Organic molecular crystal-based photosynaptic devices for an artificial visual-perception system

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Abstract

Recreating the visual-perception properties using organic electronic devices is highly desired for visual prosthetics and artificial intelligence. Although the integration of organic light-sensing components with synaptic devices can realize the recognition and memory functions for perceived images, complicated problems in device integration for practical applications are generally encountered. Here we demonstrate a new type of organic photosynaptic device based on organic molecular crystals, which can provide optical-sensing and synaptic functions together in one device by means of a unique photon-induced charge transfer effect. This device successfully emulates the working principles of human visual perception in terms of short-term plasticity, long-term potentiation, and spike-timing-dependent plasticity. Moreover, a proof-of-concept artificial image-perception system is demonstrated by integrating the photosynapses on a flexible substrate. The new devices using organic semiconductors may open up innovative application areas, such as artificially intelligent electronic and perception systems, and facilitate the integration of such devices into next-generation flexible and stretchable electronics.

Introduction

Human visual perception refers to the brain’s ability to decipher what the eyes see, which corresponds to a remarkable imaging and memory system. Figure 1 illustrates the visual-perception process in the brain. The eyes receive incoming light from the external environment and focus it onto the retina, where an image of the visual stimulus is captured. Nerve cells in the retina as photoreceptors can convert visual information (light) into electrical impulses, which can travel along the optic nerve to the visual cortex at the back of the brain. The visual cortex contains a very large number of neurons. Then the neuronal network will arrange the features of the visual information in a meaningful way. Finally, the neuronal network will interpret the visual information so that we can perceive what we see. In the neural network, synapses act as a basic unit to transmit, memorize, recognize, and learn the visual information. Visual perception is the main channel for humans to obtain information. Recreation of the visual-perception properties using electronic devices could have profound implications for visual prosthetics and artificial intelligence. For example, an artificial visual-perception system could help blind people regain their visual senses. In light of the theories of and observations on visual perception, an artificial visual-perception device requires multifunctional integration of a light-sensing device similar to the retina and a signal-management system similar to the brain.

Organic semiconducting materials with unique features of long-term biocompatibility, good mechanical flexibility, and molecular diversity are ideal candidates for bionic perception devices. Recent research efforts on artificial visual-perception systems have achieved a high level of sophistication using organic electronic materials. Mostly, organic light-sensing components and synaptic devices were integrated into these systems to realize the recognition and memory functions for perceived signals. For example, Liu and co-workers reported the integration of organic heterojunction photodetectors and organic transistors with a ferroelectric dielectric as an artificial...
visual-perception system. Lee et al. also demonstrated an optic-neural synaptic device by integrating an organic photovoltaic detector with an organic nanowire synaptic transistor. These systems successfully achieved light information processing and readout with synaptic plasticity, but they generally face complicated device integration problems for practical applications. In addition, these photodetectors made from polycrystalline films with many defects and grain boundaries exhibited small photoconduction changes under weak light illumination, leading to ambiguous signals with a low signal-to-noise ratio. Compared with polycrystalline thin films, organic molecular crystals (OMCs) with fewer defects and grain boundaries can dramatically enhance charge transport, thereby enabling a large stimulated response in OMC-based neuromorphic devices. Therefore, implementing optical-sensing and synapsis functions together in one device based on OMCs is expected to provide a new and simple approach to artificial visual-perception systems.

Herein we demonstrate OMC-based photosynapses using a single device. The high crystal quality of OMCs ensures the fabrication of high-sensitivity photodetectors with a photoresponsivity up to 1650 A W\(^{-1}\) at a low gate voltage of 5 V. In addition, photon-induced charge transfer from the OMCs to oxygen-induced deep traps occurs under light irradiation, enabling the storage of photogenerated holes to achieve the functions of synapses. Based on these unique properties, we have realized various functions of biological neural systems for visual perception in organic photosynaptic devices, including the three important forms of short-term plasticity (STP), long-term potentiation (LTP), and spike-timing-dependent plasticity (STDP). Furthermore, a proof-of-concept artificial image-perception sensor is constructed on a flexible substrate, possessing the capability to recognize and remember optical images. Given their solution-processing capability, outstanding device characteristics, and high device flexibility, organic photosynaptic devices present unique opportunities for future artificially intelligent electronic and perception systems.

**Results and discussion**

In this study, 5,11-bis(triethylsilyl)anthradithiophene (Dif-TES-ADT) crystal arrays were used as photoactive layers due to their broad light absorption spectrum (300–650 nm), excellent air stability, and high carrier mobility (~6 cm\(^2\) V\(^{-1}\) s\(^{-1}\)). Details of the fabrication and characterization of Dif-TES-ADT crystal arrays can be found in Supplementary Figs. S1 and S2. The high carrier mobility of the organic crystals can accelerate the photocarrier extraction process, while the continuous array structure is important for the scale up of the devices for future applications. A typical organic photosynaptic device is composed of a gate electrode (G), divinyltetramethyldisiloxane-bis(benzocyclobutene) (BCB) and SiO\(_2\) insulator layers, Dif-TES-ADT crystal arrays, and source (S) and drain (D) electrodes (Fig. 2a). The SiO\(_2\)/Si substrate was covered with BCB to minimize traps and guarantee good wettability for organic solvents. As shown in Fig. 2b, a light pulse can be regarded as a presynaptic spike or an external stimulus. The upper surface of the Dif-TES-ADT crystal arrays under light irradiation emulates a presynaptic membrane. The organic semiconductor channel layer with S and D electrodes acts as a postsynaptic dendrite. The holes from the S are analogous to neurotransmitters and migrate in response to presynaptic spikes, while the holes between the insulator layer and Dif-TES-ADT crystal arrays emulate synaptic clefts.
First, we investigated the photoresponsive properties of the fabricated organic photosynapses. Figure 2c shows that the device exhibits good p-type transistor characteristics in the dark. When monochromatic light of 575 nm at 10 μW cm$^{-2}$ is vertically irradiated on the device, the S–D current ($I_{ds}$) remarkably increases, along with a positive shift of the threshold voltage ($V_T$) from −15.5 to −8.2 V, indicating easier turn-on of the device under light irradiation. The photoresponsivity ($R$) is an important parameter to evaluate the sensitivity of a photodetector$^{24-27}$. It is expressed as:

$$ R = \frac{I_{pc} - I_{dark}}{P} $$

(1)

where $I_{pc}$ is the photocurrent, $I_{dark}$ is the dark current, $P$ is the power, and $E_{hv}$ is the energy of the incident photon. The $R$ versus gate voltage ($V_g$) curve of our device is displayed in Supplementary Fig. S3. The OMC-based photosynaptic device has a higher $R$ (1650 A W$^{-1}$) and a larger $I_{pc}/I_{dark}$ ratio (~104) at a low $V_g$ of 5 V compared to the polycrystalline thin-film-based device (Supplementary Fig. S4). This indicates the extremely high light sensitivity of the OMC-based device. The wavelength-dependent $R$ in Fig. 2d shows that the photosynaptic device exhibits a broad photoresponse in the visible range and thereby allows recognition of visible signals for emulation of the human vision system. In addition to the high photosensitivity, the device also exhibits a strong persistent photoconductivity behavior, that is, the photocurrent can be retained and slowly decays even after removing the light (Fig. 2c and Supplementary Fig. S5). Interestingly, this slow relaxation behavior is quite similar to the decay process of action potentials that transmit through neurons and can thus be applied in emulating the working principles of photosynapses.

Synapses allow a neuron to pass a signal to another cell. Triggering of the excitatory postsynaptic current (EPSC) is thought to be an important process in assessing neuronal transmission$^{28}$. In our photosynaptic device, a white light spike with a relatively weak light intensity of 5 μW cm$^{-2}$ and a pulse width ($W$) of 120 ms was applied to trigger a postsynaptic current at a $V_{ds}$ of −30 V and a $V_g$ of 5 V. Figure 3a shows that the EPSC rapidly reaches a peak value of 46 pA and then gradually decays back to the initial value of ~6.8 pA within 5 s. This trend emulates the transmission process of an optical signal in a biological manner, in which the incoming light generates an action spike and is transmitted across the photosynaptic device to the next photosynaptic device. At a constant light spike amplitude, the EPSC of the photosynaptic device increases almost linearly from 12 to 62 pA as the spike duration increases from 30 to 180 ms (Fig. 3b and Supplementary Fig. S6). We note that the EPSC rise for the photosynaptic device is very steep, while the EPSC fall is relatively slow. The rise time ($t_r$) and fall time ($t_f$) are estimated to be 0.8 and 4.8 s, respectively. This behavior is related to the different accumulation and release rates of photo-generated carriers. It is known that ambient oxygen can diffuse into π-conjugated OMCs, leading to the formation of deep acceptor levels in the bandgap$^{29}$. The oxygen-
related levels could be directly detected using photoemission and absorption spectra and were found to be ~0.28 eV above the highest occupied molecular orbital for pentacene. To assess the impact of O\textsubscript{2} incorporation, we performed a density of states calculation of the energy band structure of the Dif-TES-ADT crystal and indeed revealed the induction of a distinct energy level at 1.8 eV above the Fermi level (Supplementary Fig. S7).

When the device is under light irradiation, electrons and holes will be generated in the light-absorbing Dif-TES-ADT crystals. Based on the energy band diagram in Fig. 3c, the electrons quickly transfer from Dif-TES-ADT to oxygen-related levels, forming O\textsubscript{2}\textsuperscript{−} anions, while the holes remain within the Dif-TES-ADT crystals and migrate to the BCB/Dif-TES-ADT crystal interface. Meanwhile, the O\textsubscript{2}\textsuperscript{−} anions form a negative space-charge layer on the top surface of Dif-TES-ADT crystals. The negative charges generate a gating effect, which can continuously trigger hole injection from the S electrode to maintain charge conservation in the device channel (Fig. 3d). Thus the hole concentration in the device rapidly increases, resulting in a fast EPSC rise. Since the penetration depth of light in the Dif-TES-ADT crystals is only 1–5 nm, the formed O\textsubscript{2}\textsuperscript{−} anions are predominately located on the crystal surfaces under weak light irradiation. After removing the light spike, the superficial O\textsubscript{2}\textsuperscript{−} anions are unstable and will gradually vanish (Fig. 3e).

This, of course, causes the slow disappearance of the formed negative space-charge layer, leading to gradually weakening of the gating effect. As a result, the hole injection is interrupted, and the EPSC gradual declines, leading to a long \(\Delta t\). The O\textsubscript{2}\textsuperscript{−} anion release rate can be defined by the following equation:

\[
\text{Rate} = \frac{dQ}{dt} = \frac{C}{\mu_{\text{sat}}} \frac{dV_T(t)}{dt} = -\left[\frac{2LC}{\mu_{\text{sat}}W}\right]^{0.5} \frac{d(I_{ds}(t))^{0.5}}{dt}
\]

where \(C\) is the insulator capacitance per unit area, \(W\) is the channel width, \(L\) is the channel length, and \(\mu_{\text{sat}}\) is the saturation regime mobility. According to the equation, the release rate of the O\textsubscript{2}\textsuperscript{−} anions is estimated to be \(-2.5 \times\)
$10^9 \text{cm}^{-2} \text{s}^{-1}$. To further prove the validity of photoinduced charge transfer from OMCs to oxygen molecules, we compared the transfer characteristics of the device under three different conditions: air, vacuum, and an oxygen atmosphere in the dark (Supplementary Fig. S8). No significant change was observed for the transfer characteristics. However, the photocurrent under a light spike in vacuum is much smaller than that measured in air (Supplementary Fig. S9). These results unambiguously demonstrate the important role of oxygen adsorption in controlling the EPSC in OMCs.

Short-term plasticity (STP) is an important foundation of learning and memory and is believed to be the origin of the short-term memory of the brain. STP refers to the change in the strength of a synapse’s response to an external stimulus over a time range of milliseconds to a few minutes. In biological synapses, paired pulse facilitation (PPF) is a fundamental form of STP, in which the EPSC triggered by the spike increases when a second spike closely follows a prior spike. STP is successfully mimicked in our photosynaptic device, as shown by applying two successive light spikes ($7 \mu W \text{cm}^{-2}$, 120 ms) with different interspike intervals ($\Delta t_{\text{pre}}$). Figure 3f exhibits the EPSC of the photosynaptic device for a $\Delta t_{\text{pre}}$ of 300 ms. The peak value of the second EPSC spike ($A_2$) is 2.38 times that of the first EPSC spike ($A_1$). The PPF index is defined by $A_2/A_1 \times 100\%$, the peak value ratio between the second EPSC ($A_2$) and the first EPSC ($A_1$). The PPF index gradually decreases as $\Delta t_{\text{pre}}$ increases (Fig. 3g), which is similar to the neural response. For a smaller $\Delta t_{\text{pre}}$, this interval is shorter than the release rate of $O_2^-$ anions, which thus adds to the total amount of photo-generated holes in the channel during the second-light-spike period. As a result, the second EPSC is higher than the first EPSC.

Long-term plasticity (LTP) is widely regarded as the mechanism of human memory. The synaptic strength will show a persistent increase following a strong external
stimulation or a number of successive stimulations. In analogy to biological synapses, LTP in the photosynaptic device is represented by the phenomenon in which the EPSC of the device can persist for a long time after removing light stimulation. To assess the LTP of the photosynapse, a stronger light spike (white, 30 μW cm$^{-2}$, and 50 ms) was input into the Dif-TES-ADT crystals to obtain the EPSC. Figure 4a shows the EPSC retention curve after the light spike. A nonvolatile increase from the initial state of A$_1$ (9.2 pA) to the final state of A$_2$ (2.7 nA) was observed. After removing the external light stimuli, it took approximately 190 s for the device to return to the initial state (Supplementary Fig. S10). In addition, the synaptic weight change, $\Delta W ((A_2 - A_1)/A_1 \times 100\%)$, is 3.0 × 10$^4\%$ in the Dif-TES-ADT crystal-based device. In contrast, when the polycrystalline Dif-TES-ADT thin film was used as the active layer, the EPSC showed a more rapid drop after light stimulation, and $\Delta W$ (128%) was significantly reduced owing to the existence of many structural defects (e.g., misorientations, voids, and grain boundaries) in the thin film (Supplementary Fig. S11). For the Dif-TES-ADT crystal-based device, $\Delta W$ as a function of the light intensity is depicted in Fig. 4b. As the intensity of the light pulse increases from 0.001 to 53 μW cm$^{-2}$, $\Delta W$ linearly increases from ~0.4 to 1.6 × 10$^5\%$. However, when the intensity of the light pulse is ~55 μW cm$^{-2}$, $\Delta W$ starts to gradually saturate. These results indicate that our artificial photosynapses show a better memory effect when facing a more impressive activity event. $\Delta W$ can also be well modulated by $V_g$ as shown in Supplementary Fig. S12. This means that the synaptic plasticity is modulated by both electrical and light stimuli, which can mimic dopamine-facilitated synaptic activity$^{36}$. The mechanism of LTP mimicry, in this case, can be explained as follows: a stronger light stimulus can generate more carriers in Dif-TES-ADT crystals, thus increasing the concentration of O$_2$$^-$ anions (Fig. 4c, d). In this scenario, the high-concentration O$_2$$^-$ anions undergo long-range diffusion, leading to the formation of oxygen-related traps inside the crystals. As a result, the lifetime and stability of the O$_2$$^-$ anions are considerably increased compared to those of O$_2$$^-$ anions produced under weak light. Therefore, even after stopping the light spike, the O$_2$$^-$ anions can remain in the Dif-TES-ADT crystals for a relatively long time and consequently maintain the device in a highly conductive state over a long retention time (Fig. 4e). This feature makes OMCs a unique platform for emulating the LTP process in photosynapses. In addition to a stronger light spike, a higher number (N) of weak light stimulations with a short interval can also enhance the connection strength between the photosynapses. Figure 4f shows the EPSC-t curve for the aforementioned stimulation protocol with 30 weak light spikes (5 μW cm$^{-2}$, a narrow pulse width of ~20 ms, and a high frequency of ~50 Hz). An obvious enhancement in the EPSC was observed after 30 light spikes, with a total $\Delta W$ enhancement of ~9.5 × 10$^5\%$. 

**Fig. 5 Spike-timing-dependent plasticity of the photosynaptic device.** a Schematic image of photosynaptic integration. b Schematic showing two connected photosynaptic devices for the emulation of spike-timing-dependent plasticity. c, d PSC variation for different $\Delta t$ values: c $\Delta t = +5$ s and d $\Delta t = +0.2$ s. e PSC change as a function of $\Delta t_{\text{post-pre}}$. 

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Note that the EPSC can persist for hours after removing the light irradiation (Supplementary Fig. S13). Only when a large negative gate bias of $-40$ V is applied does the EPSC quickly return to a low conductivity state, realizing a fast restorability of the synaptic weight (Supplementary Fig. S14). In addition, $\Delta W$ increases as the $N$ of the presynaptic stimuli increases (Fig. 4g), suggesting that all stimulation signals are accepted by the photosynapse. These results demonstrate that many transient light signals can be transferred into long-term memory via a consolidation process in the photosynaptic devices, which is consistent with the psychological model of human memory (Supplementary Fig. S15).

In biological synapses, STDP, known as the Hebbian learning rule, is an essential function\textsuperscript{14}. It refers to the magnitude of the connection strength, which closely depends on the relative timing of prespike and postspike activities. Figure 5a schematically illustrates the photosynaptic integration. To emulate the STDP function, two organic photosynaptic devices were integrated by connecting them with a common Au electrode (Fig. 5b). To avoid light spike 1 illuminating the other photosynaptic device, we placed an aluminum foil at the middle of the two devices, as illustrated in Supplementary Fig. S16. One photosynaptic device is regarded as a presynapse, while the other one is regarded as a postsynapse. Two synaptic light spikes (white, 8 $\mu$W cm$^{-2}$, and 500 ms) were separately applied to the presynapse and postsynapse, with a varied time interval ($\Delta t$). In this case, the connection strength between the two photosynaptic devices is defined as $\Delta PSC = (PSC_2 - PSC_1)/PSC_1$, where PSC represents the postsynaptic current. Figure 5c, d show the variation in the PSC when $\Delta t$ is $+5$ and $+0.2$ s. As $\Delta t$ increases, the connection strength is weakened. A typical symmetric form of STDP induced by temporal correlations of the presynaptic and postsynaptic spikes is obtained (Fig. 5e). The symmetric STDP characteristic is important for emulating more complicated neuromorphic functions of the brain.

To simulate the visual-perception processes of humans, we further integrated photosynaptic devices on a flexible polyimide (PI) substrate (Fig. 6a). An active-matrix array with 10 $\times$ 10 photosynapses was constructed on a semi-transparent, flexible substrate ($2 \times 2$ cm$^2$) (Fig. 6b). Supplementary Fig. S17 shows a top-view optical microscopic image of the photosynaptic device array. Next, the image recognition and memory capabilities of the photosynapse array were tested. We first imaged the characters “SU” on the device array by irradiating the devices above the “SU” characters with white light (150 $\mu$W cm$^{-2}$; Fig. 6c). It is noteworthy that these characters can be clearly resolved (Fig. 6d), revealing the reliable imaging function of the device. In addition, after removing the optical signal, the “SU” characters can still be clearly read from the active-matrix array at 10 min. This verifies that the visual-memory function of human beings has been successfully mimicked. Note that the EPSC for the photosynapses under or after light exposure is distributed in a relatively narrow range with little fluctuation, confirming the high uniformity and stability of the visual-perception system (Supplementary Fig. S18). The mechanical flexibility of the OMC-based photosynaptic device was further investigated. The synaptic performances under different bending radii (from a flat state to 7.0 mm) were recorded in situ, as shown in Supplementary Fig. 19a. $\Delta W$ decreases slightly with decreasing bending radius (Supplementary Fig. 19b). Furthermore, the photosynaptic performance of the flexible device was tested before and after multiple
bending stresses were applied. At a small bending radius of 7.0 mm, the ΔW of the device slightly changes even after 2500 bending cycles (Supplementary Fig. 20). This result shows that the photosynaptic devices have outstanding flexibility and are very robust against bending strain, suggesting the great potential of the OMC-based photosynaptic devices in next-generation flexible electronics.

Conclusions
In conclusion, we successfully demonstrate a new concept of an organic photosynaptic device that features synaptic and optical-sensing functions in a single device, which successfully avoids the use of sophisticated device architectures. By taking advantage of the high photosensitivity of OMCs and the unique photon-induced charge transfer effect, the photosynaptic device is capable of directly transmitting, memorizing, recognizing, and learning a light stimulus in a manner analogous to the biological neural system. Therefore, various important biological visual-perception functions, including STP and LTP behaviors and STDP characteristics, are successfully emulated. Furthermore, as a proof of concept, an artificial visual-perception system with 10 × 10 photosynapse pixels was constructed on a flexible substrate, which shows remarkable capabilities for recognizing and memorizing optical images. This artificial visual-perception system has promise for application in future visual prosthetics and intelligent products.

Materials and methods
Preparation and characterization
A solution of 3 mg mL⁻¹ Dif-TES-ADT (obtained from Luminescence Technology Corp) was prepared in toluene (99.9%, Sigma-Aldrich). Afterwards, ~10 µL of Dif-TES-Luminescence Technology Corp was prepared in toluene (~50 nm) was used to passivate the -OH groups on the trochemically active group-free BCB insulator layer (300-nm thermally grown oxide) and a blade. The elec- 

Device fabrication and measurement
Organic photosynaptic devices were made on BCB-covered SiO₂/Si substrates. An Au (50 nm) layer was deposited through a metal mask to form S/D electrodes. All devices were measured using a semiconductor parameter analyzer (Keithley 4200-SCS). Light pulses were generated using a function generator. Monochromatic light in the ultraviolet–visible range was captured by a spectrometer with optical filters, and a power meter was used to determine the light intensity.

Photosynapse array fabrication and characterization
PI of 120-µm thickness (DuPont™ Kapton® HPP-ST) was utilized as the substrate, and it was mounted on a Si substrate to maintain a flat state for ease of handling during fabrication. Then patterned gate electrodes (Ag, 100 nm) were thermally evaporated at 0.4 nm s⁻¹ under vacuum. A crosslinked poly(4-phenylphenol) (PVP) dielec-

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Conflict of interest
The authors declare that they have no conflict of interest.
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