production of stretchable graphics, which show dynamic color changes upon the stretching and relaxation, like a chameleon.

We have developed a pragmatic technology to print structural color graphics with a high degree of freedom and unprecedented levels of controllability. The structural color graphics are promising as new design elements on apparel and accessories for the MZ generation so that they can better express their individuality. It is expected that the structural color patterns added onto contact lenses are non-toxic yet provide a visual effect, more like natural blue eyes. Also, the combination of iridescent colors and sharp peaks in spectra provides non-copiable tags for anti-counterfeiting patches. The resin for the inks can be selected to show stimuli responsiveness for temperature, humidity, pH, and specific molecules, which makes the structural colors changeable according to the stimuli. Such color-tunable graphics can be further developed to make intuitive colorimetric indicators and chameleon-like wearable displays.



**Figure 1.** Dynamic color change of chameleon graphics by mechanical deformation.



**Figure 2.** Direct writing of structurally-colored graphics composed of line segments and position-dependent orientation of crystalline arrays in the line cross-section.

**Figure 3.** Direct writing of faces through line fusion, the high reflectivity of faces with a uniform orientation of crystalline arrays, and iridescent multicolor graphics of rose and Mickey Mouse.

## Research outcomes

**Paper** J. B. Kim, C. Chae, S. H. Han, S. Y. Lee, S.-H. Kim, Direct writing of customized structural-color graphics with colloidal photonic inks, *Sci. Adv.* 7, eabj8780 (2021).

S. K. Nam, J. B. Kim, S. H. Han, S.-H. Kim, Photonic Janus balls with controlled magnetic moment and density asymmetry, *ACS Nano* 14, 15714-15722 (2020).

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