
Organic optoelectronic synapse based on photon-modulated electrochemical doping

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Mechanism comparison between different types of optoelectronic synapses

According to previously reported works, there are mainly four types of mechanisms for optoelectronic synaptic devices. In type 1, electrochemical metallization/valence change mechanism, light induces metal ions/vacancy migration between two electrodes, which makes a current pathway. As a result, a nonlinear photonic response has been mainly observed: abrupt current increase and saturation. For type 2, phase change mechanism (PC), the weight tuning is usually realized by light-induced amorphous/crystallization phase change. In many cases, switching between levels of memory requires a single pulse with varying power for amorphization and recrystallization. Type 3 is based on persistent photoconductivity (PPC) of amorphous oxide semiconductors (AOS). The PPC is related to the ionization of oxygen vacancies and the formation of metastable peroxide. AOS typically shows relatively small conductance switching, as a small number of photocarriers are created by the photoionization of oxygen vacancies and ultraviolet light is usually needed. For type 4, interfacial charge trapping mechanism, light absorber materials are coupled with charge transfer material for light harvest and photocarriers detection in a three-terminal field-effect transistor structure. The interfacial charge trapping (CT) accounts for the memory formation. Due to the charge screening effects at the interfaces, weight change is comparatively distinguishable during the initial pulses, then becomes saturated with an increased number of pulses.

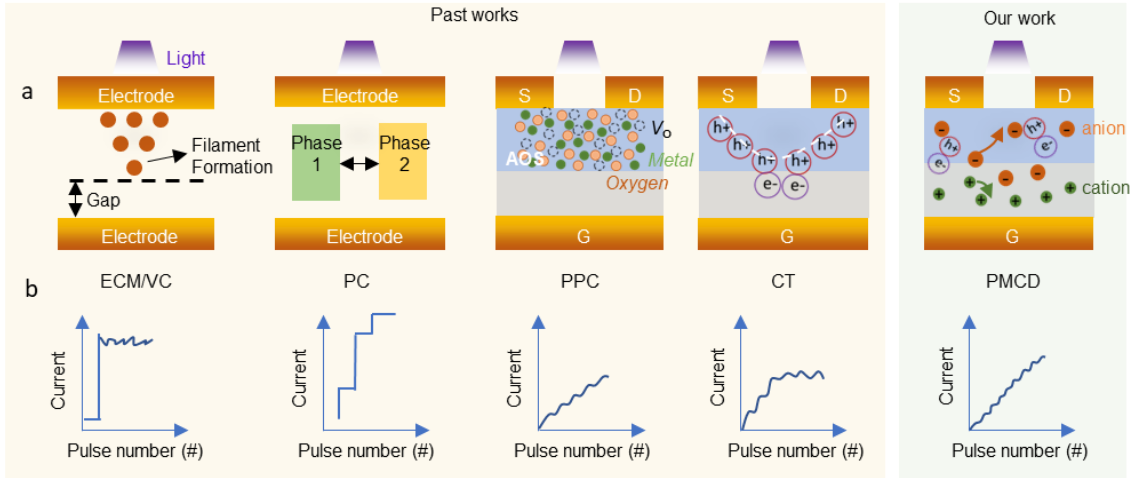


Figure S1. a. Device structures of optoelectronic synaptic devices based on working mechanism of (1) electrochemical metallization/valence change (ECM/VC), (2) phase change (PC), (3) persistent photoconductivity (PPC) of amorphous oxide semiconductors (AOS), (4) interfacial charge trapping (CT), and (5) photon-modulated electrochemical doping (PMCD). b. Typical photonic responses of the devices with these five different mechanisms.

Basic characteristics of the devices

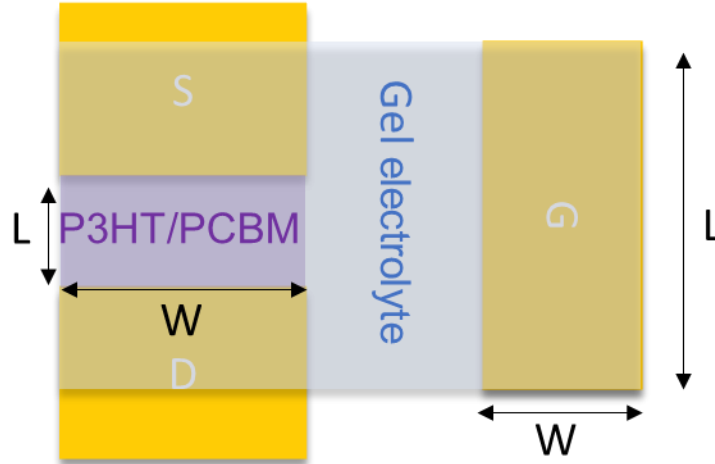


Figure S2. Device geometry and dimension parameters of the organic electrochemical optoelectronic synapse. The channel width (W) and length (L) are $1500\ \mu\text{m}$ and $100\ \mu\text{m}$, respectively. The gate width (W) and length (L) are $3\ \text{mm}$ and $7\ \text{mm}$, respectively.

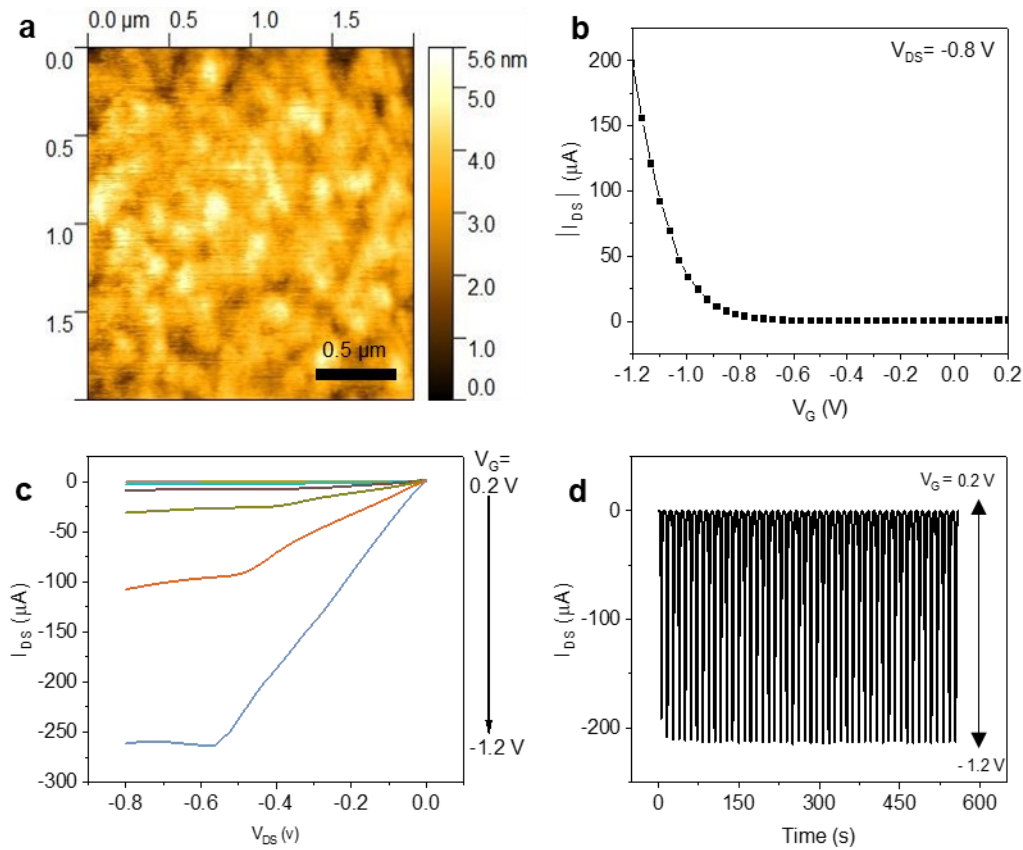


Figure S3. **a.** AFM images of P3HT/PCBM heterojunction layer. **b.** Transfer characteristics, **c.** output characteristics, and **d.** cycling stability of a typical optoelectronic synaptic device. In the cycling test, a constant V_{DS} at $-0.8\ \text{V}$ and a square wave function of V_G from $-0.2\ \text{V}$ to $1.2\ \text{V}$ were applied.

Photoresponsive characteristics of the devices

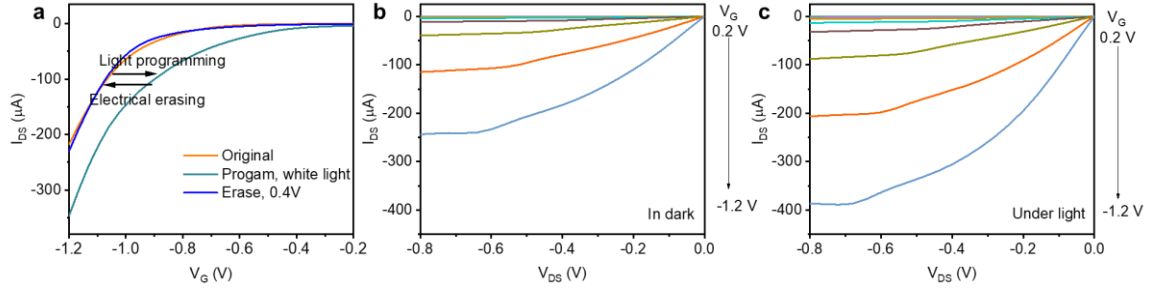


Figure S4. a. Transfer characteristics of the optoelectronic synaptic device with light programming and electrical erasing operations. Output characteristics of the device b. in dark and c. under light illumination.

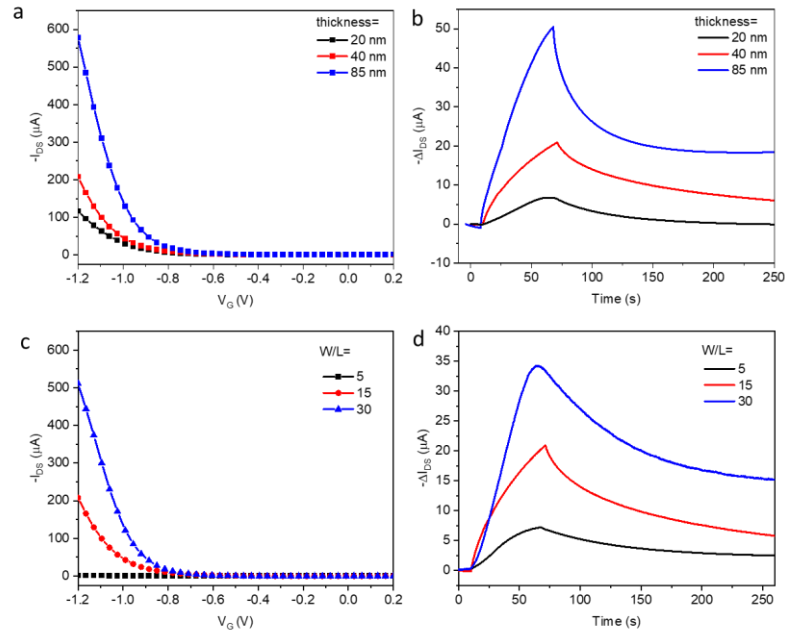


Figure S5. a. Transfer curves and b. photonic responses of the devices with different channel thicknesses. c. Transfer curves and d. photonic responses of the devices with different channel width/length ratios.

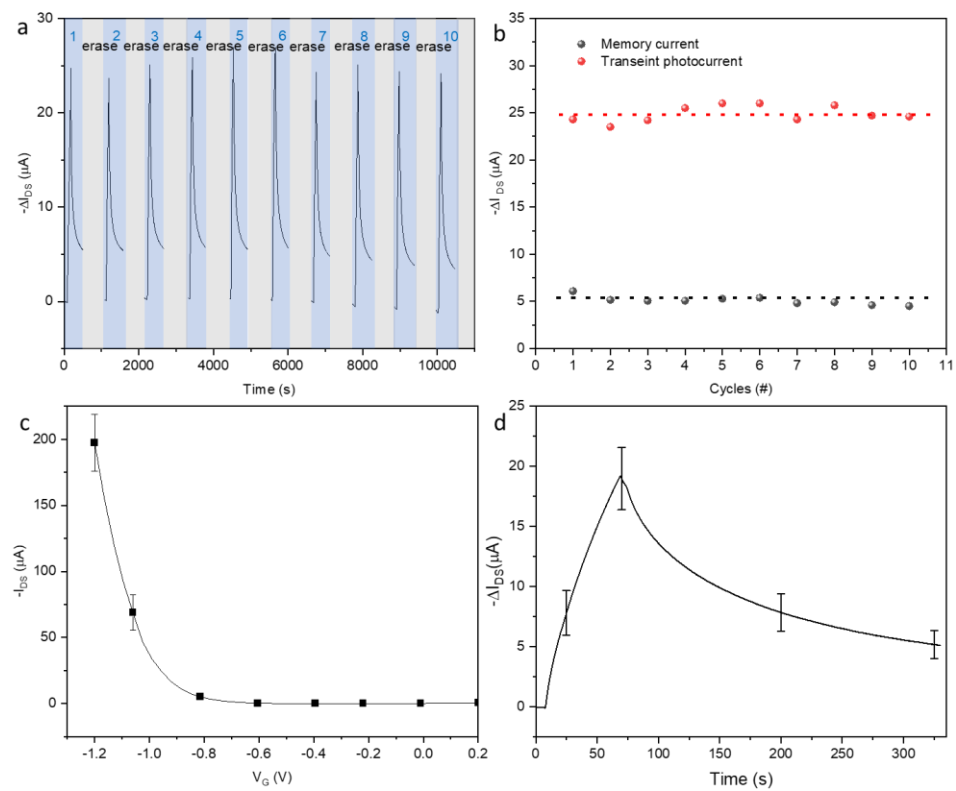


Figure S6. a. Ten cycles of writing and erasing. b. Transient photocurrents and memory currents extracted from Figure S6a. c. Transfer curve and d. photonic response with standard errors calculated from representative gate voltages and time stamps in 5 replicate devices. The error bars represent the standard errors of the mean from 5 measurements.

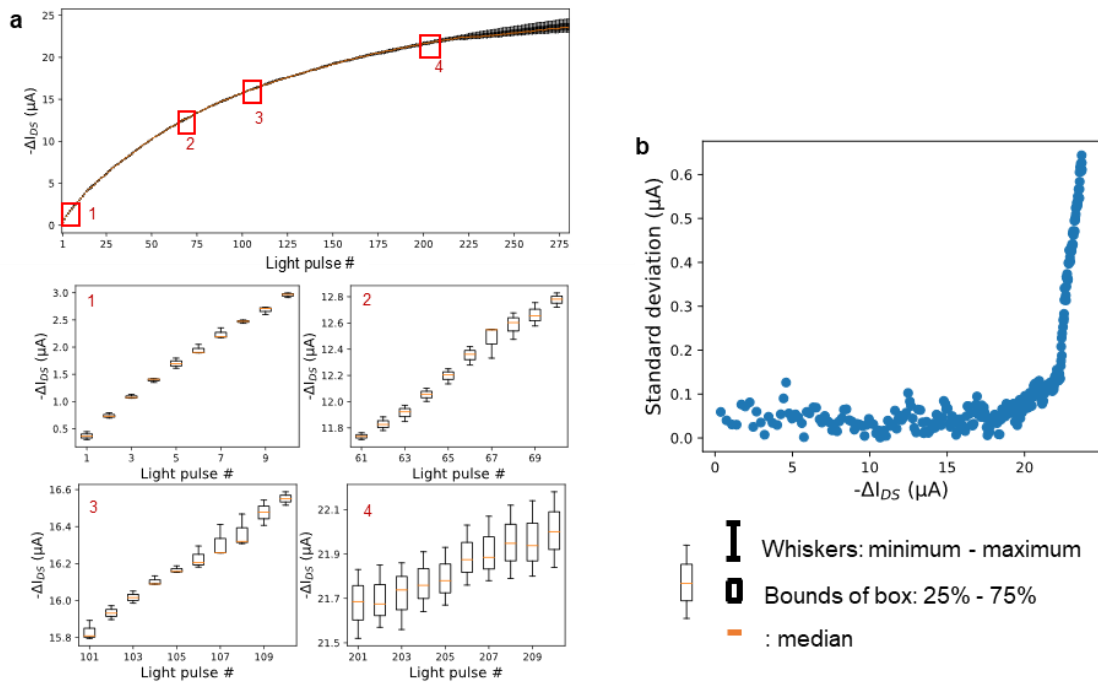


Figure S7. a. Evaluation of the distinguishability and reproducibility of the weight update in three cycles depicted in Figure 2b. The lower panels show the zoom-in views of region 1-4. Within a linear current increase range (0-15 μA), each state (indicated by organ line) is clearly distinct from others, with a small variation of around 0.1 μA . b. Standard deviation as a function of photocurrent change.

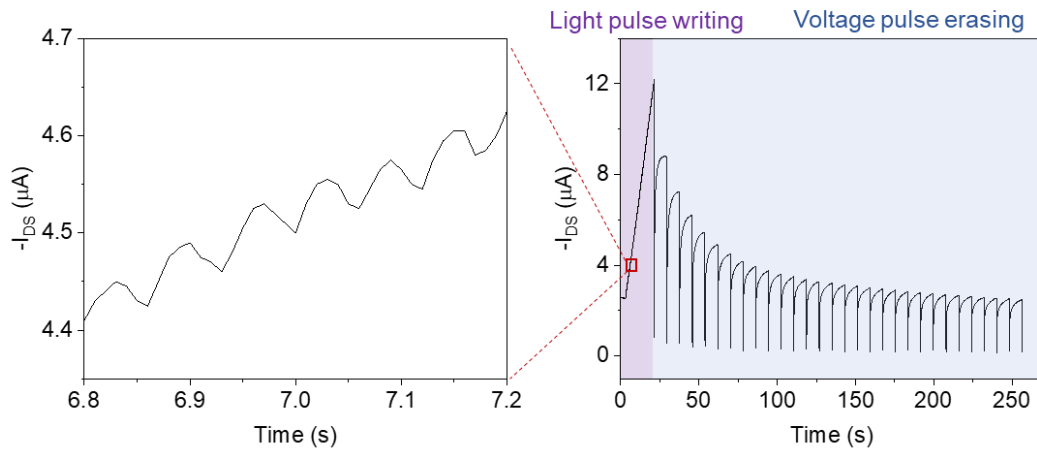


Figure S8. Photonic response under the application of consecutive light pulses and reverse voltage pulses. The light pulses have the intensity of 7.0 mW/cm², duration time of 0.04s, and interval time of 0.04s. The voltage pulses (0.4 V) have the duration time of 0.1s and interval time of 8s.

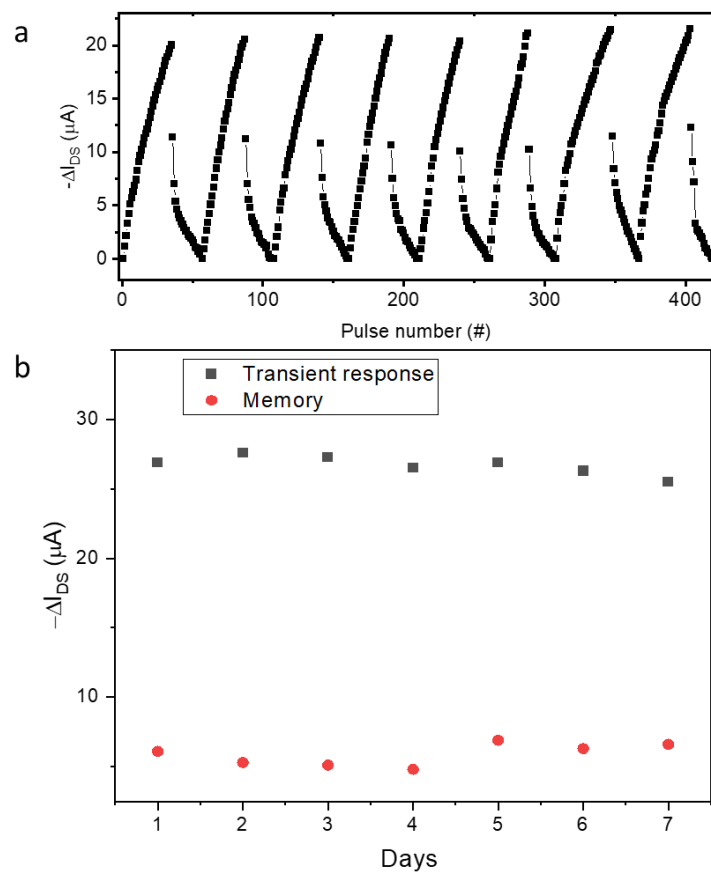


Figure S9. a. Continuous cycles of writing and erasing b. Photonic responses of the device that was stored and tested in ambient conditions across one week.

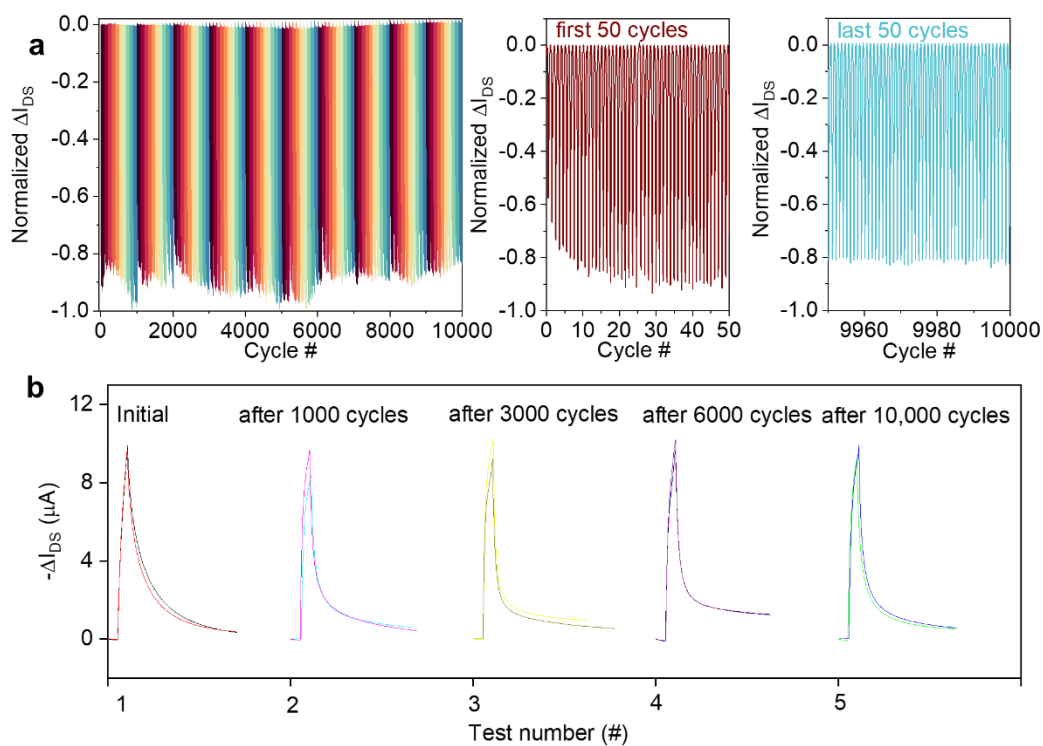


Figure S10. a. Normalized current change over 10,000 cycles of switch on (-0.8 V) and off (0.1 V). Left two panels show the zoom-in views of current change in the first and last 50 cycles. b. The photonic response of the device at the beginning, after 1000, 3000, 6000, and 10,000 electrochemical cycles.

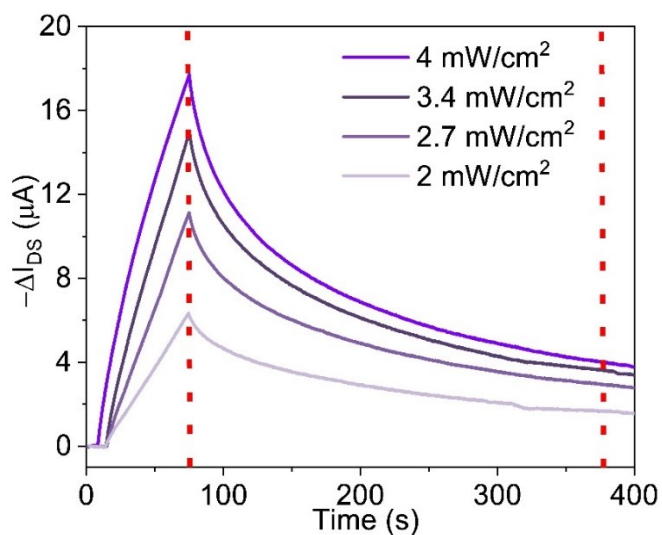


Figure S11. Light intensity-modulated photonic response. The current read at the dashed line corresponds to the transient and memory current in the main text, respectively.

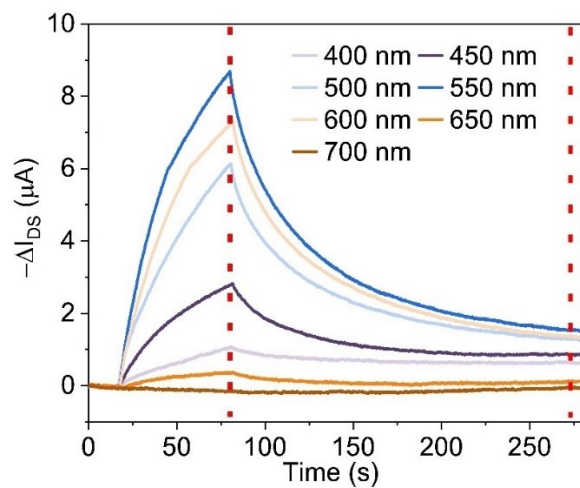


Figure S12. Light wavelength-modulated photonic response. The current read at the dashed line corresponds to the transient and memory current in the main text, respectively

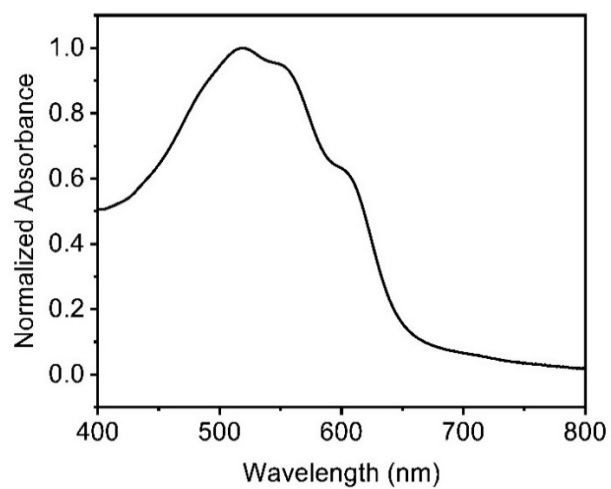


Figure S13. UV-Vis absorption spectrum of the P3HT/PCBM thin film.

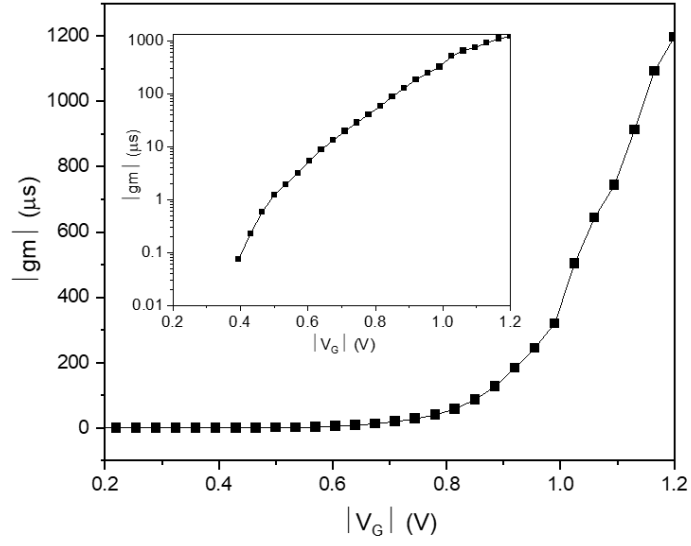


Figure S14. Transconductance values of the optoelectronic synaptic device. Inset figure shows the trend with log-scale of the transconductance values. Higher transconductance indicates that the device's signal amplification and sensitivity are enhanced.

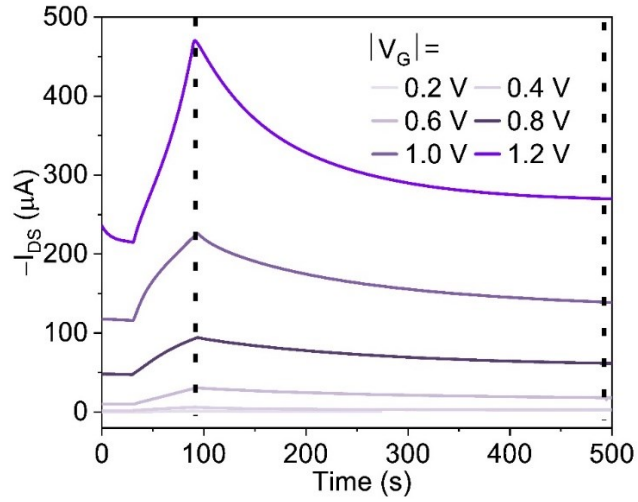


Figure S15. Gate voltage-modulated photonic response. The current read at the dashed line corresponds to the transient and memory current in the main text, respectively.

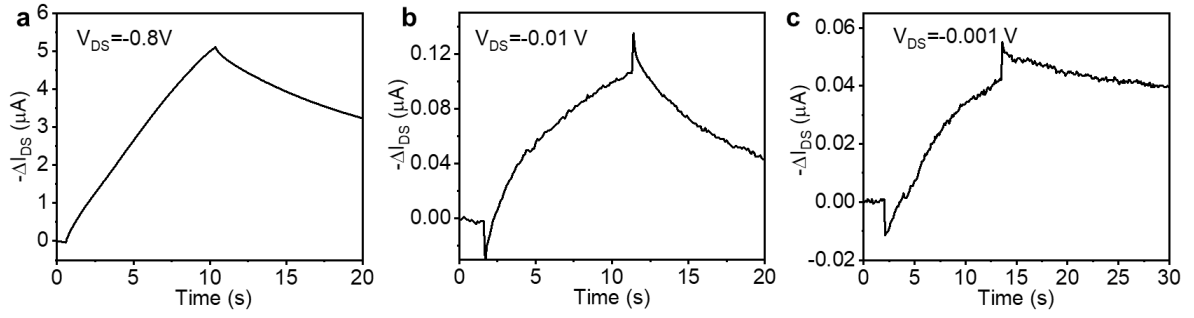


Figure S16. The photonic response of the same device to a light pulse (7 mw/cm^2 , 10 s) at different drain voltages V_{DS} . ($V_G = -0.6 \text{ V}$). These results show that the decrease of the drain voltage (from -0.8 V to -0.001 V) significantly reduces photocurrent, while the device maintains the typical photonic response and memory behavior. Thus, the power consumption of the device can be reduced by operating at lower drain voltage.

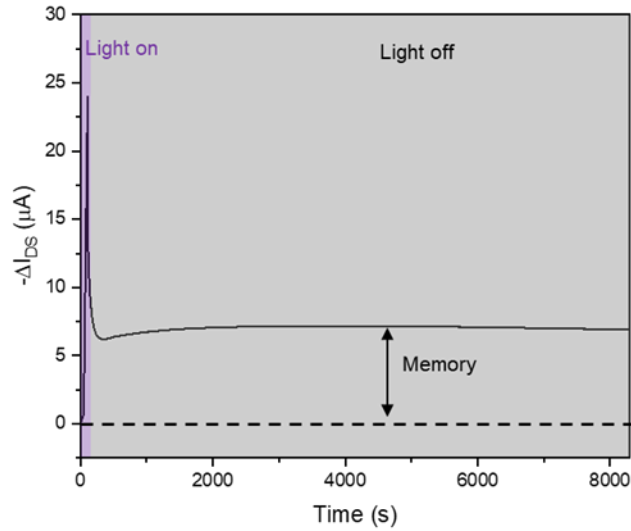


Figure S17. Memory retention characteristics of the synapse device after responding to a 60-second light pulse.

Working mechanism of optoelectronic synaptic behaviors

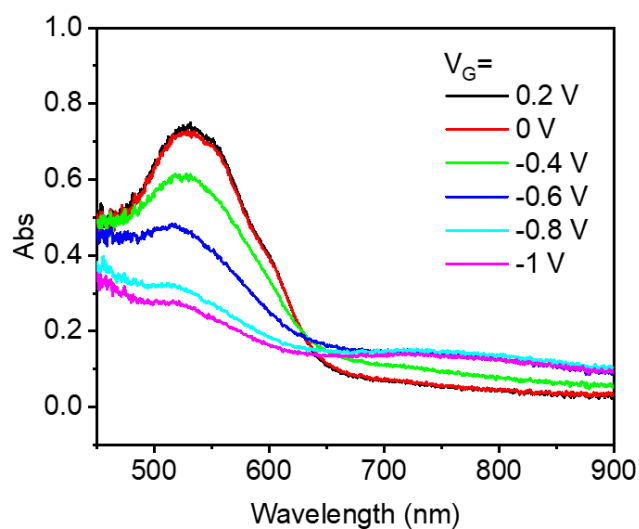


Figure S18. Spectroelectrochemistry of the P3HT/PCBM film recorded at varying gate voltages.

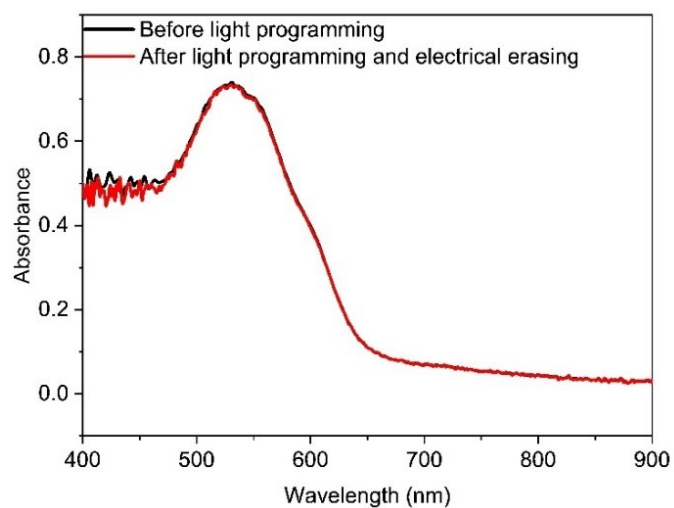


Figure S19. Absorbance spectra of P3HT/PCBM film before light programming and after electrical erasing.

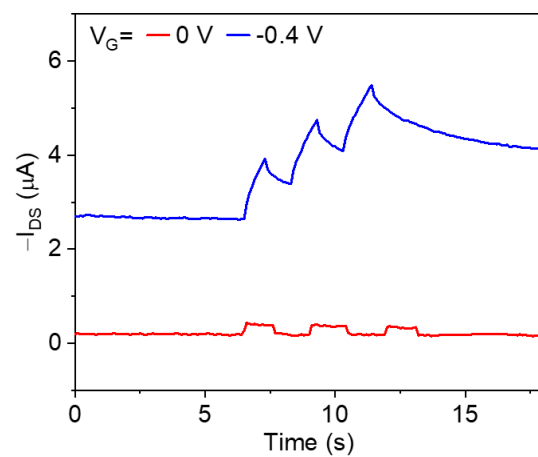


Figure S20. Photonic response to three light pulses at various gate voltages. At gate voltage of 0 V, where the ion diffusion (TFSI⁻) is limited, a negligible current change was recorded.

Pump-probe femtosecond transient absorption (TA)

As shown below, the TA spectra obtained with an excitation pulse of 532 nm show that both films, P3HT/PCBM with and without electrolyte, initially displayed broad absorption band at around 1300 nm after photoexcitation, which decays rapidly on the picosecond time scale (Figure S21a and Figure S21b). A relatively long-lived absorption band was observed at around 1000 nm at a later time stage. According to literature, the peak at around 1300 nm can be ascribed to the P3HT singlet exciton and the one at around 1000 nm to the PCBM anion radical with a contribution from P3HT polaron.^{1,2} To probe the electrolyte effect on these charge carriers' lifetime, the decay dynamics of the polaron/PCBM anion radical peak at 1049 nm was compared for the pristine film and the film with electrolyte (Figure S21c).

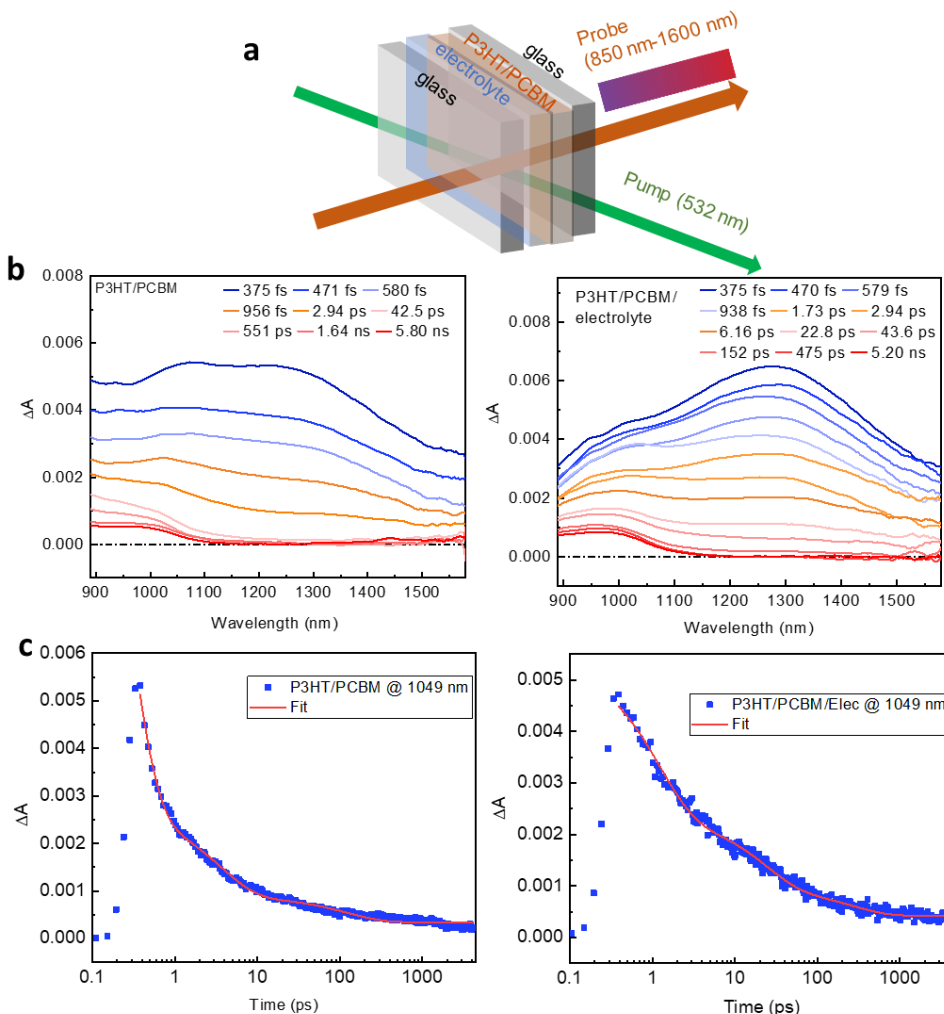


Figure S21. a. Schematic illustration of transient spectroscopy measurement. b. Transient spectra of P3HT/PCBM and P3HT/PCBM/electrolyte at various decay time. c. Decay dynamics of P3HT/PCBM and P3HT/PCBM/electrolyte are fitted using triexponential equations with a constant non-decaying component. Fitting parameters are shown in Table S1.

Table S1. Kinetic values obtained by a triexponential fit of the decay at 1049 nm^a

λ_{probe} (nm)	Film	$\tau_1(\text{ps})/(\%A)$	$\tau_2(\text{ps})/(\%A)$	$\tau_3(\text{ps})/(\%A)$	Offset(%A)
1049	P3HT/PCBM	0.2 (86.2)	3.6 (9.2)	125 (2.8)	3.4E-4 (1.8)
1049	P3HT/PCBM/Elec	1.2 (56.7)	23 (25.5)	264.6 (10)	4.2E-4 (7.8)

a: Fit formula, $Y = A1 \cdot \exp(-t/\tau1) + A2 \cdot \exp(-t/\tau2) + A3 \cdot \exp(-t/\tau3) + A_{\text{offset}}$

Reference

1. Guo, J., Ohkita, H., Bente, H. & Ito, S. Near-IR Femtosecond Transient Absorption Spectroscopy of Ultrafast Polaron and Triplet Exciton Formation in Polythiophene Films with Different Regioregularities. *J. Am. Chem. Soc.* **131**, 16869–16880 (2009).
2. Guo, J., Ohkita, H., Bente, H. & Ito, S. Charge Generation and Recombination Dynamics in Poly(3-hexylthiophene)/Fullerene Blend Films with Different Regioregularities and Morphologies. *J. Am. Chem. Soc.* **132**, 6154–6164 (2010).

Optoelectronic synaptic behaviors with various electrolyte

We studied photonic synaptic behaviors with different electrolyte: LiPF₆, LiTFSI, and TBACl. As shown in **Figure S22a**, the drain current as well as threshold voltages of the devices varies with electrolyte in transfer curve. This is mainly due to the fact that different dopant anions have various doping densities and require different potentials to be injected into the channel.¹ Thus, we anticipate this difference will also be reflected in photonic response. As shown in **Figure S22b**, under the same applied voltage and light illumination, the device with Cl⁻ anion did not display any photonic response, since Cl⁻ cannot effectively permeate into and dope the film.¹ On the other hand, in the presence of PF₆⁻, the device exhibited the largest photocurrent since PF₆⁻ can easily penetrate the polymer and perturb the electrochemical doping. The significant photoresponse enables the device to respond to fast writing pulses (120 μs) while retaining distinguishable states (**Figure S23**). Previous literature states that when conjugated polymers are immersed in concentrated LiPF₆, solvent and anions can spontaneously penetrate the polymer and form extended double layer, which may favor the electron depletion.² Consistent with the reports, we found that when LiPF₆ was employed as the electrolyte, the device exhibited a considerable current in response to the ambient light without any gate voltage stress. As demonstrated in **Figure 22c and Supporting video**, the device shows a sensible response to the hand waves, where the device was repeatedly exposed to the ambient light. Besides ambient light detection, we also show that the device is capable of responding to more dime light in **Figure S24**. This again demonstrated the importance of anion transport in photonic response and memory effects. Cations have less effect on the device performance as investigated and discussed in **Figure S25 and Table S2**.

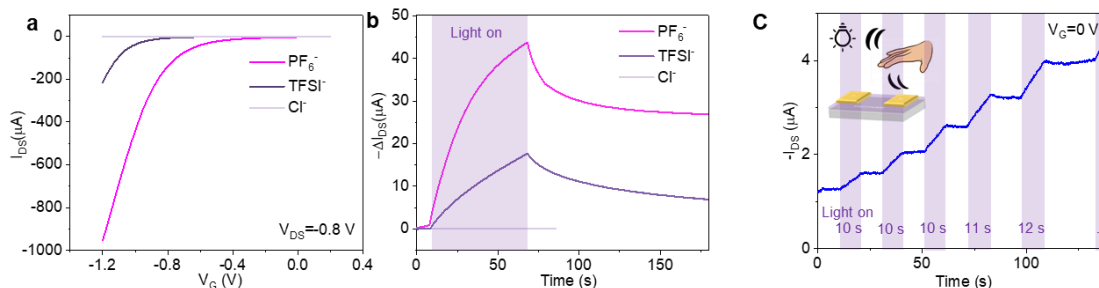


Figure S22. a. Transfer characteristics and b. photonic response ($V_G = -0.6$ V and $V_{DS} = -0.8$ V) of the devices in different electrolytes. c. Photonic response of the device to hand waves in an ambient light environment

with LiPF_6 based gel electrolyte. The purple areas indicate the periods when the device was repeatedly exposed to ambient light during the hand waves.

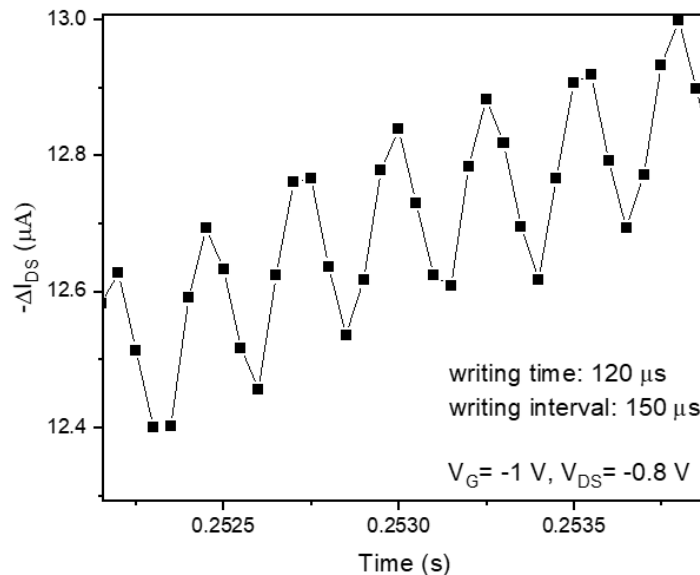


Figure S23. Drain current in response to fast light pulses.

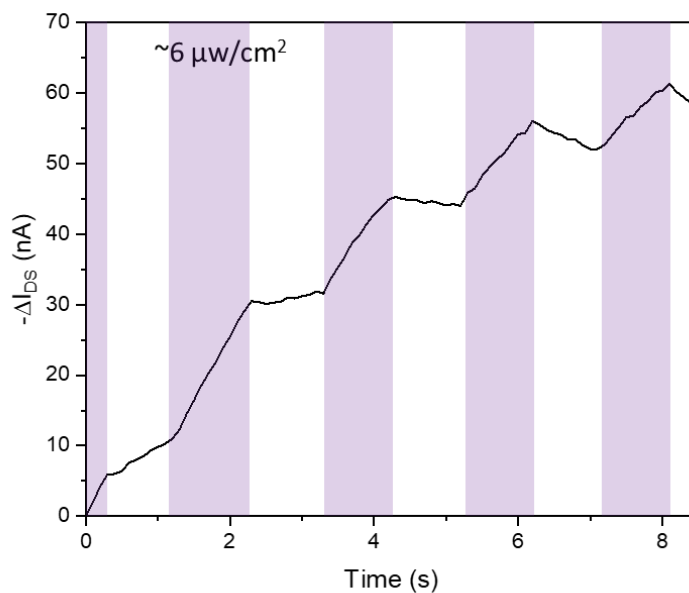


Figure S24. Photonic response to light pulses with light intensity of $6 \mu\text{W}/\text{cm}^2$.

To study the cation influence on synaptic behavior, we chose four different cations with various ionic radius and paired them with the same anion (TFIS^-) for the device test. These four cations are: Li^+ , Na^+ , tetramethylammonium (TEA^+), tetrabutylammonium (TBA^+) and their ionic radius are shown in **Table S2**. According to transfer curve in **Figure S25a**, the device shows similar threshold voltage and current with Li^+ ,

Na^+ , TEA^+ , which indicates the channel material experiences similar doping behaviors with these four cations. This is most likely due to the fact that electrochemical doping of the channel materials mainly needs anion for charge compensation. As a result, the cation has less impact on the OECT performance. Accordingly, the photonic responses of the device are also similar with these three cations, as shown in **Figure S25b**. However, when TBA^+ was used in electrolyte, the threshold voltage negatively shifted and drain current reduced in the transfer curve in comparison to the other three cations. This is mostly due to the especially large radius of TBA^+ causing high hydrodynamic resistance in the electrolyte and limiting the ionic mobility/conductivity. This trend is also reflected in photonic response, as TBA^+ has the smallest photonic response and memory.

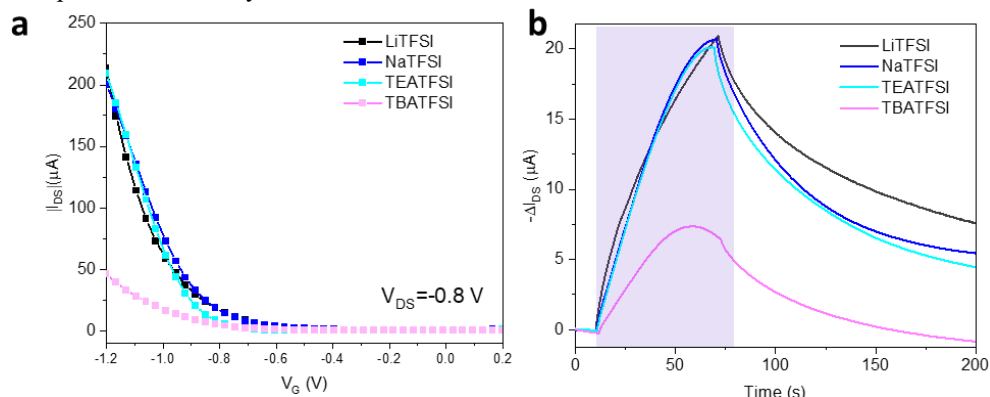


Figure S25. Transfer characteristics (a) and photonic response (b) of the devices ($V_G = -0.6$ V and $V_{DS} = -0.8$ V) in different electrolytes.

Table S2. The ionic radius of four different cations

Cation	Ionic radii, r/nm
Li^+	0.069
Na^+	0.102
TEA^+	0.337
TBA^+	0.413

Reference

1. Flagg, L. Q., Giridharagopal, R., Guo, J. & Ginger, D. S. Anion-Dependent Doping and Charge Transport in Organic Electrochemical Transistors. *Chem. Mater.* **30**, 5380–5389 (2018).
2. Li, X., Wang, X., You, L., Zhao, K. & Mei, J. Improving Electrochemical Cycling Stability of Conjugated Yellow-to-Transmissive Electrochromic Polymers by Regulating Effective Overpotentials. *ACS Materials Lett.* **4**, 336–342 (2022).

Constructing synaptic arrays for image recognition

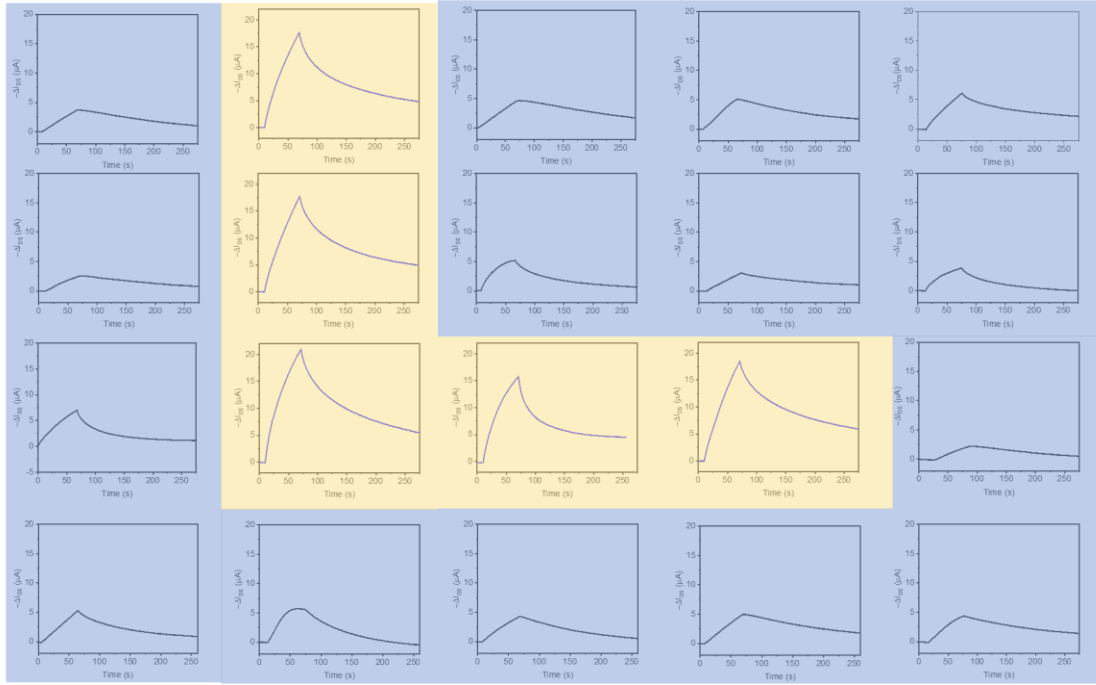


Figure S26. Photonic response of synaptic devices in a 4×5 synaptic array. The synapses at yellow regions are illuminated with light at intensity of 7.0 mW/cm², while the light illuminations at blue regions are at 2.0 mW/cm².

Reducing device's footprint for high-density integration

To reduce the device footprint, we first used the typical photolithography for microelectrode patterning, as shown in Figure S27 processes (1)-(3). Then a crosslinker molecule, bis(fluorophenyl azide) (bisFA), was blended with P3HT/PCBM (25 mg/mL in chloroform) at a weight ratio of 5% for channel materials printing, as shown in processes (4) to (6). A similar printing process has also been described in our previously published work.¹

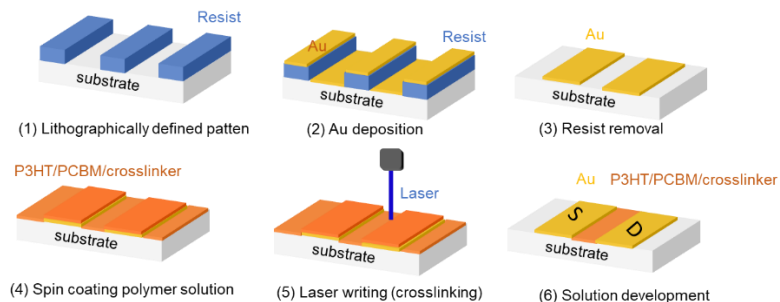


Figure S27. Schematic illustration of the photolithography process for device fabrication.

Reference

1. Chen, K. *et al.* Printing dynamic color palettes and layered textures through modeling-guided stacking of electrochromic polymers. *Mater. Horiz.* **9**, 425–432 (2022).

Simulation of a single layer of synapse array as artificial retina for face recognition

A 64×64 synaptic array was simulated as artificial retina, with each device functioning independently. In the simulation, an expectation-based thresholding model is developed for binary face recognition. Thresholding is a well-established methodology in computer vision to convert an image with continuous data into discrete format, in order to segment important features from the background. In our model, the threshold for each synaptic device is set based on the weight update expectation. In this note, we will show that the weight update of the optoelectronic synapse can be modeled as characteristic function curve and surface. Thus, the weight update expectation can be obtained from the characteristic surface and be used to adaptively set the decision-making condition for every synaptic device in the artificial retina. The weight update modeling and signal thresholding strategies have been used in other artificial synaptic applications.¹⁻³

In this work, we first modeled both postsynaptic nonvolatile and transient current of the devices as a function of light intensity and memory current using experimental data as shown below. To model the nonvolatile currents as a function of light intensity, a blank device was exposed to light pulses with same duration (60 s) but various intensities. The nonvolatile currents were recorded in each measurement and modeled with respect to light intensities using the quadratic function (Figure S28a). On the other hand, the transient current of the trained devices is dependent on both device memory and intensity of light input. To simulate this relationship, we measured the synaptic devices in response to repeating light pulses under different light intensities. Each pulse had a duration time of 2 seconds. The device memory current and current weight update for each light pulse was automatically detected. Then, we used a function surface to model the transient current change (z height with color coding) as a function of light intensity and memory current, as shown in Figure S28b.

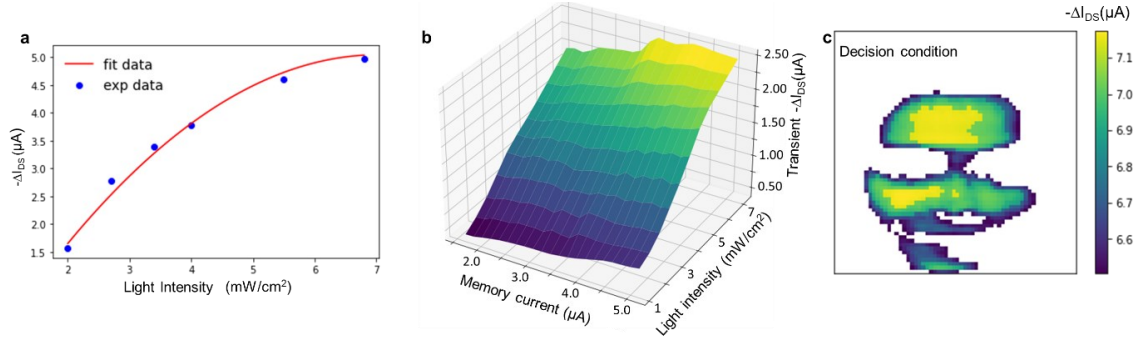


Figure S28. **a.** Light induced nonvolatile current change as a function of light intensity. **b.** A function surface showing the memory current-dependent and light intensity-dependent transient current change. **c.** A mapping of decision conditions for the feature region.

With established the functions, an Olivetti Research Laboratory (ORL) face dataset was exploited in this face recognition study. This dataset was constructed with face images of multiple people, each contains 10 images with distinct face orientations and expressions. Within the dataset, a face image of a woman is selected as recognition target. We firstly trained the artificial retina using 9 images of her face, which cover various face orientations, expresses and emotions as shown in Figure 6a. In the training, the optical signals (gray scale intensities) of facial features in images were normalized to the detection dynamic range of synaptic devices ($2-7 mW/cm^2$) and sampled to 64×64 pixels according to the dimension of our synaptic array. They were then input to the synapse array. According to fitted light intensity-nonvolatile current quadratic function (Figure s28a), nonvolatile current can be obtained for each synapse. We repeatedly this process for the same device using nine face images and captured the high-level visual features of the target woman. The learned nonvolatile current mapping is shown in Figure 6a, which clearly indicates distinct visual features in this woman's face. For instance, her hair, eye and mouth provide low light reflection, which leads to the low memory currents in synaptic array at corresponding locations. In contrast, devices at skin reigns around five senses organs show high memory currents because of strong light reflections. This analysis indicates that the artificial retina is capable of learning robust visual features from multiple face images of the target woman.

Based on the fitted function in Figure S28b, we can also automatically predict the photonic transient currents from the artificial retina when it is exposed to coming test faces. This enables us to build up an efficient face recognition model. In this model, we firstly determined the feature space of the learned memories in the synaptic arrays. We selected devices with relatively high memory currents (75% percentile as a threshold) as the feature devices, which automatically locates the characteristic outline of the target face (Figure 6b). We adaptively set the recognition decision-making criteria for feature devices based on the predictive capability for transient currents of synaptic devices under varying illumination and memory conditions (Figure S28b). If we implement the trained artificial retina with face images from the same woman (positive cases), all feature devices will provide similar current weight updates as the training process. In contrast, if the artificial retina is exposed to other face images (negative cases) which have low reflection visual features in the feature space, the photocurrents at corresponding locations will be lower than expected values in the positive recognition. To utilize the discriminative response of transient photocurrent for face recognition, we predicted the photocurrents for all feature devices in the positive case using the training face images, trained memories and the function surface, as shown in the Figure S28c. In addition, we multiplied a tolerance ratio (0.9) with predicted photocurrents and set these values as decision-making conditions.

Besides, to realize adaptive and high-speed decision making in the test environment, a protocol for dynamically updating the decision-making conditions is built, as shown in Figure S29. Real-time weight can be read out through drain current without influencing the device conductance in our three-terminal device structure. Then, they can be used for updating the decision-making conditions via transient current function surface (Figure S28b).

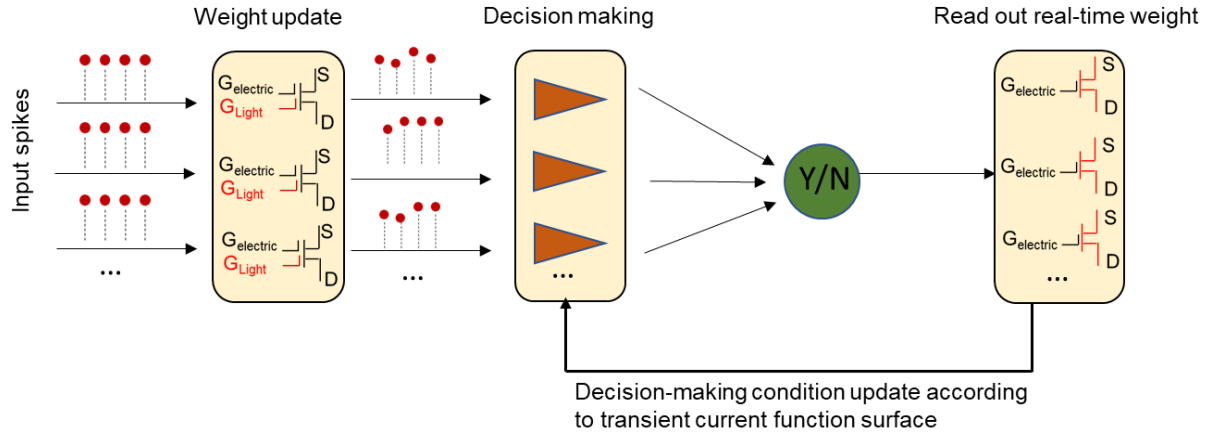


Figure S29. Protocol for dynamically updating decision-making condition using real-time weight reading.

In the test step, we showed face images from other people (negative cases) or face images from the target woman (positive cases) with different noises or orientation to the trained artificial retina device with the same learned memory and decision-making condition, as shown in Figure 6d. In the feature region, if the transient photocurrents of devices reach the condition values, we classify these devices as active status (marked as red in Figure 6c). In contrast, if the photocurrents are lower than condition values, we classify them as idle status (marked as blue in Figure 6c). In the test results of four example faces (Figure 6d), the transient photocurrents are visualized, and the color-coding heat map show the activated/idle device classifications. The percentage of activation is calculated based on device classification counts, which suggests the results for face recognitions. For true negative cases, there are distinct low-reflection facial features at the feature region (activation rates < 70%). When face image of the target woman with a different orientation was tested, a 91% activation rate is obtained. Even with a strong Gaussian noise, an 88% activation rate still renders the true positive recognition result. The face recognition test results demonstrate that we can build a robust visual system with the artificial retina device without using expensive computing frameworks.

Reference

1. Huang, H. *et al.* Electrolyte-Gated Synaptic Transistor with Oxygen Ions. *Adv. Funct. Mater.* **29**, 1902702 (2019).

2. Zhou, F. *et al.* Optoelectronic resistive random access memory for neuromorphic vision sensors. *Nat. Nanotechnol.* **14**, 776–782 (2019).
3. Ambrogio, S. *et al.* Neuromorphic Learning and Recognition With One-Transistor-One-Resistor Synapses and Bistable Metal Oxide RRAM. *IEEE Trans. Electron Devices* **63**, 1508–1515 (2016).

Simulation of hardware neural network for fashion product recognition

A variety of hardware frameworks have been reported to implement the matrix multiplication and convolution computing using the artificial synapses¹⁻³, which enables the construction of hardware neural networks for artificial intelligence applications. In this section, a single-layer perceptron and a deep LeNet⁴ convolutional neural network (CNN) were simulated using our optical synapses as shown in Figure S30. They were both trained and tested using a Fashion-MINIST dataset⁵, which is a recently developed image dataset for benchmarking machine learning algorithms. Fashion-MINIST comprises of 28x28 pixel grayscale images of 70000 fashion products from 10 categories.

Based on this object recognition task, the perceptron is constructed with 785 input neurons, corresponding to the number of image pixels (28x28+1), and 10 output neurons with a softmax layer, corresponding to 10 fashion product categories. Input and output neurons are fully connected by 7850 (785x10) synapses. LeNet is a deep CNN model, consisting of 3 convolution layers, 2 pooling layers, 2 fully connected layers and a softmax layer. Three convolution layers have kernel size of (5 × 5) with 1 input channel and 6 output channels, kernel size of (5 × 5) with 6 input channel and 16 output channels, kernel size of (5 × 5) with 16 input channels and 120 output channels, respectively. Two fully connected layers contain 120x84 and 84x10 weights, respectively. For each layer in the hardware neural network models, the input voltage signals (V) are multiplied (fully connected layer) or convolved (convolution layer) with synaptic weights (W), resulting in learned features comprising current signals ($I = W \cdot V$ or $I = W * V$). These current signals are then passed through activation function and convert into voltage signals for the computation in the next layer ($V = f_{ReLU}I$ or $V = f_{softmax}I$). In the training phase of the hardware neural network, we update the weights of optical synapses by comparing the softmax result vectors with fashion product ground-truth categories using a cross entropy criterion and Adam optimization algorithm⁶.

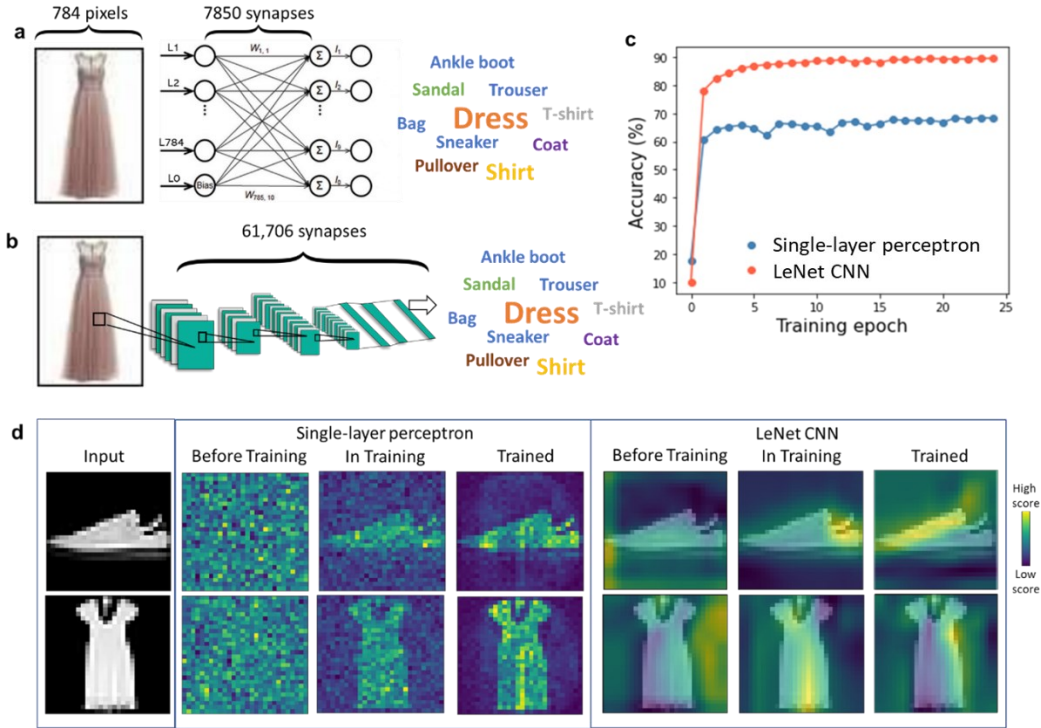


Figure S30. Simulation of hardware neural networks using optical synapses. (a) Configuration of a single-layer perceptron. (b) Configuration of a LeNet CNN model. (c) Recognition rates of two models with respect to training epochs. (d) Evaluation of model training progression using synaptic weights visualization and Grad-CAM heatmap.

To optimize these synaptic weights in the training phase, a widely used hardware-based backpropagation (HDBP) algorithm⁷⁻⁹ has been adopted. More specifically, a differential pair of optical synapses are used to represent one synaptic weight ($W = G^+ - G^-$). To update G^+ and G^- , each synapse is firstly determined whether in a potentiation or depression status based on the ΔW , which is calculated via cross entropy loss and Adam optimizer, as shown in equation (1).

$$state = \begin{cases} \text{potentiation } (G^+ \uparrow \text{ and } G^- \downarrow), & \text{if } \Delta W > 0 \\ \text{depression } (G^+ \downarrow \text{ and } G^- \uparrow), & \text{if } \Delta W < 0 \end{cases} \quad (1)$$

For the weight updating in the potentiation state, the G^+ is designed to increase and G^- to be simultaneously decreased. On the opposite depression state, the G^+ is designed to decreased and G^- to be increased as shown in equation (2).

$$\Delta G = G_{n+1} - G_n = \begin{cases} \alpha e^{-\beta \frac{G_n - G_{min}}{G_{max} - G_{min}}}, & \text{if } state = \text{potentiation} \\ -\alpha e^{-\beta \frac{G_{max} - G_n}{G_{max} - G_{min}}}, & \text{if } state = \text{depression} \end{cases} \quad (2)$$

Herein, G_n denotes the present conductance state of the optical synapses and G_{n+1} denote the updated conductance state in the next training iteration, respectively. α denotes the step size of the conductance updating and β denotes the nonlinearity. Conductance updating functions (equation 2) are fitted using the characteristic behaviors of synapse operation as shown in Figure S31a, and fitted parameters are shown in Figure S31b. To avoid the scale issue, both G^+ and G^- are initialized to G_{min} when the synaptic conductance attains the maximum (G_{max}) or minimum (G_{min}) values.

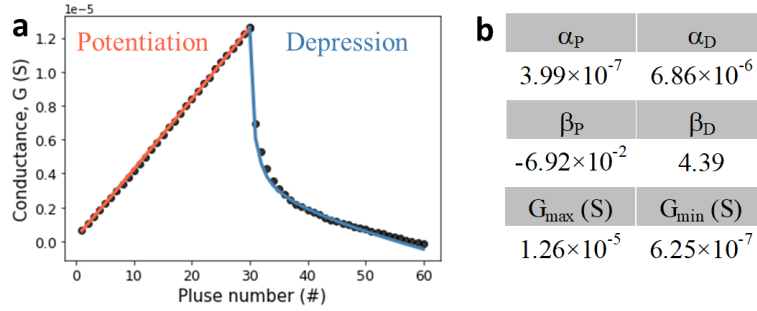


Figure S31. Parameter fitting of the device potentiation and depression pulse trains with 60 representative weight states. (a) Superimposition of experimental data points and fitted curve. (b) Fitted equation parameters.

As shown in Figure S30c, we demonstrated the object recognition performance of hardware neural networks built with optical synapses on the Fashion-MINIST dataset. In each epoch of simulation, the synaptic conductance weights were updated by the training of 60000 images of fashion products. Then we tested the network using 10000 testing images and recorded the recognition rates. Both single-layer perceptron and LeNet models converged within 25 epochs, demonstrating the excellent learning efficiency of the HDBP algorithm. As a result, recognition rates of 69.0% and 89.5% were achieved by single-layer perceptron and LeNet, respectively. To visualize the synaptic weight update and the model evolution, the synaptic weights connected to output neurons were visualized for the single-layer perceptron model. In addition, the gradient-weighted class activation mapping (Grad-CAM)¹⁰ technique was adopted to visualize the progress of the LeNet CNN model. Grad-CAM exploits the gradients of image classification in the backpropagation to locate the important regions in the image for the predicting, which is plotted using a heatmap. Herein, images of a sneaker and a dress are used for model evolution evaluation using both techniques in two models, respectively. The left panel in Figure S30d displays the synaptic weights corresponding to input images in different periods of modeling training. With the progress of the training, more clear fashion product images are mapped by the model, indicating the improved recognition performance. In the LeNet CNN model, the Grad-CAM heatmap is gradually moved to the edges of the object as shown in the superimposed images in the right panel of Figure S30d, which are key features for differentiating fashion products. The change of the heatmap indicates the CNN model has become more intelligent for locating the key visual features in the image after the model

training. This test performance and model evaluation demonstrate that our optical synapse can be effectively used as the basic computing unit to be implemented in deep neural network with a large number of parameters.

In summary, we have simulated our optical synapses in three models for visual recognition tasks: (1) a single-layer artificial retina for binary face recognition. (2) a single-layer perceptron and (3) a LeNet CNN model for fashion product recognition. We have demonstrated the increase of model complexity can result in more powerful visual recognition performance. The CNN model (61,706 synapses) provides the most robust object recognition performance, while our artificial retina (64x64 synapses) can provide highly efficient binary face recognition by fully utilizing the optical synapse integrated functions of ambient light perception, light-to-electrical signal transduction and memorization. These simulations demonstrate that our optical synapse has the promise of building a wide range of hardware sensing and computing applications. All simulations were performed using Python, a free and open-source programming language, with SciPy and PyTorch packages. Codes are available from the corresponding author upon reasonable request.

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