Supporting Information

Mixed Receptors of AMPA and NMDA Emulated Using A 'Polka Dot'-Structured Two-Dimensional Conjugated Polymer-Based Artificial Synapse

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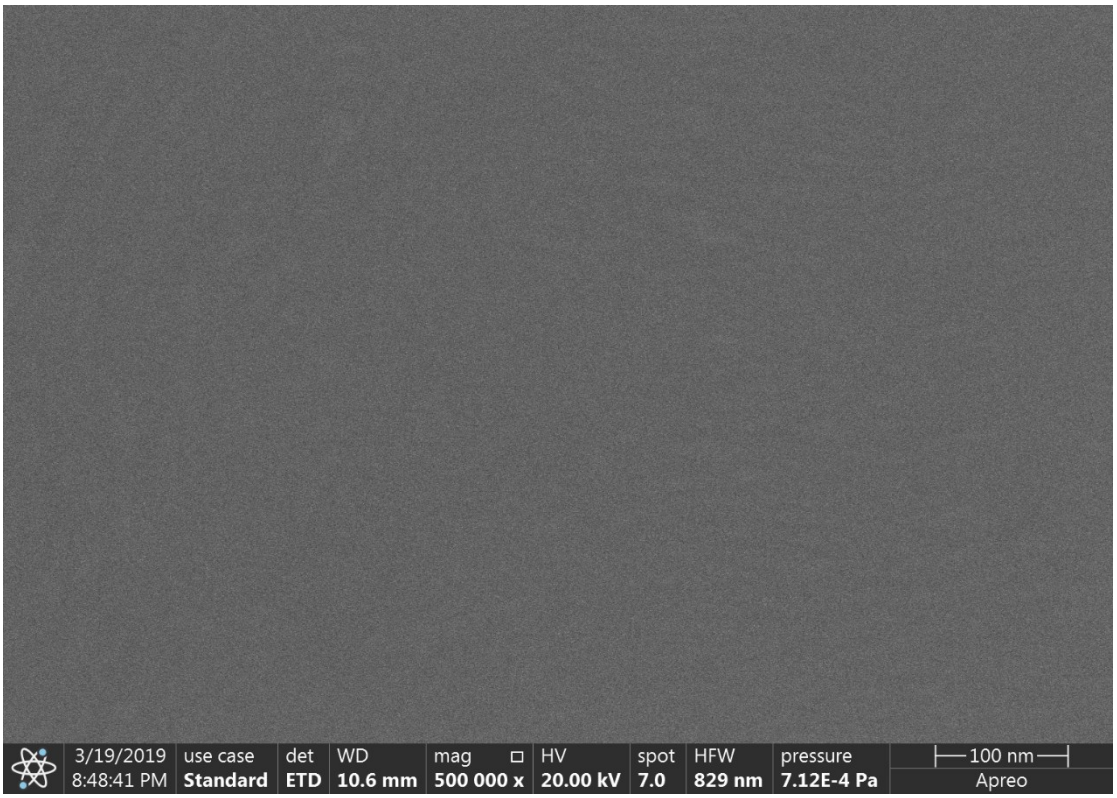

Fig. S1. SEM image of 2D P3HT film.

Equation S2. Calculate d-spacing and π–π distance of 2D P3HT film.

According to Bragg's law, d-spacing can be extracted from the peak value q_z in the out-of-plane direction, and π–π distance can be extracted from the peak value q_{xy} of the in-plane direction as

$$d = \frac{2\pi}{\theta} \tag{S2}$$

θ: x-ray beam incident angle

Equation S3. Calculation the free exciton bandwidth of 2D P3HT film.

According to Spano's model,[1, 2] interchain coupling leads to vibronic bands in the absorption spectrum. Also, the free exciton bandwidth W can be extracted from the values of vibronic bands; W is inversely related to intrachain ordering or exciton conjugation length along the backbone and its planarity. [3] Intensities of (0−0) and (0−1) transitions are used to calculate W as

$$\frac{I_{0-0}}{I_{0-1}} = \left\{\frac{1 - 0.24W/E_P}{1 + 0.073W/E_P}\right\}^2, \quad \text{(S3)}$$

where $I_{0–0}$ and $I_{0–1}$ represent the intensities of the (0−0) and (0−1) transitions, respectively. E_P – 0.18 eV is the vibrational energy of the symmetric vinyl stretch. The ultrathin film had W ~ 81 meV, and the thick P3HT film had W ~ 130 meV.

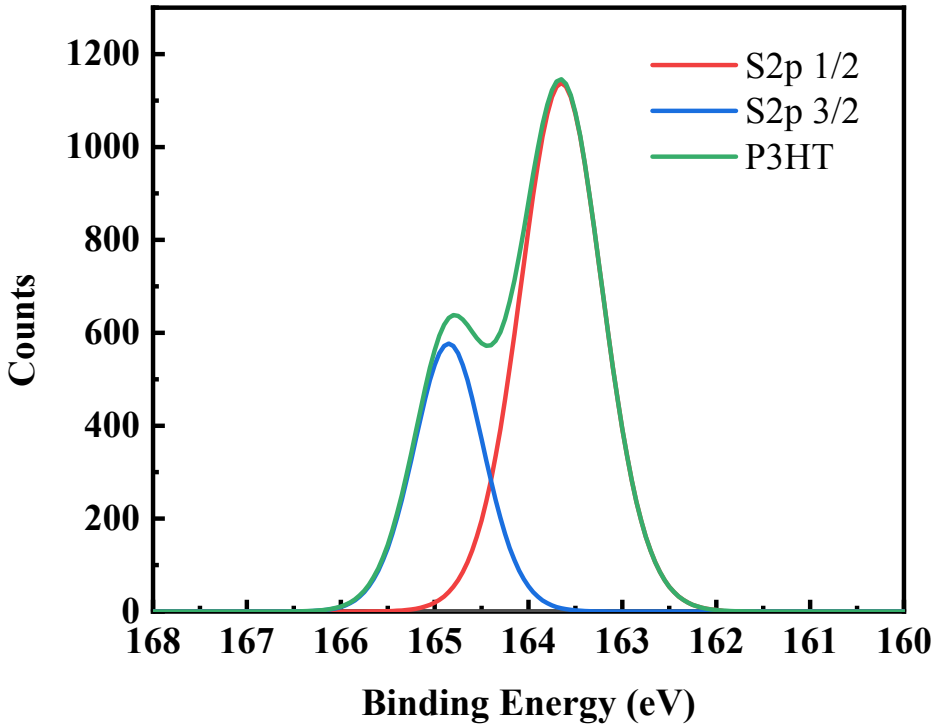

Fig. S4. XPS spectra of 2D P3HT film on SiO_2/Si substrates: S 2p photoelectron spectra.

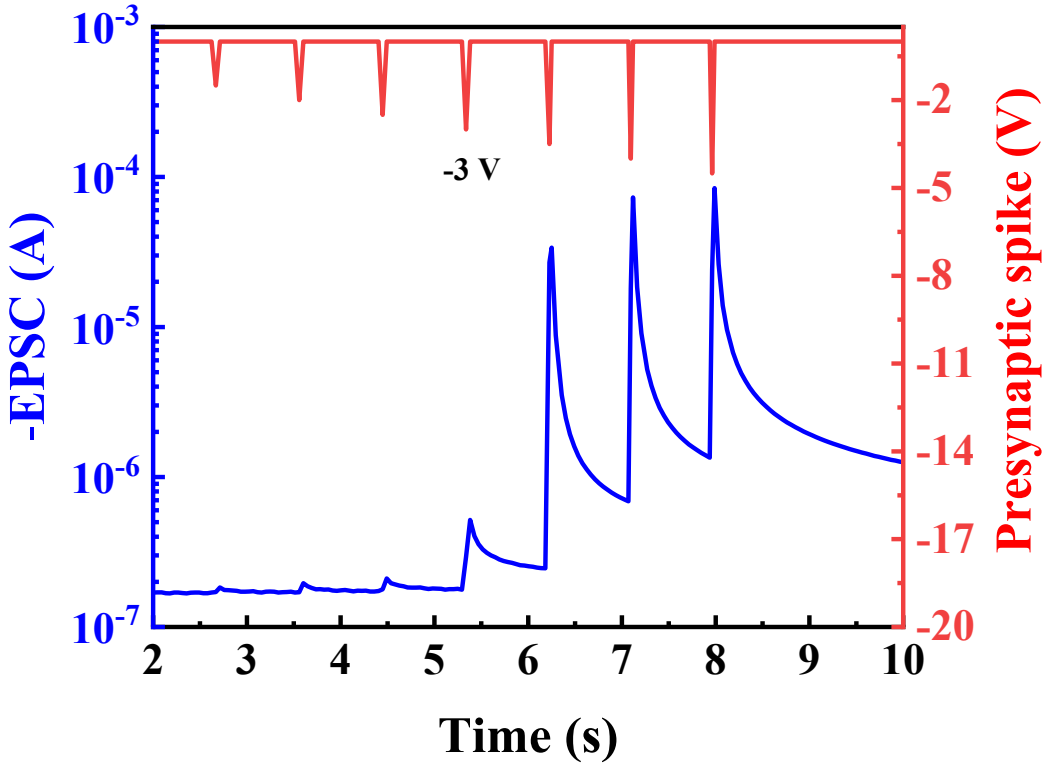

Fig. S5. EPSCs triggered by a series of gate voltage pulses with the same duration time (180 ms) and different amplitudes (-1.5 V, -2 V, -2.5 V, -3 V, -3.5 V, -4 V, -4.5 V).

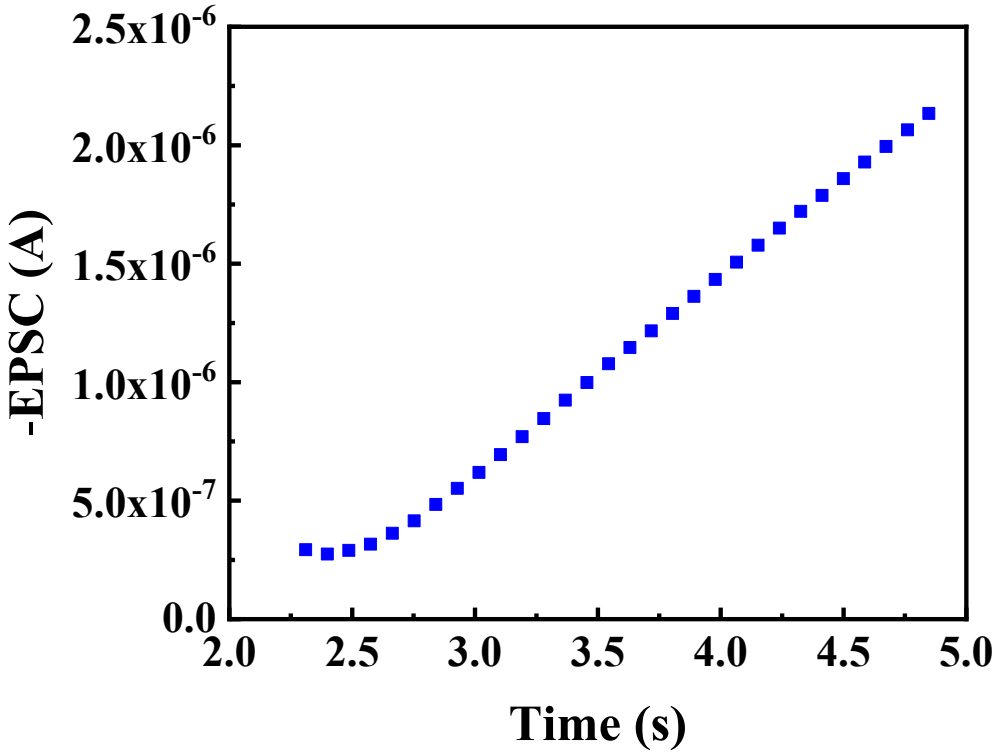

Fig. S6. Synaptic Potentiation triggered by 30 repeated gate voltage pulses (-3 V, 45 ms).

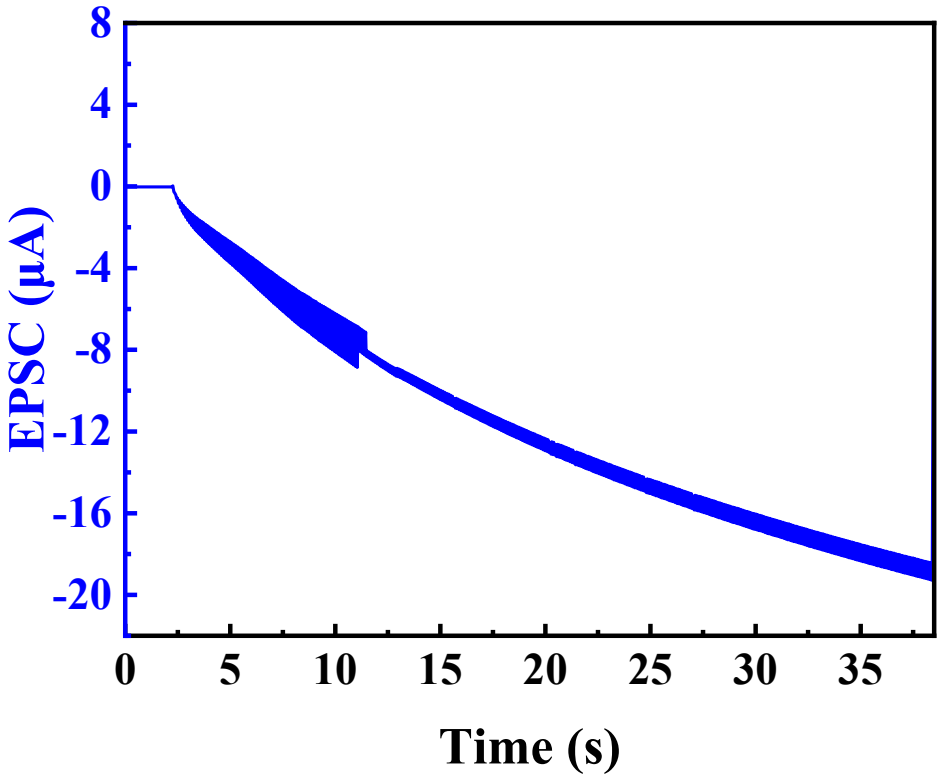

Fig. S7. Synaptic potentiation without saturation triggered by 400 repeated negative (-3 V, 45 ms) gate voltage pulses.

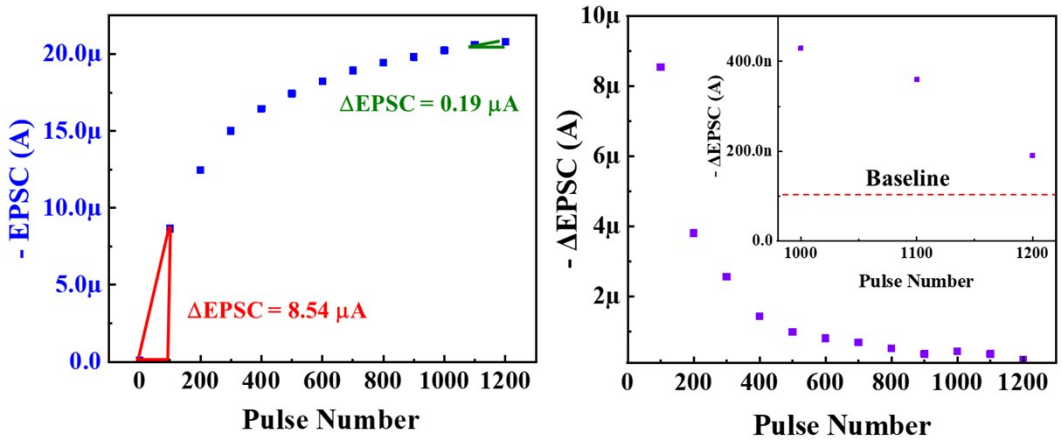

Fig. S8. EPSC and Δ EPSC triggered by 1200 repeated negative (-3 V, 50 ms) gate voltage pulses. ($\Delta EPSC = EPSC_n - EPSC_{n-100}$)

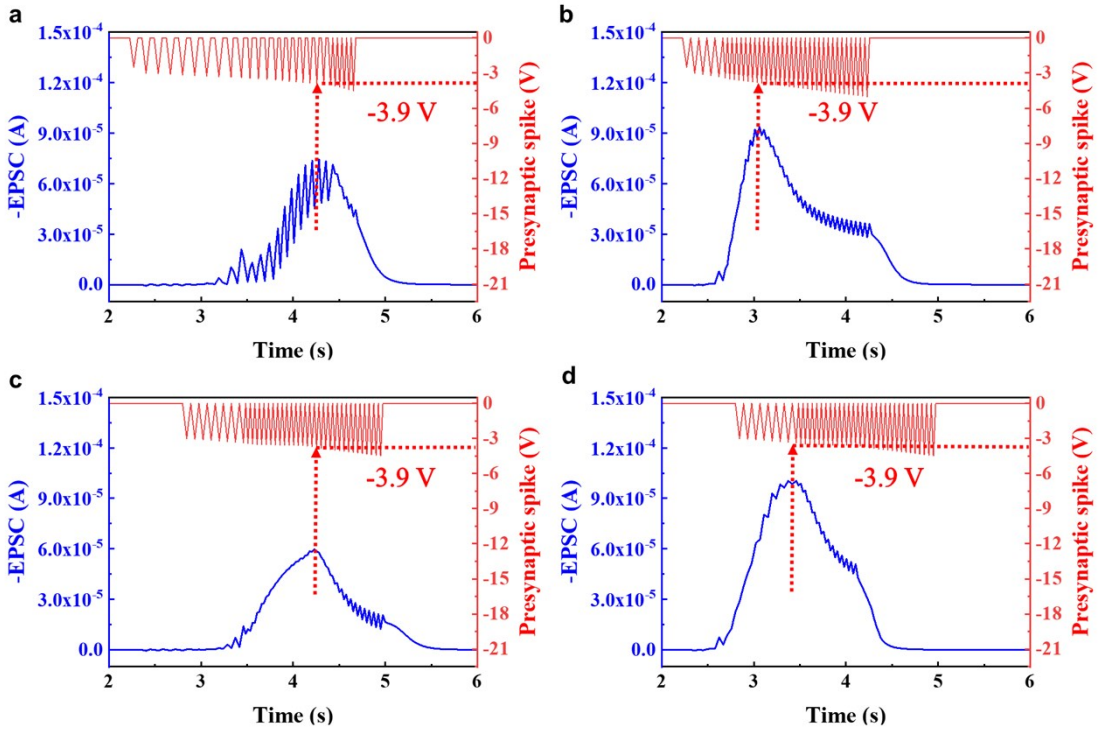

Fig. S9. Homeostatic plasticity triggered by gradually increased pulses with different intervals or steps. a) -2.5 V~ - 4.5 V gradually increased pulses. b) -2.5 V~ - 5 V gradually increased pulses. c) -3 V~ - 4.5 V gradually increased pulses. d) -3 V~ - 4.5 V gradually increased pulses. The threshold value of -3.9 V was observed in several measurements in which presynaptic spikes with incremental amplitudes were applied.

S10. Simulation Methods

First principles calculations were employed to distinguish interactions between ions and polymer chains with the projector augmented wave (PAW) method, exchange correlation functional based on density functional theory (DFT) and Perdew-Burke-Ernzerhof (PBE) as the exchange-correlation functional of choice. The cutoff energy for the plane-wave basis is set to be 400 eV. The convergence criterion of the total energy was set to be 10^{-5} eV in the self-consistent field iteration. In addition, the Monkhorst-Pack k-mesh of 3×3×1 is used for our surface structures. The tetrahedron method with Bloch corrections was used for smearing for our structures. All structures were optimized until the self-consistent force is less than 0.05 eV·Å^{-1} and the energy between two consecutive steps is less than 10^{-6} eV.

In our work, the P3HT structures were optimized based on first-principles. It is known that the amorphous structure can be obtained using ab initio molecular dynamics (AIMD) methods. In other words, the crystal structure will be transformed into an amorphous structure by simulating a liquid phase of the desired composition and quench it in order to find a non-crystalline local minimum. Therefore, the supercell has been simulated annealing to produce the amorphous P3HT with 10 ps at canonical ensemble (NVT) using AIMD, compatibly with computational limitations.

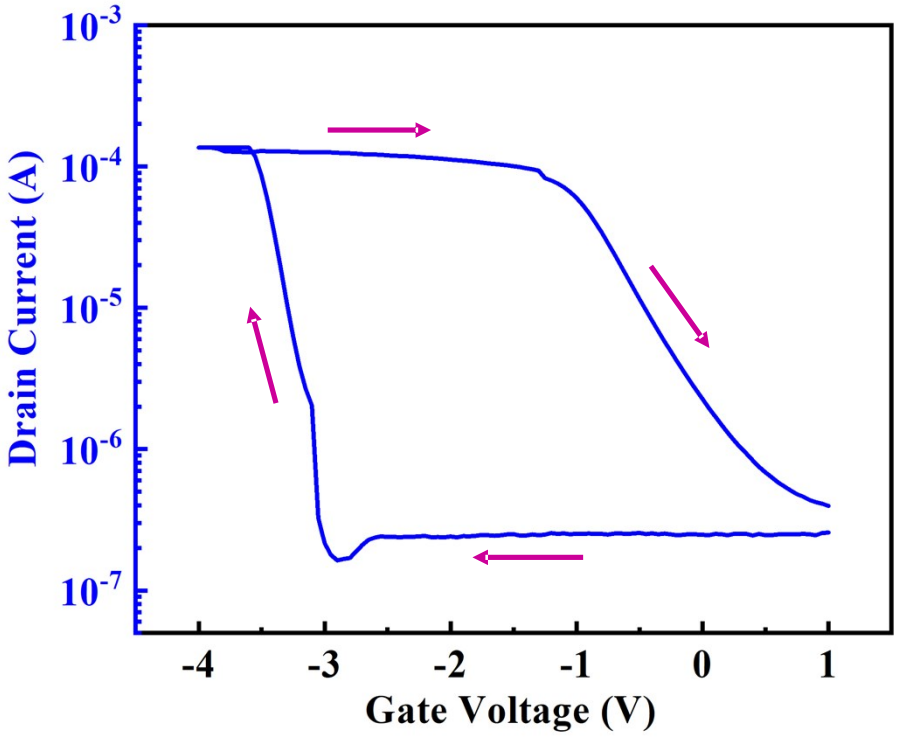

Fig.S11. Transfer curve of 2D P3HT synaptic transistor.

References

[S1]J. Clark, J.-F. Chang, F. C. Spano, R. H. Friend, C. Silva, *Applied Physics Letters*, 2009, **94***,* 117.

[S2]E. T. Niles, J. D. Roehling, H. Yamagata, A. J. Wise, F. C. Spano, A. J. Moulé, J. K. Grey, *J. Phys. Chem. Lett*, 2012, **3**, 259.

[S3]W. Wang, S. Guo, E. M. Herzig, K. Sarkar, M. Schindler, D. Magerl, M. Philipp, J. Perlich, P. Müller-Buschbaum, *J. Mater. Chem. A*, 2016, **4**, 3743.