Supporting Information

Biomimetic Structural Color Films with a Bilayer Inverse Heterostructure for Anticounterfeiting Applications

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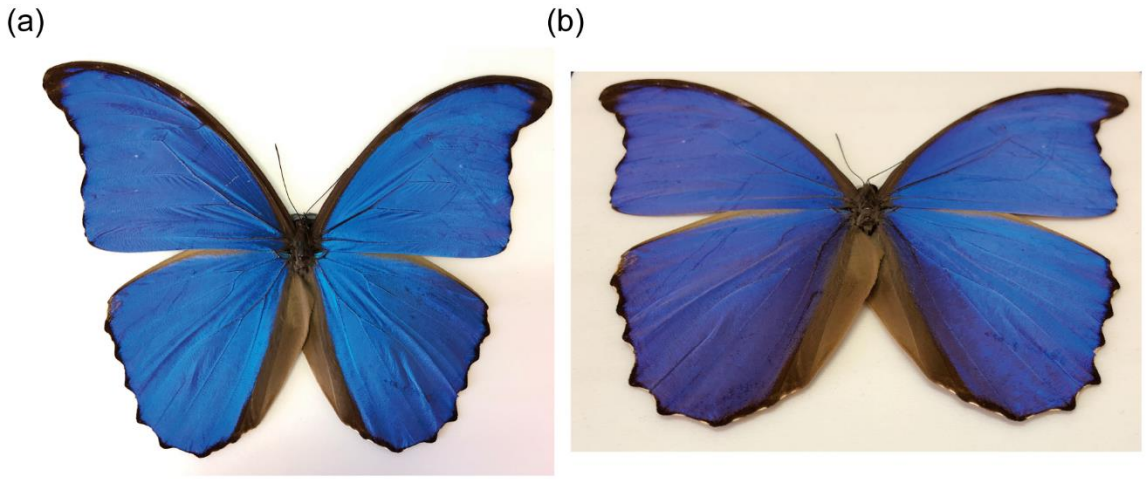
KEYWORDS: heterostructure, colloidal crystal, quasi-ordered array, bilayer inverse structure, anti-counterfeiting, structural color



Figures S1. The digital photos of *Morpho didius* butterfly at different angles (a) 5° and (b) 45°, showing iridescent angle-independent structural colors.

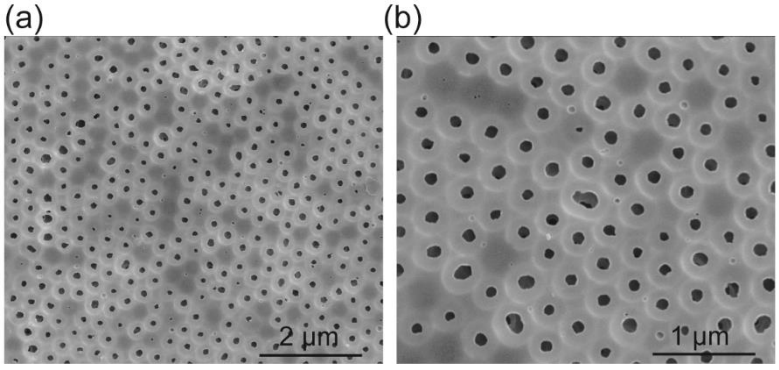


Figure S2. (a) and (b) The top-view SEM image of the BLIHS film at different magnification.

Table S1. The height, baseline and FWHM of the reflectance spectra of the four structural color films (incidence angle=50°)

	280 OALIS	280/240 BLIHS	280/280 BLIHS	280/320 BLIHS
Height (%)	40.2	37.7	33.8	38.1
Baseline (%)	11.1	10.6	10.2	10.1
FWHM (nm)	60.4	58.3	52.3	56.5

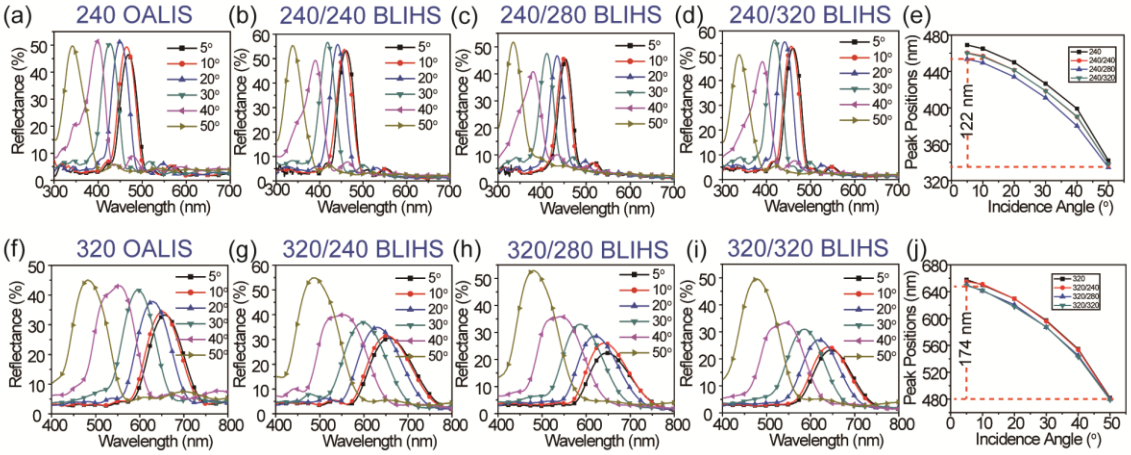


Figure S3. The optical performance and changes of the peak positions of (a) - (e) 240 nm and (f) - (j) 320 nm series samples under specular reflection mode were evaluated by their specular reflective spectra.

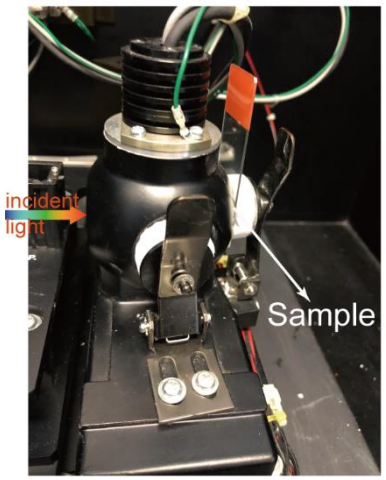


Figure S4. Reflectance spectra measured under integrating sphere mode.

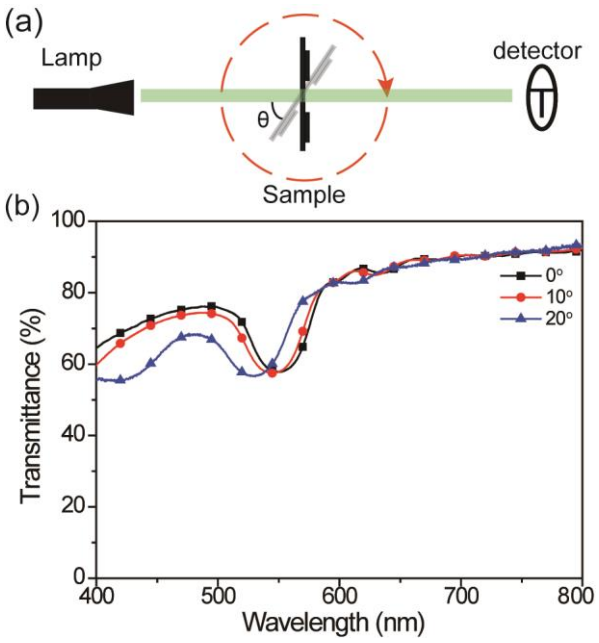


Figure S5. (a) The schematic diagram of the transmittance spectra of the PVDF films with OALIS. (b) The transmittance spectra of the structural color film at various angles of inclination (0^o-20^o)

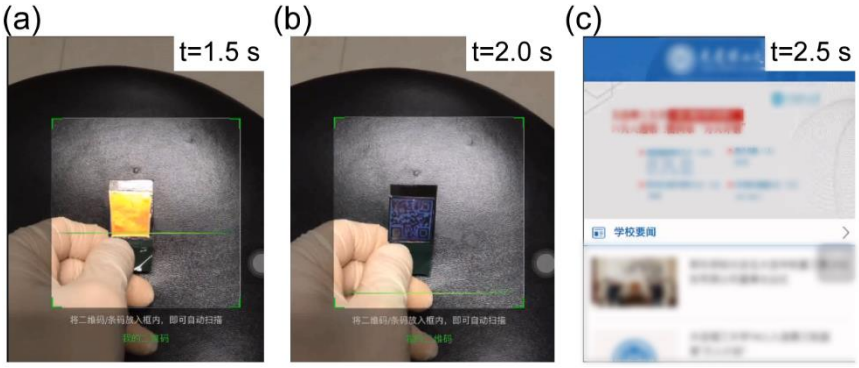


Figure S6. The printscreens of the extracting QR code process at different points in time (a) 1.5 s, (b) 2.0 s, (c) 2.5 s.

Video S1 shows the process of the extracting QR code and accessing the website (pattern displayed) by scanning the QR codes.

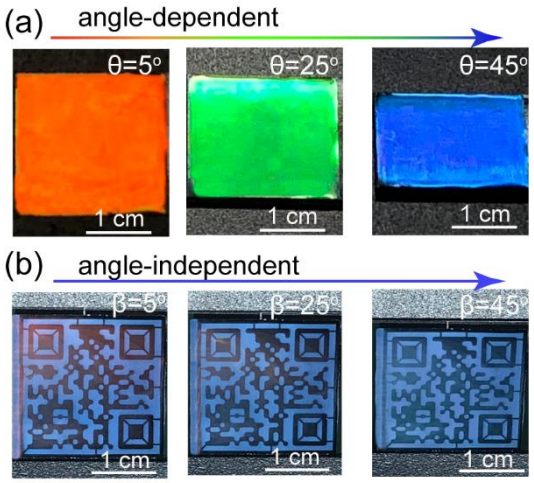


Figure S7. (a) The digital photos of the 320/240 BLIHS PVDF film exhibit different colors under specular reflection mode at different incident angles. (b) The digital photos of the 320/240 BLIHS PVDF film exhibit the non-iridescent QR-code pattern under diffuse reflection mode at different incident angles.

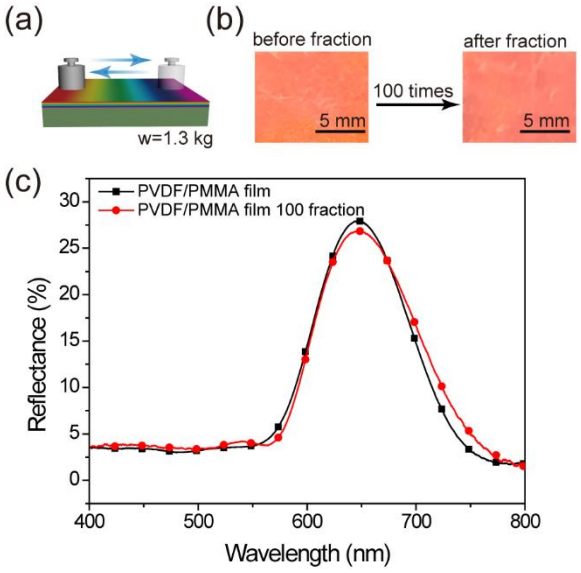


Figure S8. (a) Diagram of the friction test; (b) the digital photos of 320/240 BLIHS PVDF/PMMA film before and after 100 times friction; (c) the reflection spectrum of the PVDF and PVDF/PMMA films.

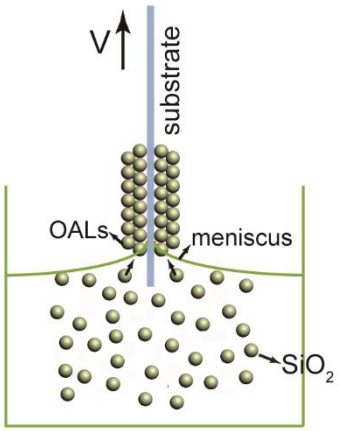


Figure S9 Schematic illustration of dip-coating method.

Table S2. Diameters and Zeta-potential of six SiO_2 Samples

Sample	Ammonia Dosage (mL)	PDI	Size (nm)		Zeta Potential (mV)
			Dh[a]	Da[b]	
(a)	4	0.02	263	240	-34.5
(b)	10	0.05	322	280	-41.6
(d)	20	0.02	359	320	-43.6

a: Hydrodynamic size of these microspheres measured by Nanoparticle size measurement. b: Actual measured diameters from the SEM.

Characterization. The morphology of the SiO_2 monodisperse nanospheres and the arrangement of the quasi-amorphous, ordered and bilayer template and PVDF inverse structure were characterized using a scanning electron microscope (Nova Nano SEM 450). The detailed information of the SiO_2 nanospheres were measure by dynamic light scattering (DLS, Malvern Zetasizer Nano ZS-90). All the specular and diffuse reflectance spectra of the colored PVDF plastic films were obtained using UV-vis spectrophotometer

(Hitachi U-4100). The tensile strength test was conducted with PT-305 Computer universe testing machine from Dongguan Precise-test Equipment Co., Ltd. A homemade device was used for the bending test and the minimum distance between the clamps was 3 mm. Digital photos were obtained by a Nikon digital camera (D7000) under normal daylight lamp.

Modification of CB. The surface CB was modified by grafting nitrobenzene free radicals from the decomposition of diazonium salt. The diazonium salt was prepared by the reaction 0.1 mol. of 4-nitroaniline, 0.1 mol. of sodium nitrite, and 12 mL of concentrated hydrochloric acid in 100 mL of deionized water. The diazonium salt was added to CB dispersion dropwise under 300 W ultrasonication for 6 h at 50°C, it was stirred overnight at ambient temperature, filtered, and washed with water. The solid was dissolved in 200 mL of acetone, ultrasonicated for 20 min, and then filtered. The solvent in the filtrate was removed by reduced pressure distillation, and the resultant CB was purified by Soxhlet extraction. The product was dried overnight in a vacuum.[1]

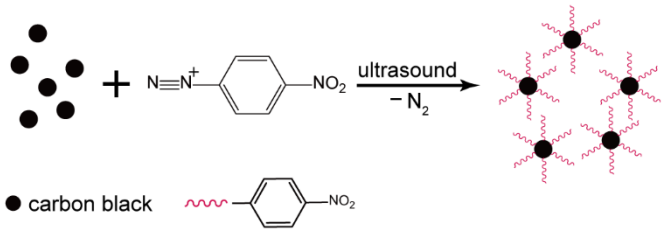


Figure S10. Schematic diagram of the CB surface-modification.

1. Wang, W.; Tang, B.; Ma, W.; Zhang, J.; Ju, B.; Zhang, S. Easy Approach to Assembling a Biomimetic Color Film with Tunable Structural Colors. *JOSA A* **2015**, *32*, 1109-1117.