Recent Progress in Photonic Synapses for Neuromorphic Systems

Junyao Zhang, Shilei Dai, Yiwei Zhao, Jianhua Zhang,* and Jia Huang*

Implementing synaptic functions with electronic devices is critical to achieve neuromorphic systems on the hardware platform, as synapses play important roles in brain computing and memory. Synapses modulated by light signals, which are also referred to as photonic synapses, can not only make effective use of the outstanding properties of light to provide devices with ultrahigh propagation speed, high bandwidth and low crosstalk but also provide a noncontact writing method, which can facilitate the evolution of optical wireless communication and operation. More importantly, real-time image processing can also be performed by photonic synapses which possess temporary memory. Thus far, tremendous efforts have been taken to design and fabricate photonic synapses. Herein, a summary of the development of different kinds of emerging materials utilized in photonic synaptic devices including memristors, field-effect transistors, and phase change memory is presented, followed by the innovative applications of photonic synapses for neuromorphic systems. Finally, some current challenges and future study directions are discussed.

1. Introduction

In the past several decades, due to excellent capacity in solving problems based on structured mathematical programs, the von Neumann architecture-based computers have rapidly expanded. However, with the increasingly obvious trend of information explosion, this conventional computing system is now facing unprecedented challenges limited to its intrinsic separated processing and memory units causing immense energy consumption.[1,2] Driven by the needs of the concept of Internet of Things and artificial intelligence, the demand for neuromorphic computing that attempts to emulate the behaviors of human brains has extremely increased. In human brains, massive information can be processed at an extraordinarily fast speed with low energy consumption of only 1–100 fJ per synaptic element, taking the advantage of parallel processing of the neural network.[3,4] More strikingly, human brains are extraordinarily efficient in handling cognitive operations, such as image recognition and processing characterized by probabilistic and unstructured problems.[5,6] Thus, inspired by human brains, neuromorphic computing simulating the neural system has been paid considerable attention until today. Nevertheless, artificial neural networks (ANNs) are still mainly constructed based on software, which still depend on the traditional computing architecture, so that software-based ANNs will also consume enormous power.[7–9] Hence, it is indispensable to imitate the neural system on physical level and first of all, synapses, the basic component in real brains, should be developed for the construction of neuromorphic computing.[10–13]

A mass of work has been done to implement artificial synapses for neuromorphic computing. Memristors,[14–16] field-effect transistors (FETs),[17,18] and phase change memory,[19–21] have been exploited to simulate synaptic behaviors. Memristors consisting of metal/insulator/metal structures have attracted immense interests due to their distinct properties, such as perfect scalability, large connectivity, low energy consumption, and excellent compatibility with complementary metal–oxide–semiconductor (CMOS) process.[22–26] In terms of memristors, signal can only be transmitted through one signal path. In contrast, FETs can not only propagate the signal but also achieve the learning process simultaneously.[27–29] Moreover, FET-based synaptic devices are more promising to achieve linear plasticity than memristors.[30,31] Certainly, FETs have more complicated structures, compared with memristors.[32] Despite rapid advances, conventional synapses are based on the electronic technology, on which the speed of neuromorphic computing is limited on account of bandwidth connection density trade-off. In contrast, synapses triggered by light pulses are more suitable for ultrahigh computing speed because they have unique advantages including high bandwidth, high interference immunity, and low power computation.[33–35] In addition, photonic synaptic devices provide a noncontact writing
method, which can facilitate the evolution of optical wireless communication. More importantly, synaptic devices modulated by photonic signals are in favor of imitating retinal neurons in real eyes, resulting in bridging the gap between brain computing and visual systems.[35–37] It is necessary to develop photonic synapses without utilizing external polarity-changing electric fields as a significant step for the next step in photogenetic-inspired circuits that embody flexibility and fidelity of biological synapses. The evolution of photonic synapses is essential to the progress of neuromorphic computing and ANNs.

In a nervous network, the synaptic weight defined as the connection strength between presynaptic neuron and postsynaptic neuron can be modulated by action potentials.[38] Synaptic plasticity, the change in synaptic weight, is considered as the foundation of memory and learning process in brains, which makes it competent to implement complicated functions of brains.[39] In general, synaptic plasticity can be divided into two main parts—short-term plasticity (STP) and long-term plasticity (LTP).[40,41] STP stored in the hippocampus is a temporal change in synaptic connection and will rapidly decay to its original state after removing the external spike, which is indispensable for information processing. When a potential pulse is applied at presynaptic membrane, neurotransmitters are released, leading to the change in the postsynaptic current. Excitatory postsynaptic current (EPSC) occurs when excitatory neurotransmitters are released, and results in the strengthening of the synaptic weight. However, inhibitory postsynaptic current (IPSC) occurs when inhibitory neurotransmitters are released, and results in the weakening of synaptic weight. Paired-pulse facilitation (PPF) is one of the typical STP behaviors, which describes the fact that an enhanced postsynaptic response is obtained by the consecutively applied pair of presynaptic spikes. In contrast, LTP is long-standing transform in synaptic connection and it is critical for memory and learning. With repetitive rehearsal, STP can be converted to LTP.[42,43] In addition to the classification of synaptic plasticity, it is also worth investigating the factors that influence synaptic plasticity. Spike timing-dependent plasticity (STDP) and spike rate-dependent plasticity (SRDP) are two main factors to change the synaptic weight, and both of them are basic learning rules in neural networks.[44,45]

Except for simulating fundamental synaptic functions, great effort has been paid to construct neuromorphic systems. These neuromorphic systems emulating simple biological senses have motivated both life and industry in various aspects. Light cognition is one of the most significant sensory functions for neuromorphic systems, which is essential for further development in image recognition, detection, optical wireless communication, and operation.[37,46,47] The development of photonic synapses for neuromorphic systems is expected to facilitate the evolution of mimicking neural networks and functions of the retina, which allows to perform the real-time image processing on the front end to improve the efficiency of image processing and the quality. The development of neuromorphic systems would be an indispensable role in bioinspired electronics. In this Review, we summarize recent progress in photonic synapses based on various candidate materials, including metal oxides, perovskites, low-dimensional materials, organic materials, and phase change materials. Moreover, the innovative applications of photonic synapses in neuromorphic systems are also discussed.

2. Emerging Materials-Based Synaptic Devices

2.1. Metal Oxides

Metal oxides, which are considered as one of the most attractive materials used in artificial synapses, are extensively studied for a long time.[48–51] Currently, researchers are focused on the materials and the device structures of three main kinds of metal oxides, including oxide semiconductors, binary oxides, and oxide heterojunctions.

2.1.1. Oxide Semiconductors

Amorphous indium–gallium–zinc oxide (α-IGZO) has outstanding advantages of transparency, low energy consumption, excellent electrical properties, and its metal-oxide network structure can be formed at room temperature.[26,52–54] IGZO-based synaptic devices are expected to emulate basic synaptic behaviors.[55–59] Critically, IGZO shows strong persistent photoconduction (PPC) behavior with continuous high photocurrent after taking away the light illumination, which is beneficial to realize photonic synaptic devices.[60–62]

Lee et al. reported an IGZO-based synaptic device to emulate major synaptic functions, such as short-term and long-term memory, neural facilitation and symmetric STDP.[63] The structure of the IGZO-based device and its optical microscope image are shown in Figure 1a. Light stimuli is worked as the action potential. The synaptic device exhibits STM behavior by adding light stimuli with a lower frequency of 0.2 Hz, whereas it demonstrates LTM behavior through a higher frequency of 1 Hz (Figure 1b,c). Figure 1d shows the simulation of neural facilitation behavior of the device. Originally, the synaptic weight has increased 2.7 nA after applying one light pulse, but at the...
30th light pulse, it has increased 9.8 nA, which is nearly 3.5 times higher than that caused by the first stimuli. The IGZO-based synaptic device also shows a symmetric STDP behavior in the emulation of neuromorphic activity (Figure 1e). Furthermore, they clarified the basic mechanism of photogenerated carrier generation or recovery behaviors by systematically analyzing the dynamics of photoinduced carriers of different kinds of amorphous oxide semiconductors. As a result, they found that the photoinduced carrier generation or relaxation is influenced by the activation energy for neutralization of ionized oxygen vacancies. With the increase in the indium in IGZO, the degree of PPC behavior increases. Additionally, during the illumination or decaying period, adding gate bias can control the photoresponse characteristics of IGZO.

In terms of three-terminal synaptic transistors, Yang et al. reported an IGZO-based electric-double-layer photoelectric transistor operated in low voltage for mimicking major synaptic behaviors.[64] They also utilized gate voltage modulation for the transition from depression mode to potentiation mode. Cheng et al. demonstrated a proton conductor gated synaptic transistor using 65 nm-thick nanogranular SiO2 as the dielectric layer and IGZO as the channel material.[65] UV light is utilized to be another control terminal to modulate the plasticity of the synaptic transistor. EPSC behaviors under illumination by electric pulses are also investigated. With a fixed illumination, the light frequency has greater influence on synaptic plasticity than the other light parameters including intensity, duration time, and wavelength. The STM to LTM transition can also be realized through adding pulse number and frequency. The EPSC gain is obtained from 126% to 148%, when the frequency is from 0.31 to 5 Hz. Moreover, by modulating the metallic composition ratio in IGZO, some important synaptic behaviors can be accordingly mediated. Recently, Wu et al. studied the effect on synaptic behaviors by regulating the element composition ratio of IGZO.[66] They found that compared with the synaptic devices of InH and GaH, the InH and GaL devices demonstrate stronger photocurrent generation, which can be explained to higher carrier concentration caused by oxygen vacancies,[67] because In element facilitates the formation of oxygen vacancies, whereas Ga element restrains its formation.

Except for IGZO, indium zinc oxide (In-Zn-O) has also been widely used as an active layer to fabricate synaptic devices due to its notable property, such as high transparency, satisfying carrier mobility, ease of fabrication, and unique PPC.[60,68] Wang et al. demonstrated a synapse transistor based on IZO with ion–gel laterally utilized as the dielectric layer.[69] In addition to simulation of EPSC, PPF, photonic potentiation, and electric habituation, four forms of STDP learning rules are also obtained through photoelectric hybrid modulation. Furthermore, the transformation of energy band diagram of the device is qualitatively discussed in combination with illumination and external electric field.

### 2.1.2. Binary Oxides

As one kind of conventional materials applied for synaptic devices, binary oxides are extensively used in memristors. Among various kinds of binary oxides, ZnO, HfO2, and AlOx have been paid growing attention in photonic synaptic devices due to their...
peculiar performance, such as their ease of fabrication, low cost, and great possibility in achieving a simple device structure.\cite{70-73}

Currently, ZnO with its unique visible-blind ultraviolet-sensitive detection has attracted great interests in designing photonic synaptic devices.\cite{74-77} Kumar et al. first reported highly transparent synaptic devices based on all metal oxide, with structure fluorine-doped tin oxide (FTO)/ZnO/In$_2$O$_3$, as shown in Figure 2a.\cite{78} In addition to emulating major synaptic behaviors including STP, PPF, and LTP, they focused on realizing the impressive photonic potentiation and electrical habituation behaviors (Figure 2b). The mechanism of photonic potentiation and electrical habituation is explained to the type-II band alignment between ZnO and In$_2$O$_3$ (Figure 2c). This finding paves the way for the application of the band alignment between ZnO and In$_2$O$_3$ in highly transparent photonic synaptic devices.

Except for ZnO thin film, ZnO nanorods can also be utilized in fabricating high-performance photonic synaptic devices. Zhou et al. for the first time used ZnO nanorods with high surface-state density in optoelectronic memristors.\cite{79} Compared with ZnO thin film, ZnO nanorods exhibit more stable resistive-switching behavior because of the distribution of oxygen vacancies along the surface of nanorods arranged vertically. Comparison of the $I-V$ sweeps of ZnO thin film and nanorods can also be observed. ZnO nanorods not only exhibit much larger switching window but also show much lower initial conductance than ZnO thin film. The device based on ZnO nanorods exhibits better endurance as compared with the thin-film device, and only the low resistance state (LRS) of the ZnO nanorods device exhibits volatile, which plays a significant role in mimicking STP. Due to the large specific surface area, the ZnO nanorods device displays much larger photo-to-dark current ratio ($10^4$) than that of the thin-film device (70). In addition, the device based on ZnO nanorods can exhibit optical shielding effect and electrical deshielding, which can be utilized in the information processing field.

2.1.3. Oxide Heterojunctions

Currently, heterojunctions have been studied on oxide synaptic devices to modulate ion diffusion or electronic tunneling.\cite{49,80} As shown in Figure 3a, a simple structure with indium-tin oxide/Nb-doped SrTiO$_3$ (ITO/Nb:SrTiO$_3$) heterojunction was built by Gao et al.\cite{81} Using pulsed light stimuli, photoresponsive

Figure 2. a) Schematic diagram of the In$_2$O$_3$/ZnO/FTO synaptic device and its original photograph. b) The current change on the synaptic devices after applying a train of light pulses (intensity: 0.4 mW cm$^{-2}$, duration and separation: 1 s) and negative electrical pulses ($-1$ V, duration and separation: 20 ms). c) Schematic illustrations of the working mechanism of the photonic synapse. Reproduced with permission.\cite{78} Copyright 2018, American Chemical Society.
characteristics of the synaptic device based on ITO/Nb:SrTiO₃ heterojunction have been deeply investigated. It is clear to see that with the increase in number or frequency of pulsed light stimuli, the synaptic weight is enhanced gradually, leading to the transition from STM to LTM (Figure 3b,c). Meanwhile, the simple device demonstrates an attractive behavior called “learning-experience” mimicking for the learning process of human brains. Referring to Figure 3d, applying the first training sequence, the synaptic weight improves to a high level, and then decays to a mediate level after removing the training. Next, only 7 pulses are enough to reach the same synaptic weight that 35 pulses are needed in the first training sequence. More importantly, with the same interval, it is obvious that the second decay of synaptic weight is much less than the first one, which much resembles the learning/forgetting/relearning experience of human. Moreover, by setting a sub-1 V external voltage, photoresponsive efficiency of this device can be regulated flexibly (Figure 3e). Thus, with proper external voltages, the same photoresponsive result can be achieved even for different kinds of illumination conditions, which is quite similar to the role of iris in human eyes (Figure 3f).

Two-terminal memristors with ITO/ZnO₁₋ₓ/AlOₓ/Pt architecture were explored by Hu et al. They successfully emulated various synaptic behaviors by utilizing both electric and photonic stimuli based on the volatile resistive-switching behavior of ZnO₁₋ₓ/AlOₓ heterojunction and the PPC behavior. To study the role played in AlOₓ, control experiments of ITO/ZnO/Pt structure are used to investigate the mechanism of the PPC behavior. The ITO/ZnO/Pt devices exhibit weak PPC effect indicating the AlOₓ layer serves as trap sites for hole, leading to the PPC behavior.

2.2. Perovskites

Among different kinds of perovskites, halide perovskites have attracted considerable attention due to their high carrier mobility,[81–85] tunable bandgap,[86,87] and solution process availability.[88–92] Due to these intensive advantages, halide perovskites are supposed to be promising materials for photonic synapses. MAPbI₃, one kind of the most outstanding organic–inorganic hybrid halide perovskites, has been widely used because of its low energy barrier to migrate iodine vacancy (VI⁻/VI⁻/C₂)[93,94] and large light absorption.[95] Zhu et al. reported a halide perovskite-based memristor with the structure of Ag/MAPbI₃/Ag inspired by optogenetics, which is prefabricated on the SiO₂/Si substrate (Figure 4a).[96] Light illumination is worked as another gate to modulate VI⁻/VI⁻ dynamics of generation and annihilation, quite similar to the Ca²⁺ dynamics controlled by light in biological synapses. As shown in Figure 4b, light illumination can inhibit the VI⁻/VI⁻ generation induced by electric field and accelerate the VI⁻/VI⁻ annihilation spontaneously in MAPbI₃ due to the increasing formation energy of VI⁻/VI⁻. In addition, by controlling the number and time of illumination, the decay rate of the conductance is tunable (Figure 4c), and the device also shows LTP in the dark, while presents LTD under light illumination stimulated by electrical pulses.

Although MAPbI₃-based photonic synapses demonstrate excellent performance, lead element in MAPbI₃ is highly toxic and it can easily pollute the environment, so that it is an enormous demand to design green devices based on lead-free perovskites. Recently, Qian et al. first fabricated a flexible photonic synaptic device based on (PEA₂)₂SnI₄, which is one kind air-stable lead-free 2D perovskite.[97] The perovskite film is developed...
through one-step solution approach and rGO/(PEDOT:PSS) is worked as the flexible electrode. They successfully emulated some typical synaptic functions, such as EPSC and STP, and these synaptic behaviors can be attributed to the Sn vacancies in (PEA)2SnI4. This work opens the application of lead-free 2D perovskites in synaptic devices. Coincidentally, (PEA)2SnI4 was also exploited by Sun et al. to design a synaptic photoconductor for regulating synaptic plasticity.[98]

Recently, inorganic halide perovskites-based photonic synaptic devices have been simultaneously reported, due to its relatively high stability.[83] Wang et al. demonstrated a CsPbBr3 QDs-based synaptic transistor through a simple solution process, which is likely to be applied in mass production.[99] As shown in Figure 4d, CsPbBr3 QDs and the organic semiconductor (PQT-12) are blended effectively, both to enhance photoresponsivity caused by the increased charge-carriers separation efficiency, and to emulate significant synaptic behaviors by the delayed decay of the photocurrent. The effects of light illumination parameters (including light pulse width, intensity, and number) on channel conductance change (ΔG) as a function of (a) presynaptic light pulse numbers, (b) presynaptic light pulse intensity, and (c) presynaptic light pulse widths. (d) EPSC of the CsPbBr3 QDs/PQT-12 synaptic transistor by 30 presynaptic light pulses with a constant light intensity of 0.1 mW cm\(^{-2}\) and light width of 0.5 s.

Figure 4. a) Schematic of the MAPbI\(_3\)-based memristor measured under light illumination. b) Illustrations indicating that light illumination can inhibit \(V_1/V'_1\) formation under bias (left) and accelerate \(V_1/V'_1\) annihilation (right). c) Conductance retention curve of the memristor in LTM regime, during which light illumination (1.29 \(\mu\)W cm\(^{-2}\)) was alternately applied and removed. Reproduced with permission.[96] Copyright 2018, American Chemical Society. d) Schematic image illustrating the neural signal transmission process through biological synapses in neuron and simplified devices structure of the CsPbBr3 QDs/PQT-12 synaptic transistor. Channel conductance change (ΔG) as a function of (a) presynaptic light pulse numbers, (b) presynaptic light pulse intensity, and (c) presynaptic light pulse widths. (h) EPSC of the CsPbBr3 QDs/PQT-12 synaptic transistor by 30 presynaptic light pulses with a constant light intensity of 0.1 mW cm\(^{-2}\) and light width of 0.5 s. i) EPSC amplitude ratio (A\(_{30}\)/A\(_1\)) plotted as a function of the presynaptic light pulse frequency. Reproduced with permission.[99] Copyright 2019, Wiley-VCH.
2.3. Low-Dimensional Materials

2.3.1. 0D Materials

In comparison with other-type materials, 0D materials have unique optical and electric properties so that they have been extensively explored for photonic synapses. Recently, Maier et al. interestingly selected InAs QDs to modulate the conductance of this memristor based on GaAs/AlGaAs heterostructure.\[101\] The synaptic weight can be controlled by both low-consumption optical pulses and electric pulses. Lv et al. exploited carbon dots combined with silk protein to build an optical synaptic transistor, which can be regulated by light not only in volatile memory mode but also in nonvolatile memory mode first.\[102\] Carbon dots/silk protein is appointed as the charge trapping medium.

In addition, Si nanocrystals heavily doped by boron were revealed to fabricate wide-broad-spectral-region photonic synaptic devices by Tan et al.,\[103\] which can be stimulated by light with the wavelength from ultraviolet to near-infrared effectively. In particular, Si nanocrystals used in this work are colloidal form, so it is proper to use the simple solution process to fabricate devices to large-scale production, which is suitable for flexible synaptic devices. More importantly, major synaptic functions have been mimicked and the mechanism of synaptic plasticity is assigned to the dynamic trapping and detrapping of photogenerated carriers at dangling bonds of the surface of Si nanocrystals. This work is of great significance to accelerate the large-scale application of Si element in neuromorphic systems.

Boron-doped Si nanocrystals were also utilized by Yin et al. to design optically-stimulated transistors to emulate demanded essential synaptic functionalities, such as EPSC (IPSC), PPF, STDP, the simulation of aversion learning and the logic functions.\[104\] Figure 5a demonstrates the structure of the synaptic phototransistor and boron-doped Si nanocrystals film, which is served as the channel of the synaptic phototransistor, is about 647 nm thickness. Both electric and optical stimulations can modulate synaptic functionalities properly. When a negative electric stimulus of $-50 \text{ mV}$ is applied, the IPSC is triggered. Although for a positive electric stimulus of $+50 \text{ mV}$, the change in the current is reversed, resulting in the EPSC (Figure 5b). From Figure 5c, it is seen that IPSC can also be induced by 1342 nm optical stimulus. Except for EPSC (IPSC), PPF, and STDP are also mimicked by the modulation of electric and optical stimulations. Interestingly, with the synergy of electric and optical stimulations, the simulation of aversion learning is well demonstrated. Aversion learning signify, after the treatment for the patients who are thirsty for alcohol, the patients will show the aversion to alcohol, which is a classical experiment in mimicking associative learning. In this article, the researchers considered the excitatory electric stimulus as the alcohol drinking and considered the inhibitory optical stimulus as the emetine taking. After the treatment, the current of the synaptic device decreases to a much lower level than the initial current before the treatment (Figure 5d). Even if a succession in electric stimulations are applied at the end of treatment, the current of the synaptic device still remain at the aversion state. Thus, the aversion learning is readily emulated through the synaptic phototransistor. More importantly, the Si nanocrystals-based synaptic phototransistor enables to implement the logic functions including “AND” gate and “OR” gate (Figure 5e). During the operation of “AND” gate, the optical stimulus is used simultaneously with a positive electric stimulus of $+50 \text{ mV}$, whereas during the operation of “OR” gate, the optical stimulus is used simultaneously with a

![Figure 5](https://www.advancedsciencenews.com/)

**Figure 5.** a) Schematic illustrations of a synaptic Si nanocrystals phototransistor. b) IPSC (EPSC) induced by a $-50 \text{ mV}$ ($+50 \text{ mV}$) electrical stimulus with the spike duration of 200 ms. c) IPSC induced by a 1342 nm laser stimulus with the spike duration of 200 ms and the power density of 30 mW cm$^{-2}$. d) Implementation of taste aversion learning with a synaptic Si-NC phototransistor. e) Implementation of logic functions including “AND” gate and “OR” gate. Reproduced with permission.\[104\] Copyright 2019, Elsevier B.V.
negative electric stimulus of −50 mV. Clearly, the logic functions including “AND” gate and “OR” gate can be successfully realized with the modulation of electric and optical stimulations.

2.3.2. 1D Materials

Carbon nanotubes have been considered as one of the emerging 1D materials that present high charge carrier mobility, excellent physical and chemical stability, and proper operating temperature.\textsuperscript{105–107} In addition to combining with semiconductors,\textsuperscript{108} carbon nanotubes assisted in lightly n-doped Si transistor can also realize the emulation of synaptic behaviors. Shao et al. showed a simple synaptic transistor based on single-walled carbon nanotubes (SWCNTs) using HfO\textsubscript{2} as the gate dielectric and lightly n-doped Si as the gate electrode (Figure 6a).\textsuperscript{109}

An interesting phenomenon can be observed as shown in Figure 6b that once the light is turned on, the current of the device decreases rapidly, whereas the current increases and maintains stable when the light is removed. This negative photoresponse characteristic can be attributed to photogating effect of n-doped Si gate. In addition, EPSC values increase as the light pulse number increases and the frequency decreases, and EPSC gain weakens rapidly when the frequency increases, so the synaptic devices can act as dynamic low-pass filters for information transmission (Figure 6c,d).

Furthermore, Li et al. used InAs nanowires with high carrier mobility in a phototransistor to emulate synaptic behaviors.\textsuperscript{110} The device shows negative photoresponse and successfully mimics some typical synaptic functions including STP, LTP, the transition from STP to LTP. The mechanism of the synaptic functions is interpreted into trapping/release of photogenerated electrons in the native indium oxide layer, which acts as a photogating layer.

2.3.3. 2D Materials

Molybdenum disulfide (MoS\textsubscript{2}), graphene and black phosphorus (BP) have been highlighted as the potential 2D materials candidates for complicated neuromorphic systems.\textsuperscript{111–116} Bulk MoS\textsubscript{2} exists an indefinite bandgap, and hence it is not really ideal for optoelectronic devices. To solve this puzzle, He et al. explored monolayer MoS\textsubscript{2} to develop an ultrathin memristor on p-Si substrate with a rectification ratio of $4 \times 10^{13}$\textsuperscript{117} thanks to its a direct bandgap of 1.8 eV, especially optical and electric characteristics.\textsuperscript{118,119} The schematic of the photonic memristor is shown in Figure 6e. The behaviors of photonic potentiation and electric habitation can be achieved, which are inferred to the PPC effect caused by light pulses and the volatile resistive switching evoked by electric pulses (Figure 6f). More importantly, the degree of the photonic potentiation and electric habitation can be accurately controlled by modulating the photonic and electric parameters. Furthermore, both EPSC and IPSC can be achieved by applying light pulses and electric pulses respectively. Then, PPF is successfully triggered by using either light pulses or negative voltage pulses and a larger stimuli interval leads to a weaker EPSC/IPSC, resulting in a smaller PPF index, which resembles the behaviors of biological synapses (Figure 6g,h). Strikingly, the authors also systematically analyzed the mechanisms of the PPC effect and the resistive switching behavior.

To further regulate and control the important synaptic functions for neuromorphic systems, the heterojunction structure with 2D inorganic semiconductor (MoS\textsubscript{2}) and 2D organic semiconductor (PTCDA) was tried in a photonic transistor by Wang et al.\textsuperscript{120} They designed two similar structures used as electric modulation and optical modulation respectively (Figure 7a,b).

For electric modulation, a novel four-terminal architecture is

![Figure 6. a) Device structure of a SWCNT-based transistor. b) Photoresponse characteristics of the SWCNT-based transistor under 520 nm pulsed light illumination with a frequency of 0.5 Hz. c) EPSCs recorded for presynaptic spike trains with different frequencies. d) The amplitude rise defined as $[\Delta A_{30} - A_1]/A_1$ corresponds to the frequency. The inset in panel (d) represents the schematic of lowpass filtering of a synapse. Reproduced with permission.\textsuperscript{109} Copyright 2019, American Chemical Society. e) Schematic of a MoS\textsubscript{2}-based photonic synapse. f) Characteristic of photonic potentiation and electric habitation. g) Photonic EPSC-induced PPF by utilizing light pulses, and h) electric IPSC-induced PPF triggered by negative voltage pulses. Reproduced with permission.\textsuperscript{117} Copyright 2018, Wiley-VCH.](image-url)
generated with the composition of \( \text{Al}_2\text{O}_3 \) as the gate dielectric and top Au electrode as the control gate. For optical modulation, a three-terminal configuration is explored without the \( \text{Al}_2\text{O}_3 \) gate dielectric and the control gate. It is remarkable that with relatively different voltage pulse, the electrons are able to transfer between MoS\(_2\) and PTCDA in both orientations due to the elaborate heterojunction structure. As a result, after adding 50 relatively negative pulses, the maximum facilitation can obtain 500%. In contrast, through 50 relatively positive pulses, the minimum facilitation of 3% is achieved (Figure 7c). Furthermore, the outstanding optical modulation demonstrates nearly 60 synaptic weight changes, which indicates the device exhibits distinguished ability to modulate the synaptic plasticity through both electric stimuli and optical stimuli.

Graphene, another star 2D material, also draws great attention for photonic synapses due to its superior properties of flexible, biosafe, good compatibility, and theoretically low cost.\(^{119,121}\)

To date, graphene has been researched in photonic synapses as the floating gate\(^{122}\) and the channel material.\(^{127}\) Sun et al. published an article about photonic synapses utilizing alkylated graphene as the charge trapping layer.\(^{122}\) Three terminal phototransistors with graphene and SWCNTs hybrid were reported by Qin et al.\(^{17}\) In addition to STP induced by the heterostructure between SWCNTs and graphene\(^{123,124}\) and LTP induced by the trap-rich interface between the substrate and mixed film, the device demonstrates superior sophisticated optical spike processing consisting of AND, OR, and NOR logic operations by applying different gate voltage levels.

Furthermore, as an emerging 2D material, BP has a peculiar advantage in broadband absorption, strong light-matter coupling, and easily tunable optoelectronic properties.\(^{17,125–127}\) Utilizing the inherent optoelectronic properties using oxidation-related defects in BP, Ahmed et al. exploited a two-terminal BP-based synaptic device to achieve all optical pathway for simulating both
excitatory and inhibitory action potentials without applying external polarity changing electric field.[128] Few-layer BP sheets are diverted onto the substrate, followed by patterning electrodes. As shown in Figure 7d, the substrates are not limited to silicon wafers; the photonic synaptic device can also be fabricated on a flexible platform, such as polyethylene naphthalate (PEN). Even at a flexure with diameter of 7 mm, the flexible synaptic device still demonstrates stable dynamic synaptic plasticity (Figure 7e).

Interestingly, under the illumination of 280 nm wavelength, the BP-based device shows positive photocurrent, whereas under 365 nm illumination, it shows negative photocurrent due to the oxidation-related defects on the surface of BP. Due to the unique optoelectronic properties of BP, both EPSC and IPSC are triggered by applying different light illumination of different wavelengths (Figure 7f,g). Furthermore, as the light pulse width increases, the EPSC response enhances, similar to the learning process in human brains. More significantly, spatiotemporally correlated dynamic logic is successfully achieved through the pristine-like BP device (Figure 7h,i). Associated with the difference of the PSC response triggered by presynapse 1 (660 nm) and presynapse 2 (280 nm) respectively, the ΔPSC as a function of ΔV_{pre2-pre1} reveals asymmetric characteristics. STDP behavior is also mimicked by the two separate BP-based devices sharing a collective electrode resulting in a symmetric ΔPSC response change (Figure 7j,k). This work paves a way for the emulation of sophisticated neural functionalities only through the optical modulation.

2.4. Organic Materials

Organic materials are considered as the most suitable candidates for designing artificial synaptic devices, due to their distinctive properties of low cost, mechanical flexibility, excellent ductility, compatibility with CMOS technology, and low energy consumption.[3,129-133] Organic materials based synaptic devices can be fabricated through simple solution processes, including printing and spin coating, which are extremely suitable for large-scale production.[132-134] Furthermore, organic materials based artificial networks will be the solid foundation for the progress of future neuromorphic systems and may open a new era for next-generation soft electronics.

Dai et al. demonstrated a simple processing strategy for building an organic phototransistor to emulate synaptic behaviors.[135] They carefully selected polyacrylonitrile (PAN) film as the gate dielectric and 2,7-dioctyl[1]benzothieno[3,2-b][1]benzothiophene (C8-BTBT) as the organic semiconductor. Major synaptic behaviors, such as EPSC, PPF, learning behaviors can be successfully realized attributed to the charge trapping effect near or between the C8-BTBT/PAN interface, rather than utilizing the unique photosensitive property of the organic semiconductor. Then, learning behavior and forgetting behavior are investigated through a T-shaped array including 20 transistors without the silicon wafer substrate. As a result, during the learning process, most synaptic weights increase to a superior level after twenty light pulses, and the memory levels rapidly decrease at early period, followed by a bit slow recession. Hence, the mechanism of mimicking the synaptic functions is further confirmed to the charge trapping effect near or between the C8-BTBT/PAN interface.

Normally, the nonvolatile memory is unstable caused by the retention time of the LTP restricted to only several minutes.[13] To settle this matter, an organic transistor modulated by both ferroelectric and electrochemical was built by Wang et al. to control the synaptic plasticity.[136] As shown in Figure 8a, the gate dielectric contains poly[(1-vinylpyrroliodone)-co-(2-ethylidimethylammonioethyl methacrylate ethyl sulfate)] [P(VP-EDMAEMAES)] worked as the electrochemical layer and poly(vinylidenefluoride-co-trifluoroethylene) [P(VDF-TrFE)] served as the ferroelectric layer. In addition, poly(isoindigo-co-bithiophene) [P(IIID-BT)], a p-type copolymer, is used as the channel material, and the photosensitive constituents on the top of the device with the series connection consist of a phthalocyanine load resistor and an organic heterojunction. By adjusting the gate voltage, both STP and ferroelectric LTP are exhibited (Figure 8b). When ten larger V_{C} pulses (−30 V) are added, the device reveals ferroelectric LTP. While lower V_{C} pulses (−4 V) are applied, STP is triggered. 30 V V_{C} pulse is used to reset the synaptic transistor to off-state between the aforementioned two operations (Figure 8c).

In addition, adding different voltages above the ferroelectric LTP threshold voltage approximately from −18 to 40 V, nonvolatile currents are dramatically increased nearly 10^4 times. Intriguingly, electrochemical LTP is also observed through low electric pulses but high frequency (−2 V, 32 Hz), resulting in a postsynaptic current retaining 10% higher than the residual current after 100 s. Moreover, they fabricated an ultra-flexible light stimulated organic neuromorphic conformal array via a water-aided peeling-off process to identify colors and then to emulate functions of the retina. Within successive NIR light stimulation, the signal current enhances ranging from 1 to 1.7 nA, followed by 0.1 nA after 600 s and the signal is almost impossible to be distinguished after 1800 s (Figure 8d-f). While with the exposure under green light, a larger signal current is evoked ranging from 0.4 to 0.67 μA, followed by nearly 78.5% amount of the beginning signal current after 600 s and the signal maintains at the least 65% after 1800 s (Figure 8g-i). This conformal device not only emulates most important synaptic behaviors but also mimics the simple functions of retinal, which promotes the further improvement in fully biologically compatible artificial devices.

2.5. Phase Change Materials

Phase change materials can be divided into two phases, namely amorphous phase and crystalline phase. Using the large characteristic difference between amorphous and polycrystalline, the devices based on phase change materials are supposed to show unique switching behaviors when enough heat is generated for phase change.[20,137] In addition, phase change materials have prominent scalability, reliability, multiple programmable resistances and low device-to-device differences, so they are ideal candidates for application in integrated electronics for neuromorphic systems. More importantly, the synaptic devices based on phase change materials have remarkable advantage of nonvolatile storages.

Cheng et al. fabricated a completely integrated photonic synaptic device based on chalcogenide Ge_{2}Sb_{2}Te_{5} (GST),[138] one kind of well-studied phase change material.[21,139]
The synapse is prepared on the Si$_3$N$_4$/SiO$_2$ platform, on which discrete GST and ITO are sputter-deposited. STDP rules are successfully mimicked with prearranged presynaptic and postsynaptic stimulations. Moreover, different synaptic weight levels are achieved by a predetermined pulse sequence and large-scale neuromorphic systems are likely to come true due to the less switching energy consumption. Constructing the synaptic devices based on phase change materials is a crucial step toward photonic neuromorphic computing.

3. Innovative Applications of Photonic Synapses for Neuromorphic Systems

Recently, in addition to simulating major synaptic functions and modulating synaptic functions, synaptic devices are also explored to build neuromorphic sensory systems. Light cognition is one of the most significant sensory functions for artificial visualization systems. Scientists have skillfully utilized light-sensitive elements combining with synaptic devices to design various photonic synapses for neuromorphic systems. Hence, researches on neuromorphic visualization systems have been explosively growing. Thus far, there are two main innovative applications of photonic synapses for neuromorphic systems: 1) mimicking the functions of human retina, including recognizing letters and numbers from the Modified National Institute of Standards and Technology (MNIST) database, identifying different colors and color-mixed patterns, and realizing the function of image preprocessing. 2) combining with motion systems to imitate both sensory and motor functions of human.

A novel photonic transistor based on IGZO-alkylated graphene oxide has been demonstrated by Sun et al. for enhancing the...
The structure of photonic synapses is shown in Figure 9a. With the application of 200 mixed pulses including light stimuli ($\lambda = 405$ nm, $100 \mu$W, $t_d = 50$ ms) and electric stimuli ($V_{WC} = -1$ V, $t_d = 50$ ms), the EPSC achieved is much bigger than that triggered only by electric pulses, gaining larger channel conductance ($\Delta G$) induced by mixed pulses at the potentiation process as well (Figure 9b,c). Importantly, with the assistance of light, nonlinearity property has not been degraded (Figure 9d). In the depression process of both mixed pulses and electric pulses, the recession trend of $\Delta G$ is almost similar. Then, the MNIST handwritten digit numbers are used, which are widely utilized to detect the pattern recognition. The neuromorphic visualization system is composed by 10 output neurons representing the number from “0” to “9” and 785 input neurons on behalf of 28 × 28 pixels of MNIST images (Figure 9e). By adding mixed pulses, the studied number “6” is well learned even at only 20 weight states, whereas by applying only electric pulses, the number “6” cannot be recognized definitely (Figure 9f). As a result, the improved recognition rate is attributed to the increasing $\Delta G$ and nearly unchanged nonlinearity property. The proposed photoelectric mixed pulses technology is expected to have potential application in future neuromorphic computing and systems.

To improve the efficiency and quality of the image processing, Zhou et al. designed a memristor with a simple structure of Pd/MoOx/ITO. The memristor array is served as preprocessing stage, and the artificial network is served as image recognition stage. The letters P, U, C are chosen from the database for the image recognition. At the preprocessing stage, the image information of the letters is processed by the memristor array. Clearly, the body signals of the letters are sharpened, whereas the noise signals are weakened. Then, two kinds of image information of both with and without the preprocessing stage are adopted to perform image recognition. The efficiency and recognition of the visual system with the preprocessing stage have great improvement compared with those of the visual system without the preprocessing stage. The recognition rate without the preprocessing stage reaches 0.980 after 2000 trainings, whereas with the preprocessing stage, the recognition rate reaches 0.986 through only 1000 trainings. As a result, the photonic synaptic device shows great potential in neuromorphic visual systems.

Except for the recognition of one-color number patterns, Seo et al. tried to recognize the different colors and color-mixed patterns (Figure 10a). They designed an optic-neural synaptic (ONS) device consisting of a synaptic element and an optic-sensing element, both based on the h-BN/WSe2 heterostructure. The optic-sensing element not only accepts light stimuli signal but also provides proper series resistance. MNIST images have also been utilized to operate training and testing. With the assistance of optic-sensing element, the recognition rate is extremely increased over 90% after 50 trainings, which is much higher than the recognition rate of conventional neural network without...
optic-sensing element obtained below 40%. Furthermore, the synaptic weight caused by blue stimuli becomes more plentiful than that triggered by red and green stimuli, and they successfully realized the recognition of color-mixed patterns. MNIST images have also been utilized by Ham et al. to investigate the learning efficiency of organolead halide perovskite synaptic devices (Figure 10b).\textsuperscript{143} The low-energy-consumption device can reach 82.7% recognition accuracy after 2000 trainings, which paves a significant way for promoting the learning and recognition capability in visualization systems and neuro-inspired computing. Moreover, Lv et al. made use of MNIST pattern to investigate learning and recognition ability in the single-layer perceptron network consisting of 785 input neurons and 10 output neurons made of carbon dots/silk-based synapses (Figure 10c).\textsuperscript{102} After 15 000 trainings, the recognitions for the letters “A” and “I” reach about 73% and 65%, respectively.

In bioinspired electronics, it is especially essential to simulate sensing and motor functions of human. Lee et al. reported an organic optoelectronic sensorimotor system based on an organic optoelectronic synaptic device and a stretchable organic nanowire synaptic transistor (s-ONWST).\textsuperscript{144} The key to this sensorimotor system is that the s-ONWST has the capability of perceiving and transmitting the inputs optical information and then generates the subsequent motor outputs. The International Morse code is utilized as the optical signals to produce the output information through the optoelectronic synapse, which is a new kind of

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**Figure 10.** (a) Integration of the h-BN/WSe\textsubscript{2} synaptic device and the colored pattern recognition based on an artificial optic-neural network. Reproduced with permission.\textsuperscript{142} Copyright 2018, Springer Nature. (b) Schematics of the organolead halide perovskite synaptic device and estimated recognition accuracy for the MNIST patterns. Reproduced with permission.\textsuperscript{143} Copyright 2019, Wiley-VCH. (c) Mapping images and recognition rate of “A” and “I” with the evolution of training phases. Reproduced with permission.\textsuperscript{102} Copyright 2019, Wiley-VCH.
optical wireless communication for human–machine interfaces. The standard emergency signal, “SOS”, is considered as the input optical signal to enable EPSC response under 100% strain, resulting in almost similar EPSC response at 0% strain. Except for visible light, different wavelengths of light, including ultraviolet and infrared are also used as the input signals. As a result, distinct EPSC responses are achieved, indicating optoelectronic synaptic devices can be applied in broad-spectrum absorption. Moreover, when applied successive light pulses, EPSC responses gained from optoelectronic synapse are transmitted to the artificial actuator. With the increase in pulses, the displacement δ and output voltage enhance as well. More importantly, even under the condition of 100% strain, the actuator can still work stably. The organic optoelectronic sensorimotor system exhibits a reliable strategy for the further development in neurorobotics and soft robotics.

4. Conclusion and Outlook

Overall, this Review has summarized recent advances in photonic synapses and their application in neuromorphic systems. Compared with synapses investigated by electric stimulus, photonic synapses have unique advantages of ultrahigh propagation speed, high bandwidth and low crosstalk, which improve computing speed and promotes optical wireless communication and operation. To date, the research of photonic synapses is still in its infancy and major synaptic behaviors, such as EPSC, PPF, STP, LTP, the transition from STP to LTP, have successfully been simulated by a great variety of emerging materials. The discussion is firstly focused on different kinds of emerging materials used in photonic synapses. Metal oxides are the most widely investigated and IGZO can be considered as the most popular material among them. Currently, perovskites have been extensively researched due to their high carrier mobility, tunable bandgap, and solution process availability. Furthermore, low-dimensional materials including 0D, 1D, and 2D materials have also been highlighted on account of their unique optical and electric properties. MoS₂, graphene, and BP are regarded as three promising 2D materials for photonic synapses, which have been actively studied by scientists. Compared with inorganic materials, polymers with their distinctive performance of low cost, mechanical flexibility, excellent ductility, compatibility with CMOS technology, and low energy consumption have attracted much more attention as well. Furthermore, phase change materials have been recently in the spotlight for their unique switching behaviors and multiple programmable resistances.

Nevertheless, most studies have only focused to mimic synaptic responses in a single device. Mimicking complex biological sensory synapses is still a tremendous challenge, which can be solved step by step through the joint effort of researchers in various fields including physics, chemistry, computer science, electronics, and biology. Although intense progress has been obtained both in materials and device structures, some issues and weak points remain unfathomed. First, the working mechanisms of photonic synapses have not been completely revealed yet. Therefore, to further enhance the performance of photonic synapses, the working mechanisms are essential to be deeply investigated. Moreover, photonic synaptic devices still face with nonlinear writing, which is a short board in image recognition. In addition, compared to electrical-stimulated synapses, photonic synapses cost more energy consumption, because relatively large illumination intensity is needed when applying light pulses. So far, each light spike still requires at least hundreds to thousands of pJ of energy, which is much higher than that of electrical-stimulated synapses and biological synapses. Power consumption is one of the most important properties of synaptic devices, so reducing power consumption is essential. Further reducing the sizes of photonic devices, improving light responses, and optimizing the structures of photonic devices may be several feasible solutions to settle the consumption problem. Furthermore, during the learning and forgetting process, light stimuli is mainly used to achieve potentiation behavior, whereas for the habituation behavior, electric stimuli is utilized. Hence, it is expected to obtain the habituation under light stimulus not only through materials with their inherent photoresponse but also through innovative architectures. Although several kinds of materials have revealed negative photoresponse and have achieved habituation under optical stimulus, almost none of them has utilized the optical habituation characteristic in neuromorphic systems. Thus, it is significant to emulate sophisticated neural functionalities only through the optical modulation, so that optical wireless communication will come true genuinely, which is an indispensable step for further information transition and operation in neuromorphic systems.

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Conflict of Interest

The authors declare no conflict of interest

Keywords

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