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## SUPPLIMENTARY INFORMATION

Title : Combined optical and electrical control of a low-power consuming (  $\sim$  fJ) two-terminal organic artificial synapse for associative learning and neuromorphic applications

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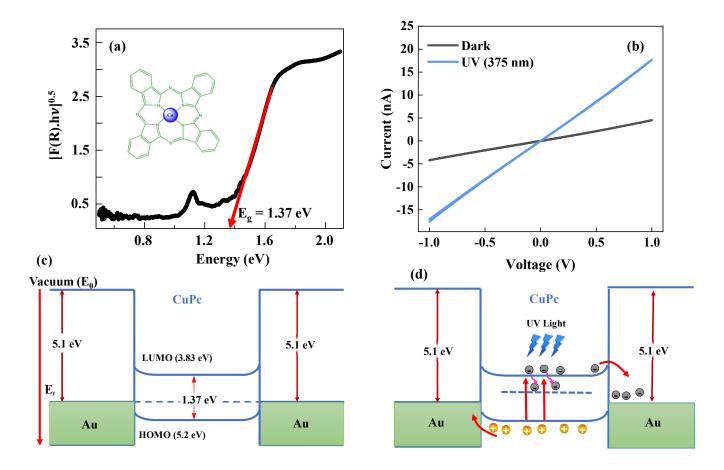


Figure S1 : (a) Kubelka – Munk function for calculation of optical energy gap of CuPc. The optical energy gap  $\sim 1.37$  eV, which is extracted from the linear extrapolation of the Kubelka – Munk function (b) Typical I-V curve at 1V in the dark and under 375 nm UV light. (c) Stationary energy level diagram of Au-CuPc-Au in the dark (d) The Energy level diagram when synaptic device is exposed to UV-light.

## Note S1. Calculation of Energy consumption per spike in the CuPc optical synapse

Energy consumption per spike can be evaluated using the standard computation

 $dE = V_{read} \times I \times dt$ ;

Here, 'V' is the read voltage of the device, 'I' is the current produced by the device and 'dt' is the duration of the optical spike. We use the read voltage here as the device is stimulated using optical pulses. In this case, the energy consumption is the energy corresponding to the electrical response of an optoelectronic synaptic device.

For Example: with the use of an optical pulse of dt = 40 ms;  $V_{read} = 0.1 \text{ Volts}$ 

Energy consumption by the device:

 $dE = V \times I \times dt = 0.1 \ V \times 0.1076 \times 10^{-9} \ A \times 0.04s = 430.4 \ fJ$ 

By reducing the dt to less than 1 ms, the energy consumption can be reduced to less than 100 fJ.

Туре	Device structure	Wavelength	Synaptic functionalities	Energy Consumption	References
Organic	Three Terminal	360 nm	STP/LTP	~ 42 nJ	1
Organic-inorganic	Two Terminal	470 nm	STP/LTP	~ 9 nJ	2
Organic-Inorganic	Three Terminal	500 nm	STP/LTP	~ 0.65 nJ	3
Organic-Inorganic	Three Terminal	532 nm	STP/LTP	~ 10 pJ	4
Organic	Three Terminal	UV.	SDDP/SNDP/light intensity dependent plasticity	~ 18.06 fJ	5
Organic-Inorganic	Three Terminal	UV–vis	EPSC/STP/LTP/Image processing	~ 1.3 fJ	6
Organic	Two Terminal	Visible	STM/LTM	~ 0.3 µJ	7
Organic	Two terminal	445 nm and 660 nm	EPSC/IPSC/PPF/Pain perception	~ 40 nJ	8
Inorganic	Two terminal	550 nm	STP/LTP/Image learning	~ 68.9 aJ	9
Organic	Two Terminal	UV-375 nm	STP/LTP/SDDP/SNDP/SRDP /light intensity dependent plasticity/Associative learning	~ 430 fJ	Our Work
	Organic Organic-inorganic Organic-Inorganic Organic-Inorganic Organic Organic Organic Inorganic	StructureOrganicThree TerminalOrganic-inorganicTwo TerminalOrganic-InorganicThree TerminalOrganic-InorganicThree TerminalOrganic-InorganicThree TerminalOrganic-InorganicThree TerminalOrganic-InorganicThree TerminalOrganic-InorganicThree TerminalInorganicTwo TerminalInorganicTwo terminal	StructureOrganicThree Terminal360 nmOrganic-inorganicTwo Terminal470 nmOrganic-InorganicThree Terminal500 nmOrganic-InorganicThree Terminal532 nmOrganic-InorganicThree TerminalUV.Organic-InorganicThree TerminalUV.Organic-InorganicThree TerminalUV.Organic-InorganicThree TerminalUV-visOrganic-InorganicTwo TerminalUV-visInorganicTwo terminal550 nm	StructureImage: StructureOrganicThree Terminal360 nmSTP/LTPOrganic-inorganicTwo Terminal470 nmSTP/LTPOrganic-InorganicThree Terminal500 nmSTP/LTPOrganic-InorganicThree Terminal532 nmSTP/LTPOrganic-InorganicThree TerminalUV.SDDP/SNDP/light intensity dependent plasticityOrganic-InorganicThree TerminalUV-visEPSC/STP/LTP/Image processingOrganicTwo TerminalVisibleSTM/LTMOrganicTwo terminal445 nm and 660 nmEPSC/IPSC/PPF/Pain perceptionInorganicTwo terminal550 nmSTP/LTP/Image learningOrganicTwo terminal550 nmSTP/LTP/Image learningOrganicTwo terminal550 nmSTP/LTP/SDDP/SNDP/SRDPOrganicTwo terminalUV-375 nmSTP/LTP/SDDP/SNDP/SRDP	StructureImage: StructureStructureOrganicThree Terminal360 nmSTP/LTP~42 nJOrganic-inorganicTwo Terminal470 nmSTP/LTP~9 nJOrganic-InorganicThree Terminal500 nmSTP/LTP~0.65 nJOrganic-InorganicThree Terminal532 nmSTP/LTP~10 pJOrganicThree TerminalUV.SDDP/SNDP/Ight intensity dependent plasticity~18.06 fJOrganicThree TerminalUV-visEPSC/STP/LTP/Image processing~1.3 fJOrganicTwo TerminalVisibleSTM/LTM~0.3 µJOrganicTwo terminal445 nm and 660 nmEPSC/IPSC/PF/Pain perception~40 nJInorganicTwo terminal550 nmSTP/LTP/Image learning~68.9 aJOrganicTwo terminalUV-375 nmSTP/LTP/SDDP/SNDP/SRDP /ight intensity dependent~430 fJ

## Table 1 : Energy consumption per spike for various devices. The organic/organic-inorganic optical synapses are listed in this table

Reference 9 is an complete inorganic synaptic device, but it has been kept for a comparative study.

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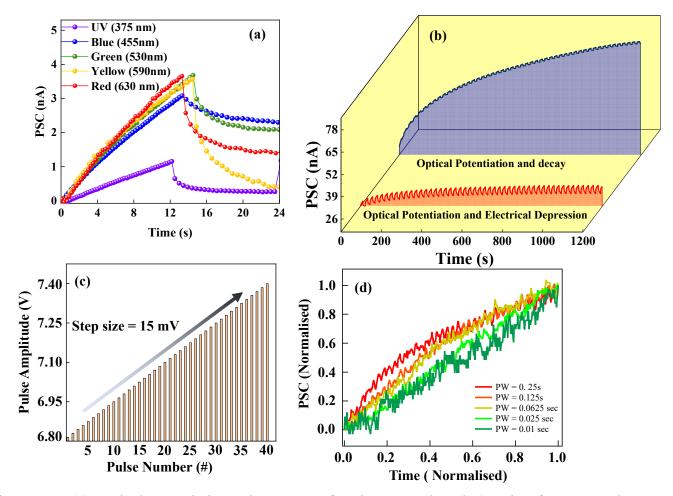


Figure S2 : (a) Optical potentiation using LEDs of various wavelength (starting from UV, Blue, Green, Yellow, Red). The intensity of UV LED was 32.45  $\mu$ W/cm<sup>2</sup> and all other LEDs were 3.16 mW/cm<sup>2</sup>. (b) 3D representation of 50 cycles of optical potentiation and normal decay along side optical potentiation and electrical depression using negative pulse (Vr = 1V, Vp = -0.5mV) (c) Effect of non-linear pulse sequence on potentiation. (a) The non-ideal optical pulses being applied for potentiation, 40 pulses having pulse amplitude varying from 6.8 V to 7.4 V with each step increasing by 0.015 V. (b) Non-linear to linear synaptic weight update by utilization of non-ideal pulse sequence. The PSC is measured for variable light pulse duration as shown. The faster pulsing ( PW = 0.01 sec) leads us a linear weight update.

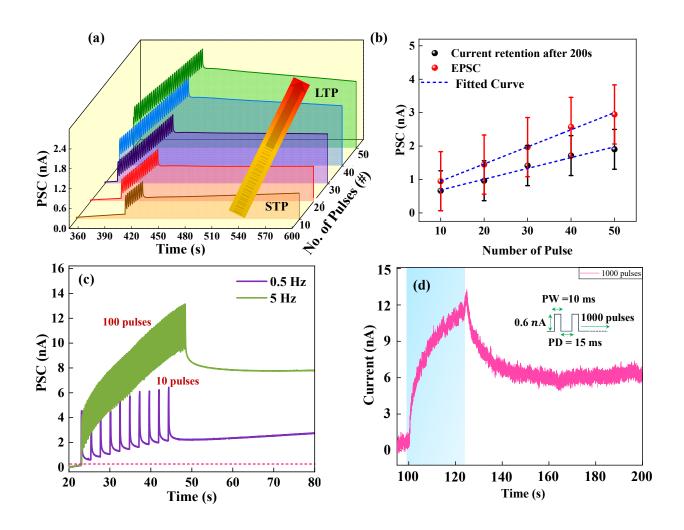


Figure S3 :(a) Transition from STP to LTP induced by increasing the number of pulses for a given UV light intensity 32.45 mW/cm<sup>2</sup> (b) The change in the EPSC and current retention (after 200s) with the number of optical light pulses (c) EPSCs with 375 nm light spiking at frequencies of 0.5 and 5 Hz for a duration of 20 s. This measurement shows by increasing the frequency by 10 folds EPSC has increased from 6.4 nm to 13.1 nm and current retention after 20 secs have increased from 2.5 nm to 7.7 nm. (d) Potentiation of the CuPc synapse by using 1000 optical pulses of intensity 32.45 $\mu$ W/cm<sup>2</sup> and pulse width 10 msec

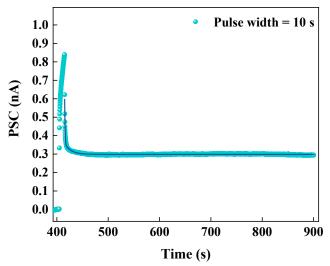


Fig. (S4) : Long-term potentiation observed in the case of optical stimulation. Pulse width =10 sec, Wavelength : 375 nm. Intensity  $32.45\mu$ W/cm<sup>2</sup>. The retained memory after 500 sec is about 35% and after one hour, it is about 26%.