

## Supporting Information

### **Optoelectronic conversion and polarization hysteresis in organic MISM and MISIM devices with DA-type single component molecules**

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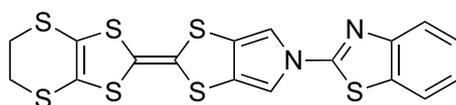
S1. NMR

S2. Thin film XRD measurements

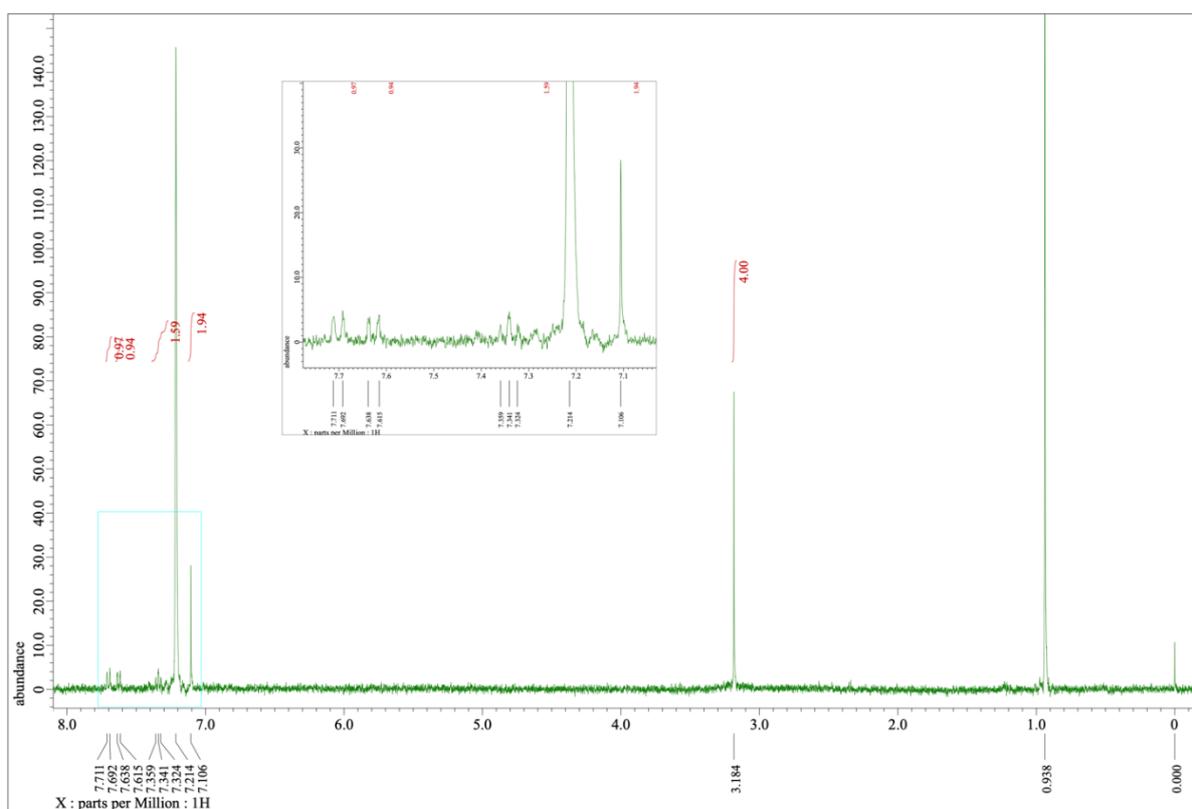
S3. Details of PUND method and ferroelectric displacement current

## S1. NMR spectra

$^1\text{H}$  NMR spectra were recorded on a Bruker Biospin AVANCE 400 spectrometers using  $\text{CS}_2/\text{C}_6\text{D}_6$  solvent. The chemical shifts were referenced to tetramethylsilane.

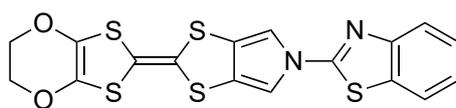


ET-PRBT

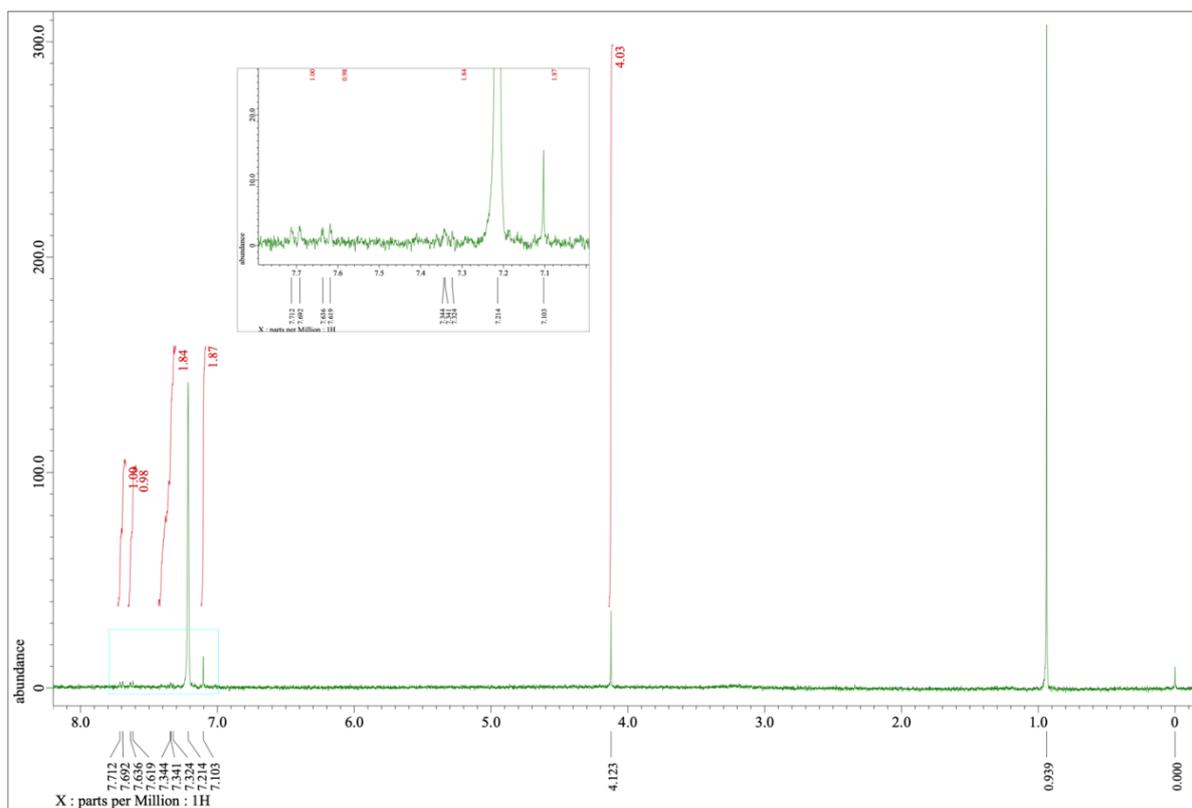


$^1\text{H}$  NMR (400 MHz,  $\text{CS}_2/\text{C}_6\text{D}_6$ , 25 °C, TMS)  $\delta$  3.18 (s, 4H), 7.11 (s, 2H), 7.32–7.36 (m, 2H), 7.63 (d, 1H), 7.70 (d, 1H) ppm.

**Figure S1.**  $^1\text{H}$  NMR spectrum of ET-PRBT in  $\text{CS}_2/\text{C}_6\text{D}_6$  (400 MHz).



EO-PRBT

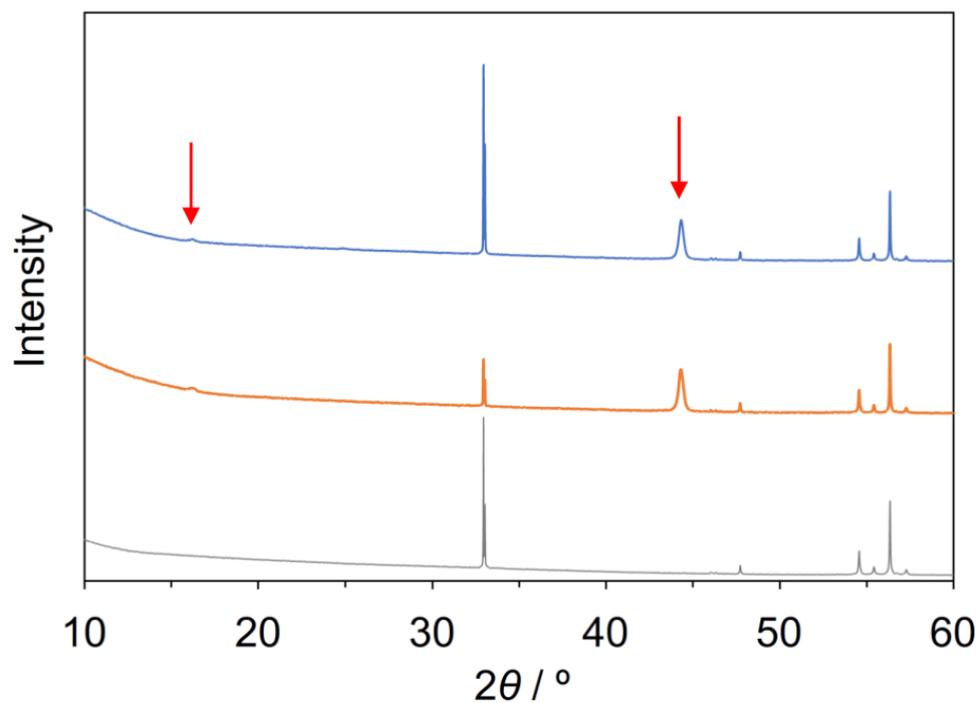


<sup>1</sup>H NMR (400 MHz, CS<sub>2</sub>/C<sub>6</sub>D<sub>6</sub>, 25 °C, TMS) δ 4.12 (s, 4H), 7.10 (s, 2H), 7.32–7.34 (m, 2H), 7.63 (d, 1H), 7.70 (d, 1H) ppm.

**Figure S2.** <sup>1</sup>H NMR spectrum of EO-PRBT in CS<sub>2</sub>/C<sub>6</sub>D<sub>6</sub> (400 MHz).

## S2. Thin film XRD measurements

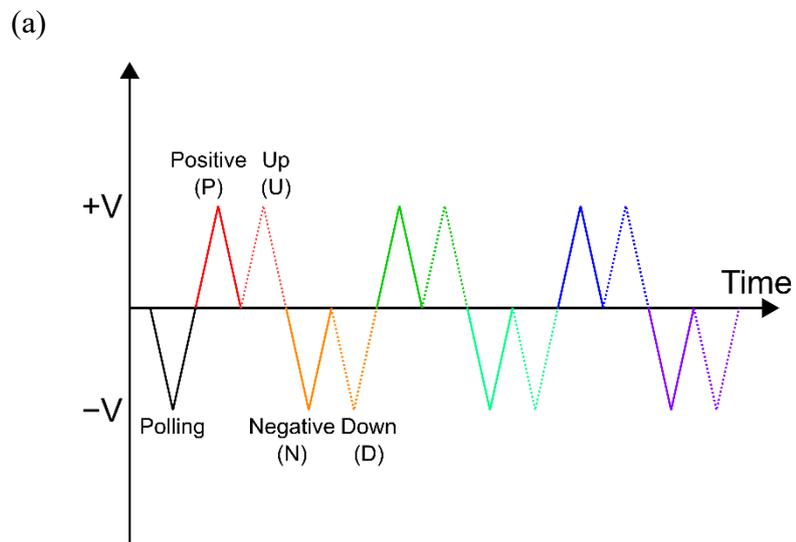
X-ray (Cu  $K_{\alpha}$ ) structural analysis was performed on thin films of ET-PRBT and EO-PRBT, using a Rigaku SmartLab diffractometer. The results are shown in Fig. S1.



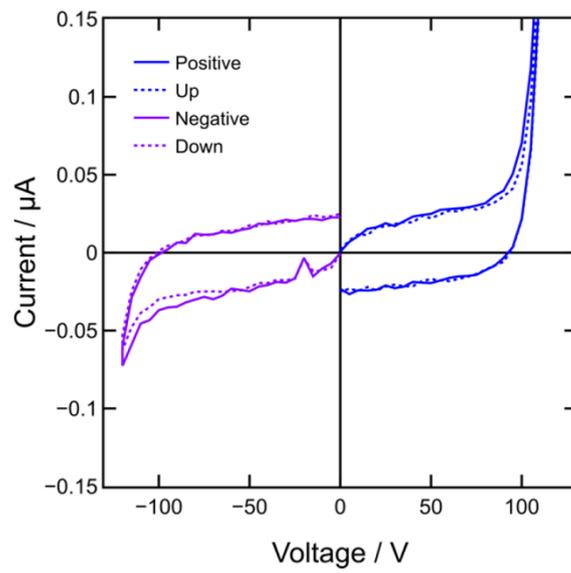
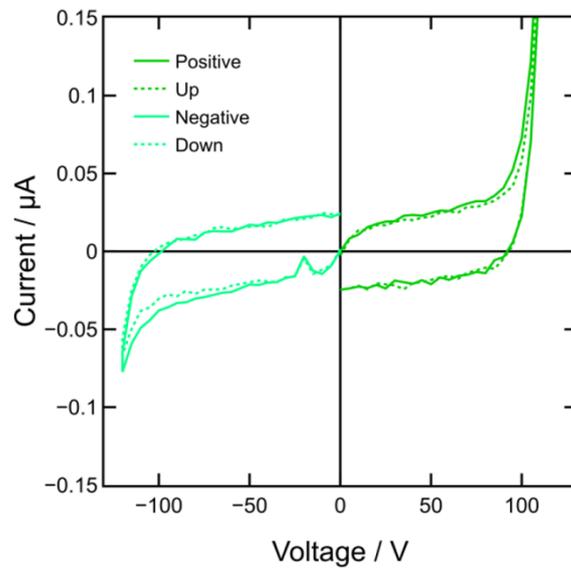
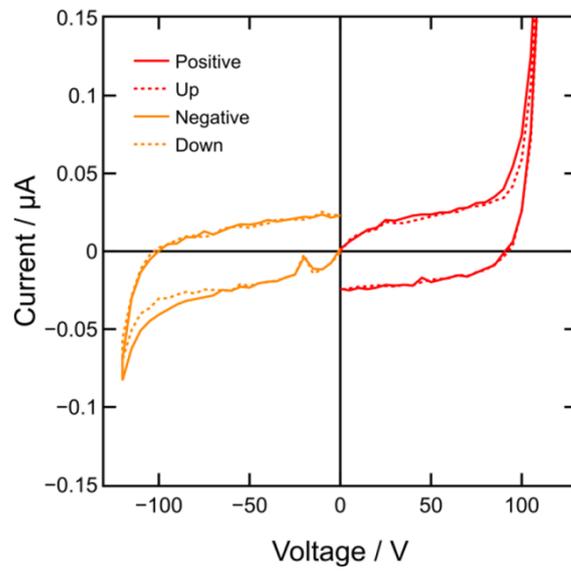
**Figure S3.** XRD patterns of the thin films (30 nm) of ET-PRBT (orange) and EO-PRBT (blue), together with Si substrate (gray). The intrinsic diffractions of ET-PRBT and EO-PRBT peaks appear at nearly the same positions,  $2\theta = 16^\circ$  and  $44^\circ$  (red arrows), indicating that they possess good crystallinity and are isostructural.

### S3. Details of PUND method and ferroelectric displacement current

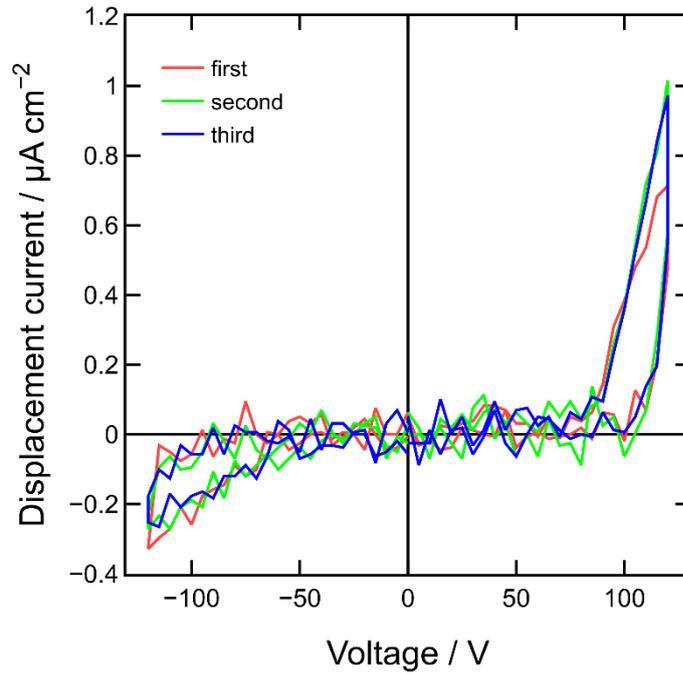
The  $P-V$  curves in Fig. 6 are obtained by the procedure described in the caption of Fig. S2.



(b)



(c)



**Figure S4.** Details of the PUND method and ferroelectric displacement current: (a) Schematic diagram illustrating the voltage scan in the PUND method. (b) Current-voltage curves obtained in the P and N scans (solid curve) and in the U and D scans (dotted curves) on the PUND method. The same measurements were done for three times (upper, middle and bottom). (c) Difference in current between the P and U scans, as well as between the N and D scans, for the three scans. Polarization hysteresis is observed in the P and N scans, but they significantly contain a capacitive component. By subtracting the results of the U and D scans from the P and N scans, respectively, the pure ferroelectric component can be extracted. The  $P$ - $V$  curves in Figure 6 are obtained by integrating the curvatures shown in Fig. S2(c).