

Direct-writing Structure Color Patterns on the Electrospun Colloidal Fibers toward Wearable Materials

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 Electronic Supplementary Information

Abstract This study presents a direct-writing structure color patterns on electrospun colloidal fibers by inkjet printing. The colloidal fiber was obtained by electrospinning the aqueous mixture of colloidal particles of poly(styrene-methyl methacrylate-acrylic acid) and poly(vinyl alcohol). The pattern was obtained by directly inkjet printing water onto the electrospun colloidal fiber. The pattern formation was attributed to the morphological transition of the colloidal fiber from the fiber aggregation to latex aggregation and the corresponding color change due to the dissolution of poly(vinyl alcohol) by water. Interestingly, a clear and clean image was successfully obtained on the ethanol-treated colloidal fibers film in comparison to a confused and blur image onto the freshly-made film. It is because the treatment process can compact the fiber structure and lower the spreading/wetting behavior of ink on the fiber structure, contributing to the formation of high-quality pattern. Various letters or quick response code were flexibly designed and printed on to colloidal fibers. Furthermore, the pattern can be easily transferred onto flexible substrate, *i.e.*, a flexible printed bracelet. This work will be of great significance for the development of novel wearable functional materials/devices based on electrospun colloidal fibers.

Keywords Electrospinning; Colloidal fiber; Pattern; Inkjet printing; Structure color

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INTRODUCTION

Colloidal crystals (CCs) have been developed for great potential applications in various optical devices based on the light manipulation properties in the past decades.^[1–16] In particular, patterned CCs constitute an important express way for various optical applications in sensing, displaying,^[2,3,9–11] anticounterfeiting, and optical devices owing to their improved response speed for detecting sensors,^[14,15] especially unique application in anti-counterfeited patterns and optical encryption. Currently, wearable optic materials are of growing research interest for next-generation artificial intelligence equipment,^[17–23] having integrated multiple functionalities, such as high bendability or stretchability, transparency, and many others. A series of research works have been developed to create wearable CC-based optic materials/devices.^[19–23] Combining CCs with fibers altogether has become an important research topic,^[24–35] such as directly electrospinning

colloidal fiber^[24–28] and coating CCs on the surface of fiber or fabric.^[31–42] For example, Sun *et al.*^[36] and Zhang *et al.*^[37] presented a reversible color change by stretching/releasing fiber based on electrophoretically depositing polymer microspheres on continuously aligned carbon nanotube sheet or dip coating the latex particles on commercially available polymer fibers. Electrospun colloidal fiber offers a more direct approach for the fabrication of structure color fiber toward wearable materials.^[24–28] Typically, Yuan *et al.*^[24] and Yuan and Zhang^[25] reported a detailed investigation on the electrospinning process of colloidal suspension (mixture of polymer microspheres and poly(vinyl alcohol) (PVA)), presenting colloidal fiber films with non-iridescent structure color. This work makes it possible to achieve dye-free textile coloration. However, it is greatly expected to develop novel applications of such highly desired structure color fibers. Particularly, few reports refer to directly writing pattern on the electrospun colloidal fiber owing to the prompt wetting and spreading of water on the as-spun fiber,^[30] although some research work is related to the melt electrospinning writing of highly ordered large volume scaffold architectures for tissue engineering and regenerative medicine research.^[43,44] Fabrication of pattern on colloidal fiber will

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greatly extend their functionality and applications toward wearable optic materials/devices. Herein, we demonstrated a direct-writing structure color colloidal patterns on electrospun fiber by inkjet printing. The colloidal fiber was obtained by electrospinning the aqueous mixture of colloidal particles of poly(styrene-methyl methacrylate-acrylic acid) (P(St-MMA-AA)) and PVA. The pattern was obtained by directly inkjet printing water onto the electrospun colloidal fiber. The pattern formation is attributed to the morphological transition of the colloidal fiber from fiber aggregation to latex aggregation and the corresponding color change owing to the dissolution of PVA by water. Interestingly, a clear and clean image was successfully obtained on the ethanol-treated colloidal fiber in comparison to a confused and blur image onto the freshly-made film. It is because of the fact that the treatment process can compact the fiber structure and lower the spreading/wetting rate of the ink on the fiber structure, which contributes to the formation of high-quality pattern. Various letters or two-dimensional code were flexibly designed and printed onto colloidal fibers. Furthermore, the pattern can be easily transferred onto a flexible substrate, *i.e.*, a flexible printed bracelet. The work will be of great significance for the development of novel wearable functional materials/devices based on electrospun colloidal fibers.

RESULTS AND DISCUSSION

Fig. 1(a) schemes the fabrication process of colloidal fiber

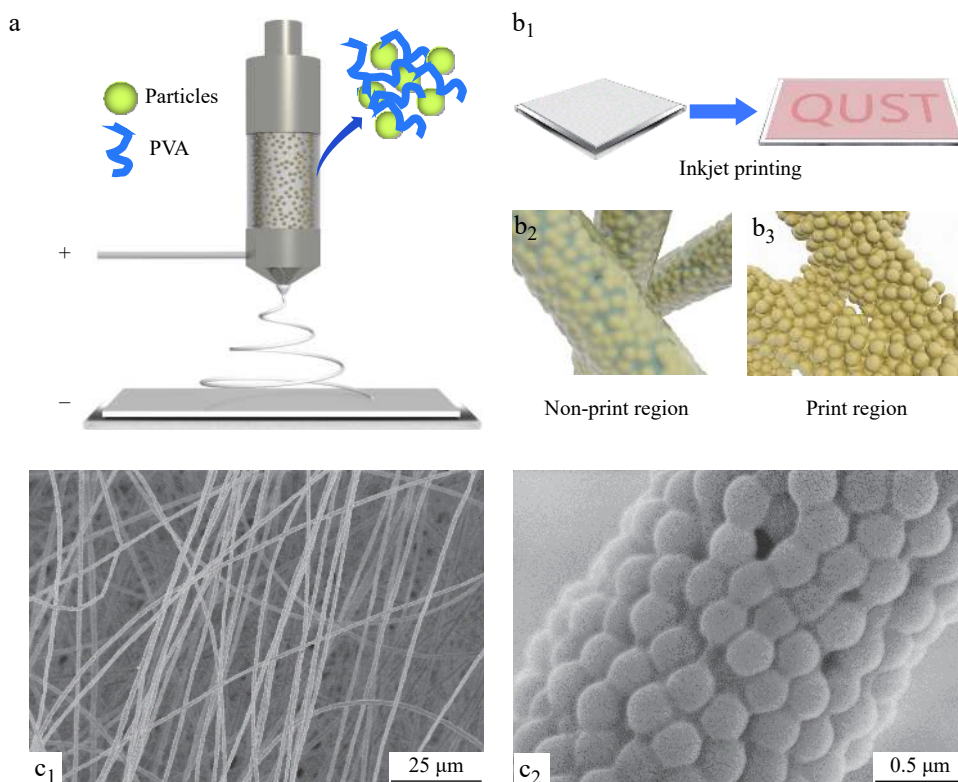


Fig. 1 Schematic illustrations of (a) electrospun set-up of colloidal fiber, and (b₁) CC pattern fabricated by inkjet printing on electrospun fibers. The pattern formation is mainly owing to the morphological transition from fiber aggregation (b₂, non-print region) to latex aggregation (b₃, print-region) and structure color change arises from water dissolution of PVA. SEM images of the as-prepared electrospun colloidal fiber: (c₁) the large-area colloidal fiber and (c₂) the magnified image of the single colloidal fiber. The latex particles are well orderly arranged in the fiber.

from electrospinning approach. It is carried out by filling the aqueous dispersion of P(St-MMA-AA) latex particles (40 wt%) and PVA (15 wt%) with weight ratio of 4:1 into a syringe, and the electrospun fiber is obtained by imposing a positive voltage of 10 kV and a negative voltage of 4.5 kV. The soft PVA acts as binder to adhere colloidal spheres.^[24–28] The wrapped colloidal spheres are packed and stretched into the fibers. The locally hexagonal packing of latex particles appears on the surface of the colloidal fibers. As further confirmed by the SEM images in Fig. 1(c₂) (Figs. S1 and S2, in electronic supplementary information, ESI), the well-ordered latex assembly produces iridescent structure color. Fig. 1(b₁) schemes the fabrication process of the CC pattern from inkjet printing on the electrospun film; the designed colorful pattern can be obtained by printing H₂O onto the colloidal fiber film. Pattern formation is mainly based on the morphological transition from fiber aggregation (non-print region, Fig. 1b₂) to colloidal aggregation (print region, Fig. 1b₃) and the corresponding color change is owing to the dissolution of PVA by water.

To clearly understand the structure color of the electrospun fiber, we investigated the single colloidal fiber by optical microscopy images as shown in Fig. 2(a). An iridescent structure color is observed from the single colloidal fibers. The red, green, and blue single colloidal fibers are assembled from latex particles with diameter of 265, 210, and 180 nm, respectively, as seen in Figs. 2(a₁)–2(a₃). The struc-

ture color of single fiber can be detected from the reflection spectra in Fig. 2(b), which shows multi-stopband characteristic for single fiber assembled from the monodispersed latex particles. These multi-stopbands are clearly confirmed from multi-color domains in the optical microscopy image (Fig. 2a) and multi-assembly structure in SEM images (Fig. S1 in ESI). Typically, the stopbands of red, green, and blue single colloidal fibers locate at 674 (465), 549 (721), and 471 (611) nm, that is, assembling from latex particles of 265, 210, and 180 nm, respectively. The main stopbands of 674, 549, and 471 nm are attributed to Bragg diffraction effect of the well-ordered latex assembly, and the values are close to the theoretical stopbands of 637, 505, and 432 nm calculated from Bragg equation (Table S1 in ESI). The difference between theoretical and experimental value is owing to the disordered latex assembly that arose from the electrospun process.^[24–28] Other spectra can be attributed to Mie scattering of either latex particles or fiber-like assembly.^[24,41,42,45–52] In our case, peak at 465 nm is ascribed to the Mie scattering of latex particles for red colloidal fiber, while peaks at 721 and 611 nm are due to the Mie scattering of fiber-like assembly for green fiber and blue fiber, respectively. Owing to the an-

isotropy of the fiber-like assembly, the corresponding Mie scattering is polarized-dependent (Fig. S3 in ESI). For instance, the reflection intensity at 611 nm changes from weak to strong when changing the polarized angle from 0° to 45°. The distinct structure color of single- or thin-layer electrospun colloidal fibers (Fig. 2c) is mainly originated from the selective reflection (Figs. 2d₂ and 2d₃) based on Bragg diffraction effect of the well-ordered assembly structure,^[53–55] similar to conventional CCs in Fig. 2(d₁). The structure colors of colloidal fibers can be attributed to the photonic band gap effect related to coherence scattering by partial ordered nanosphere arrangement and the resonant Mie scattering related to incoherence scattering by numerous disordered nanosphere scatters.^[41,42,45–52]

Although the single- or thin-layer colloidal fibers present structure color (Figs. 2a and 2c), the freshly-made thick colloidal fiber film is relatively fluffy with an increased distance among the fiber layers and appears white without any structure color (Fig. S2 in ESI). That is, a disordered aggregation of colloidal fibers introduces more interface and produces scattering/interference effect,^[49–52] which greatly degrades the selective reflection of the well-ordered assembly

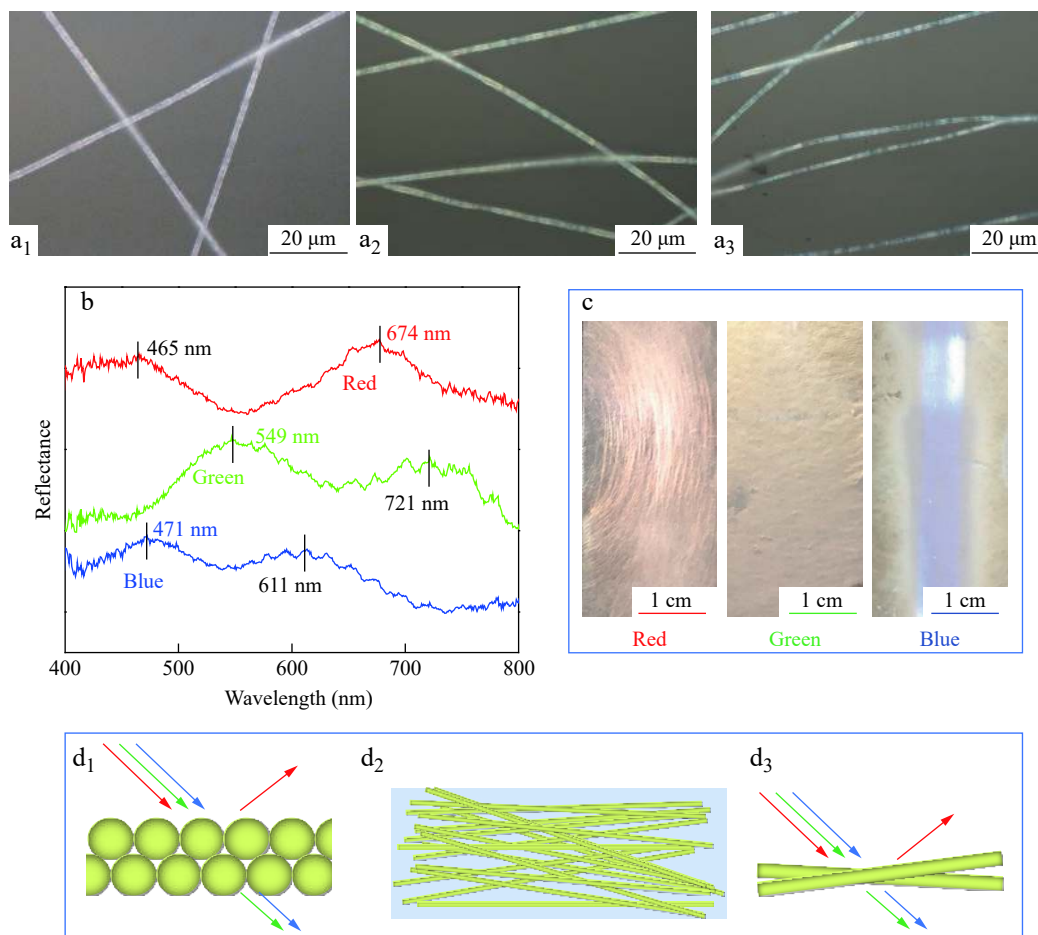


Fig. 2 Optical microscopy images of the electrospun single colloidal fiber from latex particles with diameter of (a₁) 265, (a₂) 210, and (a₃) 180 nm, and (b) the corresponding reflection spectra. There exist multi-stopbands in the spectra, indicating the combination of stopband and the Mie diffraction of latex particles and fiber structure. (c) Photos of thin-layer colloidal fiber film assembled from latex particles of 265, 210, and 180 nm showing iridescent structure color; Schemes of the selective reflection of (d₁) the conventional CCs, (d₂) the single-, and (d₃) thin layer colloidal fiber

structure, leading to the disappearance of structure color of the thick film (Fig. 3a₁). It was found that the suitable solvent treatment of the colloidal fiber generates a dense fiber structure, decreasing the interface and improving the selective reflection of the film (Figs. 3a₂ and 3a₃). The 1st-treated film refers to that the freshly-made multilayer fiber membrane is immersed in alcohol for 1 s, and then dried with cold air. The resultant sample presents a primary treatment color because the treatment process could effectively remove the air interface among fibers and decrease the scattering effect. The 2nd-treated film means that the 1st-treated sample is immersed into water for 2 min and dried with cold air. The process can compact fiber structure by removing PVA, contributing to a great improvement of the optical properties^[24] (Fig. 3a₃). It should be mentioned that directly treating the freshly-made fiber by water would make the film twisted and broken, which is infeasible for the further use. Fig. 3(c) presents the reflection spectra of as-prepared multilayer colloidal fibers that experienced the 1st- or the 2nd-treatment, respectively. The freshly-made colloidal fiber is white without any characteristic peak in the spectrum (black curve in Fig. 3c). In contrast, a slight red color is discerned and a charac-

teristic peak appears at 637 nm (pink curve Fig. 3c) for the sample that experienced the 1st-treatment. Subsequently, an obvious (red) color enhancement occurs; the intensity of the peak at 619 nm rises and its FWHM narrows (red curve in Fig. 3c) for sample which experienced the 2nd-treatment. The reflection peak intensifies gradually for the sample that experienced the 1st- and 2nd-treatment. As seen from SEM images (Fig. 3b, Figs. S4 and S5 in ESI), an intact colloidal fiber has crossed arrangement with more interfaces for the freshly-made multi-layer fiber (Fig. 3b₁). The fiber structure is twisted and deformed, forming fiber bundle after the 1st-treatment (Fig. 3b₂, Fig. S4 in ESI), which indicates a great reduction of the fiber interface and an obvious improvement of selective reflection.

The dissolution of PVA and the fusion of fiber occurred for sample which experienced 2nd-treatment process (Fig. 3b₃, Fig. S5 in ESI) led to a collapsed fiber structure and separated latex particles. This could greatly decrease the fiber interface and improve the optic properties of the film as shown in Fig. 3(c). Fig. S6 (in ESI) shows the images of the large-scale colorful electrospun film which experienced 2nd-treatment (Fig. 4d). The films present a clear non-iridescent

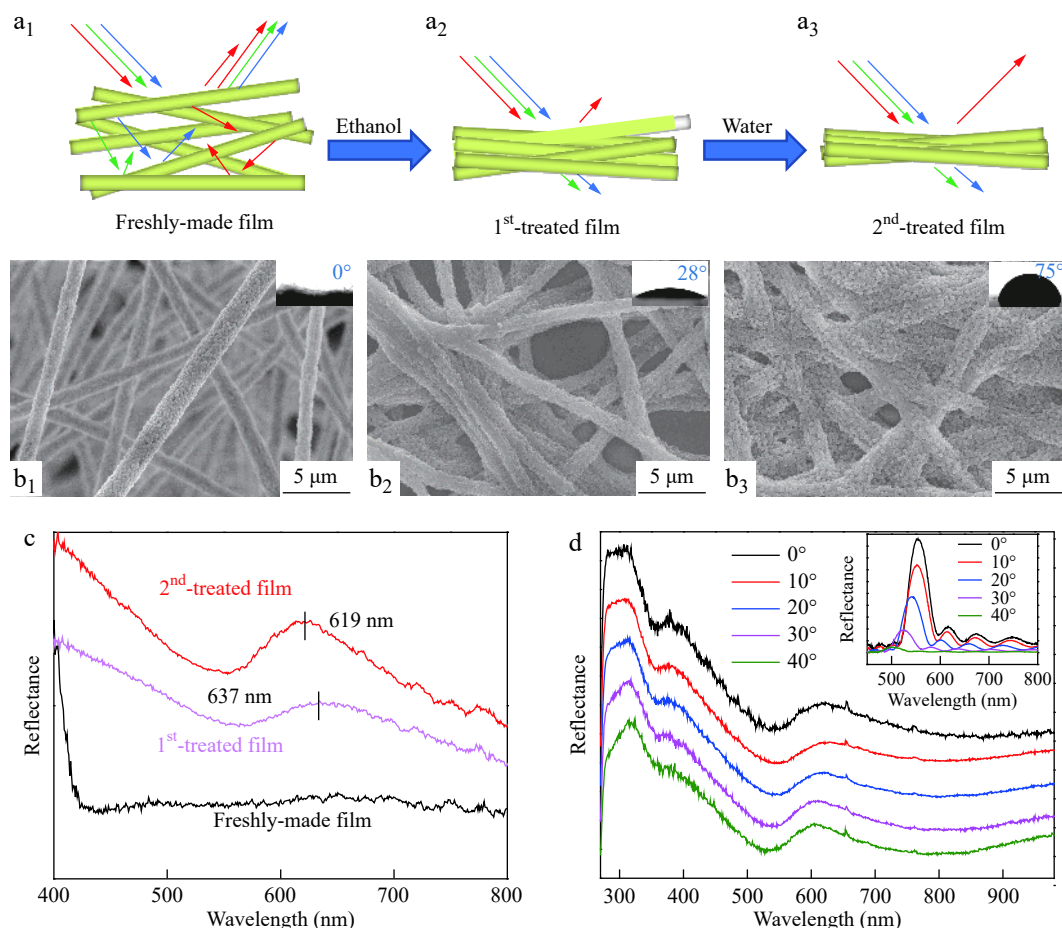


Fig. 3 Schematic illustration of (a₁) the decreased selective reflection of thick-layer colloidal fiber owing to an increased scattering and interference effect among fibers and (a₂, a₃) the reproduction of structure color of the sample which experienced (a₂) 1st-treatment and (a₃) 2nd-treatment; SEM images of (b₁) freshly-made electrospun colloidal fiber and sample which experienced (b₂) 1st-treatment or (b₃) 2nd-treatment (The insets are the photos of water droplet on the as-prepared colloidal fiber.) and (c) the corresponding reflection spectra. The structure color/optic signal is enhanced for samples which experienced 1st- and 2nd-treatment. (d) Red colloidal fiber which experienced 2nd-treatment (inserted flat CCs) with varying observing angle from 0° to 40°

structure color,^[49–52] as is proved from varying-angle reflection spectra in Fig. 3(d) and Fig. S8 (in ESI). Evidently, little change can be observed for the stopband position of as-fabricated colloidal fiber when varying the observing angle from 0° to 40°, which is very different from the dramatic change of stopband for the traditional flat CCs when varying the observing angle (inset in Fig. 3d). This non-iridescent structure color is particularly useful for the potential applications in display devices.^[46,49–55]

Importantly, the structure color pattern is easily obtained by inkjet printing H₂O onto the colloidal fiber. Fig. 4(a₁) presents the red pattern of “QUST” fabricated from inkjet printing H₂O ink onto the 1st-treated electrospun film, where a color enhancement appears upon ink touching, forming a designed pattern. The printing mechanism is mainly based on the distinct spectral signal/morphology for the sample which experienced the 1st-treatment and 2nd-treatment in Figs. 3(b) and 3(c). Green, blue, and red ‘QUST’ patterns are clearly printed on the colloidal fibers which experienced the 1st-treatment (Fig. S9 in ESI). There appears a notably clean and smooth print brim for the pattern. It is the first report of printing structure color pattern on electrospun fiber. To further understand the printing characteristic, we investigate the print (pattern) and non-print (non-pattern) region by optical microscopic images (Fig. 4b₁ and Fig. S10 in ESI) and SEM images (Figs. 4b₂–4b₄, Fig. S11 in ESI). Compared with non-print region, the print region is brighter with a narrower

reflection spectra (Fig. 4c) owing to the close-packed latex particles and the disappearance of fiber interface (Fig. 4b₄). Interestingly, an obvious boundary is observed between the print and non-print region in Fig. 4(b). It can be attributed to a washout and accumulation of PVA over the boundary as indicated by SEM images in Fig. 4(b₂). The boundary is formed owing to the coffee-ring effect during the washout process. It is also confirmed by the change of the chemical composition between print- and non-print regions (FTIR spectra, Fig. S12 in ESI), as the intensity of characteristic band at 3350 cm⁻¹ (assigned to OH group of PVA) decreases for the sample which experienced the 1st- and 2nd-treatment, indicating the removal of PVA from the fiber system during solvent treatment process. To investigate the printability of the system, a complex pattern (such as a quick response (QR) code, Fig. 4a₂) was printed on blue colloidal fiber. The QR code is clearly identified when being scanned by some device like a smartphone. Meanwhile, a high-resolution pattern (‘PC’, Fig. 4a₃) is also printed on a blue colloidal fiber film which experienced the 1st-treatment as well. Interestingly, the electrospun colloidal films can be easily transferred onto any substrate. For example, the film is transferred onto flexible substrate to fabricate a flexible bracelet with green color, and the specific character can be printed on the flexible bracelet in Fig. 4(e). Creating various patterns on electrospun colloidal fibers is of great importance for the development of novel wearable optical materials/devices based

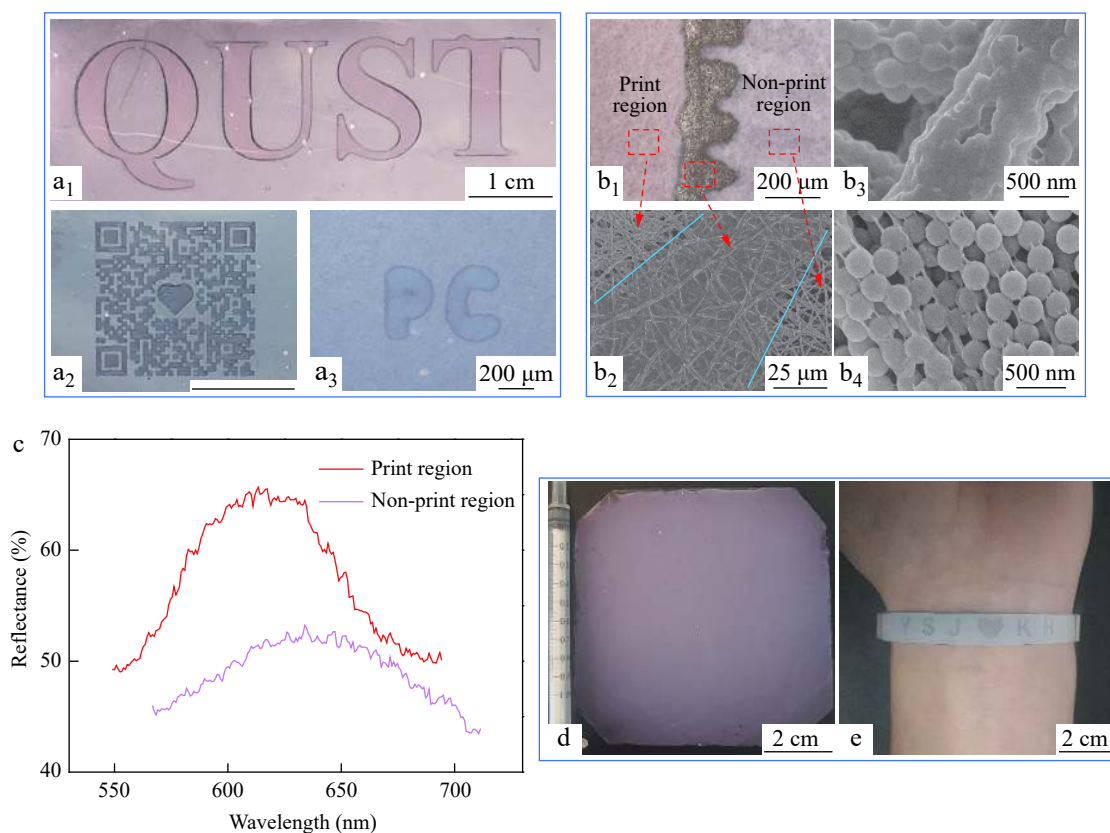


Fig. 4 Patterns of (a₁) ‘QUST’ on red colloidal fibers which experienced 1st-treatment, (a₂) a QR code (The scale bar is 1 cm), and (a₃) ‘PC’ on blue substrate which experienced 1st-treatment; (b₁) Optical microscopic images, (b₂, b₃, b₄) SEM images, and (c) reflection spectra of the boundary between non-printing region (b₃) and print region (b₄); Photos of (d) electrospun film that experienced 2nd-treated film and (e) printed bracelet from electrospun colloidal fiber film

on electrospun colloidal fiber films. This work is promising for wide applications in light-weight, flexible, wearable display materials/devices.

In fact, the coffee-ring effect is one of the important factors influencing the quality of printing pattern. Modifying the coffee-ring effect can greatly improve the printing resolution of pattern fabricated on electrospun fibers. The wettability of colloidal fiber plays an important role in the coffee-ring effect. Taking the freshly-made colloidal film (with water contact angle (CA) of 0°) and 1st-treated colloidal fiber (with water CA of 28°) as examples, we investigated *in situ* wetting of droplet spreading onto the above films and the corresponding printing process in Fig. 5. Fig. 5(a) schemes the distinct printing process of a droplet on the freshly-made film and 1st-treated film. There appears a dramatic “coffee-ring cavity” on the freshly-made film (Figs. 5a₂ and 5a₃, Fig. S13, Movie S1 in ESI) when droplet is spreading onto it, leaving little fiber/particles at the center of the ring (Figs. 5a₃ and 5b₁, Figs. S13b and S14c in ESI) but plenty of fiber and particles around the cavity (Figs. 5a₃ and 5b₂, Figs. S13a, S14a, and S14b in ESI). In comparison, a smooth coffee-ring print region has been left on the 1st-treated film (Figs. 5a₂' and 5a₃', Movie S2 in ESI) under similar droplet spreading condition. Little morphological change can be observed for the sample between brim and central part of the round print-

ing region (Figs. 5b₁' and 5b₂', Figs. S15 and S16 in ESI). The above-mentioned dramatic change of spreading condition can be attributed to the distinct wettability of droplet on the two fiber structures. Specifically, the water droplet easily wets, spreads and rapidly immerses into the freshly-made film (Figs. 5c₁–5c₃, Movie S3 in ESI) in 0.25 s; the rapid wetting/spreading process disrupts the hydrophilic and loose fiber structure of freshly-made film, forming a large coffee-ring cavity after water evaporation. All of these contribute to a confused and blur pattern printed on freshly-made fiber film as shown in Fig. 5(d). In comparison, the water droplet spreads on the dense treated film (with less hydrophilicity) in a slower way, leaving certain amount of water over the surface (Figs. 5c₁'–5c₃', Movie S4 in ESI) in 0.55 s. This process removes PVA without destroying the structure of the film (Figs. 5a₂', 5a₃', 5b₁', 5b₂', Figs. S15 and S16, Movie S2 in ESI), retaining just a clear and clean printing region. The process is beneficial to the improved printing resolution, and thus favors the fine printed pattern in Fig. 5(d').

CONCLUSIONS

In conclusion, this work presents a direct-writing structure color colloidal pattern on electrospun colloidal fibers by inkjet printing. The pattern formation is attributed to the

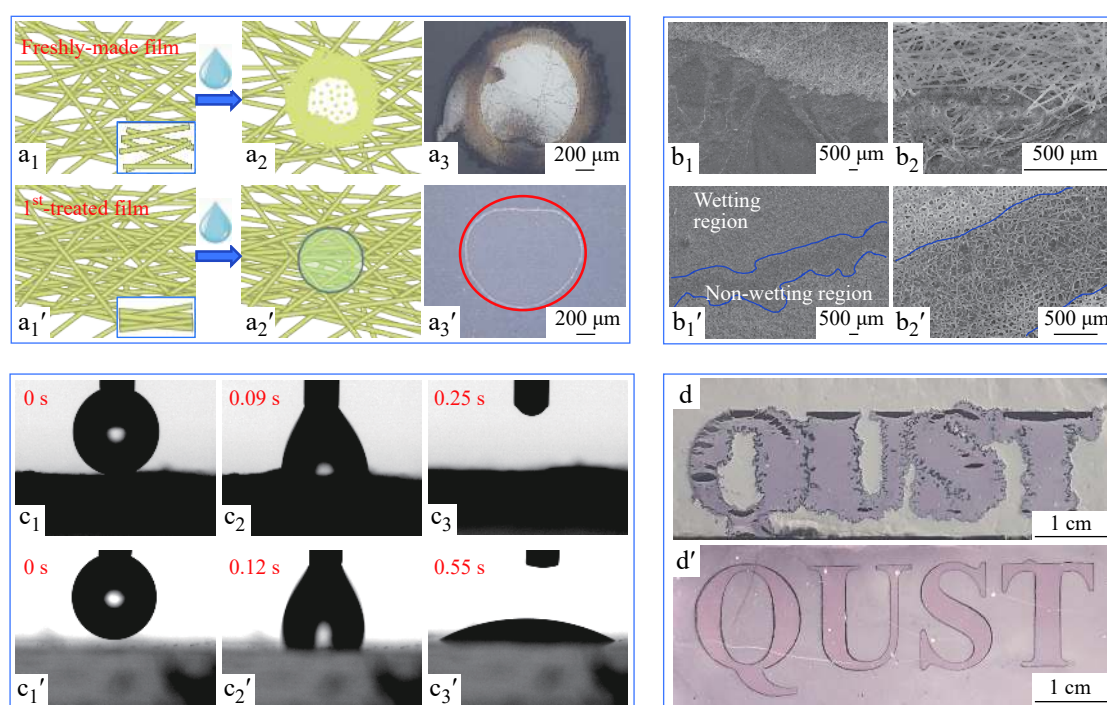


Fig. 5 Schematic illustration of the distinct water print process on (a₁, a₂) freshly-made film and (a₁', a₂') the 1st-treated film; Optical microscopy images of the water droplet on the (a₃) freshly-made film and (a₃') the 1st-treated film. A cavity structure is formed on the fresh-made film when the water droplet (0.25 µL) spreads upon it; in contrast, only a smooth print brim is left on the 1st-treated film for the same water droplet on it; SEM images of the water droplet wetting region on (b₁, b₂) freshly-made film and (b₁', b₂') the 1st-treated film. A dramatic structure collapse occurs to the freshly-made film for water droplet spreading upon it, while a slight printing brim is left on the 1st-treated film. Photos of dynamic wetting process of the water droplet (1 µL) on (c₁–c₃) the freshly-made film and (c₁'–c₃') the 1st-treated film. A rapid wetting/spreading process occurs for water droplet on the freshly-made film, while a slow spreading occurs to the droplet on the 1st-treated film; Photos of printed film on (d) freshly-made colloidal fiber film and (d') the 1st-treated film. A confused and blur image appears on the fresh-made film, while an image with smooth and fine structure shows up on the 1st-treated film.

morphological transition of colloidal fiber from the fiber aggregation to latex aggregation and the corresponding color change is owing to the dissolution of PVA by water. It was found that a confused and blur image is obtained when directly printing H₂O ink onto the freshly-made film, while a clear and clean image is successfully printed onto colloidal fiber treated with ethanol. The treated process can compact the fiber structure and lower the spreading and wetting behavior of the ink on the fiber structure, thereby contributing to the formation of high-quality pattern. Various patterns including letters and QR code are desirably printed on the electrospun fiber film. And the pattern is easily transferred onto flexible substrate, such as a flexible printed bracelet. This work will be of great significance for the development of novel wearable materials/devices based on electrospun colloidal fiber film.

Electronic Supplementary Information

Electronic supplementary information (ESI) is available free of charge in the online version of this article at <http://dx.doi.org/10.1007/s10118-019-2286-0>.

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REFERENCES

- Shiue, R. J.; Gao, Y. D.; Tan, C.; Peng, C.; Zheng, J. B.; Efetov, D. K.; Kim, Y. D.; Hone, J.; Englund, D. Thermal radiation control from hot graphene electrons coupled to a photonic crystal nanocavity. *Nat. Commun.* **2019**, *10*, 109.
- Fu, F. F.; Shang, L. R.; Chen, Z. Y.; Yu, Y. R.; Zhao, Y. J. Bioinspired living structural color hydrogels. *Sci. Robot.* **2018**, *3*, eaar8580.
- Zhang, D. J.; Liu, J.; Chen, B.; Zhao, Y.; Wang, J. X.; Ikeda, T.; Jiang, L. A hydrophilic/hydrophobic Janus inverse-opal actuator *via* gradient infiltration. *ACS Nano* **2018**, *12*, 12149–12158.
- Wang, H.; Zhao, Z.; Liu, Y. X.; Shao, C. M.; Bian, F.; Zhao, Y. J. Biomimetic enzyme cascade reaction system in microfluidic electrospun microcapsules. *Sci. Adv.* **2018**, *4*, eaat2816.
- Xu, Y. S.; Wang, H.; Luan, C. X.; Fu, F. F.; Chen, B. A.; Liu, H.; Zhao, Y. J. Porous hydrogel encapsulated photonic barcodes for multiplex microRNA quantification. *Adv. Funct. Mater.* **2018**, *28*, 1704458.
- Zhu, B. T.; Fu, Q. Q.; Chen, K.; Ge, J. P. Liquid photonic crystals for mesopore detection. *Angew. Chem. Int. Ed.* **2018**, *57*, 252–256.
- Zhong, K.; Li, J. Q.; Liu, L. W.; Cleuvenbergen, S. V.; Song, K.; Clays, K. Instantaneous, simple, and reversible revealing of invisible patterns encrypted in robust hollow sphere colloidal photonic crystals. *Adv. Mater.* **2018**, *30*, 1707246.
- Du, X.; Wang, M.; Welle, A.; Sadabad, F. B.; Wang, Y. L.; Levkin, P. A.; Gu, Z. Z. Reparable superhydrophobic surface with hidden reactivity, its photofunctionalization and photopatterning. *Adv. Funct. Mater.* **2018**, *28*, 1803765.
- Wu, L. L.; Wang, X.; Wang, G.; Chen, G. *In situ* X-ray scattering observation of two dimensional interfacial colloidal crystallization. *Nat. Commun.* **2018**, *9*, 1335.
- Guo, D.; Zheng, X.; Wang, X. H.; Li, H. Z.; Li, K. X.; Li, Z.; Song, Y. L. Formation of multicomponent sizesorted assembly patterns by tunable templated dewetting. *Angew. Chem. Int. Ed.* **2018**, *57*, 16126–1613.
- Zhang, B.; Meng, F. S.; Feng, J. G.; Wang, J. X.; Wu, Y. C.; Jiang, L. Manipulation of colloidal particles in three dimensions *via* microfluid engineering. *Adv. Mater.* **2018**, *30*, 1707291.
- Liu, X. J.; Gu, H. C.; Wang, M.; Du, X.; Gao, B. B.; Elbaz, A.; Sun, L. D.; Liao, J. L.; Xiao, P. F.; Gu, Z. Z. 3D printing of bioinspired liquid superrepellent structures. *Adv. Mater.* **2018**, *3*, 1800103.
- Zhang, J.; Zhu, Z. J.; Yu, Z. Y.; Ling, L. T.; Wang, C. F.; Chen, S. Large-scale colloidal films with robust structural colors. *Mater. Horiz.* **2019**, *6*, 90–96.
- Hou, J.; Li, M. Z.; Song, Y. L. Patterned colloidal photonic crystals. *Angew. Chem. Int. Ed.* **2017**, *130*, 2544.
- Li, Y. N.; Zhou, X.; Yang, Q.; Li, Y. D.; Li, W. B.; Li, H. Z.; Chen, S. R.; Li, M. Z.; Song, Y. L. Patterned photonic crystals for hiding information. *J. Mater. Chem. C* **2017**, *5*, 4621.
- Zhou, J. M.; Han, P.; Liu, M. J.; Zhou, H. Y.; Zhang, Y. X.; Jiang, J. K.; Liu, P.; Wei, Y.; Song, Y. L.; Yao, X. Self-healable organogel nanocomposite with angle-independent structural colors. *Angew. Chem. Int. Ed.* **2017**, *129*, 10598.
- Boto, E.; Holmes, N.; Roberts, J. G.; Shah, V.; Meyer, S. S.; Muñoz, L. D.; Mullinger, K. J.; Tierney, T. M.; Bestmann, S.; Barnes, G. R.; Bowtell, R.; Brookes, M. J. Moving magnetoencephalography towards real-world applications with a wearable system. *Nature* **2018**, *555*, 657–661.
- Markvicka, E. J.; Bartlett, M. D.; Huang, X. N.; Majidi, C. An autonomously electrically self-healing liquid metal-elastomer composite for robust soft-matter robotics and electronics. *Nat. Mater.* **2018**, *17*, 618–624.
- Qian, X.; Cai, Z. R.; Su, M.; Li, F. Y.; Fang, W.; Li, Y. D.; Zhou, X.; Li, Q. Y.; Feng, X. Q.; Li, W. B.; Hu, X. T.; Wang, X. D.; Pan, C. F.; Song, Y. L. Printable skin-Driven mechanoluminescence devices *via* nanodoped matrix modification. *Adv. Mater.* **2018**, *30*, 1800291.
- Chortos, A.; Liu, J.; Bao, Z. Pursuing prosthetic electronic skin. *Nat. Mater.* **2016**, *15*, 937.
- Gao, B. B.; Elbaz, A.; He, Z. Z.; Xie, Z. Y.; Xu, H.; Liu, S. Q.; Su, E.; Liu, H.; Gu, Z. Z. Bioinspired kirigami fish-based highly stretched wearable biosensor for human biochemical-Physiological hybrid monitoring. *Adv. Mater. Technol.* **2018**, *3*, 1700308.
- He, Z. Z.; Elbaz, A.; Gao, B. B.; Zhang, J. N.; Su, E.; Gu, Z. Z. Wearable biosensors: Disposable morpho menelaus based flexible microfluidic and electronic sensor for the diagnosis of neurodegenerative disease. *Adv. Healthc. Mater.* **2018**, *7*, 1701306.
- Zhang, Z. T.; Cui, L. Y.; Shi, X.; Tian, X. C.; Wang, D. P.; Gu, C. N.; Chen, E.; Cheng, X. L.; Xu, Y. F.; Hu, Y. J.; Zhang, J. Y.; Zhou, L.; Fong, H. H.; Ma, P. B.; Jiang, G. M.; Sun, X. M.; Zhang, B.; Peng, H. S. Textile display for electronic and brain-Interfaced communications. *Adv. Mater.* **2018**, *30*, 1800323.
- Yuan, W.; Zhou, N.; Shi, L.; Zhang, K. Q. Structural coloration of colloidal fiber by photonic band gap and resonant Mie scattering. *ACS Appl. Mater. Interfaces* **2015**, *7*, 14064–14071.
- Yuan, W.; Zhang, K. Q. Structural evolution of electrospun composite fibers from the blend of polyvinyl alcohol and polymer nanoparticles. *Langmuir* **2012**, *28*, 15418–15424.
- Lim, J. M.; Moon, J. H.; Yi, G. R.; Heo, C. J.; Yang, S. M. Fab-

- rication of one-dimensional colloidal assemblies from electrospun nanofibers. *Langmuir* **2006**, *22*, 3445–3449.
- 27 Lim, J. M.; Yi, G. R.; Moon, J. H.; Heo, C. J.; Yang, S. M. Superhydrophobic films of electrospun fibers with multiple-scale surface morphology. *Langmuir* **2007**, *23*, 7981–7989.
- 28 Moon, J. H.; Kim, S.; Yi, G. R.; Lee, Y. H.; Yang, S. M. Fabrication of ordered macroporous cylinders by colloidal templating in microcapillaries. *Langmuir* **2004**, *20*, 2033–2035.
- 29 Hou, L. L.; Wang, N.; Wu, J.; Cui, Z. M.; Jiang, L.; Zhao, Y. Bioinspired superwettability electrospun micro/nanofibers and their applications. *Adv. Funct. Mater.* **2018**, *28*, 1801114.
- 30 Zheng, S.; Du, M.; Miao, W. N.; Wang, D. Y.; Zhu, Z. P.; Tian, Y.; Jiang, L. 2D prior spreading inspired from Chinese Xuan papers. *Adv. Funct. Mater.* **2018**, 1800832.
- 31 Zhou, N.; Zhang, A.; Shi, L.; Zhang, K. Q. Fabrication of structurally-colored fibers with axial core-shell structure via electrophoretic deposition and their optical properties. *ACS Macro Lett.* **2013**, *2*, 116–120.
- 32 Huang, J. Y.; Li, S. H.; Ge, M. Z.; Wang, L. N.; Xing, T. L.; Chen, G. Q.; Liu, X. F.; Al-Deyab, S. S.; Zhang, K. Q.; Chen, T.; Lai, Y. K. Robust superhydrophobic TiO₂@fabrics for UV shielding, self-cleaning and oil-water separation. *J. Mater. Chem. A* **2015**, *3*, 2825–2832.
- 33 Ling, Z. W.; Liu, K.; Zou, Q.; Li, Q.; Zhang, K. Q.; Cui, Z.; Yuan, W.; Liu, Y. Q. Continuous and rapid fabrication of photochromic fibers by facile coating tungsten oxide/polyvinyl alcohol composites. *RSC Adv.* **2018**, *8*, 28581–28587.
- 34 Zeng, Q.; Ding, C.; Li, Q. S.; Yuan, W.; Peng, Y.; Hu, J. C.; Zhang, K. Q. Rapid fabrication of robust, washable, self-healing superhydrophobic fabrics with non-iridescent structural color by facile spray coating. *RSC Adv.* **2017**, *7*, 8443–8452.
- 35 Liu, Z. F.; Zhang, Q. H.; Wang, H. Z.; Li, Y. G. Structurally colored carbon fibers with controlled optical properties prepared by a fast and continuous electrophoretic deposition method. *Nanoscale* **2013**, *5*, 6917–6922.
- 36 Sun, X. M.; Zhang, J.; Lu, X.; Fang, X.; Peng, H. S. Mechanochromic photonic-crystal fibers based on continuous sheets of aligned carbon nanotubes. *Angew. Chem. Int. Ed.* **2015**, *54*, 3630–3634.
- 37 Zhang, J.; He, S. S.; Liu, L. M.; Guan, G.; Lu, Z. X.; Sun, X. M.; Peng, H. S. The continuous fabrication of mechanochromic fibers. *J. Mater. Chem. C* **2016**, *4*, 2127–2133.
- 38 Li, K. R.; Zhang, Q. H.; Wang, H. Z.; Li, Y. G. Red, green, blue (RGB) electrochromic fibers for the new smart color change fabrics. *ACS Appl. Mater. Interfaces* **2014**, *6*, 13043–13050.
- 39 Liu, Z. F.; Zhang, Q. H.; Wang, H. Z.; Li, Y. G. Magnetic field induced formation of visually structural colored fiber in micro-space. *J. Colloid. Inter. Sci.* **2013**, *406*, 18–23.
- 40 Liu, Z. F.; Zhang, Q. H.; Wang, H. Z.; Li, Y. G. Structural colored fiber fabricated by a facile colloid self-assembly method in micro-space. *Chem. Commun.* **2011**, *47*, 12801–12803.
- 41 Kolle, M.; Lethbridge, A.; Kreysing, M.; Baumberg, J. J.; Aizenberg, J.; Vukusic, P. Bio-inspired band-gap tunable elastic optical multilayer fibers. *Adv. Mater.* **2013**, *25*, 2239–2245.
- 42 Finlayson, C. E.; Goddard, C.; Papachristodoulou, E.; Snoswell, D. R. E.; Kontogeorgos, A.; Spahn, P.; Hellmann, G. P.; Hess, O.; Baumberg, J. J. Ordering in stretch-tunable polymeric opal fibers. *Opt. Express* **2011**, *19*, 3144–3154.
- 43 Wunner, F. M.; Wille, M.L.; Noonan, T. G.; Bas, O. D.; De-Juan-Pardo, P. D.; Hutmacher, D. W. Melt electrospinning writing of highly ordered large volume scaffold architectures. *Adv. Mater.* **2018**, *30*, 1706570.
- 44 Chen H. L.; Malheiro, A. D. B. F. B.; van Blitterswijk, C.; Mota, C.; Wieringa, P. A.; Moroni, L. Direct writing electrospinning of scaffolds with multidimensional fiber architecture for hierarchical tissue engineering. *ACS Appl. Mater. Interfaces* **2017**, *9*, 38187–38200.
- 45 Zhao, Z.; Wang, H.; Shang, L. R.; Yu, Y. R.; Fu, F. F.; Zhao, Y. J.; Gu, Z. Z. Bioinspired heterogeneous structural color stripes from capillaries. *Adv. Mater.* **2017**, *29*, 1704569.
- 46 Li, Q. S.; Zhang, Y. F.; Shi, L.; Qiu, H. H.; Zhang, S. M.; Qi, N.; Hu, J. C.; Yuan, W.; Zhang, X. H.; Zhang, K. Q. Additive mixing and conformal coating of noniridescent structural colors with robust mechanical properties fabricated by atomization deposition. *ACS Nano* **2018**, *12*, 3095–3102.
- 47 Dong, B. Q.; Liu, X. H.; Zhan, T. R.; Jiang, L. P.; Yin, H. W.; Liu, F.; Zi, J. Structural coloration and photonic pseudogap in natural random close-packing photonic structures. *Opt. Express* **2010**, *18*, 14430–14438.
- 48 Saranathan, V.; Forster, J. D.; Noh, H.; Liew, S. F.; Mochrie, S. G. J.; Cao, H.; Dufresne, E. R.; Prum, R. O. Structure and optical function of amorphous photonic nanostructures from avian feather barbs: A comparative small angle X-ray scattering (SAXS) analysis of 230 bird species. *J. R. Soc. Interfaces* **2012**, *9*, 2563–2580.
- 49 Adrian, D.; Thomas, W.; In, E. Y. A. *Light Scattering by Systems of Particles*, Springer: Berlin **2006**, *2*, pp. 99–102.
- 50 Sapienza, R.; García, P. D.; Bertolotti, J.; Martín, M. D.; Blanco, Á.; Viña, L.; López, C.; Wiersma, D. S. Observation of resonant behavior in the energy velocity of diffused light. *Phys. Rev. Lett.* **2007**, *99*, 233902.
- 51 Garcia, P. D.; Sapienza, R.; Pez, C. L. Photonic glasses: A step beyond white paint. *Adv. Mater.* **2010**, *22*, 12–19.
- 52 Takeoka, Y.; Yoshioka, S.; Takano, A.; Arai, S.; Nueangnoraj, K.; Nishihara, H.; Teshima, M.; Ohtsuka, Y.; Seki, T. Production of colored pigments with amorphous arrays of black and white colloidal particles. *Angew. Chem. Int. Ed.* **2013**, *125*, 7402–7406.
- 53 Zhang, Y. F.; Dong, B. Q.; Chen, A.; Liu, X. H.; Shi, L.; Zi, J. Using cuttlefish ink as an additive to produce non-iridescent structural colors of high color visibility. *Adv. Mater.* **2015**, *27*, 4719–4724.
- 54 Kim, Y. H.; Heo, J. S.; Kim, T. H.; Park, S.; Yoon, M. H.; Kim, J.; Oh, M. S.; Yi, G. R.; Noh, Y. Y.; Park, S. K. Flexible metal-oxide devices made by room-temperature photochemical activation of sol-gel films. *Nature* **2012**, *489*, 128–132.
- 55 Wang, J. X.; Wen, Y. Q.; Ge, H. L.; Sun, Z. W.; Zheng, Y. M.; Song, Y. L.; Jiang, L. Simple fabrication of full color colloidal crystal films with tough mechanical strength. *Macromol. Chem. Phys.* **2006**, *207*, 596–604.