

Supporting information

Ambipolar charge-trapping in self-assembled nanostructures of supramolecular miktoarm star-shaped copolymer with a zinc phthalocyanine core

Xinhao Zhong,^{a,b} Debdatta Panigrahi,^c Ryoma Hayakawa,^c Yutaka Wakayama,^{*c} Koji Harano,^d Masayuki Takeuchi,^{*a,b} and Junko Aimi^{*a}

^a Research Center for Macromolecules and Biomaterials, National Institute for Materials Science (NIMS), 1-2-1 Sengen, Tsukuba, Ibaraki 305-0047, Japan

^b Department of Materials Science and Engineering, Faculty of Pure & Applied Sciences, University of Tsukuba, 1-1-1 Tennodai, Tsukuba, Ibaraki 305-8577, Japan

^c Research Center for Materials Nanoarchitectonics (MANA), NIMS, 1-1 Namiki, Tsukuba, Ibaraki 305-0044, Japan

^d Center for Basic Research on Materials, NIMS, 1-1 Namiki, Tsukuba, Ibaraki 305-0044, Japan

Table S1. List of polymers

Polymer	Molecular structure	M_n (Kg mol ⁻¹) ^a	M_w/M_n ^a	DP
ZnPcPS ₄		14.2	1.09	36 ^b
PS ₄		16.1	1.10	39 ^b
pyPMMA		13.9	1.12	132
pyPVAc		16.1	1.27	207
pyPVK		13.0	1.22	74

^a) Number-average molecular weight and polydispersity determined by GPC analysis. ^b) Degree of polymerization (DP) estimated for each arm.

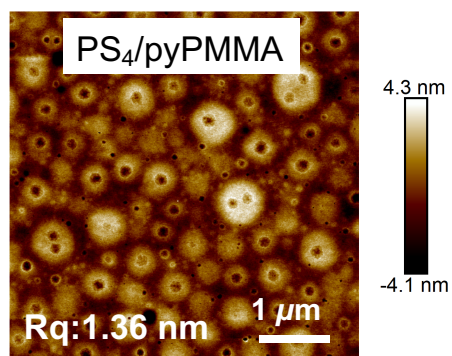


Figure S1. AFM height image of a polymer blend film of PS₄ and pyPMMA.

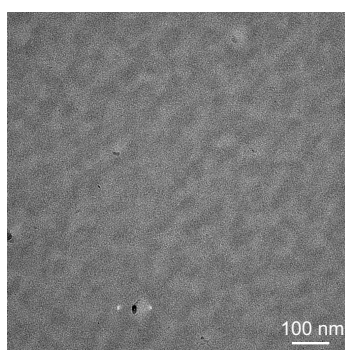


Figure S2. Bright-field TEM image of ZnPcPS₄/pyPMMA film.

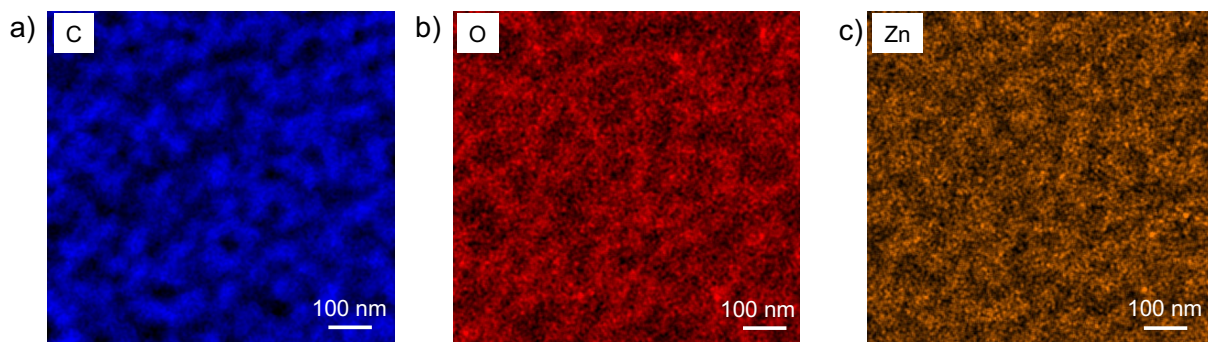
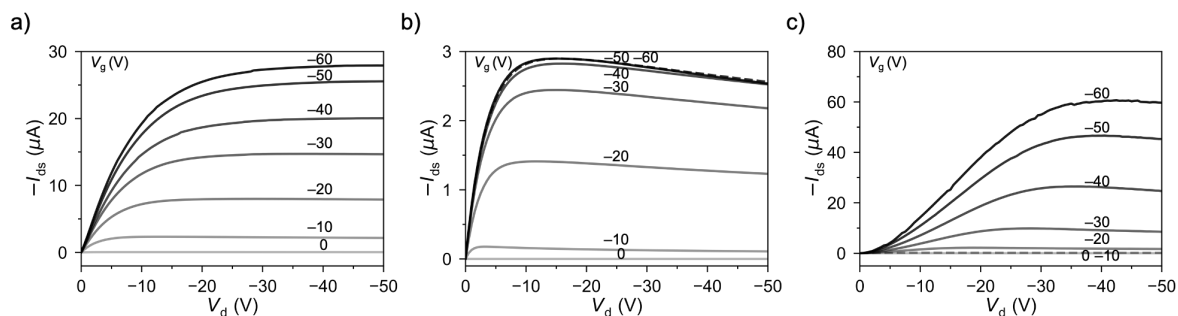
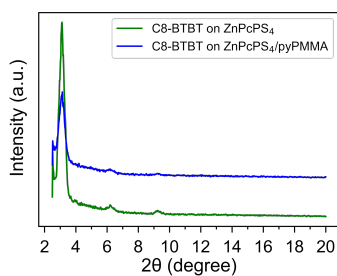


Figure S3. STEM-EDS elemental mapping image for carbon (a), oxygen (b), and Zn (c) of a polymer film of ZnPcPS₄/pyPMMA.

Table S2. Device performances of C8-BTBT-based OFET memory using polymer dielectrics

Memory layer	Initial V_{th} (V)	I_{on}/I_{off} (A)	Mobility ($\text{cm}^2 \text{V}^{-1} \text{s}^{-1}$)	Hole-trapping $V_{th(-)}^a$ (V)	Electron-trapping $V_{th(+)}^b$ (V)	Memory window (V)	Memory ratio
ZnPcPS ₄ /pyPMMA	0.57	$\sim 10^8$	0.31	-24.80	22.43	47.23	$\sim 10^7$
ZnPcPS ₄	-2.05	$\sim 10^7$	0.26	-33.54	15.07	48.61	$\sim 10^6$
pyPMMA	-12.15	$\sim 10^7$	2.96	-17.42	-1.71	15.71	$\sim 10^7$

a) The threshold voltages $V_{th(-)}$ estimated after applying $V_g = -60$ V for 1 s. b) The threshold voltages $V_{th(+)}$ estimated after applying $V_g = +60$ V under UV light for 5 s.

**Figure S4.** Output characteristics for OFET devices with ZnPcPS₄/pyPMMA (a), ZnPcPS₄ (b), and pyPMMA (c) layers.**Figure S5.** XRD profiles of the C8-BTBT films on ZnPcPS₄ (green line) and ZnPcPS₄/pyPMMA (blue line).

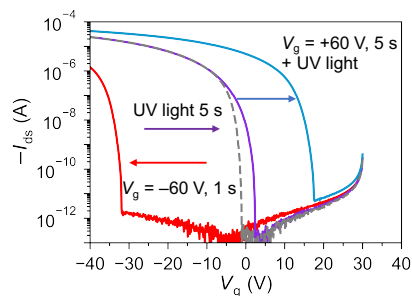


Figure S6. Transfer characteristics of OFET memory device with a polymer layer of ZnPcPS₄ at $V_d = -50$ V. Transfer curves were monitored at the initial state (gray), after electric writing (red), after photo-erasing (purple), and after photo-assisted writing (blue) operations.

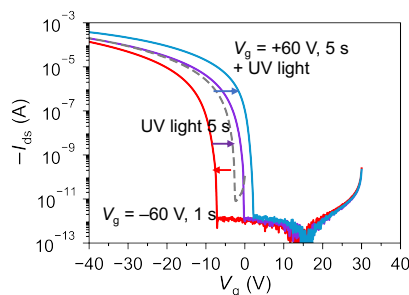


Figure S7. Transfer characteristics of OFET memory device with a polymer layer of pyPMMA at $V_d = -50$ V. Transfer curves were monitored at the initial state (gray), after electric writing (red), after photo-erasing (purple), and after photo-assisted writing (blue) operations.

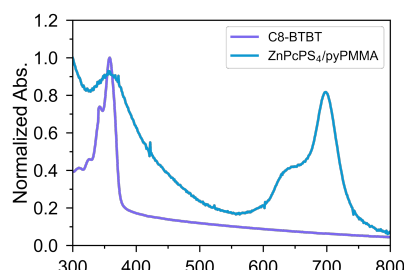


Figure S8. Absorption spectra of C8-BTBT film (purple line) and ZnPcPS₄/pyPMMA blend film (blue line).

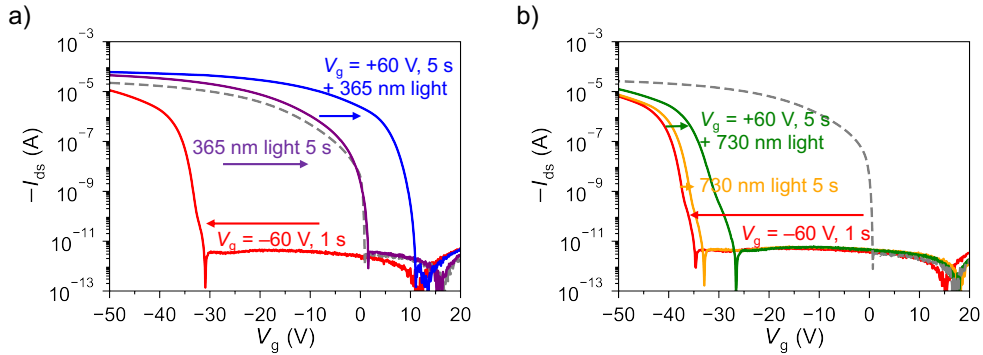


Figure S9. Transfer characteristics of OFET memory device with a polymer layer of ZnPcPS₄ at $V_d = -50$ V by LED light irradiation at (a) 365 nm and (b) 730 nm.

The transfer curve at the initial state (gray dashed line) was shifted after applying $V_g = -60$ V for 1 s (red line). Subsequent irradiation of 365 nm LED light for 5 s shifted the transfer curve to the initial state (purple line Figure S9a). Simultaneous exposure to $V_g = +60$ V and 365 nm light for 5 s shifted the transfer curve to the positive direction (blue line Figure S9a). While irradiation of 730 nm LED light for 5 s (orange line Figure S9b) and simultaneous exposure to $V_g = +60$ V and 730 nm light for 5 s (green line Figure S9b) resulted in only slight positive shift.

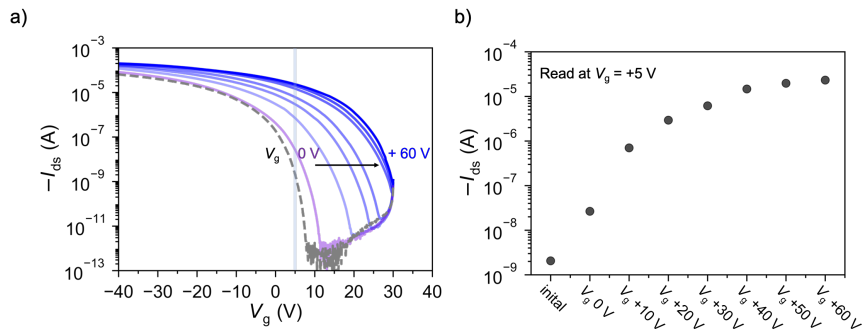


Figure S10. (a) Transfer characteristics of the OFET memory device with ZnPcPS₄/pyPMMA memory layer at $V_d = -50$ V after applying various positive V_g under the UV light irradiation for 5 s. (b) Drain current read at $V_g = +5$ V and $V_d = -50$ V after applying various positive V_g with UV light for 5 s.

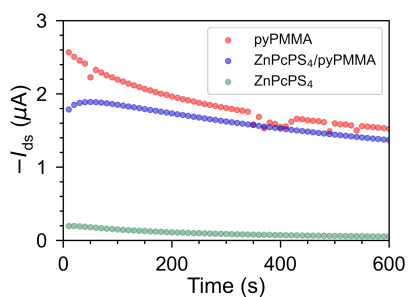


Figure S11. Retention time of I_{ds} monitored at $V_g = 0$ V and $V_d = -10$ V after photo-assisted programming of OFET memory devices with pyPMMA (red circle), ZnPcPS₄/pyPMMA (blue circle) and ZnPcPS₄ (green circle) layers.

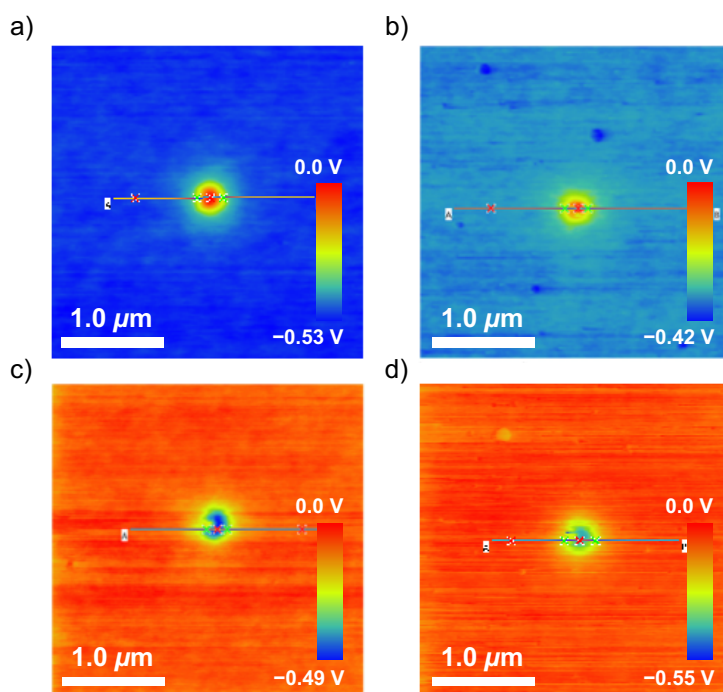


Figure S12. Surface potential images after applying $V_{tip} = +10$ V onto ZnPcPS₄ (a) and pyPMMA (b). Surface potential images after applying $V_{tip} = -10$ V of ZnPcPS₄ (c) and pyPMMA (d).

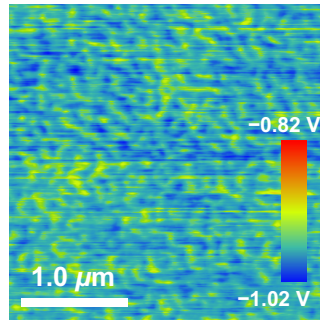


Figure S13. Surface potential image of ZnPcPS₄/pyPMMA film at the initial state.