

## Supporting Information

### **Three-dimensionally programmable soft crystals toward geometric phase photonics**

Fengyin He,<sup>a</sup> Yuxian Zhang,<sup>\*b</sup> Xuan Liu,<sup>a</sup> Cong-long Yuan,<sup>a</sup> Honglong Hu,<sup>c</sup> Xiao-Qian Wang,<sup>b</sup> Dong Shen,<sup>b</sup> Zhi-Gang Zheng<sup>\*ab</sup>

<sup>a</sup>School of Materials Science and Engineering, East China University of Science and Technology, Shanghai 200237, China.

<sup>b</sup>School of Physics, East China University of Science and Technology, Shanghai 200237, China.

<sup>c</sup>School of Chemistry and Molecular Engineering, East China University of Science and Technology, Shanghai 200237, China.

Corresponding Authors: yuxianzhang@ecust.edu.cn; zgzheng@ecust.edu.cn

#### **Other supporting information for this manuscript include the following:**

##### **Video S1 The dynamic change of PSBP color under different DC bias.**

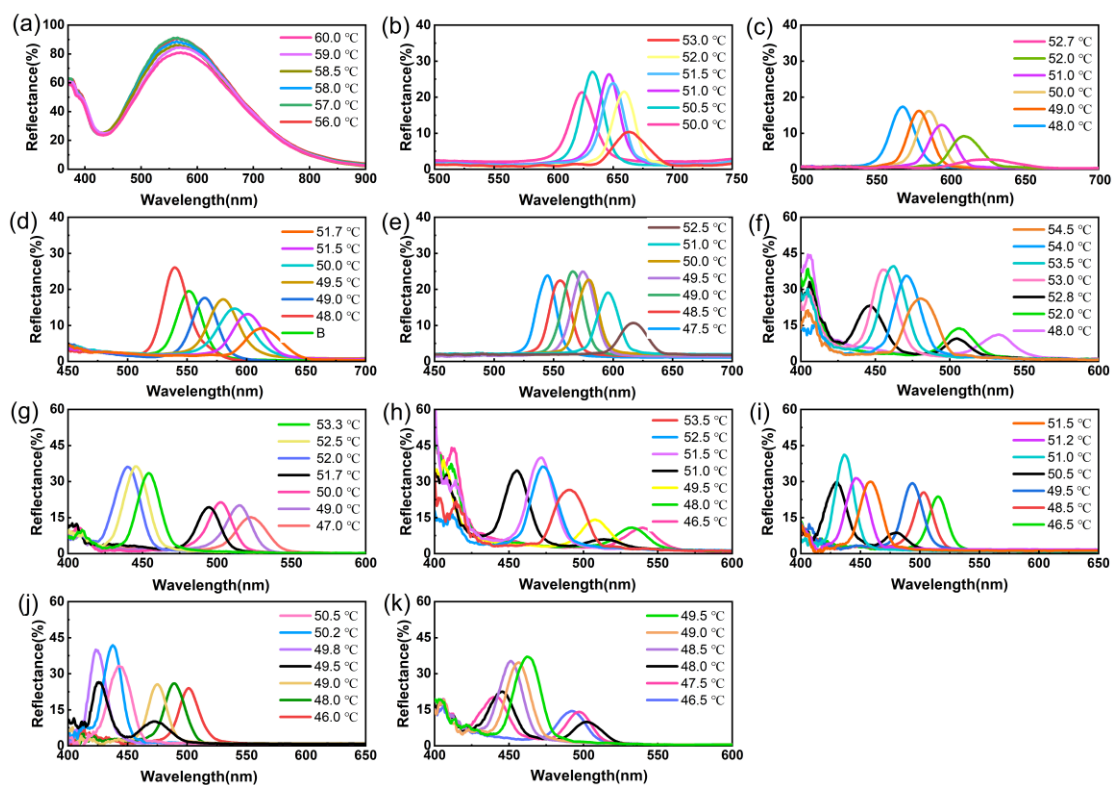
The video shows that the texture of a PSBP sample gradual change from green to blue under a negative DC bias and from green to red under a DC positive bias.

##### **Video S2 The 3D lattice deformation of the PSBPI<sub>(110)</sub> crystals suffering from the DC positive bias.**

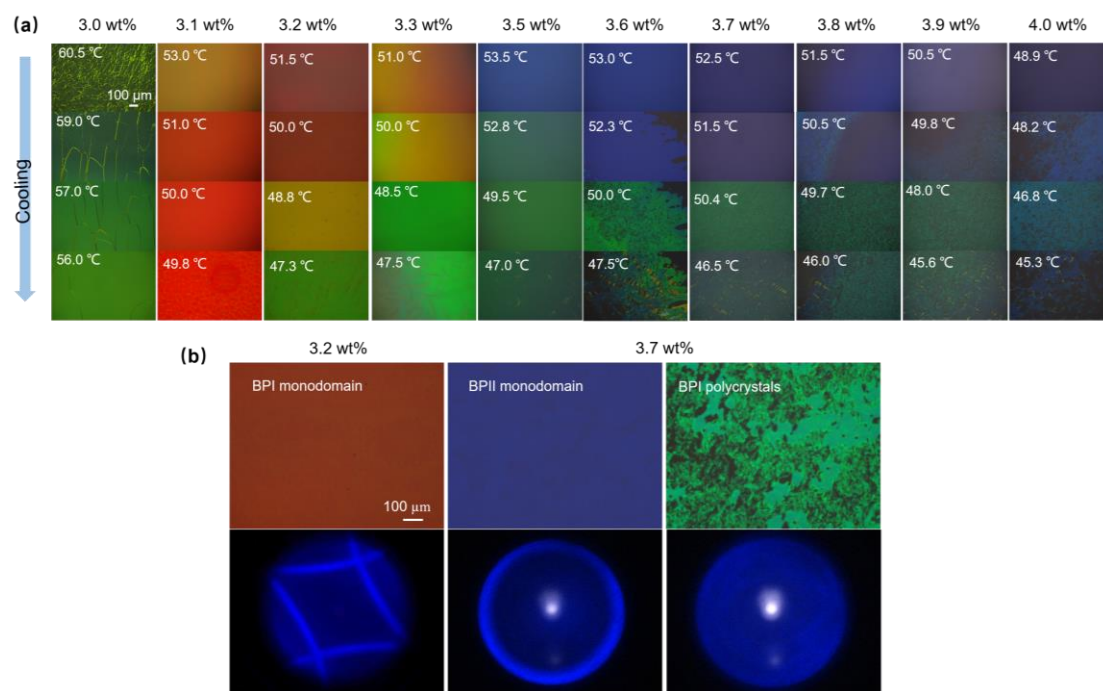
This video shows the 3D lattice deformation of the PSBPI<sub>(110)</sub> crystals by *in situ* Kossel diagrams under DC positive bias.

##### **Video S3 The 3D lattice deformation of the PSBPI<sub>(110)</sub> crystals suffering from the DC negative bias.**

This video shows the 3D lattice deformation of the PSBPI<sub>(110)</sub> crystals by *in situ* Kossel diagrams under DC negative bias.

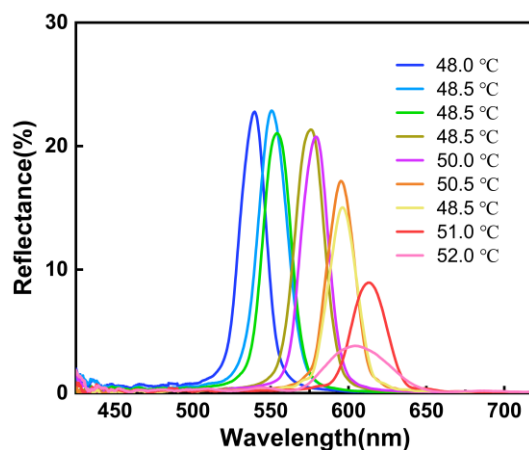


**Fig. S1.** Reflection spectra of the PSBP precursors with different R5011 concentrations during the cooling process in unidirectional photoalignment cells: (a) 3.0 wt%, (b) 3.1 wt%, (c) 3.2 wt%, (d) 3.3 wt%, (e) 3.4 wt%, (f) 3.5 wt%, (g) 3.6 wt%, (h) 3.7 wt%, (i) 3.8 wt%, (j) 3.9 wt%, (k) 4.0 wt%. The black lines in (f) through (k) represent the phase transition from BPII to BPI.

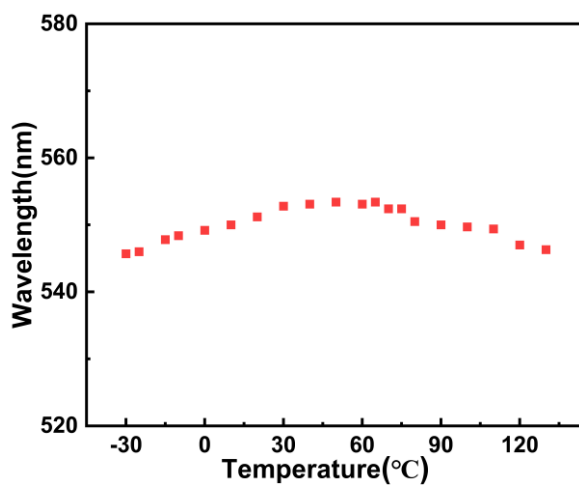


**Fig. S2.** The micrographs recorded under a polarization optical microscope. (a)

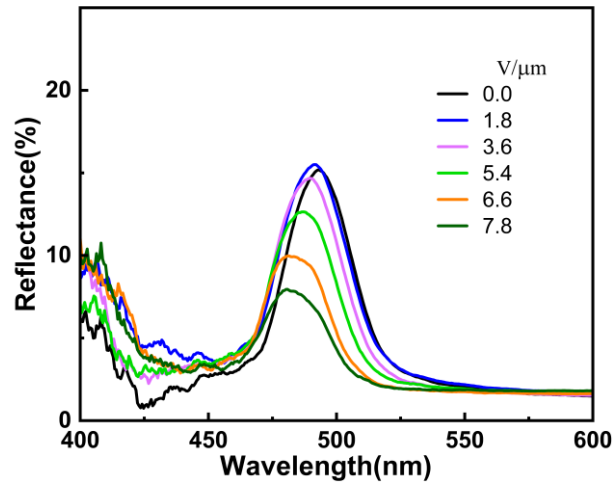
Textures of the PSBP precursor with different R5011 concentrations during cooling in unidirectional photoalignment cells. (b) Typical textures and Kossel diagrams of the PSBP precursor: BPI<sub>(110)</sub> monodomain (grown from isotropic phase), BPII<sub>(100)</sub> monodomain and BPI polycrystals (grown from BPII<sub>(100)</sub>).



**Fig. S3.** Reflection spectra of the PSBP precursor with 3.4wt% R5011 during the cooling from 52.0 ~ 48.0 °C.

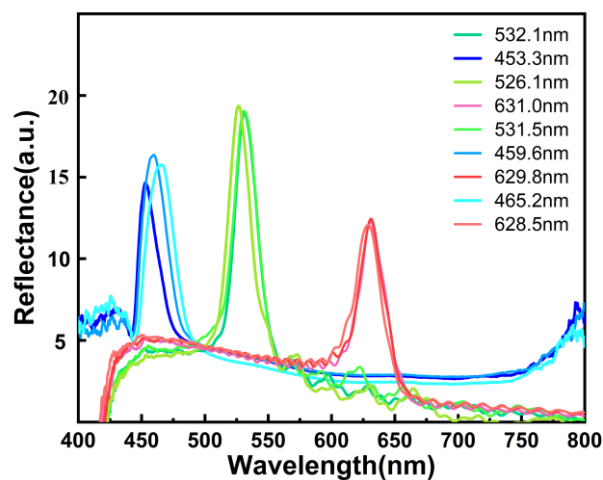


**Fig. S4.** Temperature dependence of the reflection peak of PSBP sample.

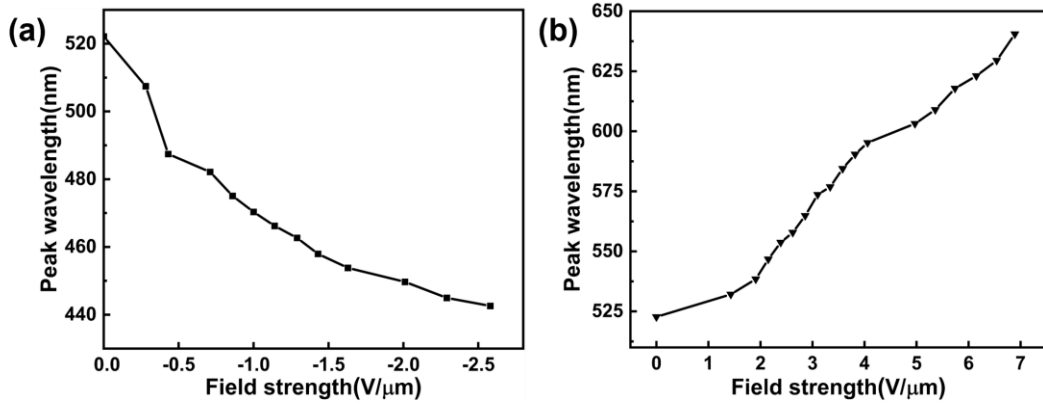


**Fig. S5.** Reflection spectra of the PSBP sample under different electric field strength of AC field (square wave, 1KHz).

The switching of PSBP reflective wavelength driving by DC is not completely reversible, as shown in Fig S6. The reflective wavelength of PSBP sample is initially 532 nm, and blueshifts to 453 nm under  $-2.7 \text{ V } \mu\text{m}^{-1}$  DC field strength but only returned to 526 nm after withdrawal field. Similarly, the reflective wavelength redshifts to 631 nm under  $+6.5 \text{ V } \mu\text{m}^{-1}$  DC field strength and is withdrawn back to 531 nm. Under both negative and positive DC bias, the reflective wavelength cannot completely return to the original position after shifting. In addition, it will move to different locations even under the same electric field strength.

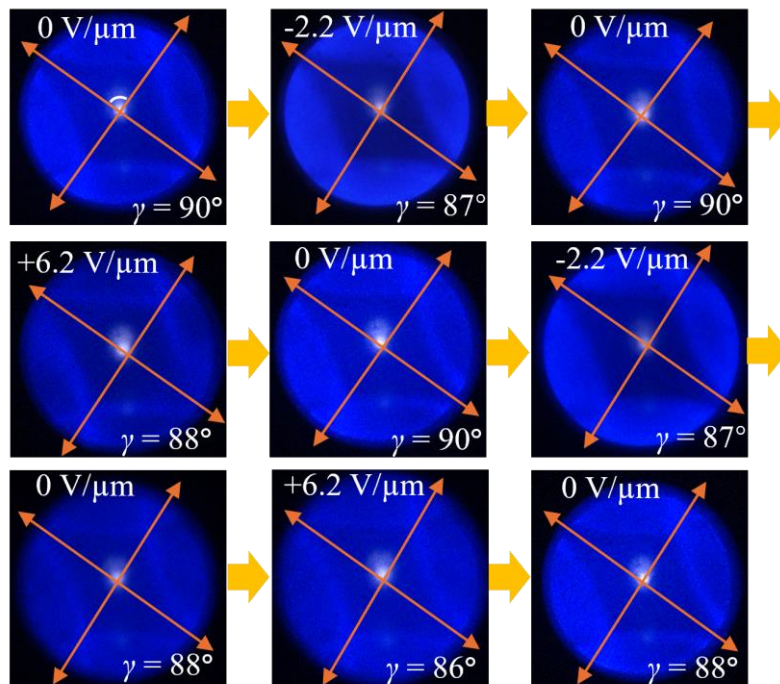


**Fig. S6.** The reversible of PSBP reflective wavelength driving under different DC electric field strength.

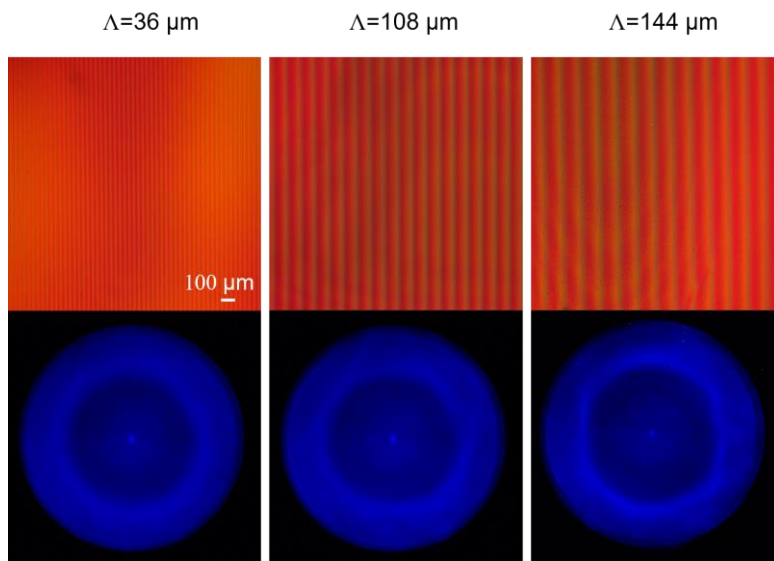


**Fig. S7.** The central reflective wavelength of the PSBP sample under the DC electric field of (a) negative and (b) positive offsets.

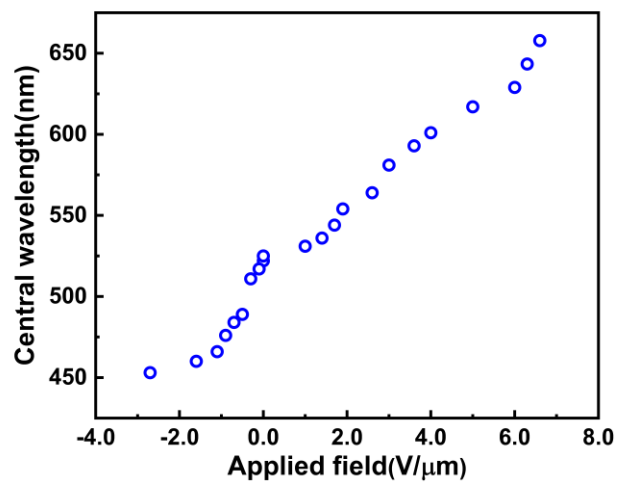
After removing the electric field, the lattice is not perfect cubic structure. Fig. S8 shows the in-situ Kossel diagrams of PSBP sample under DC field. The lattice in the initial state is a cubic structure,  $\gamma = 90^\circ$  (picture 1), and after undergoing an electric field switch (picture 2 to 5), the  $\gamma$  angle is  $90^\circ$  (picture 5), indicating that the BP lattice is still a cubic structure. However, when the DC negative bias is removed in the second cycle,  $\gamma \neq 90^\circ$  (picture 7), the lattice deforms and is no longer a perfect cubic phase.



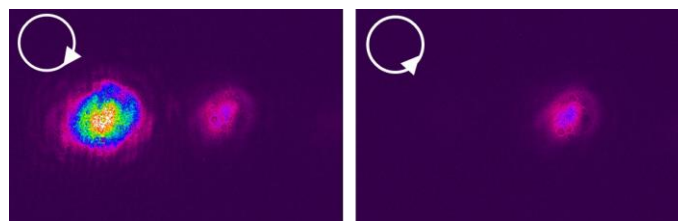
**Fig. S8.** Deformation of lattice structure after removal of electric field.



**Fig. S9.** Texture and Kossel diagram of PSBP-PG with different periods. The scale bar indicates 100  $\mu\text{m}$  for all micrographs.



**Fig. S10.** The central reflection peak position of the PSBP polarization grating under different DC field strength.



**Fig. S11.** Diffractive patterns of PSBP-PG under different CPL incidence.