2D Metal–Organic Framework Based Optoelectronic Neuromorphic Transistors for Human Emotion Simulation and Neuromorphic Computing

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1. Introduction

The explosion of information in the era of big data and the Internet of Things poses great challenges for von Neumann machines and sensors,[1–5] requiring new computing paradigms to meet the requirements of energy efficiency and big data workloads. Hardware-based neuromorphic computing, which mimics the operating principles and architectures of the brain through physical devices, is considered as one of the most promising platforms for big data computing as it has the potential to provide lower energy consumption and more efficient computing than the von Neumann machine in the future.[6–9] So far, plenty of devices have been reported with synapse and neural-like functions,[10–12] e.g., two-terminal memristors and three-/multiterminal transistors. Artificial neural networks based on an array of them have also been demonstrated for both low-level and high-level information processing.[13–18] Among these neuromorphic devices, three-/multiterminal neuromorphic transistors have attracted much attention due to their high degree of control freedom, enabling many complex neural and synaptic functions, such as dendritic integration.[19–22] Nonetheless, the development of three-terminal neuromorphic transistors is still in its infancy, and the capabilities of the devices need to be further expanded to achieve more interesting and useful applications.

On the other hand, in biosynapse, the receptors on the surface of the postmembrane can be activated by transmitters released from the premembrane.[23–26] The responsivity of the synapse to the stimulus depends on the receptor activity and the receptor number on the postsynaptic membrane. The illness of human emotions (mood disorders, depression, and stress) usually influences the synaptic plasticity in the human brain, which would further influence the memory and learning behaviors of the human.[30–32] The emulation of the receptor-tuning synaptic behavior is essential for a detailed understanding of the mechanism of synaptic behavior modulation. The multiterminal regulation ability of neuromorphic transistors gives us the ability to simulate the human brain to learn emotional regulation, but there are few related research reports.

2D metal–organic frameworks (2D-MOFs) with a periodic network structure are composed of metal clusters or metal ions and organic ligands.[33–35] The large specific surface area, stable crystal structure, and highly accessible active sites of 2D-MOFs
enable them with enormous application potential in a variety of fields, including catalysis, energy storage, gas separation, etc).

Recently, there are some reports about using MOFs as semiconducting materials and active layers for transistor and memory device fabrication, opening the door for using MOFs in electronic devices. However, few studies have explored the application potential of 2D-MOFs in optoelectronic neuromorphic computing devices.

Herein, we designed a 2D-MOF/poly(methyl methacrylate) (PMMA)-based optical-tuning dielectric layer for the fabrication of drain-tunneling neuromorphic transistors. In addition to typical light-stimulated behaviors (paired-pulse facilitation [PPF] and excitatory postsynaptic current [EPSC]), the level of source–drain voltage can be utilized to model the number of receptors on the postmembrane and control the behavior of the device in response to prestimulation. The emotion-dependent learning efficiency is also successfully demonstrated by our synaptic device via tuning the energy band alignment by changing the source–drain voltage (from −3 to −25 V). When the source–drain voltage (V_DS) is decreased to −1 V, the normal light-perception behavior of the device is completely depressed, which can be regarded as a human emotional illness. We also built a single-layer perceptron (SLP) neural network based on the extracted parameters from our device, and demonstrate the emotion-tunable learning capability of the neural network. This work can not only broaden the application scenarios of 2D-MOFs but also further advance the development of neuromorphic electronics.

2. Results and Discussions

2.1. Material Characterization of 2D-MOFs

The detailed fabrication processes of the 2D Zn_{2}(ZnTCPP) MOFs and the 2D-MOF-based neuromorphic transistors are provided in the experimental section and Figure S1a, Supporting Information. To prove that the Zn_{2}(ZnTCPP) MOF was successfully prepared, we performed X-Ray photoelectron spectroscopy (XPS) to characterize the as-prepared materials. As shown in Figure S1b,d, Supporting Information, the Zn 2p signal at ≈1017 eV and the N 1s signal at ≈399.7 eV were observed, respectively, and the C 1s signal consisted of three parts (−COOH, −C_{6}H_{2}, and −C_{6}H_{6}) was also being observed (Figure S1c, Supporting Information). The signals of these elements of the as-prepared materials are in good agreement with those observed in Zn_{2}(ZnTCPP) MOFs reported in the literature. In addition, X-Ray powder diffraction (XRD) was also performed to characterize the as-prepared materials (Figure S2, Supporting Information), which showed a similar XRD spectrum to the Zn_{2}(ZnTCPP) MOFs reported in the literature, confirming that the as-prepared materials are Zn_{2}(ZnTCPP) MOFs. The bulk Zn_{2}(ZnTCPP) MOFs were further dispersed into 2D Zn_{2}(ZnTCPP) MOFs by using ultrasonication, and the sheet structure of the 2D Zn_{2}(ZnTCPP) MOFs was confirmed by the images from transmission electron microscope (TEM) and atomic force microscope (AFM) (Figure S3, Supporting Information). 2D Zn_{2}(ZnTCPP) MOFs were mixed with PMMA and uniformly distributed in the PMMA film, as confirmed by the XPS with Ar ion etching technique (Figure S4, Supporting Information).

Our optoelectronics neuromorphic transistors were fabricated by using the 2D Zn_{2}(ZnTCPP) MOFs-PMMA film as the charge trapping layer. The cross-sectional scanning electron microscope (SEM) characterization on the device structure is shown in Figure S3b, Supporting Information. The thickness of the charge trapping layer is about 30 nm. The thickness is consistent with the AFM results (Figure S5, Supporting Information). The evaporated pentacene on the charge trapping layer displayed a typical island-growth layer-like structure, as shown in Figure S5c, Supporting Information. To characterize the optical properties of the 2D Zn_{2}(ZnTCPP) MOFs-PMMA film and the pentacene film, we performed ultraviolet-visible spectroscopy (UV–vis) analysis, as shown in Figure S6a, Supporting Information. The absorption peaks of the 2D Zn_{2}(ZnTCPP) MOFs-PMMA film and the pentacene film were located at ≈430 and ≈660 nm, respectively. The steady-state photoluminescence (PL) spectrums and the PL decay profiles of the 2D Zn_{2}(ZnTCPP) MOFs-PMMA film and the 2D Zn_{2}(ZnTCPP) MOFs-PMMA film/pentacene film are presented in Figure S6b,c, Supporting Information. Compared with the 2D Zn_{2}(ZnTCPP) MOFs-PMMA/pentacene film, the 2D Zn_{2}(ZnTCPP) MOFs-PMMA film exhibited stronger emission peaks in the orange and red regions under excitation at 405 nm wavelength. The PL decay curve of the 2D Zn_{2}(ZnTCPP) MOFs-PMMA was well fitted by a biexponential decay function, which presents two relaxation mechanisms, the lifetime of fast decay (τ_{1}) and short decay (τ_{2}). The fitting result can be quantified as τ_{1} = 1.1 ns and τ_{2} = 6.7 ns, respectively, while the profile of 2D Zn_{2}(ZnTCPP) MOFs-PMMA film/pentacene film exhibits a shorter decay time (τ_{1} = 1.0 ns and τ_{2} = 4.6 ns) due to the photoinduced charge transfer effect.

Before the demonstration of the essential emotion-tunable neuromorphic functions, the basic transfer performance (Figure S7, Supporting Information) and the synaptic behaviors of the device were characterized.

2.2. Basic Synaptic Performance of MOF-Based Device

In the biological synapse, the external stimulus is transmitted and processed by neurons and synapses (Figure 1). The transmission rate of the signal flow depends on the number and the activity of the acceptors. Therefore, the postmembrane with few and low-activity acceptors would induce a low postsynaptic current, which results in a weak signal response to the external stimulus. Compared with the low-activity synapse, the postmembrane with more acceptors would trigger enhanced postsynaptic plasticity because more acceptors can be excited by the transmitters. The photoresponsiveness of our neuromorphic device can be controlled by the source–drain voltage (V_DS), which means that the effect of V_DS on synaptic devices is similar to that of the number of receptors on the postsynaptic membrane. Therefore, we can use V_DS to modulate our device performance.

Before demonstrating the V_DS-tunable neuromorphic device performance, we first characterized the basic synaptic transistor performance of the 2D-MOF-based device (Figure 2, and S7, Supporting Information). Except the erase process of the device
(-15 V), the gate voltage applied on the device was kept at the value of 0 V to investigate the light effect of the device. In our device, the source–drain channel is regarded as the postsynaptic neuron, the $I_{DS}$ is regarded as the postsynaptic signal, and the channel conductance is treated as the synaptic weight. The photosensitive 2D Zn$_2$(ZnTCPP) MOFs-PMMA charge trapping layer can be regarded as a light-stimulated presynaptic neuron and the silicon gate can be regarded as an electrical-stimulated presynaptic neuron. In biosynapse, the presynaptic neurons contain neurotransmitters and the postsynaptic neurons contain neurotransmitter receptors. The signal reached the presynaptic neuron would open the Ca$^{2+}$ channel on the presynaptic membrane and therefore the membrane would release the neurotransmitters into the synaptic cleft. The neurotransmitters in the synaptic cleft eventually interact with the neurotransmitter receptors to induce presynaptic signals transmission to postsynaptic neurons, triggering the EPSC and the IPSC (inhibitory postsynaptic current). The investigation of the EPSC behaviors with the light spike at various wavelengths ranging from 365 to 530 nm exhibits that the 430 nm light spike can induce the largest EPSC amplitude (Figure 2b), which is consistent with the UV–vis spectrum of 2D Zn$_2$(ZnTCPP) MOFs-PMMA film (Figure 2b and S6a, Supporting Information). To assess the response of our neuromorphic device to light stimulation (430 nm), we further investigated EPSCs with different light spike durations (Figure 2c) and intensities (Figure 2d), respectively. The EPSCs could be enhanced with the increment of light spike duration or intensity.

We fabricated a device based on pentacene/PMMA structure without 2D-MOFs. The photoresponse performance of the device was characterized and compared with that of pentacene/2D-MOFs-PMMA device. As shown in Figure S8, Supporting Information, when the illumination was removed, the current of pentacene/PMMA device immediately dropped to the initial state, while the current of the pentacene/2D-MOFs-PMMA device decreased slowly. The results indicate that the generation, transportation, and trapping processes of the photogenerated charges on the MOFs polymer and organic semiconductor

![Figure 1](image1.jpg) **Figure 1.** The schematic illustration of the signal transmission behaviors is controlled by the receptor activities and receptor number.

![Figure 2](image2.jpg) **Figure 2.** a) Schematic of the light-triggered biosynapse and 2D-MOF-based neuromorphic transistors. b) The synaptic response of our device to the light spikes with different wavelengths (inset, the change of the EPSC vs light wavelength). c,d) The long-term plasticity (LTP) or short-term plasticity (STP) formation in our photonic synaptic device depends on the light-spike duration ($\lambda = 430$ nm, intensity: 100 $\mu$Wcm$^{-2}$) and the light-spike intensity ($\lambda = 430$ nm, duration: 1 s). e) The PPF ratios at various spike intervals.
PPF is an essential behavior in the biosynaptic system for temporary information processing, where the postsynaptic conductance (synaptic weight) can be enhanced via two consecutive presynaptic stimulations, resulting in the device showing higher conductance after the second spike than that after the first spike. To show that our device can simulate PPF behavior, a pair of optic-signal spikes with a certain spike interval was utilized as a presynaptic signal spike. The typical PPF behavior of the device under two consecutive light spikes was characterized. As shown in Figure S9, Supporting Information, two consecutive 430 nm 100 μW cm⁻² light spikes (1 s) with 1 s interval were applied to our synaptic device. The larger value of the EPSC triggered by the second light spike than that value triggered by the first light spike was observed. The PPF ratio can be defined by the following equation

$$\text{PPF} = \frac{A_2}{A_1} \times 100\%$$ (1)

where the $A_1$ and $A_2$ are the EPSC amplitudes of the first and the second light spike, respectively. Tuning the interval time can change the ratio of two-spike synaptic current. The PPF ratio would decrease when we increase the interval time ($t_{\text{inter}}$) (Figure 2e). The maximum value of the PPF is $\approx 149\%$, which was obtained at the minimum $t_{\text{inter}}$ of 100 ms. The electrons trapping in the 2D Zn₂(ZnTCPP) MOFs-PMMA layer induced by the first light spike have insufficient time to recombine with holes before the second light spike was applied, which results in an enhanced EPSC amplitude after the second light spike. The PPF decay can be described as the combination of rapid decay and slow decay, defined by the following equation

$$\text{PPF} = 1 + c_1 \exp\left(\frac{t_{\text{inter}}}{\tau_1}\right) + c_2 \exp\left(-\frac{t_{\text{inter}}}{\tau_2}\right)$$ (2)

where the $c_1$ and $c_2$ are the initial factors of rapid decay and slow decay, $\tau_1$ and $\tau_2$ are their corresponding relaxation durations. The experimental PPF decay result can be well fitted by Equation (2). In our case, $\tau_1$ and $\tau_2$ are 0.05 and 41 s, respectively. The property of strengthening or weakening the synaptic weight between two adjacent neurons is called the synaptic plasticity, which is supposed to be the basis of the information storage and learning functions in the brain. Here, the conductance of the device channel was regarded as the weight level in the biosynapse ($W$), which can be described with the equation below

$$W = \frac{\text{EPSC}}{V_{DS}}$$ (3)

By changing the light spike parameters, we achieved the simulation of different types of synaptic plasticity in our 2D Zn₂(ZnTCPP) MOF-based neuromorphic devices, suggesting the potential of our device for future neuromorphic computing applications.

Temperature stability is one of the key parameters when the device is practically operated. Therefore, we characterized the temperature effect on the device optoelectronic performance. As shown in Figure S10, Supporting Information, with the increase of temperature, although the device baseline current was gradually enhanced (Figure S10a, Supporting Information), our device can maintain decent photosynaptic behaviors (Figure S10b, Supporting Information), which suggests a certain degree of temperature stability of our pentacene/2D-MOF-PMMA device.

2.3. Demonstration of Emotion-Tunable Simulation

Humans have abundant emotions (such as happy, sad, plain, mild), which are essential in human learning and memory. Positive emotion would improve vitality and enhance learning efficiency. By contrast, negative emotion would depress vitality, resulting in a low learning rate (Figure 3a). The $V_{DS}$-tunable photosponse of our 2D Zn₂(ZnTCPP) MOF-based neuromorphic device can be utilized to mimic emotion-tunable memory and learning behaviors. Figure 3b displays the energy alignment controlled by $V_{DS}$ of the pentacene/2D Zn₂(ZnTCPP) MOFs-PMMA structure, which can be used to describe the mechanism of the $V_{DS}$ tuning capability. The Kelvin probe force microscope (KPFM) potential variation with the light illumination confirmed the hole accumulation in pentacene under illumination (Figure S11, Supporting Information). The level of the $V_{DS}$ determines the energy band structure. At high $V_{DS}$, the large energy difference between the drain electrode and gate electrode enhances the bending of the energy band in the pentacene and MOF layers. Light-generated carriers are easily transferred to the OSC channel, which results in a large ΔEPSC value. The recombination rate of the light-induced accumulated charge carriers in pentacene would be also depressed, delivering a long retention time. On the contrary, the recombination rate of light-generated carriers would be increased at a low $V_{DS}$ level because of the weak bending of the energy band. Furthermore, at the ultralow $V_{DS}$ or even zero-level state, the transfer process of light-induced carriers would be in chaos.

The device tests were then carried out for a $V_{DS}$-tuning demonstration. In Figure 3c, the device current ($I_{DS}$) can be defined as the vitality of the neuron, where the red face means happy, the yellow face means mild, and the blue face means sad. The presynaptic current value increases from 0.2 to 30 nA (sad, mild, and happy) when the $V_{DS}$ increases from 0.1 to 25 V, suggesting the $V_{DS}$ can be used to modulate the “vitality” of our device. As shown in Figure 3d, ten consecutive light spikes (0.3 s, 50 μW cm⁻²) were applied to the device with various $V_{DS}$ (different emotions: sad, mild, happy). The change of the device EPSC is enhanced with the increment of the $V_{DS}$ and the device at the $V_{DS}$ of −25 V exhibits the largest EPSC change (Figure 3e), while the level of the $V_{DS}$ would not change the amplification of the conductance change in the first spike (inset, Figure 3e). When a human was sad (negative emotion, low $V_{DS}$), the synapse displays a low steady signal and low responsivity, resulting in a low $A_{10}/A_1$ ratio. Humans under positive emotions can efficiently process the information and enhance memory. A high $A_{10}/A_1$ ratio was observed in our neuromorphic device under high $V_{DS}$ (happy) after ten light spikes (Figure 3f). With the increase of the applied $V_{DS}$, the change of the channel conductance was enhanced because more photogenerated holes accumulated in the pentacene channel (Figure 3g). The photinduced holes...
are hard to recombine with the electrons under large $V_{DS}$ because of large energy-band bending in the junction of pentacene/2D-MOF-PMMA. The electrons trend to be trapped in 2D-MOF and the holes trend to accumulate in pentacene, which results in high conductance and long retention time. In Figure 3h, the current variations after 100 s were extracted under various $V_{DS}$. The superlinear enhancement of the EPSC current with the increasing $V_{DS}$ can be observed. The above results show that the $V_{DS}$ can effectively control the photoresponse performance and the synaptic behavior of our device, which provides the promising platform for multifunction synaptic simulation. The above results show the $V_{DS}$-tunable neuromorphic performance of the device was successfully achieved.

Emotion illness is a kind of psychiatric disorder such as depression, anxiety disorders, and schizophrenia, which has an adverse effect on human society. In the simulation of the human emotion with device $V_{DS}$ level, the value range from $-3$ to $-25$ V refers to the normal human emotions including happy, mild, and sad. When the $V_{DS}$ is decreased to an ultralow level ($-1$ and $-0.1$ V), the device exhibits irregular responses to the same ten consecutive light stimuli ($0.3$ s, $50$ μW cm$^{-2}$) compared with the response of the device to light spikes under $V_{DS} = -3$ V (Figure 4b). The irregular response fluctuation of the device at $V_{DS} = -0.1$ and $-1$ V can be regarded as the synaptic unit in emotional illness. The damage of the depression in the human brain described in Figure 4c includes chaos, confusion, and irregular response to external stimuli. The external behaviors of the depression were successfully simulated by the current variations of our device to a certain degree.

2.4. The Emotion-SLP Simulation

To evaluate the device learning capability, the one-time potentiation and depression of our device was characterized. As shown in Figure S11, Supporting Information, the light-induced EPSC changes of our device can be effectively erased by the electric pulses, which indicates that our device can be reset to its initial value to carry out another round of new learning tasks without
need to wait for a long time. For the further demonstration of the emotion-tunable learning capability of our device, we built a neural network based on SLP by using the extracted weight updating parameters from our neuromorphic device. We severe “emotion regulation” of SLP neural network learning capability by using the simulated neural network model to recognize modified national institute of MNIST handwritten digits after training. In Figure 5a, the network would consist of 784 input neurons (the resolution of the digits image is $28 \times 28$) and 10 output neurons (the label was set from 0 to 9). The input neurons and the output neurons are fully connected through $784 \times 10$ synapse (the values are regarded as synaptic weights). The input neuron would receive one signal converted from the gray level in one pixel of the training image ($28 \times 28$). Then the input vector ($V_i$) functions through the weight values in the synaptic network ($W_{i,j}$). The calculation result was converted and transmitted to the output vector with the utilization of the sigmoid activation function. The difference between the image’s label and the output value would determine the direction of the synaptic-weight updating process via the backpropagation algorithm. Therefore, in one batch, the SLP network was trained through the input of 60,000 image. Then the recognition rate (RR) of the trained SLP network for 10 numbers from “0” to “9” was tested with 10,000 testing images. The curves and their fitting curves for the

![Figure 4.](image1.png)

**Figure 4.** a) The EPSC of the device to 10-times 430 nm light spikes at various $V_{DS}$ ($-0.1$, $-1$, and $-3$ V). b) The change of the device EPSC under light spikes at various $V_{DS}$ ($-0.1$, $-1$, and $-3$ V). c) The schematic effect of the depression on the synaptic response to the external stimulus.

![Figure 5.](image2.png)

**Figure 5.** Pattern recognition based on simulated neural network. a) The SLP network with 784 input neurons and 10 output neurons. b) 200 levels of synaptic weights were obtained from the optical-pulse and electric-pulse trains of our synaptic transistor. c) The recognition rate of the network with the increasement of training epochs. d) The hotspot graphs of synaptic weights to recognize the number “7” under various emotions.
100-weight update under various emotions ($V_{DS}$, $-5$, $-15$ V) are shown in Figure 5b. Before the simulation, the 100-times potentiation and depression at various $V_{DS}$ were tested (Figure 5b). The corresponding curves were fitting using the following equations

$$G_{n+1} = G_n + \Delta G = G_n + \alpha \Delta \frac{G_{n} - G_{max}}{G_{min}}$$

and

$$G_{n+1} = G_n + \Delta G = G_n - \alpha \Delta \frac{G_{n} - G_{min}}{G_{max}}$$

where $G_{n+1}$ and $G_n$ stand for the conductance of the device when $(n+1)^{th}$ and $n^{th}$ pulses were applied, respectively. $G_{max}$ and $G_{min}$ represent the maximum and minimum conductance values. The parameters $\alpha$ and $\beta$ indicate the step size of the conductance and the nonlinearity, respectively. The SLP network was simulated based on the fitting parameters extracted from our device (Table S1, Supporting Information). The device under higher $V_{DS}$ exhibits more linear potentiation and depression parameters than that under lower $V_{DS}$. Therefore, the network under high $V_{DS}$ (positive emotion) exhibits a higher recognition rate ($\approx$75%) than that under low $V_{DS}$ (negative emotion, $\approx$65%).

The parameters were obtained from the fitting curves, which would be applied in the following MNIST-based recognition simulation process. The recognition rate of the network was increased with the input process of training images (Figure 5c). The network under high $V_{DS}$ (positive emotion) exhibits a higher recognition rate ($\approx$75%) than that under low $V_{DS}$ (negative emotion, $\approx$65%). The instability of the training process can be observed in a low-$V_{DS}$ (negative emotion) network. Figure 5d illustrates the mapping of the corresponding conductances ($W$), which can recognize the digit “7” before and after training. It can be observed that the conductance mapping based on high $V_{DS}$ (positive emotion) exhibits the more obvious pattern “7” than that based on low $V_{DS}$ (negative emotion).

3. Conclusion

In this work, we have successfully demonstrated a novel optoelectronic neuromorphic transistor based on the design of the tunable energy band structure of the 2D-MOFs-polymer/OSC layer. The 2D-MOFs-polymer blended layer was used as the photosensing component and the uniformly dispersed 2D-MOFs were used as the charge trapping centers. The generation, transportation, and trapping processes of the photogenerated charges on the 2D-MOFs-polymer/OSC heterojunction provided the transistors with a variety of synaptic behaviors. More interestingly, we have successfully simulated human emotion-tunable learning and memory behaviors via changing the value of source–drain voltage. The study can shed light on the application of 2D-MOFs in neuromorphic computing and is also helpful to the further development of neuromorphic computing devices.

4. Experimental Methods

**Materials Preparation:** The 2D Zn$_2$(ZnTCP) MOFs were synthesized according to the previous report.[33] Tetrakis(4-carboxyphenyl)porphrin (TCP) was purchased from TCI Inc. and used without any further purification. TCPP and zinc nitrate (Zn(NO$_3$)$_2$) were dissolved in a mixed solvent (N,N-Dimethylformamide/ethanol = 3:1) and heated at 80 °C for 24 h. Purple crystals can be observed after the sample was centrifuged at 4500 rpm for 10 min and washed with ethanol in three times. In order to obtain 2D Zn$_2$(ZnTCP) MOFs, 20 mg of the as-prepared MOF (Zn$_2$(ZnTCP)) was added to 4 mL chlorobenzene (CB) and was then sonicated using an ultrasonic bath machine filled with water. The temperature was maintained at 15–20 °C. After the sonication, the resulted samples were centrifuged at 1500 rpm for 10 min to remove the large particles.

**Device Fabrication:** OFETs were fabricated using a silicon wafer with 300 nm silicon dioxide as substrate. PMMA (20 mg) was added to the 1 ml 2D Zn$_2$(ZnTCP) MOFs solution and then was stirred for 6 h to obtain a uniform solution. The solution was spin-coated on the washed substrate at 2000 rpm for 60 s. The pentacene was then thermally evaporated onto the 2D Zn$_2$(ZnTCP) MOF-PMMA film at a rate of 0.1–0.3 Å s$^{-1}$. After that, 50 nm Au was thermally evaporated onto the pentacene film through a shadow mask as source–drain electrodes. The channel length and width were 30 $\mu$m and 1 mm, respectively.

**Device Characterization:** The surface morphology of pentacene and MOF-PMMA films was investigated by AFM (Dimension Icon, Bruker). The thickness of 2D Zn$_2$(ZnTCP) MOF-PMMA film was obtained from AFM. The device characteristics and synaptic behaviors measurement were carried out using a Keithley 4200-SCS instrument at room temperature. For the characterization of the synaptic phototransistors, a light source (white light, Thorlabs MCWH55-C4) was used. The optical intensities were calibrated with an optical power meter (Thorlabs PM100D).

**Supporting Information**

Supporting Information is available from the Wiley Online Library or from the author.

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**Conflict of Interest**

The authors declare no conflict of interest.

**Data Availability Statement**

The data that support the findings of this study are available from the corresponding author upon reasonable request.

**Keywords**

metal–organic frameworks, neuromorphic computing, neuromorphic transistors, optoelectronic devices, organic transistors
