Electronic Supplementary Information

Enhancing Brightness and Saturation of Noniridescent Structural Color by Optimizing Grain Size

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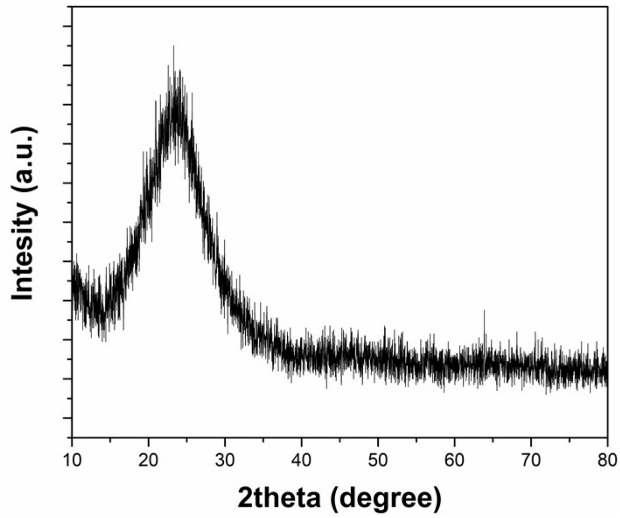

Fig. S1. XRD pattern of silica nano-spheres prepared in our laboratory.

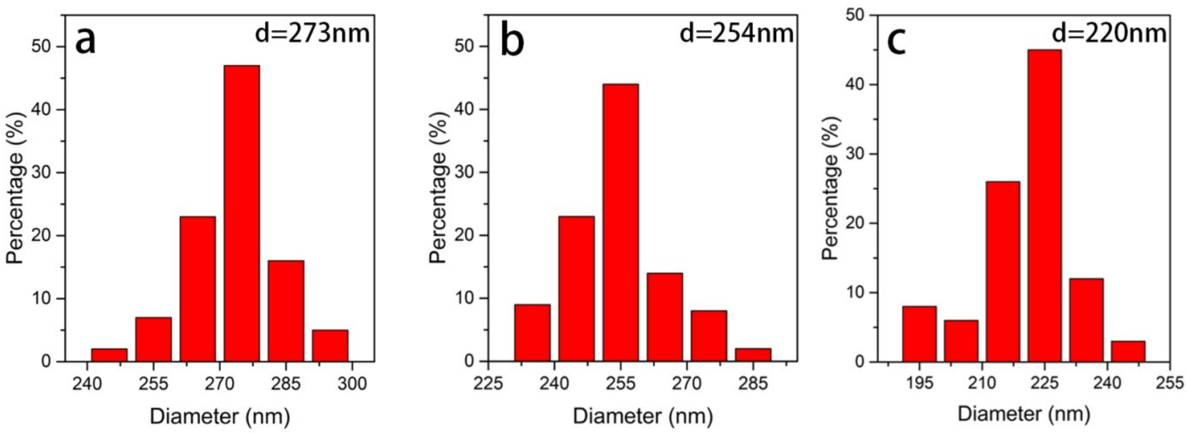

Fig. S2. (a-c) Particle size distribution histograms of the silica nano-spheres composing of the red, orange and green iridescent structural color films, respectively.

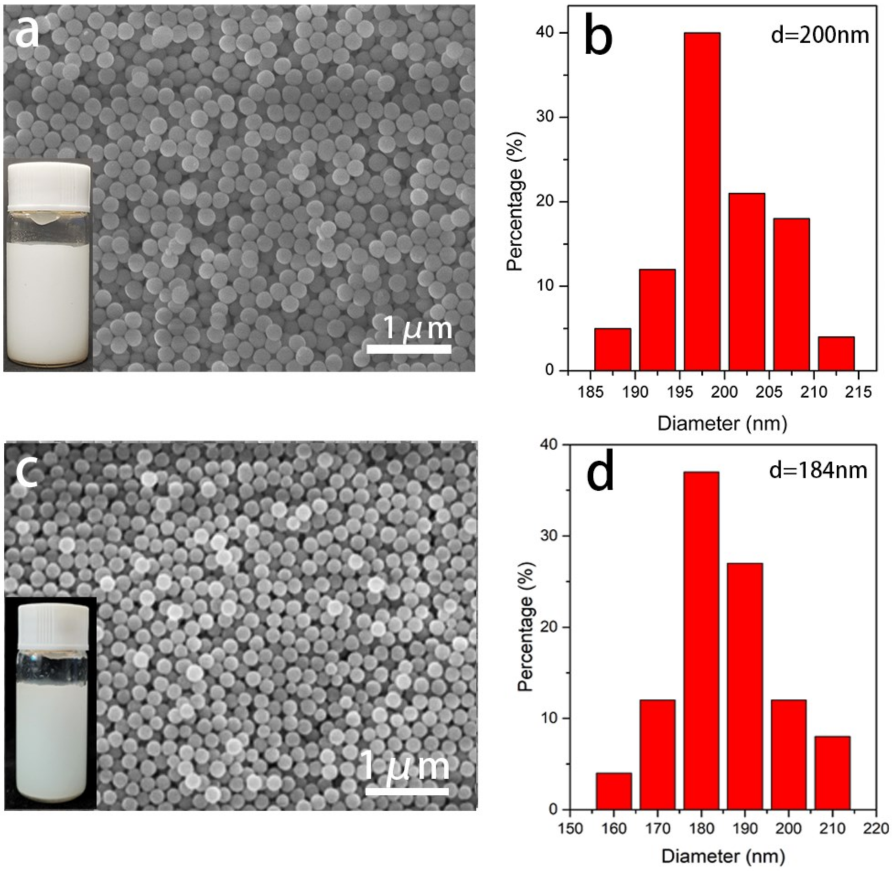

Fig. S3. SEM images and corresponding particle size distribution histograms of silica nano-spheres composing blue and blue-violet non-iridescent structural color films.

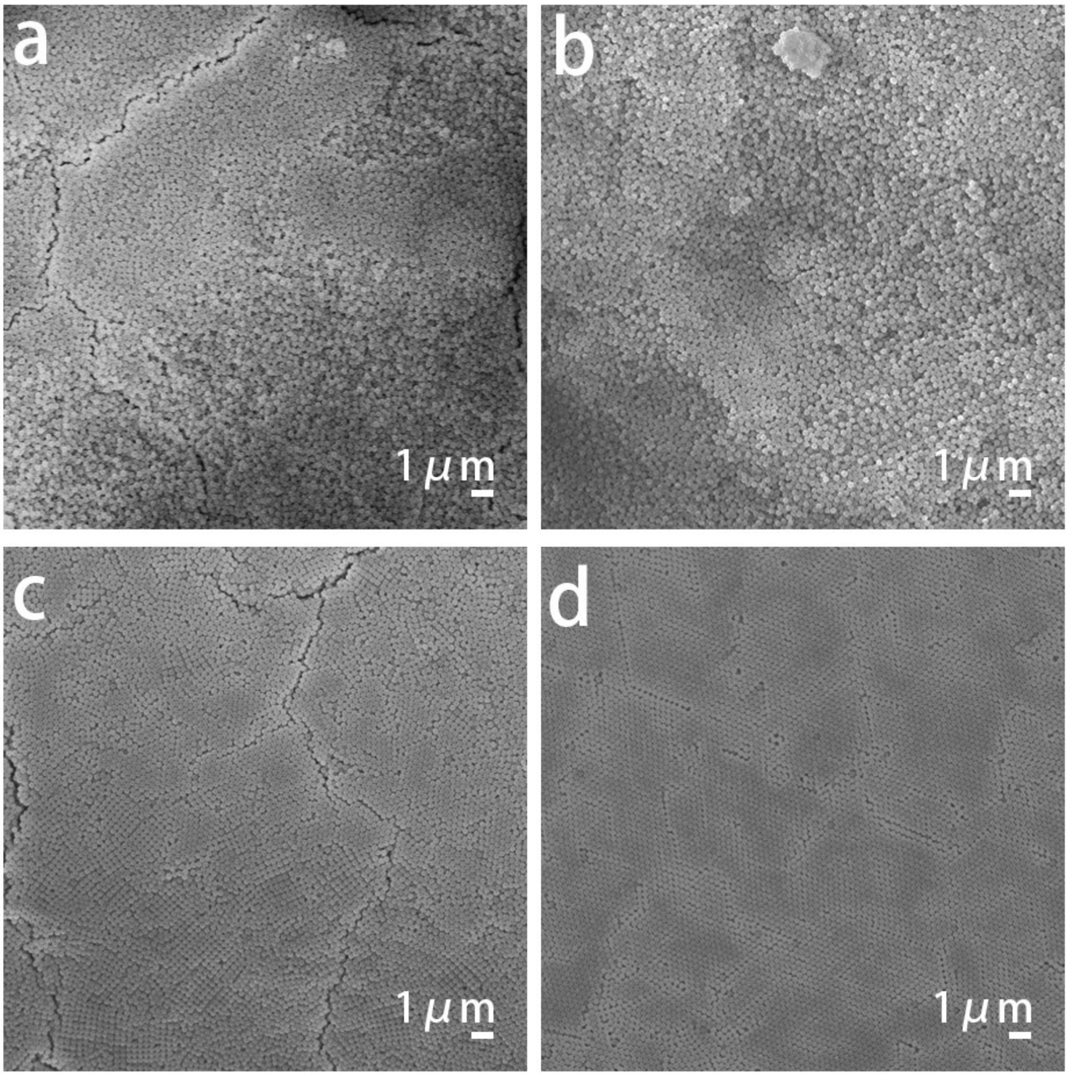

Fig. S4. (a-d) Top-view SEM of F1-F3 non-iridescent structural color films and green iridescent film used for 2D-FFT, respectively.

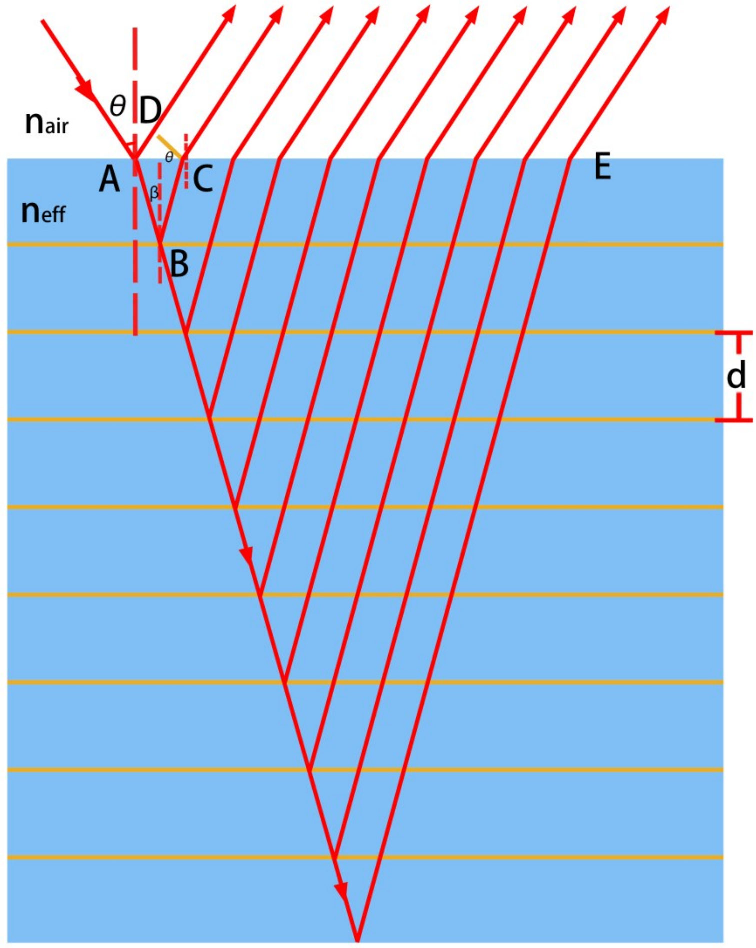

Fig. S5. Schematic diagram of light propagation inside photonic crystal.

Theoretical calculation

Photonic crystal is formed by periodic spatial arrangement of two kinds of dielectric materials with different refractive indexes. When light propagates through PC, it is affected by Bragg scattering which results in the photonic stop-gap. [9-11] Light with a certain wavelength in the range of PBG is forbidden to propagate. When the reflection peak is located in the visible light region, PC will exhibit brilliant structural colors. As we all know the relationship of reflectance peak of structural color can be expressed by Bragg-Snell's equation as：

$$\mathrm{m}\lambda = 2d\sqrt{n_{eff}^2 - \sin^2\theta} \qquad (1)$$

Where m is the order of the Bragg's diffraction, λ is the wavelength of the reflectance peak of structural color, θ is the angle between the incident light and the normal of sample surface (same as viewing angle); n_{eff} is the effective refractive index of the PC which can be expressed as equation (2):

$$n_{eff} = \sqrt{n_{\text{block}}^2 \cdot f_{block} + n_m^2 \cdot (1 - f_{block})} \qquad (2)$$

Here, n_{block} and n_m are refractive indexes of the colloidal nano-spheres and the surrounding medium respectively (in this work, n_{block} is n_{SiO2}, n_m is n_{air} and n_{air}=1); f_{block} refers to the filling ratio of colloidal nano-spheres in PC that is 0.74 for the close-packed hcp structure. And in equation (2), d is the inter-planar spacing between the (001) planes in hcp structure. For the hcp structure, the inter-planar spacing $d_{(001)}$ is given by:

$$\text{d} = \sqrt{\frac{2}{3}} D \qquad (3)$$

where D is the diameter of silica nano-spheres. Thus, the Bragg-Snell's equation can be expressed as:

$$\lambda = 1.63D\sqrt{0.74\text{n}^2_{\text{sio}_2} + 0.26 - \sin^2\theta} \qquad (4)$$

According to the results of the experiment, the reflection peak was at 521 nm at 0º viewing angle when the diameter of silica nano-spheres was 220 nm. Then plug these parameters into equation (4) and (2), we can calculate the refractive index of silica nano-spheres prepared in our lab n_{sio2}=1.58 and n_{eff}=1.45. So Bragg-Snell's equation can be written as:

$$\lambda = 1.63D\sqrt{2.11 - \sin^2\theta} \qquad (5)$$

Taking the silica nano-spheres with diameter of 273 nm as an example, the critical size of grain composed of silica nano-spheres were investigated. When D=273 nm and θ=0° are plugged into equation (5), the theoretical reflection peak λ=646 nm and the corresponding color is red. According to the visible spectrum, visible light in the range of 600-630 nm corresponds to orange color. Therefore, the structural color will shift from red to orange when the reflection peak λ=630 nm. Plug λ=630 nm and D=273 nm into equation (5) then θ=18.68°. It means that the structural color will change from red to orange as the viewing angle changes from 0° to 18.68°. So the ordered domain must be broken before the reflected light skips 18.68° in order to avoid color blue shift. We assume that the structures on both sides of the normal are symmetric. So the longest length of the ordered domain is twice of the length of area skiped by the reflected light from 0º to 18.68º.

The derivation of the Bragg-Snell's equation is based on the fact that the mixture of two dielectric materials with different refractive indexes in PC is regarded as one kind of medium. So the process of light propagation in PC can be simplified as **Figure S5** shown. S.Kinoshita et.al.[1] showed that at least nine layers are needed to obtain the maximum selectivity. Therefore, in order to facilitate, we assume that PC are composed of 9 layers of silica nano-spheres. So 2AE is the longest length of ordered domain for PC stacked by 273 nm silica nano-spheres.

$$AE = 9AC = 18\mathrm{d} \cdot \tan\beta \qquad (6)$$

$$n_{air} \sin\theta = n_{\mathrm{eff}} \sin\beta \qquad (7)$$

So in order to avoid iridescence, the longest length of the ordered region is 1748 nm; about 7 spheres are closely arranged in a line in this case. Silica nano-spheres with different diameters can get different maximum ordered region lengths and the calculation method is similar as above.

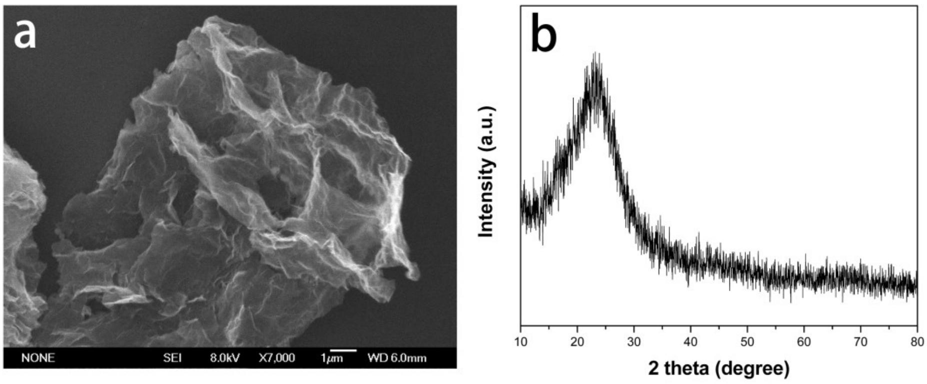

Fig. S6. (a-b) SEM image and XRD pattern of graphene nanosheets.

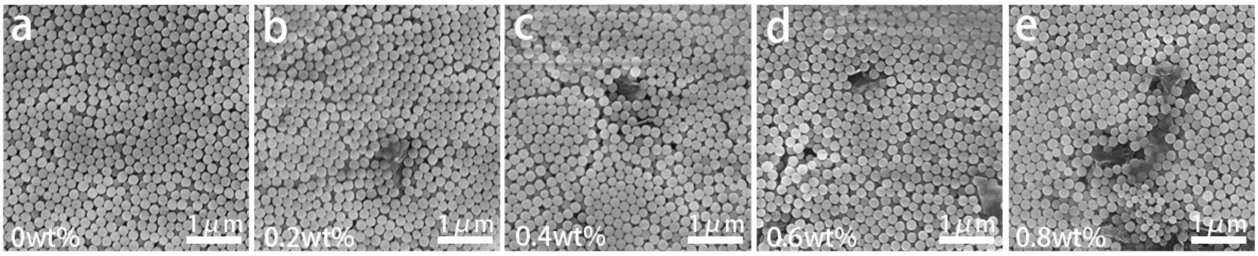

Fig. S7. (a-d) SEM images of blue-violet non-iridescent structural color films with different fractions of graphene nano-sheets.

Fig. S8. Non-iridescent structural color patterns coated on fabric.

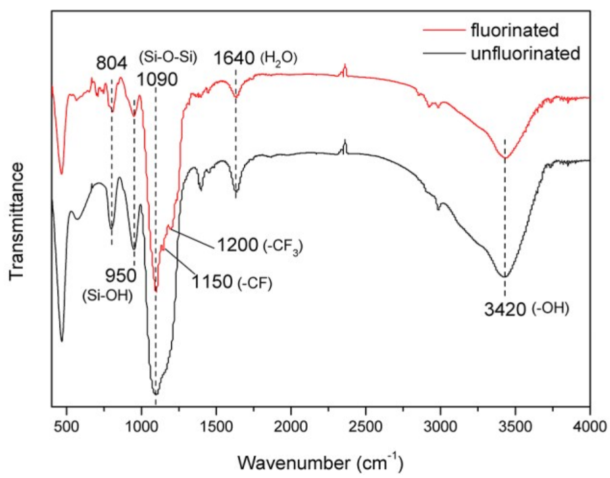

Fig. S9. Fourier-Transform Infra-red (FT-IR) spectra of the fluorinated and unfluorinated surfaces of the non-iridescent structural color films.

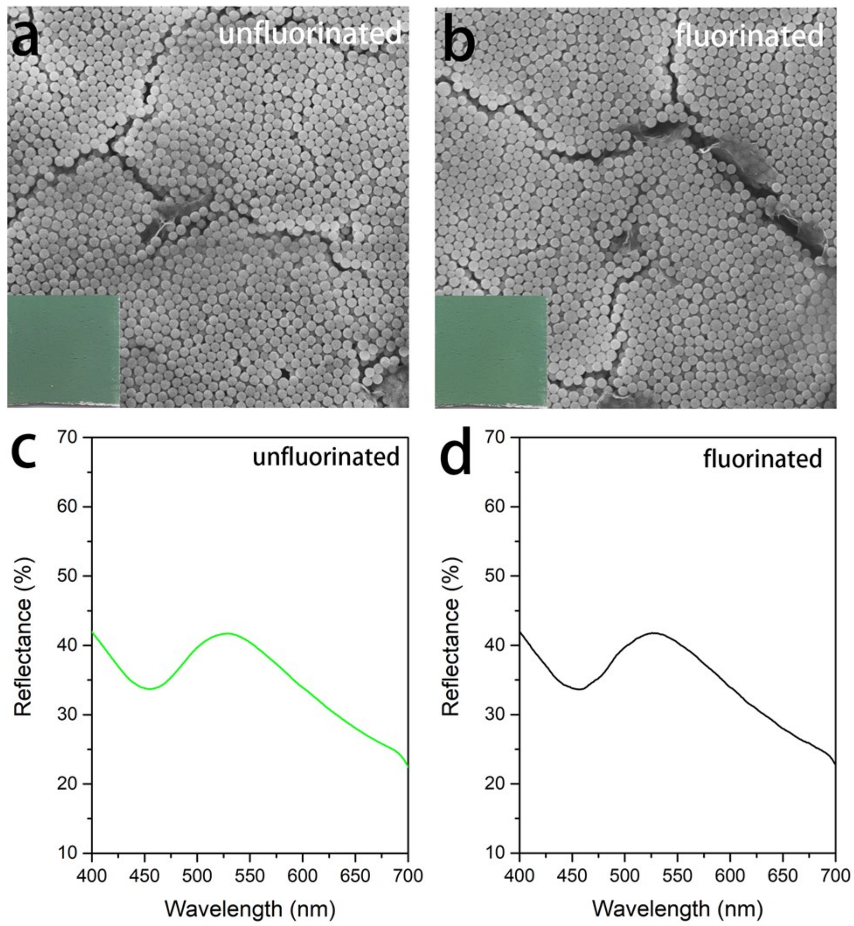

Fig. S10. (a-b) SEM images and optical photographs (insets in the left corner) of non-iridescent structural color films before and after fluorination treatment. (c-d) Reflection spectra of non-iridescent structural color films before and after fluorination treatment.

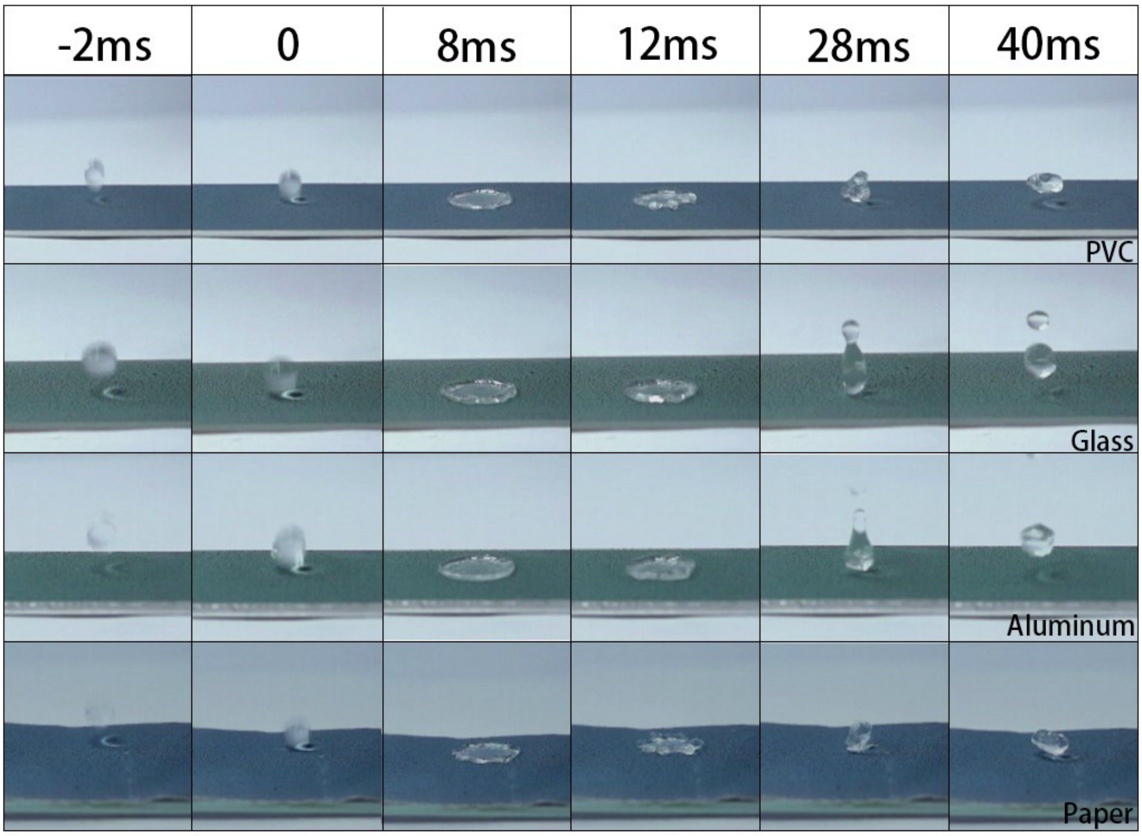

Fig. S11. Time-lapse photographs of water droplets bouncing on the fluorinated structural color films on the PVC, glass, aluminum and paper substrates.

Materials	Full Width Half Maximum (FWHM)	Reference
SiO2	126	This work
SiO2+carbon	134	[1] Fig.2b
PS+cuttlefish ink	125	[2] Fig.2b
P(St–MMA–AA) + graphene nanosheet powder+graphene quantum dots	147	[3] Fig.1E
SiO2+carbon black+poly(vinyl alcohol)	126	[4] Fig.3b
SiO2	169	[5] Fig.5c
polysulfide microspheres+ waterborne polyurea	141	[6] Fig.2b
SiO2+carbon black+poly (dimethyldiallylammonium chloride	138	[7] Fig.4b
PS@PDA+ 3-aminopropyltriethoxysilane (APTES)	155	[8] Fig.2a

Table S1 Comparison of FWHM of film F3 in this work with other non-iridescent structural color films in previous reports.

Reference

1. Y. Takeoka, M. Iwata, T. Seki, K. Nueangnoraj, H. Nishihara and S. Yoshioka, *Langmuir*, 2018, **34**, 4282-4288.
2. Y. Zhang, B. Dong, A. Chen, X. Liu, L. Shi and J. Zi, *Adv. Mater.*, 2015, **27**, 4719-4724.
3. Y. Zhang, P. Han, H. Zhou, N. Wu, Y. Wei, X. Yao, J. Zhou and Y. Song, *Adv. Funct. Mater.*, 2018, **28**, 1802585.
4. Q. Li, Y. Zhang, L. Shi, H. Qiu, S. Zhang, N. Qi, J. Hu, W. Yuan, X. Zhang and K. Q. Zhang, *ACS Nano*, **2018**, 12, 3095-3102.
5. G. Topçu, T. Güner and M. M. Demir, *Photonics and Nanostructures - Fundamentals and Applications*, 2018, **29**, 22-29.
6. M. M. Umair, Fantao Meng, Kashif Iqbal, Xin Jin, Shufen Zhang, Bingtao Tang, *ACS Appl. Mater. Interfaces*, 2019, **11**, 13022-13028.
7. K. Katagiri, K. Uemura, R. Uesugi, K. Inumaru, T. Seki and Y. Takeoka, *RSC Advances*, 2018, **8**, 10776-10784.
8. B. Yi and H. F. Shen, *Appl. Surf. Sci.*, 2018, **427**, 1129-1136.
9. S. Kinoshita, S. Yoshioka and J. Miyazaki, *Rep. Prog. Phys.*, 2008, **71**, 076401.
10. Y. Zhao, Z. Xie, H. Gu, C. Zhu and Z. Gu, *Chem. Soc. Rev.*, 2012, **41**, 3297-3317.
11. J. Sun, B. Bhushan and J. Tong, *RSC Advances*, 2013, **3**, 14862-14889.