Supplementary Materials for

Facile full-color printing with a single transparent ink

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The PDF file includes:

Figs. S1 to S22
Legends for movies S1 and S2
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Other Supplementary Material for this manuscript includes the following:

Movies S1 and S2
The transparent substrates with different wettability are prepared by the surface modification with chemical molecules and spin coating of polydimethylsiloxane (PDMS). The substrates contain the (3-glycidoxypropyl)trimethoxysilane (GPTS)-modified glass, triethoxy-n-octylsilane (TOS)-modified glass, n-decyldichlorosilane (DTCS)-modified glass, PDMS-coated glass and 1H,1H,2H,2H-perfluorodecyltrimethoxysilane (PFOS)-modified glass. After the surface treatment, we can obtain the transparent substrates with different wettability.
Fig. S2. The morphology control of the polymer droplets deposited on a hydrophilic (A) and hydrophobic (B) substrate. When the polymer droplet is deposited on the solid substrate, it will experience four typical processes including contact, spreading, retraction, and balance. At the spreading process, the droplet spreads homogeneously because the surface heterogeneity is overcome by the large kinetic energy until it reaches the maximum spreading. Then the liquid starts to recoil. The surface energy of the hydrophilic and hydrophobic substrate determines the pinning liquid boundary and receding liquid boundary (45). On the hydrophobic substrate with low adhesion, the three-phase contact line (TCL) recedes continuously and the droplet film can retract into the domed morphology. While on the hydrophilic substrate with high adhesion, the TCL is pinned easily and the film morphology is preserved, finally.
Fig. S3. The droplet-by-droplet impact dynamics. (A) The sequenced images of a polymer droplet impacting on a hydrophobic surface (scale bars, 1 mm). (B) The sequenced images of the second polymer droplet impacting on the first-balanced droplet. When the second droplet falls on the top surface of the first microdome, the squeeze deformation will appear. For that the first microdome is not cured, it can keep the liquid state and fuse with the falling droplet. Then the same compression, max-spreading, retraction and balance processes will happen. (C) The sequenced images of the third polymer droplet impacting on the merged microdome. For the homogeneous hydrophobic effect of the substrate, all the droplets will retract into the domed morphology with the constant curvature angles.
Fig. S4. The fusion control between ink droplets. The fusion between droplets depends on whether the two droplets can contact (46), which is controlled by the relative size of the maximum spreading radius and the initial depositing distance (47). As the maximum spreading distance ($R_m$) of the droplet is fixed, we can set the initial depositing distances ($d_d$) to control the separation or fusion of different droplets. (A) The separation of two ink droplets with $R_m = 2.86$ mm and $d_d = 2.95$ mm. Because $R_m$ is smaller than $d_d$, the two droplets will not touch or merge. (B) The contact fusion of two ink droplets with $R_m = 2.86$ mm and $d_d = 2.75$ mm. (C) The fusion of two ink droplets with $R_m = 2.86$ mm and $d_d = 1.80$ mm. As long as $R_m$ is larger than $d_d$, the two polymer droplets will contact and merge into a larger microdome.
Fig. S5. The diameter statistics of the microdomes by large-area printing. (A) Top-view SEM image of the microdome arrays. (B) Statistical diameter distribution of the microdome array in the SEM observation, showing their average diameter is 16.23 ± 0.40 μm. (C) Statistical table of the diameters of the printed microdomes with different ink volume.
Fig. S6. Schematic illustration of the built observing system to record the real-time coloration of the microdomes during printing. The coloration process of printed microdomes is recorded using a custom microscope system. A portable electronic digital microscope integrated with four white LEDs (UM02-B, Vitiny) is employed. The transparent substrate is placed on the upper side of the lens with controlled space. The illumination intensity can be regulated by the switch knob. Then the printer nozzle (Microplotter II, Sonoplot) is precisely positioned on the substrate surface for printing. During printing, the color signal is collected and recorded in real time.
Fig. S7. The real-time coloration of the microdomes during printing. (A-D) The observed real-time dark-field microscopic images of the printed microdomes. The printed yellow dot is set as the initial printing site. With the continuous movement of the nozzle, uniform blue dot array is prepared with single-pixel resolution.
Fig. S8. The different microdome arrays by depositing the same ink droplets on the substrates with different wettability. It is clear the diameters of the printed microdomes can be decreased with the increase of substrate hydrophobicity.
Fig. S9. The curvature angles ($\theta_{\text{cua}}$) of different microdomes on the same hydrophobic substrate. The various microdomes are fabricated by printing the ink droplet with different volume on the same hydrophobic substrate. For the homogeneous hydrophobic property, the curvature angles of microdomes can keep basically constant.
Fig. S10. The geometric relation of the microdome volume \( V_m \) with the diameter \( d \), the height \( h \) and the curvature angle \( \theta_{cua} \). (A) The side-view SEM image of the typical microdome morphology. (B and C) The mathematical geometric models of the microdome. \( V_m \) can be presented as

\[
V_m = \pi \times h^2 (3R - h)/3
\]

According to the relations of \( h \) and \( R \) with \( d \) and \( \theta_{cua} \), as

\[
h = d \times (1 - \cos \theta_{cua})/(2\sin \theta_{cua})
\]

\[
R = d/(2\sin \theta_{cua})
\]

Then \( V_m \) can be expressed as

\[
V_m = \pi \times d^3 \left(\frac{2 + \cos \theta_{cua}}{24 (\sin \theta_{cua})^3}\right)(1 - \cos \theta_{cua})^2
\]

We defined \( A \) as \( \frac{24 (\sin \theta_{cua})^3}{(2 + \cos \theta_{cua})(1 - \cos \theta_{cua})^2} \) then \( A \times V_m = \pi \times d^3 \). \( V_m \) can also be expressed as \( V_i \times X \) (\( V_i \) is the printed ink volume and \( X \) is the retention rate controlled by ink evaporation.), thus

\[
\pi d^3 = V_i \times A \times X
\]
Fig. S11. The retention rate of the printed microdomes. (A) Top-view SEM images of the microdomes. With the increase of ink volume, the microdome diameters are enlarged continuously and the retention rates are basically fixed. (B) The retention rate of different microdomes. The retention rates of different microdomes can keep constant for the controlled processing.
Fig. S12. The color regulation of microdomes controlled by the physical morphology. (A) The simulated microdome color mapping with the change of curvature angle and diameter. We can obtain the full-color regulation by controlling the curvature angle and diameter of the microdome. (B and C) The range of the curvature angles to create the color, which are larger than 46° and smaller than 145°. At this scope, the microdome-based interface can induce the effective TIR paths.
Fig. S13. The comparison between the simulated spectra (A) with measured spectra (B). We compared the simulation spectra with the measured data for the printed microdomes with the diameters of about 17.64 μm, 25.99 μm, 27.98 μm and 29.75 μm. The curvature angles of the four microdomes are about 60 degree. The printed palettes composed of different microdomes were illuminated at the incident angle of 35° and received vertically with the receiving angle of 0°. From the integrated spectra, we could clearly observe the shift of interference peaks caused by the change of microdome diameter, which resulted in the different reflected colors. But there existed error between the simulated and measured spectra with a peak discrepancy of about 50 nm. The discrepancy is from the deviations in preparing the microdomes and measuring the spectra in the experiment. In simulation, the calculated object is a single microdome with perfect morphology and ideal test condition. However, in the experiment, a microdome array (about 10×10 dots array) instead of a single microdome was tested to collect the size-dependent spectra with the macroscopic spectrometer, for the spectra intensity of the single microdome is too weak to be detected. Additionally, the incident and receiving angles are hard to be exactly the same as the simulated ones due to the limitation of the spectrometer. Meanwhile, although the inkjet printing technology is clearly the accurate manufacturing system, the error in preparing the microdomes still exists, which would induce the defects in microdome morphology and affect the consistency of the microdomes in the array. Deviations in microdome preparation and spectra measurement resulted in the peak discrepancy in the simulated and measured spectra. However, from the contrast of the simulated and measured spectra, the similar shift trend of interference peaks was clearly exhibited with the change of microdome diameter.
Fig. S14. The preparation of multicolor micro-scale patterns. (A-C) The dark-field micrographs of the printed multicolor patterns (Scale bars, 60 μm).
Fig. S15. The programmable editing of the color microdomes towards color mixing. (A) Schematic illustration of the editing process via droplet-by-droplet printing. (B-D) Dark-field microscopic images of the programmed structure-color pixels including the pure green array (C), green and red array after the second editing (B), and green-red-blue array after the third editing (D). (E) The integrated-pixel arrays composed of various color microdomes (scale bars, 60 μm). Insets are the corresponding macro-images (scale bars, 1 mm). It is clear the printed macro-images can exhibit typical color mixing effects with the editing of microdome pixels.
Fig. S16. CIE chromaticity coordinates of the blend colors. The dashed green and red arrows indicate the continuous evolution of b-colors. The chromaticity values are transformed continuously from I (0.4506, 0.3418), II (0.4230, 0.3369), III (0.3864, 0.3306), IV (0.3209, 0.3076), to V (0.1847, 0.2169).
Fig. S17. The printing resolution of different ink volume with the continuous inkjet printing (DMP-2831, Dimatix Fujifilm). (A) The schematic diagram of the resolution setting in the printer by controlling the printing pitch ($p$). (B) The optical micrographs of the printed microdome array with different pitches. (C) The optical micrographs of the printed microdome array, showing the maximum printing resolution with different ink volume. (D) The summary chart of the maximum printing resolution with different ink volume and pitch.
Fig. S18. The lightness distribution of the printed structural-color palette. Lightness represents the degree to which the light reflected or emitted by an object. The lightness can be converted from sRGB color space, as \( \ell = (\max(R, G, B) + \min(R, G, B))/2 \). In the experiment, the lightness of the structural-color palette is the summation of microdome pixels in the region, which can be regulated by the microdome density. We prepared six structural-color palettes with different densities on one substrate and collected their reflection images. Then we performed the analysis by MATLAB software and calculated the lightness distribution of the different palettes. The lightness distribution from I to VI clearly demonstrates the lightness of the printed palette can be gradually altered by regulating the microdome density.
Fig. S19. The printed various structural-color images. (A) The printed large-area structure-color film. (B) The printed structure-color green QR codes of different size. (C) The printed complex pseudo-homochromatic images (down) from the original digital pictures (up). (D) The direct exhibition of the structure-color images by using a smart phone.
The mapping relationship between image depth and color is mainly controlled by the color pixels composed of three primary color components (Red, Green, and Blue). Each primary color component directly determines the primary color intensity. Grayscale can directly describe the visual feature of the images, which refers to the color depth. When a color image is converted to a grayscale image, the effective brightness value of each pixel in the image needs to be calculated as:

$$I_{\text{gray}} = (0.3 \times R) + (0.59 \times G) + (0.11 \times B)$$

Thus, each printed color pixel in the colorful map corresponds to a fixed gray value.
Fig. S21. The grayscale regulation of the printed structural-color portrait. (A) The designed distorted gray-scale image of Mona Lisa. (B and C) The preparation of distorted image of Mona Lisa, which contains the marked SEM image to show the distribution of printed pixels (B) and distorted digital photograph (C) (scale bars, 60 μm and 5 mm). In the distorted image, the profile pixels are directly printed on a bare substrate. The printed profile together with the black background composes the image. (D) The grayscale distribution histograms of the distorted image. For that the grayscale of profile (II) is higher than the black background (I), the printed image is distorted. (E) The designed realistic gray-scale image of Mona Lisa. (F and G) The experimental realistic image of Mona Lisa, which contains the marked SEM image to show the distribution of printed pixels (F) and realistic digital photograph (G) (scale bars, 60 μm and 5 mm). In the realistic image, the profile pixels are precisely printed on a microdome-based substrate. The printed profile together with the yellow background composes the image. (H) The grayscale distribution histograms of the realistic image. Since the grayscale of profile (III) is lower than the grayscale of microdome-based background (IV), the printed image can become realistic.
Fig. S22. Angle-dependent demonstration of the printed structural-color palette. A, The images of the printed structural-color palette at different incident angles from 30° to 60°. The palette is composed of the printed microdomes with the diameter of about 16.2 μm. B, Angle-resolved reflectance spectra of the printed structural-color palette.

The reflected colors from TIR are controlled by the TIR trajectories and optical path length ($l$) differences that cause the interference. The angle dependence of the color from TIR is similar to the thin film interference, highly depending on the optical path. The optical path length from TIR can be calculated as:

$$l = n_1md \cos\left(\pi/2 - (\pi - \theta_{in} + \theta_{out})/2m\right)$$

Where $n_1$ refers to the refractive index of the microdome, $m$ is the number of total reflections, $d$ is the microdome diameter, $\theta_{in}$ is the incident angle, and $\theta_{out}$ is the observed angle. When the microdome only allows one time total internal reflection ($m = 1$), the optical path length can be simplified to:

$$l = n_1d \cos((\theta_{in} - \theta_{out})/2)$$

In the experiment, we fixed the $\theta_{out}$ ($\theta_{out} = 20^\circ$) and changed the $\theta_{in}$ (from $30^\circ$ to $60^\circ$). The initial interference wavelength was relatively large, which induced the red structural color. Increasing the $\theta_{in}$ would reduce optical path $l$. Thus, blue shift happened with the increase of incident angle.
Descriptions of movies S1 to S2

**Movie S1. The direct coloration of the transparent polymer ink during printing.** The coloration process of printed microdomes is recorded using a custom microscope system. A portable electronic digital microscope integrated with four white LEDs (UM02-B, Vitiny) is employed. The transparent substrate is placed on the upper side of the lens with controlled space. The printer nozzle (Microplotter II, Sonoplot) is precisely positioned on the substrate surface for printing. During printing, the color signal is collected and recorded in real time.

**Movie S2. The programmable structural-color printing via droplet-by-droplet printing.** By the droplet-by-droplet printing, the microdomes with different morphology are prepared. These microdomes can exhibit different colors and serve as the color pixels. We can design the microdome morphology and distribution to achieve the programmable structural-color printing.
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