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## Supporting information for

## Molecular Template Growth of Organic Heterojunction to Tailor Visual Neuroplasticity for High Performance Phototransistor with Ultra-Low Energy Consumption

Ender Ercan,\* Chih-Chien Hung, Guan-Syuan Li, Yun-Fang Yang, Yan-Cheng Lin, and Wen-Chang Chen\* **Table S1.** Morphological and transport properties of the biocomposite films in the GISAXS and phototransistor characterizations.

Sample	<i>d-spacing</i> (nm) <sup>a</sup>	crystallite size (L <sub>c</sub> ) (nm) <sup>b</sup>	$\mu_{FET} \times 10^{-3}$ (cm <sup>2</sup> V <sup>-1</sup> s <sup>-1</sup> ) <sup>c</sup>	
DNTT/p-6p (2nm)	15.5	8.0	$0.287\pm0.0066$	
DNTT/p-6p (10nm)	15.3	5.8	$0.107\pm0.0034$	
DNTT/p-6p (20nm)	15.2	8.0	$0.043\pm0.0074$	

<sup>*a*</sup> *d*-spacing distances calculated based on the 1-D GISAXS profiles using  $d = 2\pi/q^*$ . <sup>*b*</sup> Crystallite sizes calculated based on the out-of-plane (100) diffraction using Scherrer equation: L<sub>c</sub> = 0.9 × 2 $\pi$ /FWHM. <sup>*c*</sup> The field-effect mobility ( $\mu_h$ ) is calculated at the saturation regime of transfer curve.

	μ <sub>h</sub> (cm <sup>2</sup> V <sup>-1</sup> s <sup>-1</sup> )	I <sub>ON</sub> /I <sub>OFF</sub> <sup>a</sup>	<i>ΔV</i> <sub>TH</sub> [V]		
Heterojunction			Program	Erase	ΔV <sub>TH</sub> [V] <sup>b</sup>
DNTT/p-6p (2nm)	$\begin{array}{c} 0.287 \pm \\ 0.0066 \end{array}$	8.09×10 <sup>3</sup>	2.2	-3.7	5.9
DNTT/p-6p (10nm)	$\begin{array}{c} 0.107 \pm \\ 0.0034 \end{array}$	1.97×10 <sup>5</sup>	3.9	-10.6	14.5
DNTT/p-6p (20nm)	$0.043 \pm 0.0074$	1.66×10 <sup>4</sup>	3.5	-19.9	23.4

**Table S2.** Photonic FET device performance of the studied BCP films.

a  $I_{\rm ON}$  and  $I_{\rm OFF}$  currents were estimated from the transfer curves before and after light irradiation at a reading  $V_{\rm GS}$  of 0 V. b Memory window ( $\Delta V_{\rm TH}$ ) was calculated from transfer curves of the photonic memory devices.

Numbers of pulse	τ <sub>m</sub> (s)	Light intensity (mW/cm²)	τ <sub>m</sub> (s)	Pulse width (s)	τ <sub>m</sub> (s)
100	10.73	21.2	7.19	10	4.24
50	10.02	14.6	6.00	5	2.69
20	2.64	11.8	3.97	1	1.49
10	1.16	8.25	3.25	500	0.99
5	0.85	5	2.90	300	0.92
1	0.04	1.4	1.98	50	0.52

**Table S3.** Summary of the fitted time constants from the forgetting curves with different measurement conditions of pulse numbers, light intensity and pulse width.

The respective average time constant ( $\tau_m$ ) can be calculated based on Eq. 1 and Eq. 2.

$I = I_0 + A_1 \exp\left(-\iota/\tau_1\right) + A_2 exp_{100}\left(-\iota/\tau_2\right)$	(Eq. 1)
$\tau_m$ (mean weight decay time) = $(A_1\tau_1 + A_2\tau_2)/(A_1 + A_2)$	(Eq. 2)

**Table S4.** Summary of the device parameters of the reported organic and heterojunction synaptic transistors.

Material system	Light wavelength (nm)	Channel area W(μm)×L(μm)	Operating voltage (V)	Energy consumption
PDPP4T/ Chlorophyll <sup>[13]</sup>	430	300 × 5	-10 <sup>-5</sup>	0.25 fJ
P3HT-b-P2VP <sup>[14]</sup>	450	25 × 1500	$-3 \times 10^{-4}$	0.56 fJ
Natural carotene/ PDPP4T <sup>[26]</sup>	405	-	$-1 \times 10^{-5}$	0.0034 fJ
SWCNT/ chlorophyll-a <sup>[27]</sup>	665	Aspect ratio 1000	-10-4	17.5 fJ
DNTT/TPP <sup>[28]</sup>	450	14000 × 50	$-7 \times 10^{-5}$	1.4 fJ
Pentacene/ $C_3N_4 ND^{[29]}$	365	1500 × 50	-0.3	18.06 fJ
Pentacene/PMMA /2DP <sup>[30]</sup>	400	1000 × 30	-0.1	0.29 pJ (290 fJ)
PTCDA/MoS <sub>2</sub> <sup>[31]</sup>	532	2 × 5.3	0.1	10 pJ
Bilayer C8-BTBT <sup>[32]</sup>	365	40 × 20	-2	13.6 pJ
DNTT/ p-6P (This Work)	450	1000 × 20	-10 <sup>-3</sup>	0.54 fJ

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**Figure S1.** (a) AFM topography of the DNTT film on the wafer without a p-6P template. 2D GIXD patterns of the p-6P films with different thickness spanning the range of (b) 2 nm (c) 10 nm, and (d) 20 nm.



**Figure S2.** Normalized UV-Vis absorption spectra of the DNTT-only, DNTT/p-6p (2nm), DNTT/p-6p (10nm), and DNTT/p-6p (20nm) heterojunctions.



**Figure S3**. Transfer characteristics and temporal  $I_{DS}$  change of the (a, b) DNTT/p-6p (2nm), (c, d) DNTT/p-6p (10nm), and (e, f) DNTT/p-6p (20nm) heterojunction phototransistors at  $V_{DS} = -30$  V under blue light illumination (450 nm) with various pulse-width exhibiting memory mode.



**Figure S4**. EPSC variations of the photosynaptic transistors comprising the DNTTonly, DNTT/*p*-6*p* (2nm), DNTT/*p*-6*p* (10nm), and DNTT/*p*-6*p* (20nm) heterojunctions receiving 50 presynaptic light pulses (450 nm; pulse width: 50 ms) under an operating  $V_{DS}$  of -1 V.



**Figure S5**. The summarized statistical synaptic performance of the cells from the the studied photosynapse devices. ( $A_1$  and  $A_{50}/A_1$  ratio are defined as the EPSC change under a single light pulse (450 nm; width: 50 ms) and the EPSC ratio after the fiftieth pulse to the first pulse, respectively.)