# Direct writing of customized structural-color graphics with colloidal photonic inks

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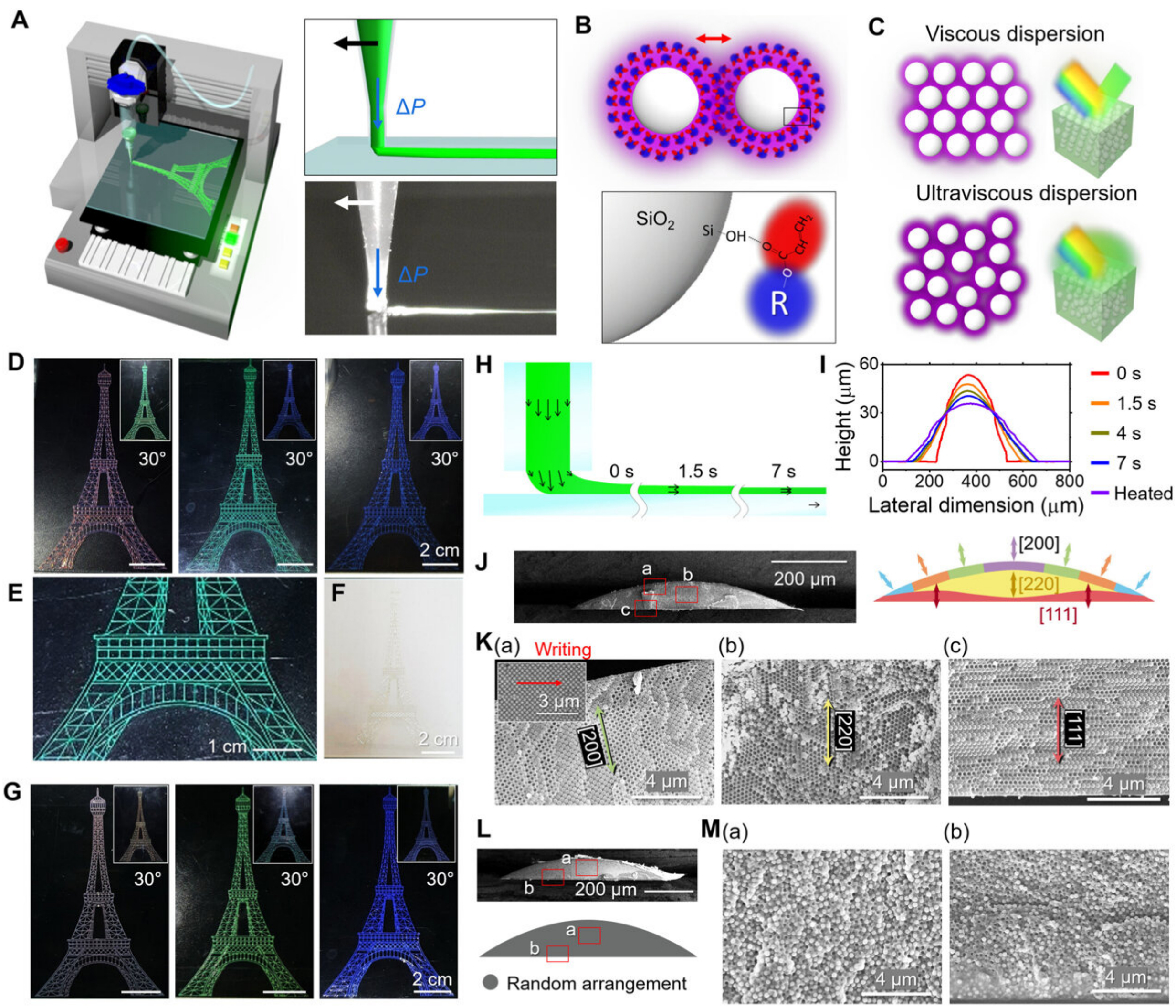



Direct writing of structurally colored lines. (A) Dispenser for line drawing with a moving nozzle. (B) Formation of a solvation layer on the silica surface by hydrogen bonds. (C) Crystalline and glassy packing in moderately viscous ethoxylate acrylate (EA) and ultraviscous urethane acrylate (UA). (D to F) Eiffel Tower patterns drawn using EA inks: red, green, and blue colors (D), magnified view (E), and the pattern viewed under off-reflection with a white background (F). The insets are off-normal views for specular reflection. (G) Eiffel Tower patterns drawn using UA inks. (H and I) Velocity profiles of inks (H); temporal evolution of the surface profiles of an EA line (I). (J and K) Cross-sectional scanning electron microscopy (SEM) images of an EA line and schematic showing position-dependent crystal orientations (J); (200) planes of a face-centered-cubic (fcc) lattice in the top [K(a)], (220) planes in the middle [K(b)], and (111) planes in the bottom [K(c)]. The inset in [K(a)] shows a square array on the top surface. (L and M) The same set of subfigures as those in (J) and (K) but for a UA line. A consistent amorphous array is formed in the whole volume. Credit: Jong Bin Kim, KAIST.

Colloidal crystals and glasses are tunable, iridescent, nonfading and nontoxic materials that can be used to develop structural colors. In a new report now published in *Science Advances*, Jong Bin Kim, and a team of researchers in chemistry and advanced materials in the Republic of Korea, developed direct writing of structural color graphics with high brightness and saturation using colloidal inks.

The team prepared the inks by dispersing silica particles in acrylate-based resins where they optimized the volume fraction to simultaneously provide pronounced colors and satisfactory printing rheology. Using the inks, they could directly write any macroscopic design of lines on various substrates, while setting the microscopic colloidal arrangement to be either crystalline or amorphous based on the resin viscosity to control the iridescence of colors. The resulting graphics could be surface-transferred, origami-folded or elastically stretched to provide a direct writing approach with versatility and regulation.

## Bioinspired artificial periodic nanostructures

Nature has created periodic nanostructures to enhance or reduce the visibility of animals during mating competition or camouflage. Plants that are living in light-deficient environments can use nanostructures to improve their efficiency of photosynthesis.

Researchers are inspired by nature to develop artificial periodic nanostructures with tunable, iridescent, nonfading, nontoxic colors. Structural-color printing can be customized by design, color combination, and the type of target substrate. In this work, Kim et al. described direct writing with colloidal photonic inks for customizable structural color printing. The scientists optimally formulated the inks by dispersing silica particles in photocurable resins for macroscopic printing and microscopic colloidal structuring. Using the inks, they drew lines and faces, and could directly write multi-color graphics on various substrates, such as glass, plastics, metals, paper and some fabrics. The graphic containing paper could also be folded for origami and released from the substrates to prepare freestanding films or transfer them onto other surfaces to create structural-color graphics for many purposes.

**Formulating the inks and forming the Eiffel tower.**

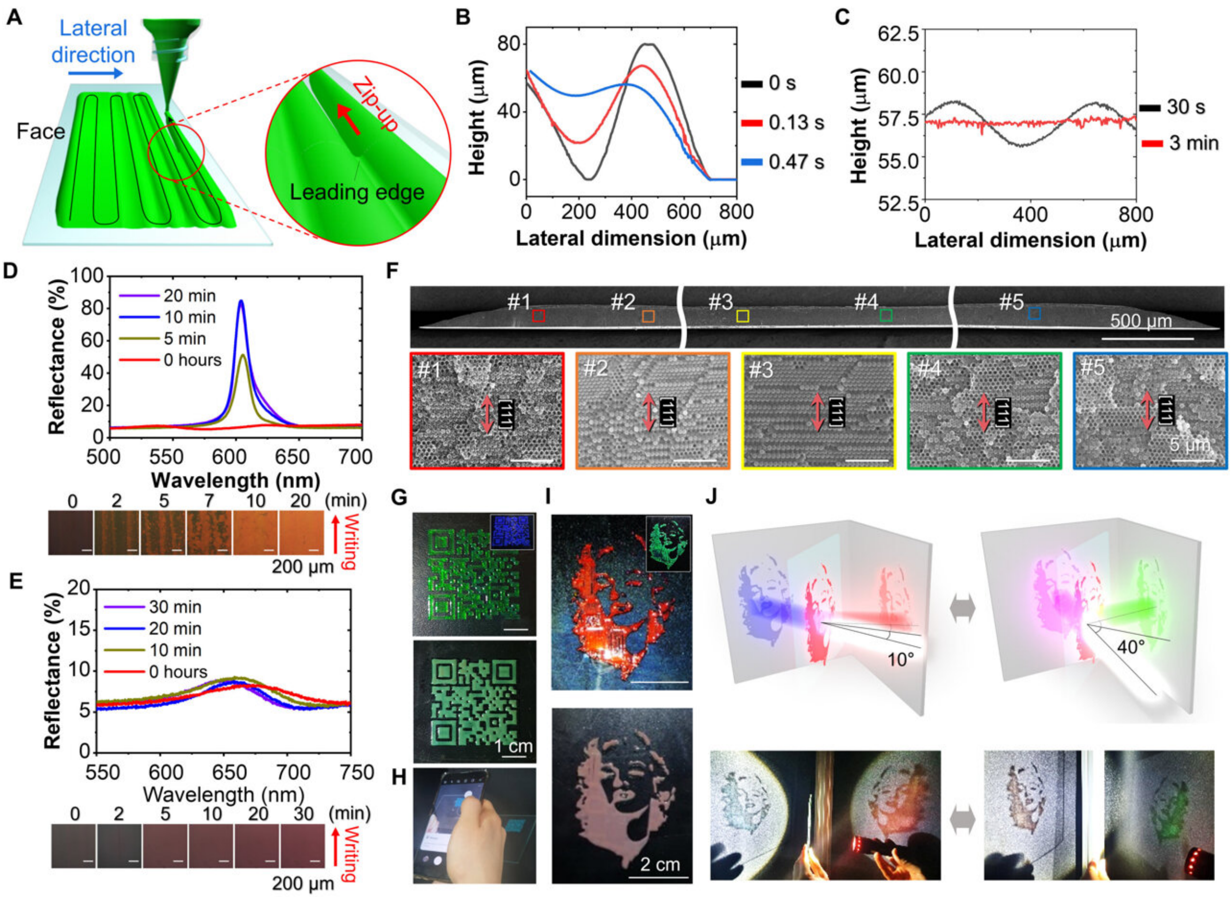
The direct writing process could deposit ink materials along predetermined trajectories to produce customized graphics and patterns without masks to minimize the use of valuable ink. The team governed the printing speed and pattern resolution on the rheological properties of the ink where a well-defined, colloidal arrangement in the form of crystals or glasses were a prerequisite for structural color. The ink formulations were important for direct writing of structural color graphics to achieve high-resolution, high quality, fast printing. To create the inks, Kim et al dispersed monodisperse silica particles in acrylate-based photocurable resins in the absence of volatile components. They then used carbon black nanoparticles on the acrylate resin to reduce Mie scattering. The team screened and optimized the ethoxylate acrylate

(EA) resins, phenyl ether acrylate (PEA), and urethane acrylate (UA) resins and optimized versions of both EA and UA inks to draw a graphic of the Eiffel tower on a glass slide. During the experiments, the team loaded the inks into a tapered nozzle for ejection, while the nozzle moved horizontally in a 100-µm gap between the nozzle and slide. They maintained the writing speed at 8 mm per second, where the constituent crystalline structures made the printed graphics with EA (ethoxylate acrylate) inks highly transparent.

## Printing lines and faces on substrates

The scientists next tuned the line width during the printing experiments by adjusting the writing speed to form cursive texts such as "Happy Birthday." By changing the pressure and speeds for the two types of inks (EA and UA) they could write texts with comparable variation of line widths. Kim et al. did not restrict the target surfaces to glass, drawing graphics on various non-absorbing surfaces including plastics, metals and silicon wafers. For absorbing surfaces such as paper, they devised a higher ejection rate, or slower printing process.

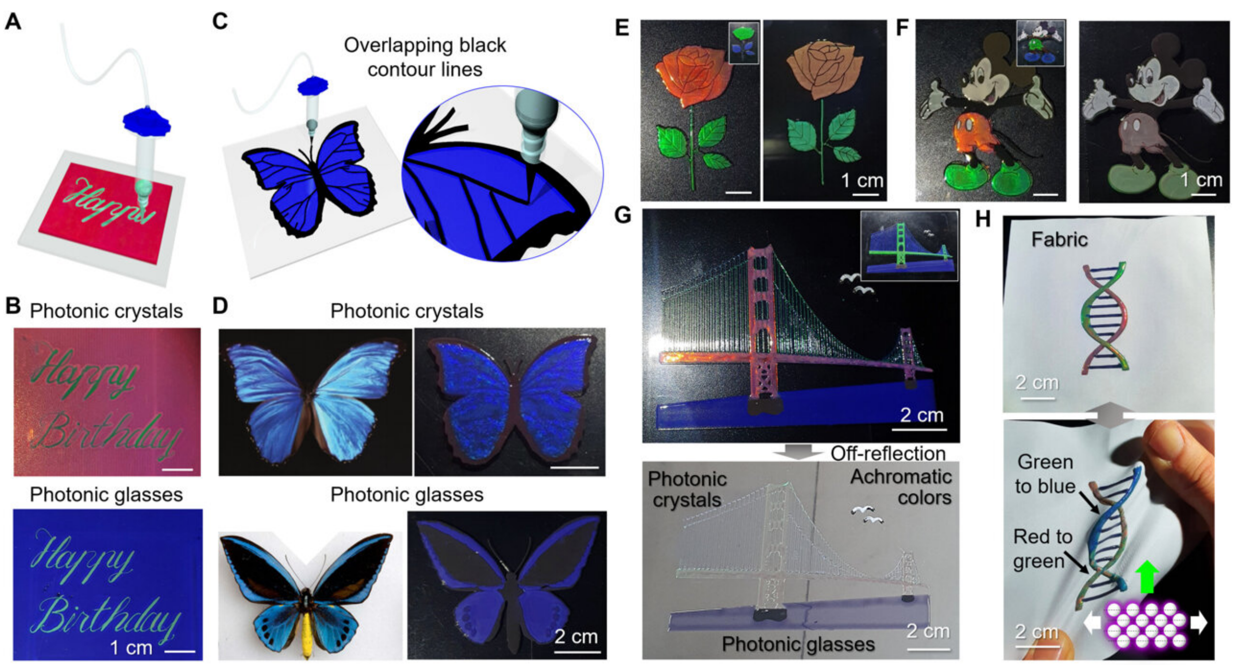
The team next printed faces by fusing lines drawn side by side. The as-printed faces were optically homogenous and gradually revealed color-stripes along the direction of writing within five minutes at 85 degrees Celsius. The films were optically homogenous during the whole process of thermal incubation for UA inks due to the absence of crystallization. Comparatively, the high reflectivity at the resonant wavelength of faces printed using the EA ink originated from the high crystallinity and uniform crystal orientation in the entire film. The scientists used the setup to draw various color graphics.




Direct writing of structurally colored faces. (A) Schematic showing the formation of a face by fusion of alternately drawn lines, where the gap with an adjacent line is rapidly healed by zipping. (B and C) Rapid evolution of a surface profile by fusion (B) and gradual flattening of the surface (C). (D) Temporal evolutions of the reflectance spectrum and optical texture for a face drawn using an EA ink at 85°C. (E) Same set of images as in (D) but for a face drawn using UA ink. (F) Cross-sectional SEM images showing high flatness and thickness homogeneity and the uniform crystal orientation of the (111) plane along the top and bottom interfaces throughout the entire thickness. (G and H) QR code patterns produced using EA and UA inks (G) and recognition of the code with a smartphone (H). The inset is an off-normal observation. (I and J) Patterns of a Marilyn Monroe image produced using EA and UA inks (I) and projection of the images for reflection and transmission of the EA pattern at two different angles of illumination, as denoted (J). Credit: Jong Bin Kim, KAIST.

## Multicolor printing

Kim et al. sequentially used distinctly colored inks on a single surface without photocuring, and formulated black inks by dispersing a large quantity of black nanoparticles in the EA and UA inks that already contained silica particles. The larger particles did not show structural resonance in the visible region, while providing the desired rheological properties for printing. As proof-of-concept, the scientists printed graphics of a morpho butterfly and Troides urvillianus using blue EA and UA inks alongside additional black inks. The outcome indicated a Morpho butterfly with a sparking blue regular structure and a T. urvillianus butterfly with matte blue and random sphere-packing.




Multicolor patterning. (A and B) Schematic (A) and photos (B) showing the overwriting of the cursive “Happy Birthday” using EA and UA inks on uncured faces of the same inks but with different colors. (C and D) Schematic for drawing black lines on structurally colored faces (C) and photos of a Morpho butterfly (top left) and a graphic drawn with EA ink (top right) and a T.

urvillianus butterfly (bottom left) and a graphic drawn with UA ink (bottom right) (D). Reproduced with permission from (43) (2012 Wiley-VCH, top left) and (44) (2006 the Company of Biologists, bottom left). (E) A rose and leaves direct-written using EA and UA inks. The inset is an off-normal observation. (F) Mickey Mouse printed using EA and UA inks. (G) Golden Gate Bridge printed using both EA and UA inks for specular reflection and off-reflection with a white background. The bay drawn with UA ink is noniridescent and translucent, whereas other parts colored with EA inks are iridescent and transparent. (H) Nucleic acid double helix drawn on fabric using PEA inks. The insets show lattice deformation by stretching. Credit: Jong Bin Kim, KAIST.

The team simultaneously drew a red rose with green leaves using the inks, while depositing black ink to outline the shape as contour. Although the red roses and leaves drawn using EA inks were strongly iridescent, those drawn with the UA ink were not. Kim et al. also developed a Mickey Mouse graphic using five different inks, followed by the Golden Gate Bridge in San Francisco drawn with six different inks and sequential printing. They used red for the mainframe of the bridge, green and blue EAs for the front and rear wires, blue for the bay and black for parts of birds alongside white for bird wings. All parts of the illustration were iridescent and transparent, except the bay drawn with the UA ink, which remained consistently blue.

**Outlook**

In this way, Jong Bin Kim and colleagues developed direct writing of structurally colored graphics with viscoelastic photonic inks. The team optimally formulated the inks to achieve macroscopic rheological properties suited for high printability, alongside microscopic colloidal arrangements desired for structural color. The team explored the controlled direct writing method to directly print on various target surfaces including glass, metals, paper and fabrics, to provide unprecedented levels of versatility and color graphics. The researchers envision the use of color-tunable graphics, which are much-like chameleon skin for wearable displays. They also propose the development of stimuli-responsive patches as intuitive colorimetric indicators for temperature, humidity, pH and specific molecules of interest for applications in daily life.

**More information:** Jong Bin Kim et al, Direct writing of customized structural-color graphics with colloidal photonic inks, *Science Advances* (2021). DOI: 10.1126/sciadv.abj8780

Matthew Jacobs et al, Photonic multilayer structure of Begonia chloroplasts enhances photosynthetic efficiency, *Nature Plants* (2016). DOI: 10.1038/nplants.2016.162



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