

An Optoelectrochemical Synapse based on a Single-component n-type Mixed Conductor

Corresponding Author: Professor Sahika Inal

This file contains all reviewer reports in order by version, followed by all author rebuttals in order by version.

Attachments originally included by the reviewers as part of their assessment can be found at the end of this file.

Version 0:

Reviewer comments:

Reviewer #1

(Remarks to the Author)

The paper presents a compelling design concept for an organic ion-tunable optoelectrochemical synapse, integrating a single-component n-type mixed conductor into an electrochemical transistor. The innovative approach to mimicking ion-based cellular communication through the use of organic mixed ionic-electronic conductors (OMIECs) is particularly noteworthy. The study effectively demonstrates the potential of the developed synapse in emulating biological synapses, showcasing features such as synaptic plasticity, pair-pulse facilitation, and transition from short-term to long-term memory at low operating voltages. The device's ability to perceive multi-spectral visual information, extending even beyond the human visual system's capabilities, is a significant advancement in optogenetics-inspired technology. The integration of an 8x8 array of these devices for image recognition, preprocessing, and memory tasks showcases the scalability and practical applicability of the proposed technology. Furthermore, the device's capability for in-sensor reservoir computing highlights its potential for decoding complex spatiotemporal events in biological media.

Overall, this paper makes a valuable contribution to the field of bio-inspired electronics and optogenetics-inspired neuromorphic systems. I highly appreciate this study and in principle agrees with the publication after addressing the following specific points.

1. In the introduction, the authors highlighted the requirement for high operating voltages attributed to the properties of the channel materials utilized. Nonetheless, in the case of employing P3HT:PCBM as the OECT channel, the operational voltage stays below 1V, signifying a notably lower operational voltage threshold. Accuracy in describing the voltage levels associated with the specific channel materials used in organic electrochemical transistors is crucial.
2. Please ensure consistency in the formatting of the paper. For example, V of V_{TH} in line 153 should be italics.
3. The simulation methods used in "motion perception" and event-based in-sensor reservoir should be described in detail in Supplementary Info. For example, in Fig. 6(g), the authors show bioinspired sensor under real-world scenes. However, it is unclear how the system determines the precise speed of motion, particularly for fast or slow movements in a mosquito. Can the authors provide more details on how the speed of motion is calculated and how the bio-inspired sensor works?
4. For this rigid of lactone-based backbone, more linear longer or branched chains can make the mobility higher. I advise the authors need to investigate more longer chains to improve performance in their future work.

Reviewer #2

(Remarks to the Author)

This manuscript presents an investigation of an optoelectronic organic electrochemical transistor (OECT) featuring an n-type photoactive organic mixed ionic-electronic conductor (OMIEC) channel, which exhibits neuromorphic behavior and computational capabilities. However, the manuscript requires significant improvements, particularly in providing more detailed information on the experimental procedures and ensuring the reproducibility of the presented data. Given these concerns, the reviewer suggests that the authors consider submitting the manuscript to a more specialized journal rather than Nature Communications.

Below some major points that needs to be addressed:

1. In the title and in the introduction the device has been presented as an example of optogenetics-inspired synapse, but it

would be more appropriate to include in the definition the word 'optoelectronic', because this one clarifies better the operation of the device. Even in the mentioned works of the same field the word 'optoelectronic' is always present in the title. Naming it 'optogenetics-inspired' could be a bit misleading, since there is no stimulation of cells.

2. In Fig. 1a it is a bit misleading to put "synaptic plasticity (synaptic weight)". Indeed, the synaptic plasticity refers to modulation during the time of the synaptic weight.

3. In Fig.1b there is the microscope image of the channel of the device. It would be better to substitute it with another representation of the device geometry since it is not visible from the chosen picture.

4. In Fig.1c the transfer curve has been reported and it would be necessary to mention in the graph or in the description of the figure the drain voltage used. Moreover, an explanation of the reason behind the choice of the gate voltage window (from 0V to 0.6V) and the drain voltage value is required.

5. In Fig.1d the label of the x-axis is missing.

6. In the manuscript the following text has been inserted "At $V_D=0.4$ V and $V_G=0.45$ V, the maximum geometry-normalized transconductance ($g_{m,n}$) reaches 25.94 S cm^{-1} ". A description about the calculation method of the the maximum geometry-normalized transconductance should be reported in the Methods section.

7. The Methods regarding the acquisition of the transfer curves and the electrical characterization of the OECTs have to be added.

8. From the manuscript is clear that an Ag/AgCl gate has been used for the electrical characterization of the OECT, while a planar Au electrode has been used for the transfer curves in response to light. It is not clear the reason behind this choice. Moreover, it would be more consistent to show the transfer curves of the device without light with the gold electrode too.

9. In Fig. 2b and in other parts of the manuscript, ΔI_D has been used instead of the absolute value of the channel current. What is the reason behind this choice? A brief description of the way in which this quantity has been extracted should be added in the manuscript. At the same time, it is required an explanation of the reason why in Fig. S12 absolute values of the channel current have been used.

Moreover, which values have been used for the voltages at the drain and at the gate for this graph in which light pulses have been applied?

10. In Fig S14, it is necessary to quantify the recovery of the memory effect, after the removal of the light stimulation. In fact, it seems that for certain values of gate voltage, there is not a recovery of the conductance state after the removal of the light, or at least the temporal dynamic is very slow compared to other cases. Some hypothesis behind this phenomenon should be added.

11. In Fig.2c how many samples have been use to extract PPF index? Are they mean values? Error bars of PPF index values should be included. Details about the statistics should be included.

12. In the manuscript, it is reported: "Moreover, increasing the pulse number or the training period improved the device's learning ability, suggesting spike-number and spike-time-dependent plasticity (Fig. S15a and Fig. 2d). These findings demonstrate the device's ability to easily transform plasticity from short-term (STP) to long-term (LTP) through repetitive rehearsal events occurring multiple times". A quantitative evaluation of the spike-time-dependent-plasticity should be carried on and more samples should be tested with the same delays.

13. In Fig. 2 g the description is "The comparison of photocurrent generated by OECTs bearing various OMIECs in the channel. All the devices were illuminated for 5 s with a light intensity of 200 mW cm^{-2} . In all experiments, the gate electrode is gold." What is the wavelength used in this experiment? This information should be added. In addition, the graph reports the values of current in nanoampere, while in the text the values are expressed in microampere. It would be better to be coherent with the units used.

14. In the manuscript, it is reported "These memories can be consolidated into long-term memory (LTM) through rehearsal learning. Our device exhibited a similar pattern, wherein after the initial learning phase (50 excitation pulses) and subsequent forgetting, the current could be restored to its previous state using only 25 pulses (Fig. 3a)." In Fig. 3a it is not clear which is the initial learning phase and the subsequent one. Moreover, in the graph seems that the channel conductance cannot be recovered after the forgetting state, and this should be the reason why a continuous increase of the baseline is highlighted. Overall, it is not clear which is the purpose of this experiment.

15. In the manuscript, it is reported : "When either Light 1 (375 nm) or 291 Light 2 (455 nm) was turned ON, the excitatory postsynaptic current I_D surpassed the threshold 292 (12.6 μA), indicating the "OR" logic operation (Fig. 3d and Fig. 3f). The I_D exceeded the threshold 293 (15.7 μA) only when Light 1 and Light 2 were applied simultaneously, indicating the AND logical 294 operation (Fig. 3e-f)."

In Fig.3d-3f, thresholds are defined according to the experiment but it is not clear how much 0 and 1 states can be discriminated by using these thresholds. The threshold values used for the OR logic operation are quite different from the one used for the AND operation. The choice of the threshold should be explained better, and more experiments could be performed to prove the consistency of these computing capabilities.

16. In Fig.4e intensity values are missing. They should be added to the graph.

17. Is the cell viability test for 24h enough to demonstrate the biocompatibility of the material? An explanation regarding the choice of this timing should be included.

Reviewer #3

(Remarks to the Author)

I co-reviewed this manuscript with one of the reviewers who provided the listed reports. This is part of the Nature Communications initiative to facilitate training in peer review and to provide appropriate recognition for Early Career Researchers who co-review manuscripts.

Reviewer #4

(Remarks to the Author)
The comments are attached.

Version 1:

Reviewer comments:

Reviewer #1

(Remarks to the Author)
The authors fully addressed my concerns during the revision. I also go through the other reviewers' comments and found that they are responded properly.

Reviewer #2

(Remarks to the Author)
All comments have been addressed.

Reviewer #4

(Remarks to the Author)
I believe the authors have sufficiently addressed my previous concerns and comments. The revised manuscript now raises novel scientific and engineering advancements compared to other OECTs device systems, making it suitable for publication in Nature Communications.

Open Access This Peer Review File is licensed under a Creative Commons Attribution 4.0 International License, which permits use, sharing, adaptation, distribution and reproduction in any medium or format, as long as you give appropriate credit to the original author(s) and the source, provide a link to the Creative Commons license, and indicate if changes were made.

In cases where reviewers are anonymous, credit should be given to 'Anonymous Referee' and the source.

The images or other third party material in this Peer Review File are included in the article's Creative Commons license, unless indicated otherwise in a credit line to the material. If material is not included in the article's Creative Commons license and your intended use is not permitted by statutory regulation or exceeds the permitted use, you will need to obtain permission directly from the copyright holder.

To view a copy of this license, visit <https://creativecommons.org/licenses/by/4.0/>

Response to reviewers' comments:

Reviewer 1:

The paper presents a compelling design concept for an organic ion-tunable optoelectrochemical synapse, integrating a single-component n-type mixed conductor into an electrochemical transistor. The innovative approach to mimicking ion-based cellular communication through the use of organic mixed ionic-electronic conductors (OMIECs) is particularly noteworthy. The study effectively demonstrates the potential of the developed synapse in emulating biological synapses, showcasing features such as synaptic plasticity, pair-pulse facilitation, and transition from short-term to long-term memory at low operating voltages. The device's ability to perceive multi-spectral visual information, extending even beyond the human visual system's capabilities, is a significant advancement in optogenetics-inspired technology. The integration of an 8x8 array of these devices for image recognition, preprocessing, and memory tasks showcases the scalability and practical applicability of the proposed technology. Furthermore, the device's capability for in-sensor reservoir computing highlights its potential for decoding complex spatiotemporal events in biological media.

Overall, this paper makes a valuable contribution to the field of bio-inspired electronics and optogenetics-inspired neuromorphic systems. I highly appreciate this study and in principle agree with the publication after addressing the following specific points.

We appreciate the reviewer's time for reviewing our work and recommendation. We will address the specific comments below.

1. In the introduction, the authors highlighted the requirement for high operating voltages attributed to the properties of the channel materials utilized. Nonetheless, in the case of employing P3HT:PCBM as the OEECT channel, the operational voltage stays below 1V, signifying a notably lower operational voltage threshold. Accuracy in describing the voltage levels associated with the specific channel materials used in organic electrochemical transistors is crucial.

We appreciate the reviewer's comment. We have removed the description of high operating voltages. We have revised the sentence:

“The only demonstration of a miniaturized OEECT-based optoelectronic synapse, which generated a non-volatile light-induced memory, used conventional organic semiconductors¹⁰ (a bi-component donor/acceptor system, P3HT:PCBM), in the channel, which are not compatible with aqueous electrolytes and require a toxic ionic liquid as the electrolyte.”

2. Please ensure consistency in the formatting of the paper. For example, V of V_{TH} in line 153 should be italics.

We have corrected our text for such errors.

3. The simulation methods used in “motion perception” and event-based in-sensor reservoir should be described in detail in Supplementary Info. For example, in Fig. 6(g), the authors show bioinspired sensor under real-world scenes. However, it is unclear how the system determines the precise speed of motion, particularly for fast or slow movements in a mosquito. Can the authors provide more details on how the speed of motion is calculated and how the bio-inspired sensor works?

We appreciate the feedback and have added further details to Supplementary Note 6.

“ The device’s ability to capture spatiotemporal dynamics was further demonstrated using real-world video data, such as the movement of a mosquito (Fig. 7g). A video of the mosquito was recorded at a fixed frequency, and its movement velocity was estimated based on time intervals. Adjusting the video frame rate allowed us to simulate varying motion speeds; for example, increasing the frequency by a factor of 10 simulated a tenfold increase in perceived motion speed. The bio-inspired sensor encoded this motion information into spatiotemporal frames, breaking the video into sequences that captured both the spatial position and temporal evolution of the mosquito’s movement. The feature map illustrates the sensor’s ability to distinguish between fast and slow motions by encoding the temporal dynamics of objects. This example underscores the sensor's effectiveness in capturing and interpreting dynamic information for motion detection and classification tasks.”

4. For this rigid of lactone-based backbone, more linear longer or branched chains can make the mobility higher. I advise the authors need to investigate more longer chains to improve performance in their future work.

We acknowledge this advice on the further investigation of this type of polymer backbone. We are currently unable to synthesize/acquire new materials for this project, but we will definitely keep this idea in mind for follow-up work.

Reviewer 2:

This manuscript presents an investigation of an optoelectronic organic electrochemical transistor (OECT) featuring an n-type photoactive organic mixed ionic-electronic conductor (OMIEC) channel, which exhibits neuromorphic behavior and computational capabilities. However, the manuscript requires significant improvements, particularly in providing more detailed information on the experimental procedures and ensuring the reproducibility of the presented data. Given these concerns, the reviewer suggests that the authors consider submitting the manuscript to a more specialized journal rather than Nature Communications. Below some major points that needs to be addressed:

Given that this work has various components in chemistry, materials science, device physics, and data science, we believe that it should reach an interdisciplinary and broad audience as this journal has. We thank the reviewer for all their constructive suggestions that have significantly improved this manuscript. We will address the specific comments below:

1. In the title and in the introduction the device has been presented as an example of optogenetics-inspired synapse, but it would be more appropriate to include in the definition the word ‘optoelectronic’, because this one clarifies better the operation of the device. Even in the mentioned works of the same field the word ‘optoelectronic’ is always present in the title. Naming it ‘optogenetics-inspired’ could be a bit misleading, since there is no stimulation of cells.

We agree with the reviewer that the term ‘optogenetics-inspired’ in our manuscript may lead to misconceptions. We have changed our title to “An **Optoelectrochemical** Synapse based on a Single-component n-type Mixed Conductor”. We have replaced ‘optogenetics-inspired’ with

‘optoelectronic or optoelectrochemical’ throughout the manuscript. For instance, we have revised “optogenetic-inspired synapse” to “**optoelectronic synapse**” in the first and third paragraphs of the introduction, and also in the conclusion, we now refer to “**optoelectronic**” or “**optoelectrochemical**” rather than “optogenetics-inspired”. We thank the reviewer for making this very important distinction.

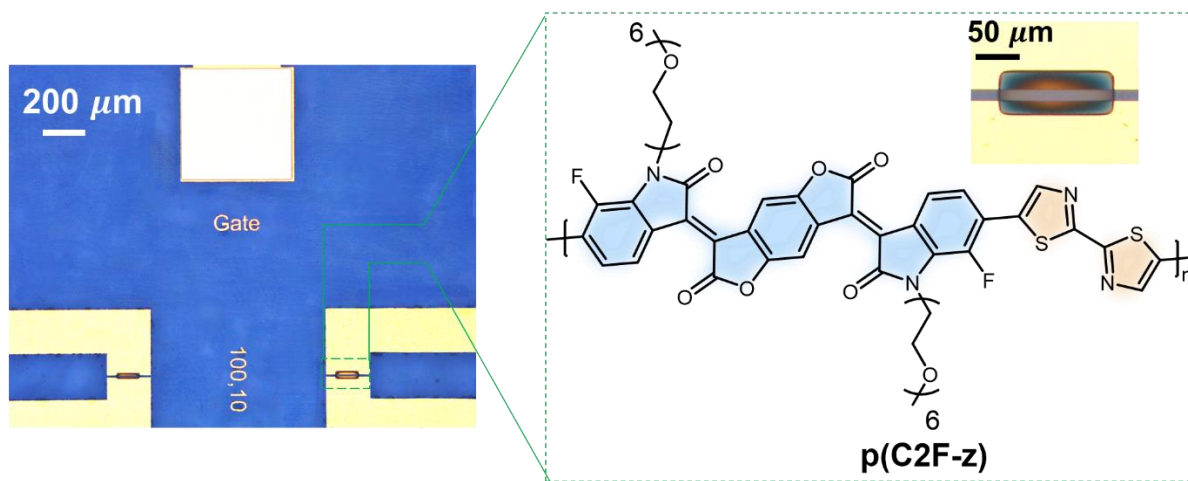
2. In Fig. 1a it is a bit misleading to put “synaptic plasticity (synaptic weight)”. Indeed, the synaptic plasticity refers to modulation during the time of the synaptic weight.

We agree with the reviewer regarding the description of synaptic plasticity. We have revised the caption of Fig. 1a:

“In synapses, synaptic weight is responsible for learning and storing visual information and memory.”

3. In Fig.1b there is the microscope image of the channel of the device. It would be better to substitute it with another representation of the device geometry since it is not visible from the chosen picture.

To show the full device architecture with the source, drain, and gate contacts, we replaced Fig. 1b with a new microscope image. We now also zoom into the channel region of the device and added an inset to allow the reader to clearly understand the device geometry and the channel coating.



b) Chemical structure of p(C2F-z) with the microscope image of its film in the microscale OECT channel (length = 10 μm, width = 100 μm, d = 79 nm) and the lateral Au gate electrode. The schematic representation and a microscope image of the OECT are shown in Fig. S7a-b.

4. In Fig.1c the transfer curve has been reported and it would be necessary to mention in the graph or in the description of the figure the drain voltage used. Moreover, an explanation of the reason behind the choice of the gate voltage window (from 0V to 0.6V) and the drain voltage value is required.

We have added the V_D information both in Fig. 1c and its caption ($V_D = 0.6$ V). For the V_G selection, since the channel comprises an n-type material, V_G is always positive ($V_G > 0$). The selection of V_G is then based on the CV curve in Fig. S6a, which shows reduction currents at and beyond 0.2 V vs. Ag/AgCl, indicating electroactivity within the $V_G = 0.2 - 0.6$ V range. The doping is also

obvious from the in situ UV-VIS spectrum (Fig. S2c) from the polaronic features observed starting from -0.1 V. Therefore, we set V_G within this range (0-0.6 V). Note also that we now added operating conditions details to all figures where this information was missing.

5. In Fig.1d the label of the x-axis is missing.

We have added the x-axis label in Fig. 1d. The updated Fig. 1 is shown in the revised manuscript.

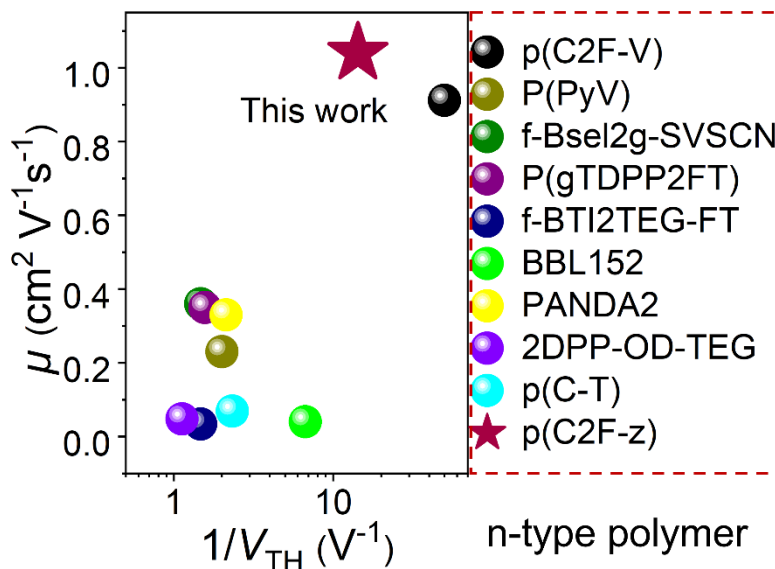


Fig.1 d) Comparison of p(C2F-z) device performance metrics (μ and V_{TH}) with those of other high-performance n-type OECTs.

6. In the manuscript the following text has been inserted “At $V_D=0.4$ V and $V_G=0.45$ V, the maximum geometry-normalized transconductance ($g_{m,n}$) reaches 25.94 S cm⁻¹”. A description about the calculation method of the the maximum geometry-normalized transconductance should be reported in the Methods section.

We thank the reviewer for pointing out the issue with the calculation method for the maximum geometry-normalized transconductance. We need to first correct a small error in the main text: when calculating the maximum transconductance, V_D is not 0.4 V but 0.6 V. The sentence “At $V_D=0.4$ V and $V_G=0.45$ V, the maximum geometry-normalized transconductance ($g_{m,n}$) reaches 25.94 S cm⁻¹ with a drain current (I_D) of a few sub-mA.” has been changed to “At $V_D=0.6$ V and $V_G=0.45$ V, the maximum geometry-normalized transconductance ($g_{m,n}$) reaches 25.94 S cm⁻¹ with drain current (I_D) of few sub mA.”

We added the calculation method to the caption of Table S1a: ^a calculated according to $g_{m,norm} = g_m W^{-1} L d^{-1}$. To enhance clarity, we have also included this calculation method for maximum geometry-normalized transconductance in the Methods section under device fabrication: “The maximum geometry-normalized transconductance is calculated according to $g_{m,norm} = g_m W^{-1} L d^{-1}$.”

7. The Methods regarding the acquisition of the transfer curves and the electrical characterization of the OECTs have to be added.

We have provided additional details on OECT characterization in the Method section under Device Fabrication and Characterization.

“A 0.1 M NaCl aqueous solution was used as the electrolyte, with a silver pellet serving as the gate. A PDMS well was utilized as a reservoir to contain the aqueous electrolyte. OECT device characterization, including transfer and output characteristics, along with repetitive pulsing measurements, was performed in air at room temperature using a dual source-meter unit (Keithley 2636A) with a scan rate of 250 mV/s.”

Additionally, we included electrical characterization details in the caption of Table S1 and added them to the Device Fabrication and Characterization section.

“The threshold voltage (V_{TH}) was determined by extrapolating the linear regime of the $I_D^{1/2} \sim V_G$ plot. The product μC^* was extracted from the slope of the linear fit to g_m vs. $(Wd/L)(V_{TH} - V_G)$ plot, where C^* was extracted from electrochemical impedance spectra, and mobility (μ) was calculated using μC^* divided by C^* value. All OECTs were operated in a 0.1 M NaCl aqueous solution under ambient conditions. Average values were calculated from 5 operational channels.”

8. From the manuscript is clear that an Ag/AgCl gate has been used for the electrical characterization of the OECT, while a planar Au electrode has been used for the transfer curves in response to light. It is not clear the reason behind this choice. Moreover, it would be more consistent to show the transfer curves of the device without light with the gold electrode too.

We thank the opportunity to clarify the rationale for choosing the gate electrode. In our initial OECT performance comparison, we chose Ag/AgCl as the gate electrode. Our study involves a novel semiconductor, and we need to establish a baseline for performance comparison with other published materials. The devices using these other materials have used Ag/AgCl gate electrode, a common practice in OECT materials research. Therefore, when we discuss the OECT performance of this new material in Fig. 1 and the associated SI figures, we use an Ag/AgCl gate electrode. We now added the description in the main text:

“To characterize the performance of this new polymer in OECTs and benchmark its mixed conduction properties, we used an Ag/AgCl pellet as the gate electrode.”

Later in the study, we transitioned to gold electrodes to investigate light response. Gold electrodes were preferred because they are polarizable, and they are used as the source/drain metal of the channel and interconnects, so it is straightforward to have Au as the lateral gate electrode. Moreover, the Ag/AgCl electrode is externally introduced, i.e., suspended in the electrolyte above the channel, which can hinder the light path to the channel and make the whole device large. In contrast, the planar gold gate electrodes could be integrated directly with the channel during photolithography process as described. Gold electrodes are advantageous for array integration in subsequent applications. We now add the gate electrode type information to all our figures when needed, making the device architecture clearer.

The dark line in Fig. 2a represents the transfer curves of the channel gated with the planar gold gate electrode. As per reviewer’s request, we now have this data separately in Fig. S9a in addition to the transconductance values.

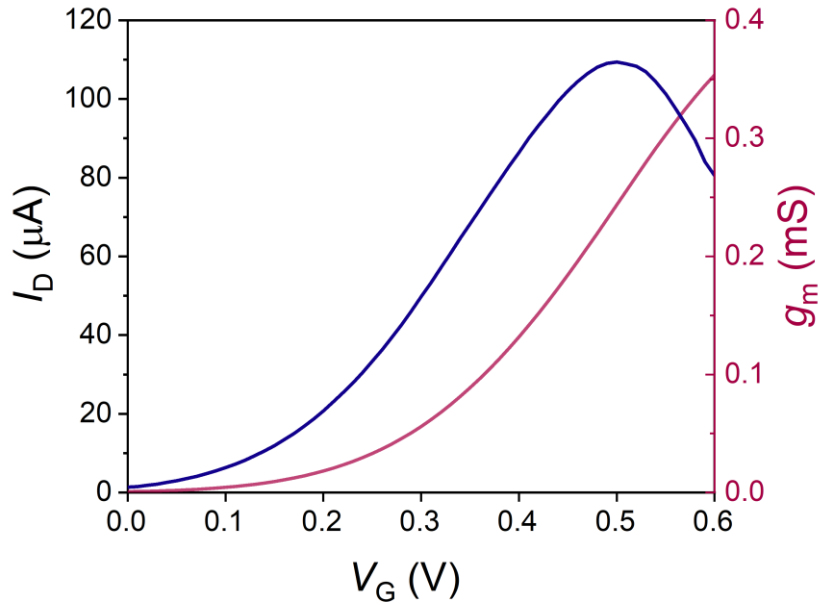


Fig. S9. a) The transfer characteristics of Au gated p(C2F-z) OEET measured in the dark and the corresponding g_m vs. V_G . $V_D = 0.6$ V.

9. In Fig. 2b and in other parts of the manuscript, ΔI_D has been used instead of the absolute value of the channel current. What is the reason behind this choice? A brief description of the way in which this quantity has been extracted should be added in the manuscript. At the same time, it is required an explanation of the reason why in Fig. S12 absolute values of the channel current have been used. Moreover, which values have been used for the voltages at the drain and at the gate for this graph in which light pulses have been applied?

When a voltage is applied to our device without illumination, it shows an initial current of approximately 40 μA . Introducing light causes an increase in current, and the magnitude of this increase varies based on specific testing conditions such as light intensity. In some cases, the current increase is smaller, while in others, it is larger. To make these changes more visually intuitive, we represent them using relative values (ΔI_D). For most sensors, eventually, the interesting value is not the absolute current but the change in the current. We now changed the text so that we clearly describe the value reported (photocurrent or photonic current or, simply, current).

In Fig.S12, we use ΔI_D instead of absolute values. Conversely, in Fig.S14a, absolute values are presented for aesthetic purposes, as this format better visualizes the optical response across different voltages.

During the light pulse application, the applied voltages were $V_D = V_G = 0.4$ V. We have now added voltage details in the caption of Fig. S10-S13.

10. In Fig S14, it is necessary to quantify the recovery of the memory effect, after the removal of the light stimulation. In fact, it seems that for certain values of gate voltage, there is not a recovery of the conductance state after the removal of the light, or at least the temporal

dynamic is very slow compared to other cases. Some hypothesis behind this phenomenon should be added.

We now included the recovery time in Fig. S14a. We magnified the initial current values at different gate voltages and included them as an inset. We also added the recovery times in the figure as well as in the caption of Fig.S14.

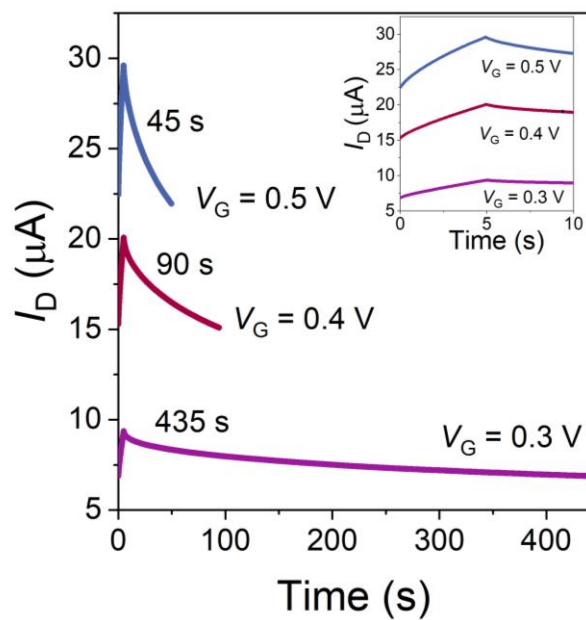


Fig. S14. a) The device current-time profile recorded at different gate voltages as a light source at 375 nm with an intensity of 200 mW cm^{-2} is switched on for 5 s. $V_D = 0.4 \text{ V}$. The inset demonstrates a zoomed in profile of the first 10 seconds. The recovery time of the current is noted as 45 s, 90 s, and 435 s for V_G 0.5, 0.4, and 0.3 V, respectively.

The associated text is:

“Additionally, the photonic synaptic weight of the device could be controlled by adjusting the gate voltage, with higher biasing values leading to larger changes in I_D but a shorter recovery time (Fig. S14a-b and Supplementary Note 1).”

We added the discussion about the speed of recovery in Supplementary Note 1:

Supplementary Note 1. The effect of V_G on the current recovery after the light pulse

“To quantify the memory capability, we recorded the current change over time post-light pulse (Fig. S14a). As the gate voltage increases, the time required for the current to return to its initial value decreases from 45 s ($V_G = 0.5 \text{ V}$) to 90 s ($V_G = 0.4 \text{ V}$) and further to 435 s ($V_G = 0.3 \text{ V}$). Once the light is off, the separated charge carriers generated by light need to travel to the corresponding contacts, and their motion is affected by interactions with electrolyte ions inside the film.¹⁶ Different gate voltages can influence the rate and extent of ion migration within the device. Higher gate voltages can increase the speed of ion migration as the stronger electric field drives ions more effectively, improving the speed of the recovery process.¹⁷ We postulate that varying gate voltages lead to different levels of interference of the separated charges and electrolyte ions, resulting in distinct recovery times.”

New references:

¹⁶Nat. Photon. 17, 629–637 (2023)

¹⁷Adv. Theory Simul. 2022, 5, 2100563

11. In Fig.2c how many samples have been use to extract PPF index? Are they mean values? Error bars of PPF index values should be included. Details about the statistics should be included.

To improve reproducibility, we calculated the PPF index as an average across four samples and added error bars; please refer to the updated Fig. 2c below. We revised the caption by adding: “Each data point represents the average of four samples.”.

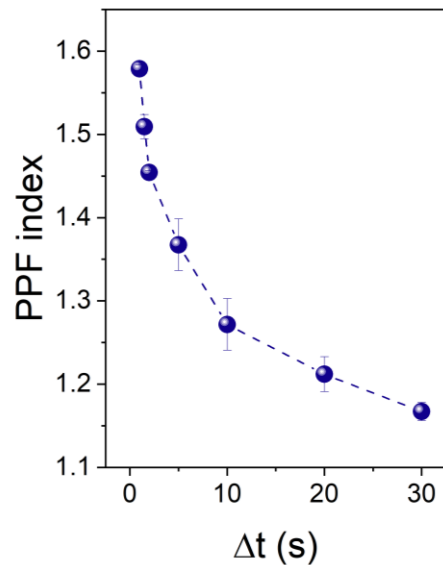


Fig. 2 c) PPF index as a function of pulse interval time with pulse width of 1 s. Each data point represents the average of four samples.

12. In the manuscript, it is reported: “Moreover, increasing the pulse number or the training period improved the device’s learning ability, suggesting spike-number and spike-time-dependent plasticity (Fig. S15a and Fig. 2d). These findings demonstrate the device’s ability to easily transform plasticity from short-term (STP) to long-term (LTP) through repetitive rehearsal events occurring multiple times”. A quantitative evaluation of the spike-time-dependent-plasticity should be carried on and more samples should be tested with the same delays.

As per reviewer’s comment, we conducted four independent tests for each pulse condition and pulse width, calculated the average values, and included error bars. This data is now presented in Fig. S15. We identified an error in the pulse number (Fig. S15b-c) and width (Fig. S15d-e) and have made the necessary corrections.

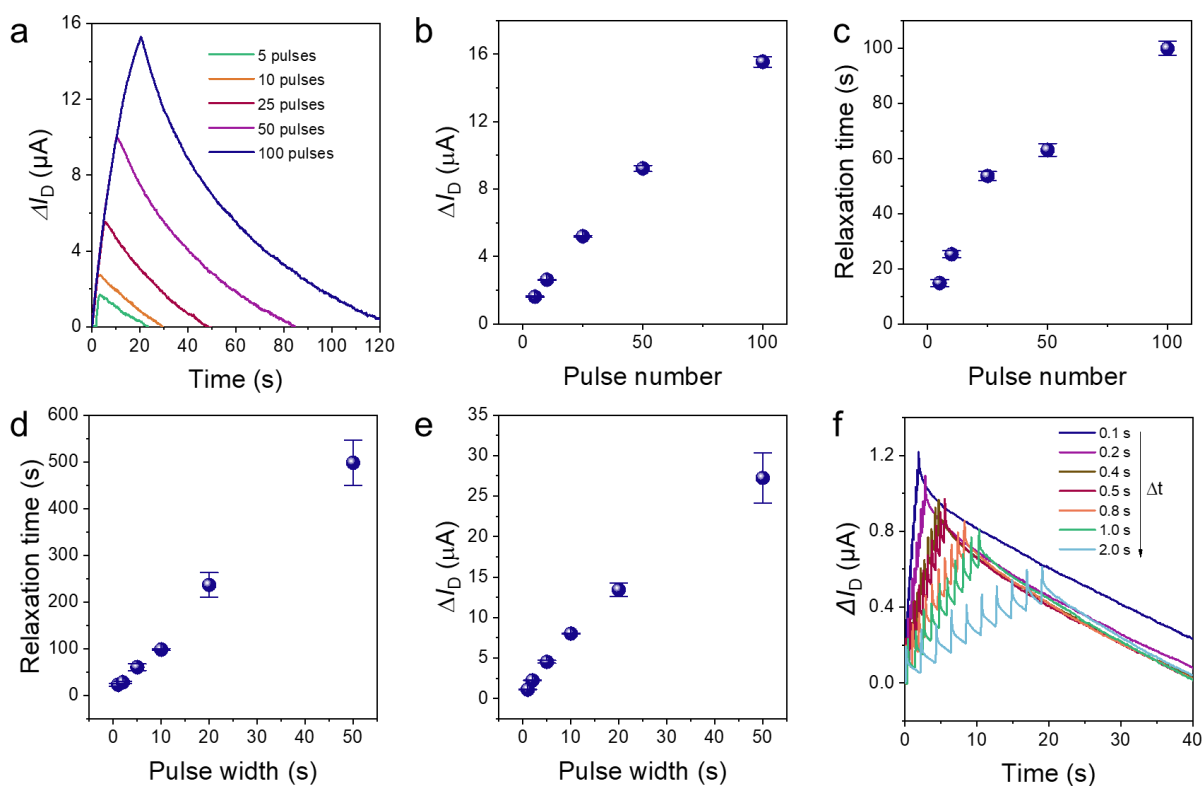


Fig. S15. a) The photonic current-time profile in response to various light pulse numbers (200 mW cm^{-2} , $t_p = 0.1 \text{ s}$, $\Delta t = 0.1 \text{ s}$). **b)** the photonic current as a function of pulse number, **c)** the current relaxation time as a function of pulse number. **d)** the current relaxation time as a function of pulse width, and **e)** the photonic current as a function of pulse width. **f)** The photonic current-time profile in response to various pulse intervals (200 mW cm^{-2} , $t_p = 0.1 \text{ s}$) In all plots, the error bars represent data from four measurements.

13. In Fig. 2 g the description is “The comparison of photocurrent generated by OECTs bearing various OMIECs in the channel. All the devices were illuminated for 5 s with a light intensity of 200 mW cm^{-2} . In all experiments, the gate electrode is gold.” What is the wavelength used in this experiment? This information should be added. In addition, the graph reports the values of current in nanoampere, while in the text the values are expressed in microampere. It would be better to be coherent with the units used.

We have added further details about the experiment to the caption of Fig. 2g. We also thank the reviewer for pointing out our mistake regarding unit consistency, the reported values’ unit are now changed to μA in Fig. 2g:

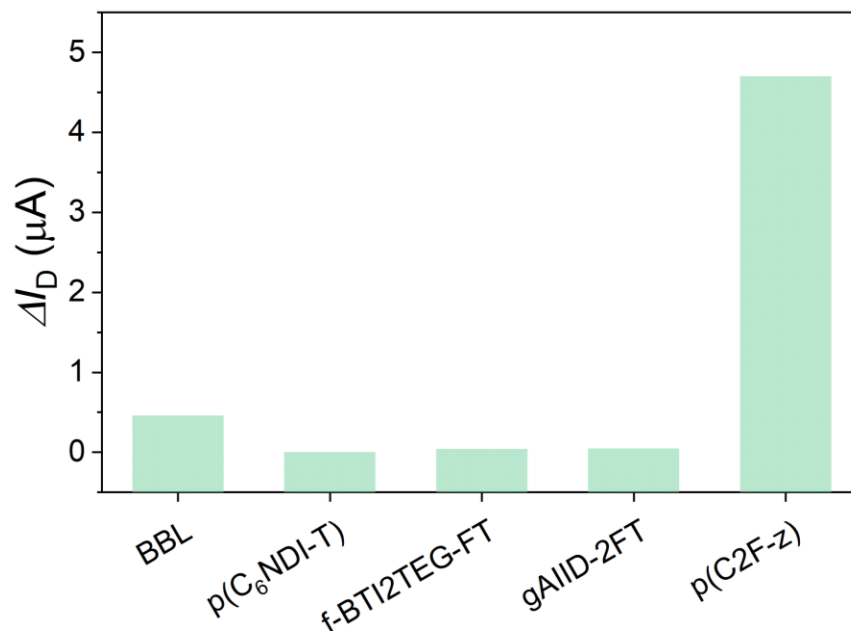


Fig. 2 g) The comparison of photocurrent generated by other n-type OEFTs. The devices were illuminated by 375 nm light for 5 s at a light intensity of 200 mW cm⁻². The devices were operated in 0.1 M NaCl using Au gate electrode and had identical channel size.

14. In the manuscript, it is reported “These memories can be consolidated into long-term memory (LTM) through rehearsal learning. Our device exhibited a similar pattern, wherein after the initial learning phase (50 excitation pulses) and subsequent forgetting, the current could be restored to its previous state using only 25 pulses (Fig. 3a).” In Fig. 3a it is not clear which is the initial learning phase and the subsequent one. Moreover, in the graph seems that the channel conductance cannot be recovered after the forgetting state, and this should be the reason why a continuous increase of the baseline is highlighted. Overall, it is not clear which is the purpose of this experiment.

We understand the reviewers’ confusion and appreciate their careful evaluation which allowed us to correct this figure. We now clearly show the initial learning phase and the subsequent one. We have replaced Fig. 3a with the following figure and caption:

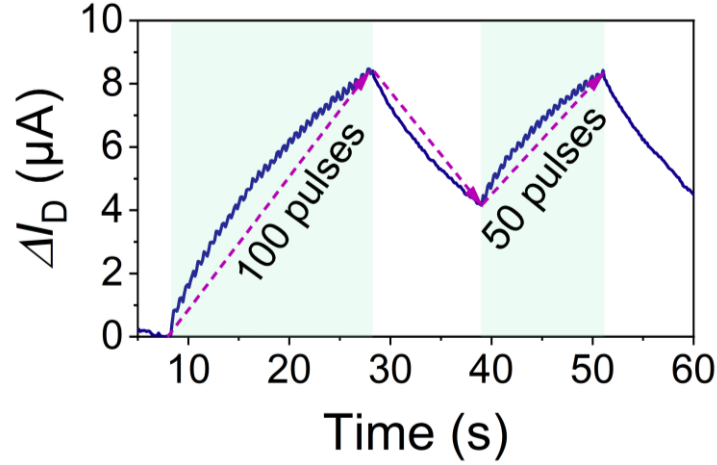


Fig. 3. a) Learning behavior in which only a few pulses (50) restore the memory gained through training with more pulses (100). The light pulse was at 375 nm with 200 mW cm⁻² intensity.

The figure now corresponds to the description: “Our device exhibited a similar pattern: after the initial learning phase (100 excitation pulses) and subsequent forgetting, the current could be restored to its previous state using only 50 pulses (Fig. 3a). Continuous sequential learning can enhance the consolidation of long-term memories (Fig. S20a).”

Additionally, we have moved the original Fig. 3a to the supporting information as Fig. S20a, which corresponds to the description: “Continuous sequential learning can enhance the consolidation of long-term memories (Fig. S20a).”

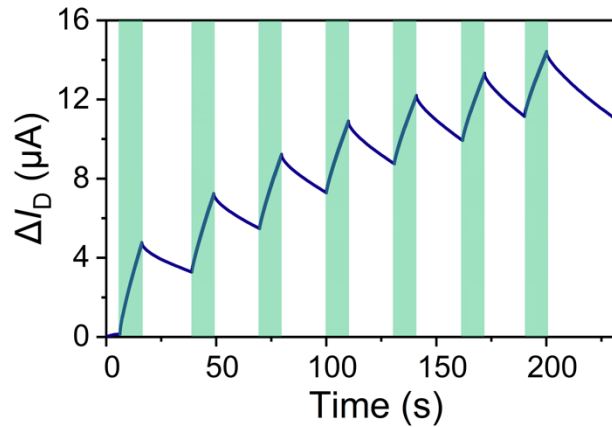


Fig. S20. a) The learning-forgetting-relearning protocol repeated seven times. The green highlighted area (50 pulses) represents the period under the light (200 mW cm⁻², λ=375 nm).

Moreover, in the graph seems that the channel conductance cannot be recovered after the forgetting state, and this should be the reason why a continuous increase of the baseline is highlighted

After the forgetting state, the channel conductance is actually recovered but this time with much less stimulation. A continuous increase in the baseline in Fig S20a is because a new learning cycle is introduced, which increases the strength of connections. Moreover, the photoresponse is

reversible, as shown in Fig. 3i and Fig. 2e. Nevertheless, Fig. 3a and Fig S20a are not intended to demonstrate the recovery of the conductive state of the material or the stability of the device. Instead, these two experiments aim to simulate the process by which the human brain acquires optical information: learning, forgetting, and relearning (see the Fig. R1 below). Currently, there are two ways to represent the transition from short-term memory to long-term memory in devices. One approach involves reducing the required pulse count by training to achieve the same memory level (Fig. 3a and Nat. Photon. 17, 629–637 (2023)). Usually, the relearning process takes a shorter amount of time compared with the first-time learning. This behavior was also achieved in our device. After first-time learning (100 pulses of excitation) and forgetting, the current can be restored to its previous state with only 50 pulses. The other method relies on continuous training to enhance memory (Fig. S20a, Adv. Mater. 2023, 35, 2300446).

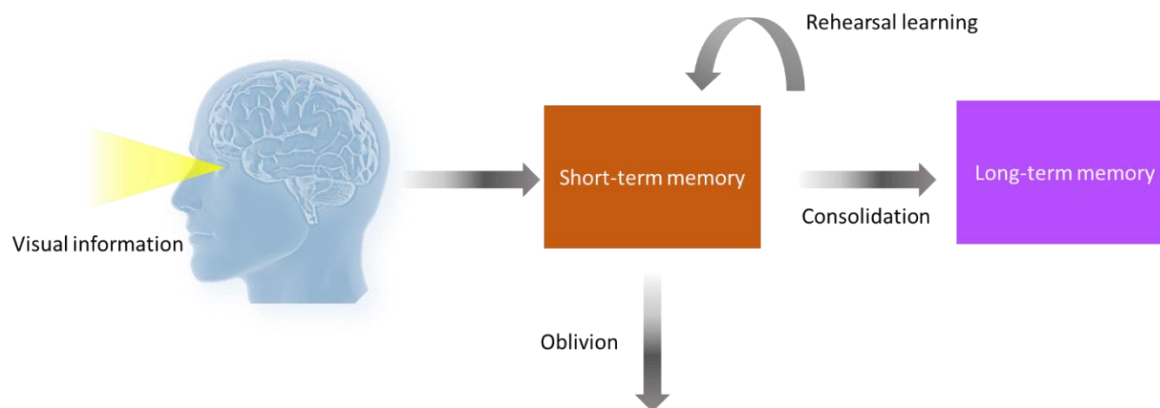


Fig. R1. Schematic of the memory-formation process in the human brain.

We now added a few introductory sentences to let the reader follow our aims:

“Our device exhibited a similar pattern: after the initial learning phase (100 excitation pulses) and subsequent forgetting, the current could be restored to its previous state using only 50 pulses (Fig. 3a). Continuous sequential learning can enhance the consolidation of long-term memories (Fig. S20a). Another way the human brain acquires information is through associative learning, where a connection is made between two stimuli or between a behavior and a consequence. Associative learning helps in forming long-term memories by creating strong connections between different pieces of information. We used long-term potentiation to simulate Pavlovian associative learning, as shown in Fig. 3b”

15. In the manuscript, it is reported:”When either Light 1 (375 nm) or 291 Light 2 (455 nm) was turned ON, the excitatory postsynaptic current ID surpassed the threshold 292 (12.6 μ A), indicating the “OR” logic operation (Fig. 3d and Fig. 3f). The ID exceeded the threshold 293 (15.7 μ A) only when Light 1 and Light 2 were applied simultaneously, indicating the AND logical 294 operation (Fig. 3e-f).” In Fig.3d-3f, thresholds are defined according to the experiment but it is not clear how much 0 and 1 states can be discriminated by using these thresholds. The threshold values used for the OR logic operation are quite different from the one used for the AND operation. The choice of the threshold should be explained better, and more experiments could be performed to prove the consistency of these computing capabilities.

We thank the reviewer for the comments on the threshold current for our logic function. We completely agree that this threshold current is defined based on experimental results. To improve the accuracy of our experiments and demonstrate the reproducibility of the device's computational capabilities, we conducted multiple experiments, performing four independent tests. The results are shown in Fig. R2 below. We can see that the data from the device is highly reproducible, indicating that the computing capability is repeatable.

Additionally, we have now redefined the threshold current, setting it at $14.6 \mu\text{A}$. Both the AND and OR logic operations use this the same threshold current. This definition clearly distinguishes between '1' and '0', demonstrating the excellent discriminative ability of the defined threshold current. We have updated the results in Fig. 3d-e. We hope this experiment addresses the consistency of the threshold current and clearly distinguishes between state '1' and state '0' based on this threshold current.

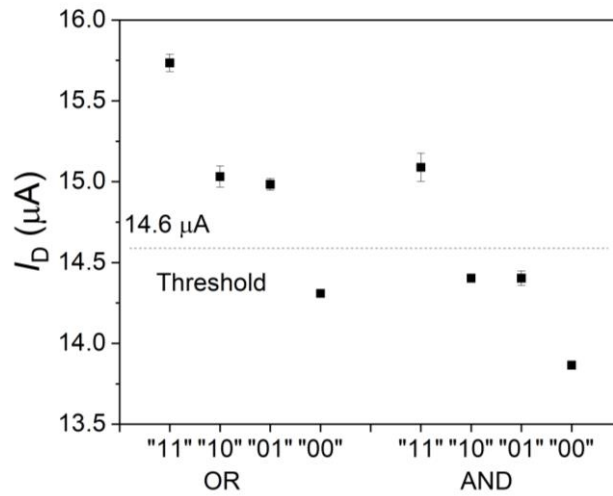


Fig. R2. The input (Light 1, 375 nm and Light 2, 455 nm) and output characteristics of the “OR” operation and “AND” operation.

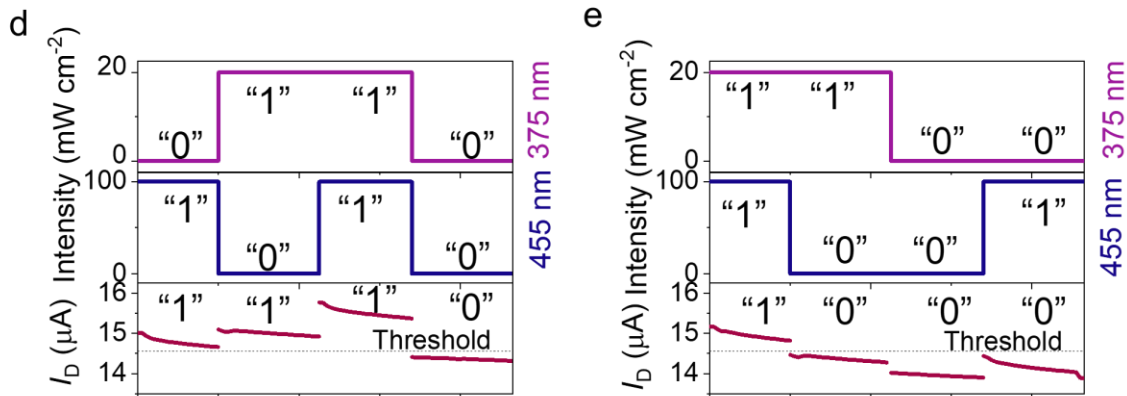


Fig. 3. The input (Light 1, 375 nm and Light 2, 455 nm) and output characteristics of the **d)** “OR” operation and **e)** “AND” operation. Light 1, “1”: a low illumination intensity (20 mW cm^{-2}), and a

high output current and “0”: no light, a low output current; Light 2, “1”: a high illumination intensity (100 mW cm^{-2}), and a high output current and “0”: no light, a low output current.

The text reads as:

“When either Light 1 (375 nm) or Light 2 (455 nm) was turned ON, the excitatory postsynaptic current I_D surpassed the threshold ($14.6 \mu\text{A}$), indicating the “OR” logic operation (Fig. 3d and Fig. 3f). The I_D exceeded the threshold ($14.6 \mu\text{A}$) only when Light 1 and Light 2 were applied simultaneously, indicating the AND logical operation (Fig. 3e-f). Furthermore, the device effectively responded to both optical and electrical stimuli through the adjustment of the values of the bias and light intensity, demonstrating OR/AND gate operations regulated by a combination of the two input types (Fig. S20c-e).”

16. In Fig.4e intensity values are missing. They should be added to the graph.

We have added the light intensity in the caption of Fig. 4e (but also to other figures where necessary).

17. Is the cell viability test for 24h enough to demonstrate the biocompatibility of the material? An explanation regarding the choice of this timing should be included.

We appreciate the reviewer for pointing out the cell viability description. There was an error in our description. The correct duration for cell culture is 48 hours, not 24 hours. We have updated the caption in Fig. S21 to: “A live and dead assay was performed on MDCKII cells cultured for 48 hours on polymer-coated glass and regular glass used as a control.”

We decided on 48 hours of cells in direct contact with the material/agent to be tested since it is often the standard duration for a cytotoxicity assay with this cell type. The optimal time depends on different factors including the cell type, the testing material/agent, and the endpoint. In this case, we are using MDCKII cells in which cytotoxicity has been widely tested for different materials/agents with exposure times ranging between 24 -72 hrs (Part Fibre Toxicol 8, 9 (2011), Toxicology in Vitro 22 (2008) 1846–1852 and Toxicology in Vitro 12 (1998) 209-217). Our endpoint was to do a general assessment of early apoptosis or cell death to indicate possible toxicity, which is often tested with material exposure between 24-48 hours (Assay Guidance Manual, Internet]. Bethesda (MD): Eli Lilly & Company and the National Center for Advancing Translational Sciences; 2004. PMID: 31070879 and Sci Rep 12, 9047 (2022)). Furthermore, in compliance with the International Standard ISO 10993-1 and 10993-5, cytotoxicity of medical devices/materials can be tested with extracts or by direct contact, and cell exposure to the material or extracts should take place at 37°C for 24-72 hours for interaction with both polar and nonpolar constituents from the test (10.3389/ftox.2023.1337468).

Reviewer 3:

I co-reviewed this manuscript with one of the reviewers who provided the listed reports. This is part of the Nature Communications initiative to facilitate training in peer review and to provide appropriate recognition for Early Career Researchers who co-review manuscripts.

We thank the reviewer for their comments which helped us improve the quality of our work. We hope that they appreciate the changes made in response to these comments.

Reviewer 4:

This manuscript introduces an organic electrochemical transistor (OECT) and its array utilizing a photoactive mixed conductor, p(C2F-z), as the channel material. The device exhibits multispectral visual perception across a broad range of wavelengths, surpassing the capabilities of human vision. It also displays synaptic behaviors like PPF and synaptic plasticity, simulating the learning process of then human brain. The switching mechanism involves light-induced doping of the channel and interaction with separated charges within the electrolyte. However, certain explanations lack clarity or accuracy, and the device structure and array schematics don't align perfectly with the optical images. Additionally, the applications demonstrated using the OECT array seem somewhat generic and lack distinct advantages over similar devices. Also, it looks that they are not proper applications. It looks like they did not reflect their own strengths driven by this selected material and device.

We appreciate these comments of the reviewer, which helped us to improve our manuscript by writing with more clarity, correcting for certain claims and improving our figures. We also made sure to connect the applications section to the polymer discovery section. We now explain better why this particular device was used in the chosen application and how its properties were leveraged. We also introduce a new application that specifically relies on light sensitivity. We have substantially revised the manuscript as per the suggestions of the reviewer, even in parts where they have not raised any concerns.

There are several suggestions and questions on this manuscript as follows,

(1) In Fig. 1, the right of Figure 1a (device schematic) did not match Fig. 1b. It is better to indicate which parts (source, drain, gate, electrolyte, channel) are. I did not find the VTH in Fig. 1d.

We have taken new photographs of actual exemplary devices, demonstrated the source-drain and gate regions (Fig. 1b), and labeled the dimensions directly on the device. Additionally, to make our device schematic more representative of the actual device, we have modified the right-hand side schematic side of Fig. 1a.

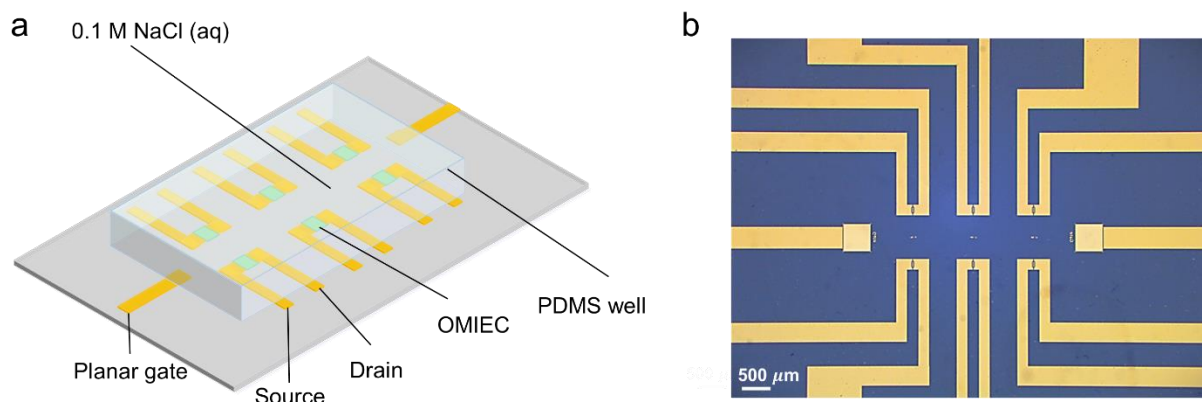


Fig. S7. a) Schematic representation of OEETs with six channels and two lateral gate electrodes, **b)** the microscope image of OEET device...

In addition, we have also revised Fig.1 d and added a label of $1/V_{TH}$ as the x-axis.

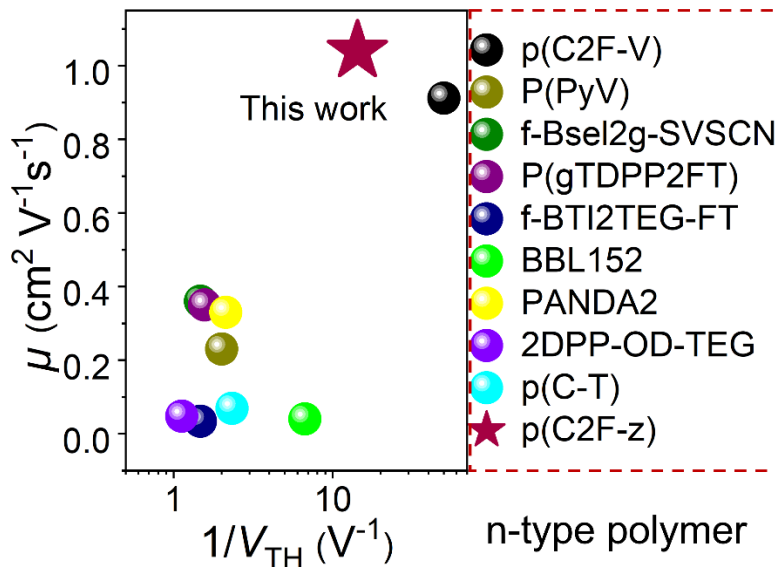


Fig.1 d) Comparison of p(C2F-z) device performance metrics (μ and V_{TH}) with those of other high-performance n-type OEETs.

(2) Figure 1f shows the $ID/ID,0$ as a function of time. Considering this device is OEET that does not memorize the states when the voltage is turned off (Fig. 1e), it should be careful to define and characterize “the retention property”. They did not retain their programmed state. It looks the cycling endurance.

We have removed all retention claims, as discussed later in comment 3. In the original sentence “These features are accompanied by high operational stability (Fig. 1f), with the device exhibiting a channel current retention of 92.9% after 1 hour of doping/dedoping cycles.” The term “current retention” however refers to remaining current after 1 h doping/dedoping cycles. We have revised the sentence

“These features are accompanied by high operational stability (Fig. 1f), with the device maintaining 92.9% of its channel current after 1 hour of doping/dedoping cycles.” We have also changed the figure as shown above highlighting that this is a stability test.

(3) In Fig. 2, the author insisted the device achieved high-density linear weight update. Considering this spontaneous erasing process, this linearity differs from the change of the synaptic weights of the synaptic devices that can retain their states upon the time. They cannot be updated and retained. And in-situ and ex-situ learning process would be failed. Therefore, I believe that most of the applications that the author introduced, such as logic gates, motion detection (for FNN), and accuracy estimation, are not proper tasks for highlighting this device.

We agree that making small, precise updates to a system’s stored values reflects or creates a type of memory or characteristic that doesn’t get erased when the system is powered off. Only when the updated weight values are retained can they be used in subsequent training and inference. Therefore, we have removed the description that our device implements high-density weight updates. The caption of Fig. 2b “**b) The device current was written with 512 light pulses (200 mW cm⁻², the pulse width t_p = 0.1 s, the pulse interval Δt = 0.05 s), which achieved high-density linear weight updates and was spontaneously erased.**” has changed to “**b) The device current change as a function of time as the device was modulated with 512 light pulses, and the current was spontaneously erased.**” In addition, the description “The device exhibited precise linear weight updates within a wide dynamic current range of 21.96 μ A, achievable through 512 subsequent light pulses” has changed to

“The device achieves these conductance states within a wide dynamic current range of 21.96 μ A. Only after 340 seconds spent in the dark (ca. 3 times more time spent in recovery compared to stimulation) did the current return to its initial level, suggesting that the device shows somewhat charge storage ability induced by light (Fig. 2b).”

Regarding image recognition and memorization, we corrected the text as:

“The current profile changed following the shape of these letters, showcasing image recognition capability of the device. Although weakly, the ‘KA’ shape can still be observed after the light was OFF (Fig. S28).”

Non-volatile properties are crucial for matrix computation in in-situ and ex-situ learning. However, in our work, we harness the temporal dynamics with volatile abilities to adjust the synaptic weight for video (motion) information decoding, and we do not utilize the non-volatility for reservoir computing. Recent studies about in-sensor computing were also based on temporal dynamics of the devices (see Nat Commun 14, 468 (2023) and Nat Electron 6, 870–878 (2023)).

To emphasize which property of the device we are relying on, we have revised the text as follows:

“Reservoir computing is an emerging approach inspired by neuromorphic systems, particularly suited for efficient edge learning in dynamic signal processing. It maps temporal signals onto a high-dimensional feature space through a fixed dynamic system with fading memory, enabling effective analysis of time-dependent data. We leverage the spatiotemporal dynamics of our light responsive OECT, which exhibits nonlinear current responses to light stimuli, to design a reservoir encoder that effectively preprocesses and extracts information from event-based data..”

“ To demonstrate the capability of the device to integrate spatial and temporal dynamics, it was applied to high-accuracy motion recognition in machine vision applications. To evaluate its motion perception abilities, we used the device to classify the direction and speed of moving balls (Fig. 7d, see Fig. S33 and Supplementary Note 6 for details). By extracting a feature map from event-based data, the sensor array provided input to a compact artificial neural network (ANN) for motion detection (Fig. 7e). The sensor’s ability to capture both spatial and temporal dynamics enabled the creation of a spatiotemporal feature map, which provided comprehensive information about movement behavior, including direction and velocity, critical for accurate classification. In contrast, conventional sensors acquire spatial information from individual frames but do not encode temporal dynamics within the data (see Supplementary Note 6 for details).”

The accuracy estimation highlights the device's ability to dynamically encode and respond to temporal information with spontaneous adaptability.

“Furthermore, the neural network exhibited robustness to device-to-device variations, maintaining a training recognition accuracy above 80% even with a 30% variation in device conductance (Fig. S34), demonstrating the network's tolerance to non-ideal conditions”

In experiments simulating Pavlovian conditioning and logic functions, we utilized wavelengths of 375 nm and 455 nm, exploiting the material’s spectral sensitivity. To emphasize this point, we included a description in the Pavlovian conditioning:

“Another way the human brain acquires information is through associative learning, where a connection is made between two stimuli or between a behavior and a consequence. Associative learning helps in forming long-term memories by creating strong connections between different pieces of information. We used long-term potentiation to simulate Pavlovian associative learning, as shown in Fig. 3b. Since this material has broad and distinct spectral sensitivity, we used two sets of consecutive spikes under different wavelengths of light ($\lambda=455$ nm and $\lambda=375$ nm) representing the “bell” and “beef” stimulus, respectively.”

and logic function:

“Besides the ability of dynamic learning, a synaptic device could process information using logic functions. We leveraged our OMIEC’s distinct current response to individual wavelengths of light and used two wavelengths as inputs to modulate the conductance of a single channel, generating OR and AND operations (Fig. 3c)..... Furthermore, the device effectively responded to both optical and electrical stimuli through the adjustment of the values of the bias and light intensity, demonstrating OR/AND gate operations regulated by a combination of the two input types (Fig. S20c-e)”

These works, including logic gates, motion detection, and accuracy estimation emphasize the device’s inherent ability to adjust and reset states in real-time sensing scenario, which can lead to more efficient and adaptive processing.

To further highlight the particular functionality of the selected material and device, we now demonstrate the material’s ability to differentiate RGB colors, showcasing its potential for color-discriminative applications.

We have added the following description to the main text.

“Leveraging the device’s sensitivity to specific wavelengths, we next explored its differential

response to R, G, and B light for full-color image pre-processing. A sample color image from the CIFAR-10 dataset was decomposed into R, G, and B channels, with Gaussian random noise added to the R and G channels to simulate noisy conditions (Fig. 6a). During the mapping process, the R, G, and B pixel values were assigned to corresponding light pulse intensities at their respective wavelengths. The device's noise reduction capability was influenced by its wavelength-specific photoresponsivity, with a notably higher photocurrent response to blue light (455 nm) compared to longer wavelengths. We evaluated the photoresponsivity using varying pulse counts (10, 20, 50, and 100), as shown in Fig. 6b. With increasing pulse numbers, the contrast between blue features and other components of the image was enhanced, significantly improving the clarity of both the original and noisy images (Fig. 6c, Fig. 6d). This result demonstrates that repeated light pulses selectively emphasize blue features within the image, effectively isolating them due to the device's wavelength-specific sensing properties. The methodology for this color image preprocessing approach is described in further detail in Supplementary Note 4.”

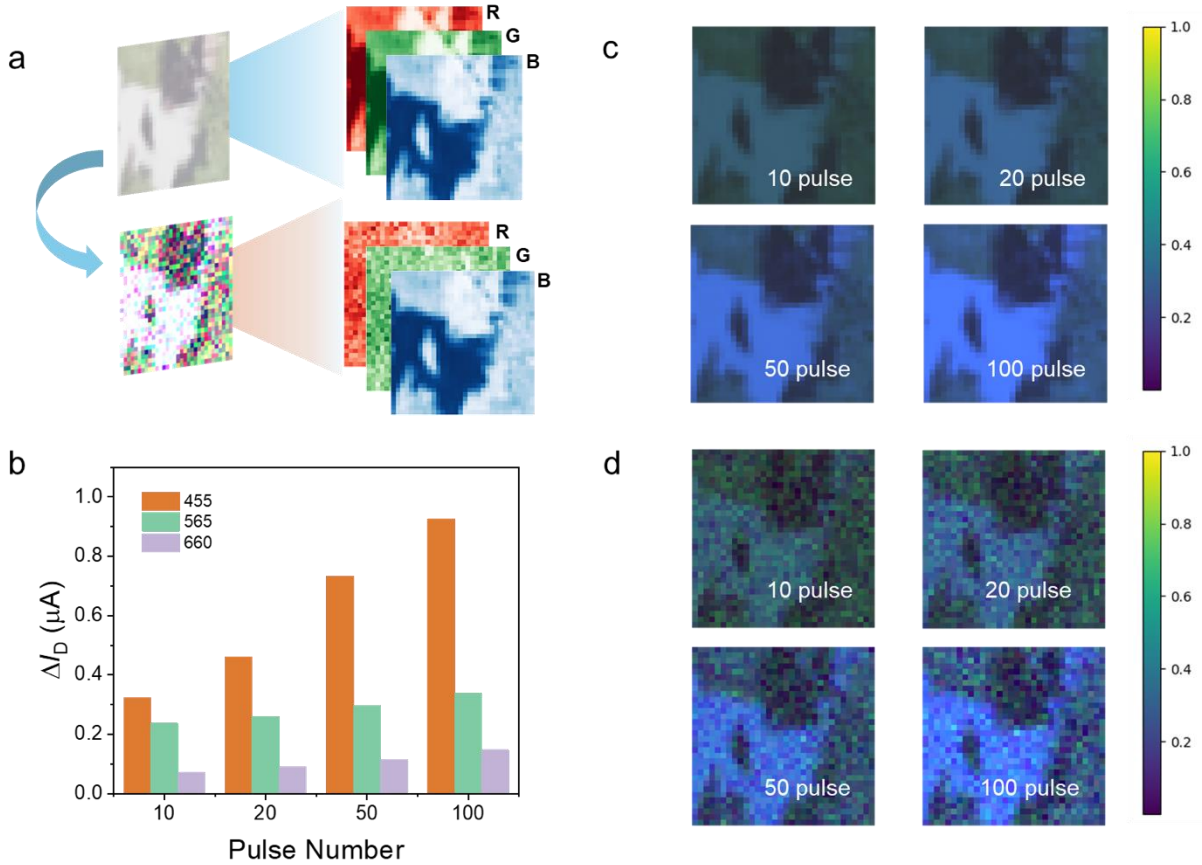


Fig. 6. **a)** A schematic for the color information encoding process. **b)** The channel current change as a function of pulse number under different light sources: red (660 nm), green (565 nm), and blue (455 nm). The contrast between the blue and other features increases in the **c)** original image and **d)** noisy image with the number of light pulses.

We added the methodology for color image processing to Supplementary Note 4.

Supplementary Note 4. Multispectral sensing for full-color image processing

In color image processing, the selected color image was first sampled into the R, G, and B components. Since the device exhibits a wavelength-dependent current response, the image

processing result based on the device characteristics is the sum of the responses to light of different wavelengths. The equation used in the simulation is as follows:

$$f_X(X) = a \times X^b + c$$

$$f_{output} = f_R(R) + f_G(G) + f_B(B)$$

where X represents the grayscale value of each channel, a is the weight associated with the corresponding photocurrent response, and b and c represent the exponential term and bias, respectively.

We now also changed the flow of the work where we first show applications that rely on the light response of the OECT and then show two applications that rely on the response generated upon electrical response (e.g., graded neuron). We hope that the text we have now added in each section, that clearly connects the material property to the application and the new application we are involved in will help the community understand the opportunities (as well as limitations) of this type of device.

(4) In Fig. 2b, it needs to address why they show asymmetric change of programming and erasing process. Also, I suggest that the position of 512 states and their counts are better displayed clearly. It looks that the number of states is dependent on the number of light pulses. How much can the device increase the states? When the light pulses increase, does the change of ID saturate?

The asymmetric changes in programming and erasing mentioned by the reviewer pertain to the device operation mechanism, which we explained in the section “The mechanism of light-based doping and memory generation” with Figures 4 and S24. We hypothesized that the asymmetry arises because the ions are injected into the material quickly during the programming phase under light exposure but are released more slowly in the erasing phase due to interactions with the other residual charges in the material. This leads to a slower "erasing" than "programming" process.

A per reviewer's suggestion, to enhance the visibility of the 512 data points, we now plotted the relationship between the pulse number and the current variation and included it in the Supplementary Information (Figure S9g). As shown in this figure, the current values (i.e., the number of states) has a steady increase over time and depend on the number of light pulses. We hope that the position of 512 states and their counts is now better displayed.

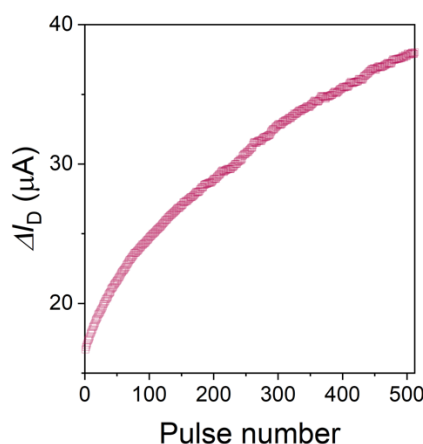


Fig. S9 g) The device current was written with 512 light pulses (200 mW cm^{-2} , the pulse width $t_p = 0.1 \text{ s}$, the pulse interval $\Delta t = 0.05 \text{ s}$)

When the pulse count increases to a certain level, the current saturates. When the number of current pulses reaches 1000, the current rising slope becomes smaller and nonlinear. Continuing to increase the number of pulses, the current rises very slowly. When the number of pulses increased to 1673, the device current nearly approached saturation, as shown in the figure below.

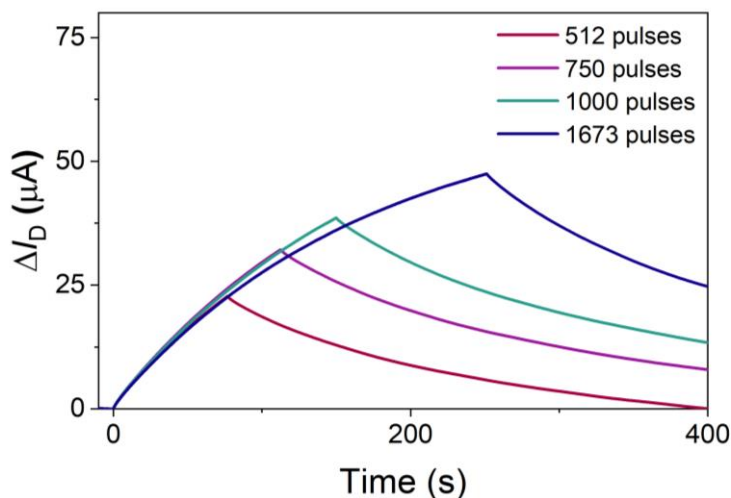


Fig. R3. The photonic response to various pulse number (200 mW cm^{-2} , the pulse width $t_p = 0.1 \text{ s}$, the pulse interval $\Delta t = 0.05 \text{ s}$)

(5) If the author wants to argue that this device is non-volatile, it needs to show the retaining capability over long time. Returning current to its original level after 340 s indicated the volatile switching. Furthermore, the current is continuously decreasing its value.

We agree with the reviewer's insightful comment regarding the volatile nature of our device, we have removed any non-volatility claim from the text as the light response is not retained more than this value. As the device has several other features to highlight, we show that the device has only

this asymmetric response and hence a strong memory behavior is not achieved. Please see the changes made written in response to the comment above.

(6) For the logic application, the author used two different wavelengths of lights. I don't see why the author used different light pulses. These basic logic gates can be achievable even if the same light pulses use. Therefore, there seem to be lacking in evidence for the following paragraph "the wavelength-dependent programmable response of the optoelectronic synapse can improve visual perception capability and enhance the performance of optoelectronic Boolean logical applications. (page 13)" Also, the fundamental logic gates can not be implemented by AND and OR gate. Universal logic gates (NAND and NOR) need to be demonstrated if the author wants to sustain this argument. And, real device images (for Fig. 3c) how they are measured and connected need to be shown. Showing just device schematic is not enough to understand them.

We agree that basic logical functions can be implemented using single-wavelength pulses but the point is that it can also be made by two wavelengths, enhancing the functionality of the system. Our initial choice of using two wavelengths (375 nm and 455 nm) was driven by our intention to leverage the material's inherent multispectral response. To emphasize this point, we included a description in the logic function "We leveraged our OMIEC's distinct current response to individual wavelengths of light and used two wavelengths as inputs to modulate the conductance of a single channel, generating OR and AND operations (Fig. 3c)." in the main text.

To improve the accuracy, we revised the description by removing the sentence "the wavelength-dependent programmable response of the optoelectronic synapse can improve visual perception capability and enhance the performance of optoelectronic Boolean logical applications." Additionally, we modified the description "Besides regulating dynamic learning processes, the wavelength-dependent programmable response of the optoelectronic synapse can improve visual perception capability and enhance the performance of optoelectronic Boolean logical applications." to "Besides the ability of dynamic learning, a synaptic device could process information using logic functions."

We also agree with reviewer that fundamental logic gates cannot be implemented by AND and OR gate, which should include NAND and NOR. Therefore, we adjusted the description and removed the words of logic gates with logic function or operation. The caption of Fig.S20 "c) The truth table of AND/OR logic gates." changed to "e) The truth table of AND/OR logic function."

Furthermore, we have added a real image of the setup for optical logic functions of "AND" and "OR" operations of the optoelectronic synapse in Fig. S20b. The caption for Fig.3c now states: "The setup of optical logic functions of "AND" and "OR" operations is shown in Fig. S20b." Our device configuration includes two LED lights (375 nm and 455 nm) positioned above as the input. The device was tested using a semiconductor analyzer connected via alligator clips, with a PDMS reservoir for storing the electrolyte.

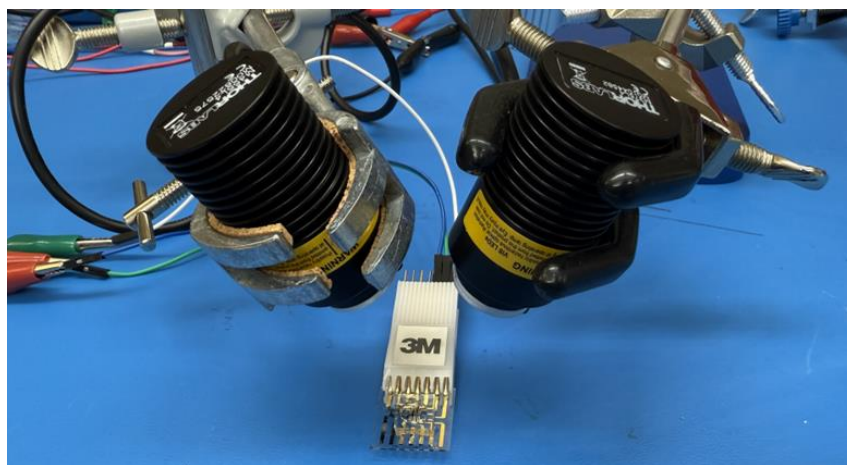


Fig. S20. b) The setup evaluating optical logic functions of the optoelectrochemical synapse.

(7) Similarly, there is no sufficient information of Fig. 5a. Where is the electrolyte? Is electrolyte used covering all the channel (like common dielectric layer)? Where are the gate lines? How do they function as artificial neural networks for human visual system?

We added another figure of the synaptic array structure in the supporting information (Fig. S27) showing clearly gates and channels, what the electrolyte is and the area that it covers. and included a note in the main text:

“We first recorded the transfer and transconductance characteristics of these 64 transistors throughout the entire area (Fig. S27a, see Supplementary Note 2 for experimental details) to assess the uniformity of channel response across the array.”

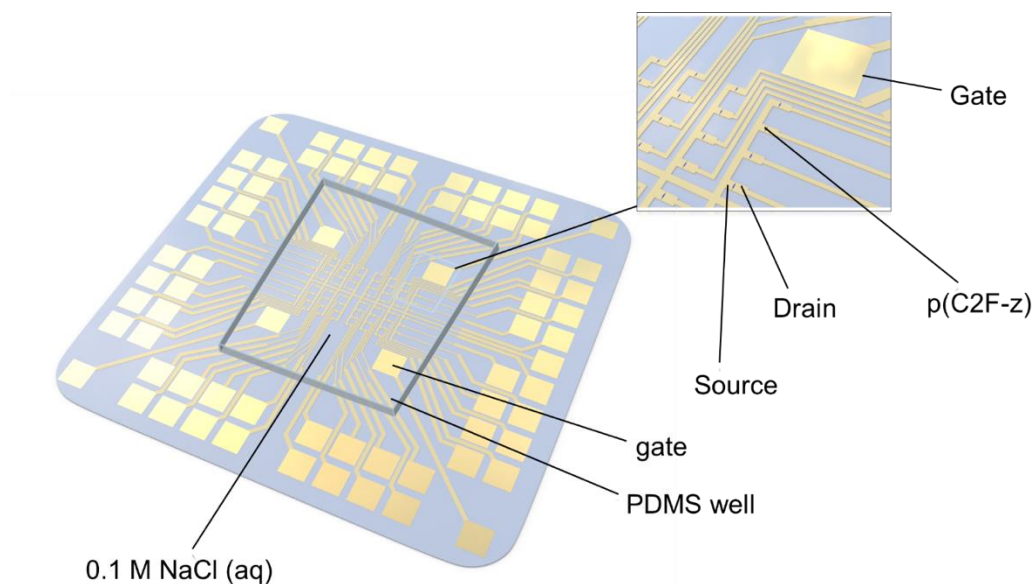


Fig. S27. a) The schematic of the 8x8 synaptic array. The inset is a zoomed-in view of a part of the device.

We also added more details about the architecture in the supplementary information.

Supplementary Note 2. The OECT synaptic array characterization

We used PDMS as a reservoir to contain 0.1 M NaCl solution. The electrolyte is in contact the channels and the large gold electrodes that serve as the gates exclusively, while the source, drain, and gate lines are shielded with Pa-C insulating layer, fabricated following the same fabrication process as for single devices. During current recording, a common gate electrode was employed, and we recorded the current of each transistor one after the after. In between each measurement, the Au electrode's electrochemical potential was monitored to avoid polarization, which may otherwise affect the next measurement.

As for the question regarding how this system functions as artificial neural networks for human visual system, we wrote a section describing that synaptic arrays designed for image perception and memorization function similarly to artificial neural networks (ANNs) in the human visual system by mimicking the way biological synapses process and store information (Nat Commun 14, 468 (2023) and Nat. Photon. 17, 629–637 (2023)). These synapses integrate optical information capturing, preprocessing, and memorization functions. To assess the retina-like signal pre-processing capability of synaptic array, we applied a light transistor array through a photomask with a “KA” pattern. The device responds to light stimuli in a neuromorphic manner, which means the channels can emulate the way biological synapses in the human brain process visual information. As a result, the letter “KA” is sensed by the array.

The related description in the main text “

The array was then illuminated through a photomask with a pattern displaying the letter ‘KA’, with each transistor channel corresponding to one image pixel (Fig. 5e). We measured the current change of 20 transistors immediately after light exposure (Fig. 5f) and 30-second post-light exposure (Fig. 5g). The current profile changed following the shape of these letters, showcasing image recognition capability of the device. Although weakly, the ‘KA’ shape can still be observed after the image was removed (Fig. S28).”

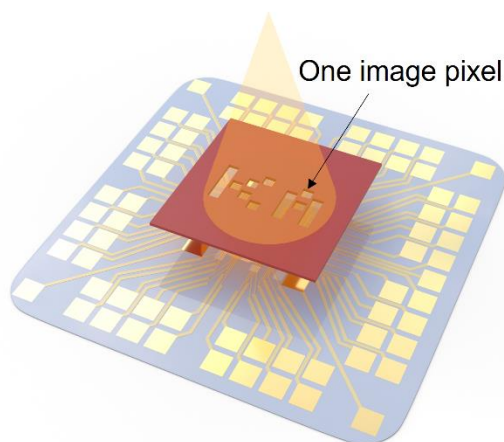


Fig. 5. e) Schematic illustration of synaptic active-matrix arrays with a photomask patterning the letters ‘KA’.

(8) The author compared the visual processing between their device and conventional sensor. However, there is missing the information of the conventional sensor devices. What does the training epoch mean for the image processing using “conventional sensor array”? Which parts in Fig. 6e are applied with the designed device? It looks vague and sufficient descriptions are missing.

We appreciate the reviewers’ feedback on the details of our simulation experiments.

In this study, the conventional image sensor is an image sensor that operates on a frame-based sensing principle that captures images at fixed intervals, such as a CMOS image sensor. A conventional image sensor captures images at fixed time intervals, meaning it takes a complete picture or frame at regular intervals (such as 30 frames per second). It doesn’t continuously track motion or changes within the scene in real-time; instead, it captures a series of still frames that, when played back, create the appearance of motion. This frame-based approach limits its ability to capture dynamic events as efficiently as a sensor designed for continuous or event-based sensing. (Adv. Mater. 9, 2203830 (2022) and Nat. Electron. 3, 546–553 (2020)). So the images for the video we used were taken by such a sensor.

We added in the caption of Fig. 7d:

“The sample motion video captured by a sensor array. The bioinspired sensor can extract temporal and spatial information, while the conventional image sensor only generates spatial information because it captures images at fixed time intervals.)”

We also added the description of this part in the main text:

“To demonstrate the capability of the device to integrate spatial and temporal dynamics, it was applied to high-accuracy motion recognition in machine vision applications. To evaluate its motion perception abilities, we used the device to classify the direction and speed of moving balls (Fig. 7d, see Fig. S33 and Supplementary Note 6 for details). By extracting a feature map from event-based data, the sensor array provided input to a compact artificial neural network (ANN) for motion detection (Fig. 7e). The sensor’s ability to capture both spatial and temporal dynamics enabled the creation of a spatiotemporal feature map, which provided comprehensive information about movement behavior, including direction and velocity, critical for accurate classification. In contrast, conventional sensors acquire spatial information from individual frames but do not encode temporal dynamics within the data (see Supplementary Note 6 for details).”

Fig. 7e (the original Fig. 6e) shows the motion detection task using our bio-inspired device, aimed at classifying the direction and velocity of a moving ball. The network input (also known as the extracted feature map) is applied with the designed device. The device decodes input information by extracting both spatial and temporal features. Specifically, the spatiotemporal information captured by the sensor array is transformed into a feature map, which serves as input to a compact artificial neural network for motion classification. The value of the feature map in the temporal frame is converted into the current. The feature map of our bio-inspired sensor is represented as discrete action events across temporal frames, with each pixel value converted into the corresponding current as shown in Fig. 7c. The final layer of seven-output neurons represents seven specific classes of ball movement, each with distinct directions and speeds: high, medium,

and low speeds for both clockwise and counterclockwise rotations, as well as a static state. The highest value among seven neurons represents the most possible classification result.

We have added all experimental details in Supplementary Note 6.

“For the ball movement detection tasks, we developed a customized video dataset using OpenCV. Each video frame was configured to dimensions of 32×32 pixels, with the ball's radius fixed at 5 pixels, and a frame rate of 30 frames per second. To enhance dataset diversity, the ball was randomly positioned at the start of each sequence and assigned velocities of 0, 1, 3, and 5 m/s. The dataset includes two rotation directions: clockwise and counterclockwise, with each class comprising 60 samples. The dataset was split into training and test sets at a ratio of 4:1. A sample video frame representation is shown in Fig. S33.

The feature map extracted from temporal frames was converted into electrical current values by the bio-inspired sensor (Fig. 7c). This feature map, represented as discrete action events across temporal frames, encodes essential spatial and temporal patterns. Each pixel value in the temporal frame corresponds to a specific current value. These feature maps were used as inputs to a neural network for classification. The network's final layer consisted of seven output neurons, representing seven classes of ball movement: high, medium, and low speeds for both clockwise and counterclockwise rotations, as well as a static state. The class with the highest neuron value was selected as the predicted result.

For comparison, we trained the feature maps generated by a conventional sensor array, where individual frames captured at fixed intervals were used as input. These feature maps were fed into the neural network, as illustrated in Fig. 7d, and the network was trained using backpropagation to optimize weights and minimize prediction errors. The difference in accuracy between the conventional and bio-inspired sensors is attributed to the latter's ability to integrate both spatial and temporal data, which provides a richer representation of motion.

The device's ability to capture spatiotemporal dynamics was further demonstrated using real-world video data, such as the movement of a mosquito (Fig. 7g). A video of the mosquito was recorded at a fixed frequency, and its movement velocity was estimated based on time intervals. Adjusting the video frame rate allowed us to simulate varying motion speeds; for example, increasing the frequency by a factor of 10 simulated a tenfold increase in perceived motion speed. The bio-inspired sensor encoded this motion information into spatiotemporal frames, breaking the video into sequences that captured both the spatial position and temporal evolution of the mosquito's movement. The feature map illustrates the sensor's ability to distinguish between fast and slow motions by encoding the temporal dynamics of objects. This example underscores the sensor's effectiveness in capturing and interpreting dynamic information for motion detection and classification tasks.”

This manuscript introduces an organic electrochemical transistor (OECT) and its array utilizing a photoactive mixed conductor, p(C2F-z), as the channel material. The device exhibits multispectral visual perception across a broad range of wavelengths, surpassing the capabilities of human vision. It also displays synaptic behaviors like PPF and synaptic plasticity, simulating the learning process of the human brain. The switching mechanism involves light-induced doping of the channel and interaction with separated charges within the electrolyte. However, certain explanations lack clarity or accuracy, and the device structure and array schematics don't align perfectly with the optical images. Additionally, the applications demonstrated using the OECT array seem somewhat generic and lack distinct advantages over similar devices. Also, it looks that they are not proper applications. It looks like they did not reflect their own strengths driven by this selected material and device. There are several suggestions and questions on this manuscript as follows,

- (1) In Fig. 1, the right of Figure 1a (device schematic) did not match Fig. 1b. It is better to indicate which parts (source, drain, gate, electrolyte, channel) are. I did not find the V_{TH} in Fig. 1d.
- (2) Figure 1f shows the $I_D/I_{D,0}$ as a function of time. Considering this device is OECT that does not memorize the states when the voltage is turned off (Fig. 1e), it should be careful to define and characterize “the retention property”. They did not retain their programmed state. It looks the cycling endurance.
- (3) In Fig. 2, the author insisted the device achieved high-density linear weight update. Considering this spontaneous erasing process, this linearity differs from the change of the synaptic weights of the synaptic devices that can retain their states upon the time. They cannot be updated and retained. And in-situ and ex-situ learning process would be failed. Therefore, I believe that most of the applications that the author introduced, such as logic gates, motion detection (for FNN), and accuracy estimation, are not proper tasks for highlighting this device.
- (4) In Fig. 2b, it needs to address why they show asymmetric change of programming and erasing process. Also, I suggest that the position of 512 states and their counts are better displayed clearly. It looks that the number of states is dependent on the number of light pulses. How much can the device increase the states? When the light pulses increase, does the change of I_D saturate?
- (5) If the author wants to argue that this device is non-volatile, it needs to show the retaining capability over long time. Returning current to its original level after 340 s indicated the volatile switching. Furthermore, the current is continuously decreasing its value.
- (6) For the logic application, the author used two different wavelengths of lights. I don't see why the author used different light pulses. These basic logic gates can be achievable even if the same light pulses use. Therefore, there seem to be lacking in evidence for the following paragraph “*the wavelength-dependent programmable response of the optoelectronic synapse can improve visual perception capability and enhance the performance of optoelectronic*”

Boolean logical applications. (page 13)” Also, the fundamental logic gates can not be implemented by AND and OR gate. Universal logic gates (NAND and NOR) need to be demonstrated if the author wants to sustain this argument. And, real device images (for Fig. 3c) how they are measured and connected need to be shown. Showing just device schematic is not enough to understand them.

- (7) Similarly, there is no sufficient information of Fig. 5a. Where is the electrolyte? Is electrolyte used covering all the channel (like common dielectric layer)? Where are the gate lines? How do they function as artificial neural networks for human visual system?
- (8) The author compared the visual processing between their device and conventional sensor. However, there is missing the information of the conventional sensor devices. What does the training epoch mean for the image processing using “conventional sensor array”? Which parts in Fig. 6e are applied with the designed device? It looks vague and sufficient descriptions are missing.