Optoelectronic Synaptic Devices for Neuromorphic Computing

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1. Introduction

Computing based on the von Neumann architecture has witnessed rapid development for more than half a century due to its capability of solving complex and structured mathematical problems.[1] However, it will soon struggle to meet the increasing demand of intelligent computing with low energy consumption.[2] The potential of the computing based on the von Neumann architecture is in fact limited by the physical partition of memory and central processing unit (CPU), which leads to high energy consumption in data-intensive operation. Moreover, the computing based on the von Neumann architecture can only execute prespecified programs, hardly handling unstructured, probabilistic, and real-time problems.[3,4] By contrast, a human brain can be considered as the most efficient biological processor. It is capable of high-speed parallel information processing with ultralow energy consumption and considerable fault tolerance.[5–7] In addition, the human brain can perform a series of intelligent activities such as self-learning, language processing, and image recognition.[8,9] Therefore, a computing system that is as efficient and fast as the human brain should circumvent the von Neumann bottleneck. To this end, scientists have been studying neuromorphic computing for its ability to emulate the biological computing of the human brain, ushering into a new era of computing.[10–15]

The human brain has a complex network of $\approx 10^{11}$ neurons, which are efficiently connected via $\approx 10^{15}$ synapses.[8] A neuron receives signals in the form of action potentials from other neurons. These signals are integrated and then transferred to neighboring neurons. The transfer of all the signals is enabled by synapses.[6,10,16] Therefore, the mimicking of the biological functionalities of synapses is critical to neuromorphic computing.[17–20] Many efforts were made to mimic the complex synaptic functionalities using graphics processing units (GPUs) with backpropagation algorithm. Since Carver Mead proposed the term of “neuromorphic” in the late 1980s,[21] neuromorphic circuits based on silicon complementary metal–oxide–semiconductor (CMOS) have been rapidly developed to mimic synaptic functionalities, as evidenced by those in IBM’s TrueNorth chip[22] and Intel’s Loihi chip.[22] Nevertheless, all the achievements have the limitation of complex architectures, which are extremely hungry for power and occupy too much space.[23]

First demonstrated by HP Labs in 2008,[24] nonvolatile memristors (NVMs) have recently become popular as synaptic devices.[25–32] Phase-change memory devices,[33–35] atomic-switch memory devices,[36,37] and ferroelectric memory devices[38] have all been used as novel NVMs to mimic synaptic functionalities. However, these devices are often purely electronic, having

Neuromorphic computing can potentially solve the von Neumann bottleneck of current mainstream computing because it excels at self-adaptive learning and highly parallel computing and consumes much less energy. Synaptic devices that mimic biological synapses are critical building blocks for neuromorphic computing. Inspired by recent progress in optogenetics and visual sensing, light has been increasingly incorporated into synaptic devices. This paves the way to optoelectronic synaptic devices with a series of advantages such as wide bandwidth, negligible resistance–capacitance (RC) delay and power loss, and global regulation of multiple synaptic devices. Herein, the basic functionalities of synaptic devices are introduced. All kinds of optoelectronic synaptic devices are then discussed by categorizing them into optically stimulated synaptic devices, optically assisted synaptic devices, and synaptic devices with optical output. Existing practical scenarios for the application of optoelectronic synaptic devices are also presented. Finally, perspectives on the development of optoelectronic synaptic devices in the future are outlined.
electrical input and electrical output. They face challenges such as narrow bandwidth and interconnect-related resistance-capacitance (RC) delay and power loss. With the emergence of optogenetics in neuroscience, light has been incorporated into synaptic devices. Several fully optical synaptic devices have been demonstrated in photonic chips based on phase-change materials and fiber-based laser systems. Optical neuromorphic computing based on the fully optical synaptic devices needs to face serious issues such as complicated wavelength conversion and sophisticated laser systems. Optoelectronic synaptic devices may enable the bidirectional conversion between electrical and optical signals. This can significantly facilitate the integration of optoelectronic artificial neural systems. Optoelectronic synaptic devices have advantages such as wide bandwidth, excellent interconnect with negligible RC delay and power loss, and global regulation of multiple synaptic devices. In addition, optoelectronic synaptic devices are capable of combining visual sensing, signal processing, and memory. This is significant for neuromorphic computing given the fact that human beings obtain most information via a biological visual cortex system. Therefore, considerable efforts have been devoted to the investigation of optoelectronic devices in the past few years. A series of materials with remarkable electronic and optical properties have been used in the fabrication of optoelectronic devices.

In this work, we discuss the progress of optoelectronic synaptic devices. First, we introduce the basic functionalities of synaptic devices (e.g., short-term plasticity [STP], long-term plasticity [LTP], spike-timing-dependent plasticity [STDP], and spike-rating-dependent plasticity [SRDP]). We then discuss a variety of optoelectronic synaptic devices by categorizing them into optically stimulated synaptic devices, optically assisted synaptic devices, and synaptic devices with optical output. Scenarios that have already been demonstrated for the use of optoelectronic synaptic devices (e.g., emulation of neural activities, image recognition, filtering, and logic functions) are then presented. Finally, a few perspectives are provided. The overview of this Review is shown in Figure 1.

2. Basic Functionalities of Synaptic Devices

Synaptic devices mimic biological synapses and therefore realize the basic functionalities of biological synapses. It is well known that various synaptic functionalities stem from synaptic weight, which is essentially the connection strength between a presynaptic neuron and a postsynaptic neuron. There is a simple analogy that can be drawn between a biological synapse and a synaptic device: the change in synaptic weight of the biological synapse is analogous to the variation of a specific parameter (e.g., resistance or conductivity) of the synaptic device. In practice, it is routine to measure the current across the synaptic device to examine the simulation of synaptic functionalities upon stimulation. This current corresponds to the postsynaptic current (PSC), which may be either excitatory or inhibitory.

Figure 2a shows the typical inhibitory PSC (IPSC) and excitatory PSC (EPSC), which were observed from an optoelectronic synaptic device based on the hybrid structure of graphene and carbon nanotubes (CNTs). Please note that both the IPSC and EPSC could be tuned by the gate voltage.

Synaptic plasticity refers to the change of synaptic weight. According to Hebbian’s theory, STP, LTP, STDP, and SRDP are the typical forms of synaptic plasticity. Synaptic plasticity is critical to neural activities such as learning, forgetting, image recognition, and logic functions. Hence, synaptic devices must be able to exhibit synaptic plasticity.

2.1. STP, LTP, and STP-to-LTP Transition

STP is related to the recognition and processing of input information that only relies on the properties of neural response. Thus, the recognized information can be easily forgotten. Paired pulse facilitation (PPF) and paired pulse depression (PPD) are the most renowned functionalities for synaptic devices to reflect STP. For two successive stimuli, the response of synaptic devices to the second stimulus may increase (decrease) compared with that to the first stimulus, leading to PPF (PPD). Please note that for biological PPF, residual Ca$^{2+}$, Na$^+$, and K$^+$ in the active regions of a synapse enhance subsequent action potential. Biological PPD indicates the voltage-dependent inactivation of Ca$^{2+}$ channels or the temporary depletion of accumulated neurotransmitter vesicles in presynaptic neurons. The PPF (PPD) index is obtained by calculating the ratio of the EPSC (IPSC) evoked by the second stimulus ($A_2$) to that evoked by the first one ($A_1$). Moreover, the change of the
 dos PPF/PPD en función del intervalo de tiempo entre dos estimulaciones sucesivas \( \Delta t \) se puede usar para evaluar el STP de dispositivos sinápticos. La figura 2b,c muestra un comportamiento PPF típico y la dependencia del índice PPF en \( \Delta t \), respectivamente. El índice PPF usualmente decrece con el aumento de \( \Delta t \), lo que puede cumplir con la ecuación:

\[
\text{PPF decay} = c_1 \cdot \exp(-\Delta t/\tau_1) + c_2 \cdot \exp(-\Delta t/\tau_2)
\]

Donde \( \tau_1 \) (\( \tau_2 \)) es el tiempo característico de relajación del fase rápida (lenta) del PPF y \( c_1 \) (\( c_2 \)) es el magnitud inicial de la fase rápida (lenta). Es importante notar que \( \tau_2 \) es usualmente un orden de magnitud mayor que \( \tau_1 \) en sinapsis biológicas regulares, lo que es característico de la sinaptización rápida.\(^{20,66}\) Algunos dispositivos sinápticos óptoelectrónicos han imitado el PPD. Wang et al.\(^{60}\) lograron PPD después de PPF mediante la modulación del intervalo de tiempo de los estímulos ópticos en un dispositivo sináptico basado en cuantos de perovskita \( \text{CsPbBr}_3 \). La transición de PPF a PPD se demostró al aumentar \( \Delta t \) de 1 a 10 s, como se muestra en la figura 2d.

LTP actúa como el cambio a largo plazo de la sinaptización, el cual está asociado con el aprendizaje y la memoria que continúa durante varias horas en el cerebro humano. En el cerebro humano, el cambio de sinaptización tiene polaridad. LTP incluye potenciación a largo plazo y depresión a largo plazo, que representan los cambios excitatorios e inhibitorios durante la estimulación repetitiva, respectivamente.\(^{40}\) En el hipocampo, la potenciación a largo plazo es un fenómeno biológico, donde un gran flujo de iones \( \text{Na}^+ \) y \( \text{Ca}^{2+} \) fluye en las membranas sinápticas resultando en una sinaptización aumentada. Este aumento es debido al aumento de potenciación a largo plazo.
activation of Mg$^{2+}$-sensitive receptors and the phosphorylation of α-amino-3-hydroxy-5-methyl-4-isoxazolepropionic acid (AMPA) receptors.[67] In contrast, depression decreases synaptic weight due to the internalization of AMPA receptors by low-frequency activation of phosphatases. This pair of contrary changes in synaptic weight depolarizes and hyperpolarizes the membrane potential. Long-term depression is likely to reverse the previous long-term potentiation. Thus, the counterbalance between long-term potentiation and long-term depression maintains synaptic weight in a linear range, enabling the frequency-based signals to be processed in an agile manner.[68] According to He et al.'s work,[63] long-term potentiation and long-term depression may be implemented simultaneously (Figure 2e,f) although long-term depression is realized with the help of electrical stimulation.

In addition, STP can be gradually transformed into LTP after a few iterations of external information training.[69] Normally, synaptic devices are investigated using spike frequency- or number-varying stimulation to render the change from STP to LTP.[70,71] Gao et al. demonstrated the STP-to-LTP transition of a Schottky-junction synaptic device by changing the number and frequency of electrical stimuli (Figure 2e,f).[59] Generally, the decay of the final EPSC/IPSC induced by consecutive stimuli can be fitted using the following Kohlrausch stretched exponential function[72]

$$I(t) = I_0 \exp \left[ -\frac{(t - t_0)^\beta}{\tau} \right] + I_\infty$$

where $I(t)$ is the decay function, $\tau$ is decay time, $I_0$ is the pre-exponential factor, $I_\infty$ is the value of the EPSC/IPSC after the decay, and $\beta$ is the stretch index ranging from 0 to 1. The increase in the decay time (i.e., the increase in retention time) indicates more intensive memory and lower forgetting rate, signifying the accomplishment of STP-to-LTP transition. The decay of the EPSC/IPSC induced by consecutive stimuli has been shown to be similar to the well-known Ebbinghaus forgetting curve.[73,74]

### 2.2. STDP, SRDP, and Learning Rules

STP and LTP are the basic characteristics of individual biological synapses. However, for the understanding on the interaction between neurons, it is necessary to explore the interconnection among multiple synapses through STDP and SRDP. STDP concerns the dependence of the change of synaptic weight on the temporal order and time interval between presynaptic and postsynaptic spikes.[75–77] As shown in Figure 2i,[62] there are four forms of STDP that can be derived from different changes of synaptic weight ($\Delta$wt%), which are caused by varying the spike order and spike interval ($\Delta t$). For the presynaptic spike before (after) the postsynaptic spike, i.e., $\Delta t > 0$ ($\Delta t < 0$) $\Delta$wt% can be positive (negative), indicating the potentiation (depression) of synaptic weight. The change of synaptic weight with the timing of the spikes usually complies with a STDP function, from which the so-called learning window can be obtained.[78] Symmetric STDP can enable associative learning, in which synaptic weight is strengthened upon learning a pattern so that the original pattern may be subsequently recalled even with the presence of an incomplete or noisy pattern. Asymmetric STDP can give rise to sequence learning, in which future events may be anticipated based on the temporal sequences of previous events.[79] In contrast, SRDP modifies the sign and magnitude of synaptic plasticity by varying the firing rate of presynaptic spikes. A high-frequency firing rate leads to potentiation, whereas a low-frequency firing rate gives rise to depression.[80]

The Hebbian learning rule identifies not only the competition between different synapses but also the activity-dependent synaptic plasticity. The aforementioned STDP and SRDP are two paradigms of the Hebbian learning rule. In particular, STDP can be regarded as a competitive spike-based formulation of the Hebbian learning rule because it introduces competition between different synapses to control the timing of postsynaptic action potentials.[81] SRDP mainly reveals activity-dependent synaptic plasticity, indicating that low (high) postsynaptic activity reduces (enhances) synaptic efficacy.[80] The window for the depression in SRDP and STDP could be considered as an extension of the Hebbian learning rule.[17,82,83] The simulation of the Hebbian learning rule has been adopted in artificial neural networks (ANNs) to handle sophisticated scenarios with excellent learning efficiency.[84,85]

### 3. Categories of Optoelectronic Synaptic Devices

Various types of synaptic devices with electrical stimulation and output have been demonstrated for relatively long time. Optoelectronic synaptic devices are now newly developed. The functionalities of optoelectronic synaptic devices are realized with a variety of material systems, device structures, and stimulation modes. Optoelectronic synaptic devices based on different operating mechanisms are fabricated to target different applications.

#### 3.1. Optically Stimulated Synaptic Devices

Pina et al. once investigated the mimicking of some elementary properties of neurons by using a chemical solution in 2000.[86] They found that the changes of the absorption spectrum of the solution showed the integration effect of two distinctive UV light inputs due to chemical reactions. Such a phenomenon was actually similar to synaptic summation. Such a phenomenon was actually similar to synaptic summation. This early work demonstrated the usefulness of light for artificial neural devices. The incorporation of light into synaptic devices was first reported by Pilarczyk et al. in 2016. They used the hybrid structure of CdS/multiwalled carbon nanotube (MWCNT) to fabricate the devices.[87] The device structure consisted of a photocell assembled with a photoactive layer of the hybrid structure, two-terminal indium-doped tin oxide (ITO) electrodes and a semisolid electrolyte (Figure 3a). The synaptic devices successfully exhibited the STDP when the optical stimulation induced photocurrent was examined. In the past five years more and more optically stimulated synaptic devices have been fabricated. Different working mechanisms have been proposed for them.

#### 3.1.1. Ionization/Neutralization of Oxygen Vacancies

Oxide semiconductors are the most common materials used for all-electronic synaptic devices. They are also used for optoelectronic synaptic devices. The inherent persistent photoconductivity
(PPC) of oxide semiconductors enables synaptic devices to mimic synaptic plasticity upon optical stimulation. Yu et al.\cite{88} proposed a model to explain the origin of PPC, as shown in Figure 3b. An oxygen vacancy ($V_O$) is assumed to have two different states in oxide semiconductors. One is the neutral ground state ($V_0^0$), which is insulating. The other is the metastable ionized state ($V^+_O$ or $V^{2+}_O$), which is conducting. When the energy of photons is larger than the bandgap of an oxide semiconductor, light causes the oxide semiconductor to switch from the neutral ground state to the metastable ionized state. Thereafter, slow backward relaxation occurs presumably because electrons in the metastable ionized state need to circumvent a barrier ($E_a$), inducing the PPC.

Lee et al.\cite{89} fabricated a series of optoelectronic synaptic devices based on amorphous oxide semiconductors (AOSs) such as indium gallium zinc oxide (IGZO), indium strontium zinc oxide (ISZO), indium strontium oxide (ISO), and indium zinc oxide (IZO), as shown in Figure 3c. The relationship between the PPC and synaptic functionalities was systematically investigated. They found that IGZO-based synaptic devices showed the most successful simulation of LTP, whereas IZO synaptic devices did not exhibit any obvious synaptic plasticity due to the weak PPC (Figure 3d). This difference is induced by the different values of $E_a$ for different AOSs. With the increase in $E_a$, the recombination time of photocarriers increases, leading to more pronounced PPC (Figure 3e).

Yu et al.\cite{88} used the wide bandgap insulator of SnO$_x$ to cover IGZO, enabling the extraction of weakly bound oxygen from IGZO. This resulted in more oxygen vacancies in IGZO. Moreover, it was found that $E_a$ shifted from 0.84 to 0.90 eV after the SnO$_x$ coverage. Therefore, more remarkable PPC was observed, improving synaptic simulation (Figure 3f). Wu et al.\cite{90} alternatively enhanced the PPC effect by tuning the composition of IGZO. They found that a low (high) fraction of Ga (In) led to higher photocurrent and the slower decay of the photocurrent (Figure 3g). This was essentially explained by the fact that Ga more strongly bonded with oxygen than In to retrain the formation of oxygen vacancies. As relatively long time was required to neutralize ionized oxygen vacancies, the devices based on IGZO with a low (high) fraction of Ga (In) exhibited the slow decay of the photocurrent. Such slow decay process was similar to those observed for optically stimulated synaptic devices based on Nb:SrTiO$_3$,\cite{59} InGaZnO$_x$/Al$_2$O$_3$,\cite{91} InZnO,\cite{92} ZnO,\cite{93} and InGaCdO,\cite{94} facilitating the simulation of long-term memory (LTM).
3.1.2. Trapping/Detrapping of Photogenerated Carriers

**Defect-Induced Trapping/Detrapping:** Defects such as dangling bonds and local structural distortions may act as traps that capture carriers. Trapped carriers usually take certain time to be released. This well simulates the decay of PSC in biological synapses. The carrier-trapping process readily occurs when electrons and holes are generated by optical stimulation. After the optical stimulation, the release of trapped photogenerated carriers leads to the decay of photocurrent. If a synaptic device is optically stimulated again before the complete release of the trapped photogenerated carriers, it is assumed that traps with even lower energy would be occupied. This can give rise to longer decay time and even nonvolatile residual current, which is desired for the mimicking of LTM.

Tan et al. [95] studied two-terminal synaptic devices based on silicon nanocrystals (Si NCs) (Figure 4a). It was found that defects such as dangling bonds were at the surface of Si NCs, as evidenced by the electron paramagnetic resonance (EPR) signal. Scanning tunneling spectroscopy (STS) revealed that the dangling bonds introduced deep energy levels, which may trap photogenerated electrons. The photogenerated holes contributed to the increase in the device current due to nearest-neighbor hopping in the film of Si NCs. After the optical stimulation stops, trapped electrons at dangling bonds were slowly released to the conduction band through thermal fluctuation, giving rise to the decay of the photocurrent (Figure 4b). Several other researchers have also taken advantage of the defects of photosensitive materials as traps to fabricate optically stimulated synaptic devices. For example, Qian et al. [99] used (PEA)$_2$SnI$_4$ perovskite with Sn vacancies to make synaptic devices. The trapping/detrapping of photogenerated electrons at Sn vacancies enabled the devices to simulate synaptic plasticity upon optical stimulation.

**Interface-Induced Trapping/Detrapping:** Carrier trapping/detrapping can also occur at a semiconductor/dielectric interface via defects, polymer polar groups, and electric-double-layers (EDLs). Hu et al. [96] reported a two-terminal synaptic device based on the structure of ITO/ZnO/Al (Figure 4c). The device could work with both electrical and optical stimulations. They showed that PPC induced by optical stimulation at the wavelength

![Figure 4](https://www.advancedsciencenews.com/wasui.png)

**Figure 4.** a) Schematic of an array of Si NC-based synaptic devices. b) Diagram for the electronic structure and carrier behavior of SiNCs. The trap state is denoted by $E_T$. Reproduced with permission. [95] Copyright 2018, Elsevier Ltd. c) Device structure of an ITO/ZnO/AlO$_x$/Al two-terminal optoelectronic synaptic device. d) Band diagram of the ZnO$_{1-x}$/AlO$_x$ junction and operating mechanism under optical stimulation. Reproduced with permission. [96] Copyright 2018, American Chemical Society. e) Schematic of the InAs NW phototransistor. f) Energy band of excitation, thermalization, recombination, and trapping processes in an InAs NW. Reproduced with permission. [97] Copyright 2018, IOP Publishing Ltd. g) Structure of MoS$_2$ EDL phototransistor. Band diagram of the MoS$_2$ phototransistor under negative gate bias h) with and i) after light stimulation. Reproduced with permission. [98] Copyright 2019, The Royal Society of Chemistry.
of 310 nm enabled the simulation of synaptic functionalities. X-ray photoelectron spectroscopy (XPS) on the ZnO/Al interface was used to elucidate the mechanisms of the PPC. It was indicated that Al captured O from ZnO to form an interfacial AlO\textsubscript{x} layer.\textsuperscript{[109]} Defects in the interfacial AlO\textsubscript{x} layer were traps, leading to the PPC. To verify the effect of the interfacial AlO\textsubscript{x} layer, they made a device based on the structure of ITO/ZnO/Pt. It was observed the PPC of the ITO/ZnO/Pt device was absent, indicating that the traps in the interfacial AlO\textsubscript{x} layer were indeed responsible for the PPC. Upon optical stimulation, photogenerated holes might be trapped in the interfacial AlO\textsubscript{x} layer, while they would be slowly released once the optical stimulation stops (Figure 4d).

In contrast to the usual optical-stimulation-induced increase in current, Li et al.\textsuperscript{[97]} reported an optical-stimulation-induced decrease in current for a synaptic device based on a n-type InAs nanowire (NW) (Figure 4e). This enabled their demonstration of IPSC. It was believed that a trapping level was in InO\textsubscript{y} at the surface of InAs (Figure 4f). Photogenerated electrons might be trapped by the trapping level in InO\textsubscript{y} or recombine with holes in the valence band after thermalization. Given the high mobility of the InAs NW, the photogenerated electrons had enough free path to reach InO\textsubscript{y} and get trapped there before thermalization. Meanwhile, photogenerated holes recombined with intrinsic electrons in the InAs NW, reducing the current. After the removal of optical stimulation, there was an increase in the current because detrapped electrons recombined with the photogenerated holes in the InAs NW. The slow detrapping process of electrons in InO\textsubscript{y} led to the slow recovery of the current, facilitating the mimicking of LTP. A similar working mechanism has also been demonstrated for devices based on the thin-film structure of IZO/HfO\textsubscript{2}.\textsuperscript{[101]} Analogously, both Qin et al.\textsuperscript{[58]} and He et al.\textsuperscript{[61]} hypothesized that the trapping sites originated from dangling Si–O bonds on the substrate surface (SiO\textsubscript{2} layer). The gate electric field could effectively control the charging/discharging of trapping sites thereby emulating the potentiation or depression alternatively (Figure 2e,f).

Organic polar groups may also be responsible for the carrier trapping/detrapping, as demonstrated by Dai et al.’s work\textsuperscript{[102]} on synaptic transistors based on C8-BTBT. A polyacrylonitrile (PAN) film was used as a dielectric layer. Photogenerated holes in C8-BTBT might be trapped and then detrapped at the C8-BTBT/PAN interface, where polar groups exist. In addition, the hydroxyl groups of chitosan at the ITO/chitosan interface were proposed to trap and then release photogenerated electrons. This facilitated the fabrication of optically stimulated ITO/chitosan-based synaptic transistors.\textsuperscript{[103]}

At the electrolyte-semiconductor interface, EDLs are formed and act as a nanogap capacitor. This capacitor could create large electric fields at the interface.\textsuperscript{[104]} An EDL-based phototransistor is usually gated by an electrolyte, which is full of mobile ions. High polarization readily occurs under an electric field.\textsuperscript{[105]} It has been realized that the gating of EDLs plays an important role in regulating channel carrier density via trapping/detrapping or doping/dedoping processes.\textsuperscript{[106–108]} Thus, EDL-based phototransistors have become important optoelectronic synaptic devices. Jiang et al.\textsuperscript{[98]} have taken advantage of the strong electrostatic coupling of an EDL to mimic long-term potentiation and long-term depression. Their device was a transistor with the channel material of 2D molybdenum disulfide (MoS\textsubscript{2}) and the electrolyte gate of liquid sodium alginate biopolymer, as shown in Figure 4g. Under a negative gate electric field, the electrolyte was polarized so that electrons in the electrolyte and positive charges in MoS\textsubscript{2} accumulated at the electrolyte/MoS\textsubscript{2} interface. Upon optical stimulation, photogenerated electrons in MoS\textsubscript{2} were attracted to the electrolyte/MoS\textsubscript{2} interface by the positive charges in MoS\textsubscript{2} (Figure 4h). This reduced the Schottky barrier at the metal/MoS\textsubscript{2} interface, increasing the efficiency of electrons tunneling into the channel (Figure 4h). Thus, the device exhibited potentiation plasticity due to the increase in the conductance of the channel. Moreover, upon optical stimulation, the trapped photogenerated electrons at the electrolyte/MoS\textsubscript{2} interface formed new trapping states for holes to increase the Schottky barrier, leading to depression plasticity (Figure 4i). The EDL-related carrier trapping and detrapping also occurred to optically stimulated synaptic transistors with a different channel material or electrolyte film.\textsuperscript{[109,110]} Please note that these synaptic devices might operate at a relatively low voltage, which was conducive to low energy consumption.

**Heterojunction-Induced Trapping/Detrapping:** For synaptic devices intentionally designed with type-II energy band structures, photogenerated carriers may be trapped in a potential well induced by the energy band alignment.\textsuperscript{[60,111–116]} Wang et al.\textsuperscript{[111]} showed a synaptic phototransistor, in which a hybrid film of CsPbBr\textsubscript{3} QDs and poly (3,3-didodecylquaterthiophene) (PQT-12) was the channel (Figure 5a). The energy band diagram of the hybrid film of PQT-12 and CsPbBr\textsubscript{3} QDs in Figure 5b. The photogenerated holes in CsPbBr\textsubscript{3} QDs were injected into PQT-12, whereas the photogenerated electrons in CsPbBr\textsubscript{3} QDs were retained due to the potential well at each CsPbBr\textsubscript{3} QD. PQT-12 actually assisted the separation of photogenerated carriers to reduce their recombination. This was evidenced by the fact that the photocurrent of the device based on CsPbBr\textsubscript{3} QD/PQT-12 decayed much more slowly than that of the device based on CsPbBr\textsubscript{3} QDs (Figure 5c). Wang et al.\textsuperscript{[114]} fabricated optoelectronic synaptic devices using a molybdenum disulfide/perylenetetracarboxylic dihydride (MoS\textsubscript{2}/PTCDA) hybrid heterojunction (Figure 5d). The type-II energy band alignment of the heterojunction allowed photogenerated electrons to move from PTCDA into MoS\textsubscript{2}, leading to the increase in the channel current (Figure 5d). After removal of the optical stimulus, these electrons gradually returned to PTCDA and recombine with photogenerated holes, resulting in gradually decreasing channel currents. As shown in Figure 5e, Ni et al.\textsuperscript{[113]} took advantage of the synergy of the excellent broadband optical absorption of boron (B)-doped SiNCs and the efficient charge transport of 2D WSe\textsubscript{2} to hybrid optoelectronic synaptic devices. Thus, the SiNC/WSe\textsubscript{2} synaptic devices exhibit important synaptic functionalities under the optical stimulation in a broad spectral region from the ultraviolet (UV) to near-infrared (NIR). In addition, the energy consumption per synaptic event of the SiNC/WSe\textsubscript{2} synaptic devices may be as low as \textasciitilde75 fJ.

Wang et al.\textsuperscript{[60]} demonstrated an optoelectronic synaptic transistor with the channel of CsPbBr\textsubscript{3} QDs/poly(methyl methacrylate) (PMMA)/pentacene (Figure 5f). Upon optical stimulation, photogenerated holes in CsPbBr\textsubscript{3} QDs readily moved to pentacene by tunneling through PMMA due to the type-II energy band alignment (Figure 5g), giving rise to current. Photogenerated electrons in CsPbBr\textsubscript{3} QDs could not go into pentacene to
recombine with holes unless an electric bias was used to change the energy band alignment (Figure 5h). Therefore, an electric field might be used to modulate the decay of the EPSC. A similar working mechanism was demonstrated with a synaptic device based on the heterostructure of In$_2$O$_3$/ZnO, whose transmittance was greater than 70% in the measured visible range (400–750 nm). However, the transmittance decreased sharply around 400 nm, showing the fundamental absorption of the devices. Due to the type-II band alignment between ZnO and In$_2$O$_3$, the photogenerated electrons moved from ZnO to In$_2$O$_3$ through the Fowler–Nordheim (F–N) tunneling under a positive bias, whereas the photogenerated electrons moved from In$_2$O$_3$ to ZnO via direct tunneling under a negative bias (Figure 5i).

The heterojunction-based optoelectronic synaptic devices can be fabricated using various hybrid structures with elaborately designed energy band alignment. These hybrid structures inherit the advantages of their component materials. The resulting synergistic effect may give rise to higher optical sensitivity and lower energy consumption. Particularly, the hybrid structures prevail over those with the carrier traps in terms of robustness and variability of devices. Moreover, electrical means can be additionally used to control the recombination of photogenerated carriers, improving the freedom of tuning the performance of optoelectronic synaptic devices.

3.1.3. Optical Resistance Switching

Resistive random-access memory (RRAM) devices are widely used for nonvolatile memory processing. They exhibit a transformation between high and low resistivity states under external stimuli. From the perspective of visual memory, optoelectronic nonvolatile resistance switching holds the potential for collocalization sensing, real-time processing, and memory of visual
information. Therefore, among various RRAM devices, optically triggered resistance switching is a suitable candidate for visual memory processing. Zhou et al.\cite{117} used MoO$_x$ to fabricate optoelectronic RRAM (ORRAM) devices, whose volatile and nonvolatile resistance switching were used to mimic the short- and long-term synaptic plasticities, respectively. As shown in Figure 6a, an ORRAM device had the two-terminal structure of Pd/MoO$_x$/ITO. A high resistance state (HRS) appeared during voltage sweeping (Figure 6b, black line). Switching to a low resistance state (LRS) took place under UV light stimulation (Figure 6b, blue line). After removing the optical stimulation, the ORRAM device was reset to the HRS under a negative voltage sweep (Figure 6b, red line). Figure 6c shows the pulse switching characteristics of optical setting and electric resetting for this ORRAM device. The resistance switching characteristics were investigated using the XPS of MoO$_x$ before (Figure 6d) and after (Figure 6e) the UV stimulation. The results revealed that the UV stimulation led to resistance switching by changing the valence states of Mo ions from $6^+$ to $5^+$. In the optical setting process, the MoO$_x$ absorbed the UV light to generate electrons and holes. The photogenerated holes reacted with the absorbed water molecules and produced protons (H$^+$). The photogenerated electrons and protons resulted in the transition of the valence state of the Mo ions. In the electric resetting process, the electric field extracted the protons from the MoO$_x$, reverting the resistance state to HRS. Thus, the ORRAM device mimicked synaptic functionalities by taking advantage of optical inputs and light-dosage-dependent resistance-state outputs. The STP, LTP, and STP-to-LTP transition under the optical stimulation were observed in the ORRAM device (Figure 6g–i). Synaptic devices based on optoelectronic resistance switching exhibit the potential for visual memory because of their long endurance and tunable forgetting processes.\cite{118} In addition, the crossbar structures of ORRAM devices vastly downscale the device area, facilitating large-scale integration for neuromorphic visual systems.

3.2. Optically Assisted Synaptic Devices

In physiology, optogenetics shows that intracellular Ca$^{2+}$ influx is controlled by not only electrical impulses but also light.

Figure 6. a) Device structure of MoO$_x$ ORRAM. b) Optical set and electrical reset process of ORRAM. c) Pulse-switching characteristics. Narrow scans of the Mo 3D peak for the MoO$_x$ layer d) before and e) after UV illumination. f) Proposed switching mechanism in the MoO$_x$ ORRAM. g) Light-intensity-dependent STP with a pulse width of 200 ms and 2 s. h) LTP with the pulse number increased to 500. i) STP-to-LTP transition after 300 pulse stimuli at different light intensities. Reproduced with permission.\cite{117} Copyright 2018, Springer Nature.
With additional light stimulation, biological neurons and neurotransmitters gain modified recognition and learning capabilities by changing the threshold of synaptic plasticity.\cite{119} Optically assisted optoelectronic synaptic devices provide opportunities for the simulation of light-assisted intracellular Ca\textsuperscript{2+} dynamics. Unlike the optically stimulated synaptic devices, optically assisted synaptic devices use the effect of light on an electrically induced ion/vacancy migration process. Light stimulation alone may not be sufficient to induce ion/vacancy migration in optically assisted synaptic devices, which may be based on ion-conducting materials such as organometal trihalide perovskites.\cite{55,56,120} In the early stages, Xiao and Huang\cite{56} successfully demonstrated a two-terminal polycrystalline CH\textsubscript{3}NH\textsubscript{3}PbI\textsubscript{3} memristor with synaptic functionalities under electrical stimuli. They observed the phenomenon of photo-read SRDP when sun illumination was used along with electrical stimuli. It was proposed that the drift of ions/vacancies was induced not only by applied external electrical bias, but also by photovoltage due to the light poling effect of the perovskite devices.\cite{121} Based on the influence of light on ion/vacancy migration, Zhu and Lu\cite{120} reported a two-terminal lateral CH\textsubscript{3}NH\textsubscript{3}PbI\textsubscript{3} based memristor that exhibited optically assisted synaptic annihilation dynamics. The device structure is shown in Figure 7a. Under electrical bias, the increasing concentration of iodine vacancies (\(V_{\text{I}}/V_{\text{I}}^\text{x}\)) in their device was induced by \(\text{I}^-\) migration in MAPbI\textsubscript{3}, causing the device to switch to the LRS. \(\text{I}^-\) ions were then stored in an Ag anode temporarily and diffused back to MAPbI\textsubscript{3}, leading to the decrease in conductance after the removal of the electrical bias. Hence, the switching to the HRS was observed. Please note that with the increase in light intensity it became more difficult for the device to switch to the HRS. But it was easier to switch to the HRS (Figure 7b). This phenomenon indicated that under an electric field, light could inhibit \(V_{\text{I}}/V_{\text{I}}^\text{x}\) generation by increasing the formation energy of \(V_{\text{I}}/V_{\text{I}}^\text{x}\) (Figure 7c). On the contrary, light facilitated the recombination of \(V_{\text{I}}/V_{\text{I}}^\text{x}\)’s and \(\text{I}^-\) after the removal of the electrical bias and expedited the conductance decay process (Figure 7c). It was proved that the decay current was decreased (increased) by alternatively applying (eliminating) optical stimulation, as shown in Figure 7d. Thus, the competition between electrically induced formation and light-induced annihilation rendered the long-term potentiation or depression in the same device. In other words, the memory

![Figure 7](image-url)

Figure 7. a) Schematic of the device and the current–voltage (I–V) measurement setup. b) Switching voltage dependence on illumination intensity. c) Light illumination can inhibit formation under bias and accelerate annihilation. d) Conductance retention curve of a device during alternately applied and removed optical stimulation. e) Long-term depression and long-term potentiation processes are achieved with and without optical stimulation. Reproduced with permission.\cite{120} Copyright 2018, American Chemical Society. f) Mechanism of a conductive path formed by the \(\text{I}^-\) vacancy migration in the absence and presence of light, respectively. g) \(E_a\) for the case of light off (black) or on (red), and the change in \(E_a\) as a function of the migration distance of iodine vacancies. Reproduced with permission.\cite{55} Copyright 2018, Wiley-VCH.
formation or loss could be controlled by tuning the light illumination.

For the further modification of synaptic behavior, Ham et al.[55] fabricated a two-terminal vertical Ag/CH₃NH₃PbI₃/ITO synaptic device with optically assisted synaptic strengthening dynamics. Unlike the former lateral structure, the filament has the same direction with the diffusion of the separated electrons (holes) driven by photogenerated electric field in the vertical structure. As shown in Figure 7f, iodine vacancies moved to the ITO under an external electrical field ($E_{\text{ext}}$). When the device was stimulated by an optical spike, the separation of photogenerated electron–hole pairs produced photovoltage ($E_{\text{ph}}$), which had the same direction as $E_{\text{ext}}$. Due to the increase in the electrical field ($E = E_{\text{ext}} + E_{\text{ph}}$), the driving force $zeE$ (z is the charge state, and $e = 1.60 \times 10^{-19}$ C) of the iodine vacancies increased. This led to the increased current induced by the electrical field. Hence, the activation energy ($E_a$) of the iodine vacancies for migration might be effectively reduced by $-e/2 (v \propto zeE)$.[122] As a result, in contrast to the former synaptic device, under light illumination, the lower $E_a$ enabled the accelerated migration of the iodine vacancies. This allowed a smaller external electric field to enable the high-order tuning of synaptic plasticity, expediting learning and recognition with substantially lower energy consumption.

3.3. Synaptic Devices with Optical Output

Optoelectronic synaptic devices with electrical inputs and optical outputs may be useful for signal visualization, which indicates how individual neurons integrate synaptic inputs to function together.[123] In addition, this type of devices complete bidirectional conversion between optical and electrical signals, which is required for the full optoelectronic integration of neuromorphic systems. Zhao et al.[54] demonstrated electroluminescent (EL) synaptic devices based on Si NCs with logic functions. The EL synaptic devices had the multilayer structure of ITO/ZnO/Si NCs/CBP/MoO₃/Au with the EL peak at the wavelength of $\approx 740$ nm, as shown in Figure 8a. The EL lifetime was $\approx 20$ ms (Figure 8b), which was in the typical range (1–$10^4$ ms) for the decay time of a signal in a biological synapse.[66,125] Based on EPR analysis, the area density of dangling bonds on the NC surface was $\approx 8.4 \times 10^{11}$ cm$^2$. Deep energy levels induced by the dangling bonds were close to the conduction band minimum (CBM) of Si NCs. As shown in Figure 8c, the deep energy levels might be involved in the recombination of carriers injected via an electrical stimulus. Electrons and holes could directly recombine to emit light (process $\circ$). Meanwhile, some electrons might be trapped by the deep energy levels. After the electrical stimulus was removed, the trapped electrons could be detrapped and tunneled to the CBM of a neighboring Si NC (process $\circ\circ$) and then recombined with holes there (process $\circ\circ\circ$), contributing to the light emission.

Zhao et al.[124] subsequently reported the use of NIR QD light-emitting diodes (QLEDs) as optoelectronic synaptic devices (Figure 8d) given the advantage of the low loss of NIR light in several technologically important materials. These NIR QLEDs had the multilayered structure of Ag/ZnO/Si NCs/PFN/P3HT/PEDOT:PSS/ITO/glass with the EL peak at the wavelength of $\approx 850$ nm. The hole-transport layer (HTL) of P3HT with negligible absorption in the NIR region had a high hole mobility, mitigating the carrier injection imbalance of the devices. An interlayer of PFN between the P3HT layer and Si QDs layer was used to block electron leakage from the Si QD layer. These NIR QLEDs had successfully simulated important

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**Figure 8.** a) Schematic of Si NCs-based EL synaptic device. b) EL decay of the synaptic device stimulated by an electrical spike. The electrical spike and the output optical power are shown in the inset. c) Band alignment between the ZnO, Si NCs, and CBP layers under a bias voltage. The trapping/detrapping process in Si NCs and the dangling-bond-induced deep energy level are represented by the dashed line. Reproduced with permission.[53] Copyright 2018, Elsevier Ltd. d) Schematic of an NIR Si QD-based QLED. e) PPF of the NIR QLED. f) LTP of the NIR QLED. Reproduced with permission.[124] Copyright 2019, Science China Press and Springer-Verlag GmbH Germany.
synaptic functionalities such as STP and LTP with a lower energy consumption (Figure 8e,f).

4. Application Scenarios

4.1. Emulation of Neural Activities

The emulation of neural activities is of great significance for the development of ANNs.[23] Among all kinds of neural activities, memorizing and forgetting are fundamental ones.[36] According to the psychological model of human memory proposed by Atkinson and Shiffrin (Figure 9a),[111] biological memory levels can be divided into sensory memory (SM), STM, and LTM.[127] They differ in terms of the retention time of the memory. The SM is gained by sensory organs. It is transferred to the STM if the brain pays attention to the SM. The STM typically lasts from a few milliseconds to minutes. Before the complete loss of the STM, frequent rehearsal can lead to the LTM, which lasts for several hours or more. Forgetting can be described using the well-known Ebbinghaus forgetting curve, which indicates the decay of human memory over time.[73,74]

Most optoelectronic synaptic devices are capable of simulating memorizing and forgetting processes. The memorizing (forgetting) is usually realized by the long-term change (retention) of device conductance. Various factors, such as the number, frequency, duration, and intensity of stimulations may affect the memorizing and forgetting. For example, Wang et al.[111] showed that memory/forgetting could be efficiently simulated by using optical stimuli with varying quantity, intensity, and duration (Figure 9b). When the quantity, intensity, or duration of the optical stimulus increased, both the memory level and memory retention time increased. In addition, learning experiences are associated with the rememory and reforgetting process of the human brain. It often takes less time to remember the forgotten information, whereas it usually costs more time to forget the relearned information.[128] Gao et al.[59] demonstrated the effect of the learning experience using a simple ITO/Nb-doped SrTiO3 (ITO/Nb:SrTiO3) optoelectronic synaptic device (Figure 9c). Thirty-five cycles of optical stimulus were used to obtain certain potentiation of synaptic weight in the first learning process. The learned information was partially forgotten 100 s after the learning. However, in the relearning process, only seven cycles of optical stimulus were needed to reach the

Figure 9. a) Schematic of the human memory model proposed by Atkinson and Shiffrin. b) Channel conductance change as a function of presynaptic optical stimulus number, intensity, and duration. Reproduced with permission.[111] Copyright 2019, Wiley-VCH. c) Learning-experience behavior under optical stimulus. Reproduced with permission.[59] Copyright 2019, American Chemical Society. d) Classical conditioning demonstrated by Pavlov’s dog experiment. Reproduced with permission.[126] Copyright 2018, Wiley-VCH. e) Aversion learning demonstrated with Si NCs phototransistor. The drinking of alcohol and administering of treatment are emulated by the electrical and optical stimuli, respectively. Reproduced with permission.[54] Copyright 2019, Elsevier Ltd.
previous synaptic weight. In the meantime, less relearned information was forgotten 100 s after the relearning.

The human brain has been adept at learning from the surroundings and using logical thinking to predict future events. Learning capacity may be signified by interesting phenomena such as associative learning, which is often based on the STDP learning rule. Associative learning is related to cognitive behaviors and logical thinking, which may be well demonstrated by the famous classical conditioning of Pavlov’s dog experiment. Mathews and coworkers implemented this classic conditioning on a single MoS₂ thin-film synaptic transistor. Optical pulses were used as food, i.e., unconditioned stimuli (UCS). Electrical pulses were used as bell ringing, i.e., conditioned stimuli (CS). Current of the device above the threshold was used as salivation. Before training, only food could activate salivation (unconditioned response), whereas bell ringing alone did not lead to any salivation, as shown in Figure 9d. After training the dog with a few cycles of paired bell ringing and food stimuli, the bell ringing was associated with food. This caused the current of the device to be higher than the salivation threshold. The trained dog then salivated when only hearing the bell ringing (conditioned response). Hence, the classical conditioning was well simulated.

Aversion learning also belongs to the associative learning. Yin et al. have recently mimicked aversion learning using a synaptic SINC phototransistor, as shown in Figure 9e. A patient was initially craving for alcohol. The drinking of alcohol led to an excited brain activity. To treat the alcoholism, the patient was required to repeatedly drink alcohol and promptly expelled it via emetine-induced emesis. Before the treatment, an electrical spike gave rise to an EPSC, signifying the craving for alcohol. During the treatment, the optical spikes were repeatedly applied

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**Figure 10.** a) Schematic of two intercoupled synaptic devices with a symmetric configuration. Histograms of the values for the logic functions of b) AND and OR, and c) NAND and NOR. The threshold value is indicated by dashed lines. Reproduced with permission. Copyright 2018, Elsevier Ltd. d) Schematic operation diagram for the “AND” and “OR” gates. Histograms of the values for the logic functions of e) AND and f) OR. The threshold value is indicated by dashed lines. Reproduced with permission. Copyright 2019, Elsevier Ltd. g) Synaptic weight as a function of presynaptic spike frequencies. The red solid line is the fitting model. h) Schematic of MoS₂ synaptic devices as both high-pass and low-pass photonic filters. i) Magnitude of the gain as a function of presynaptic spike frequencies. The gain was defined as \(|A_{10} - A_1|/A_1|\) corresponds to the frequency. A₁₀ and A₁ are EPSC amplitudes of the last and first spikes, respectively. The inset in panel (i) is the schematic of a low-pass filter in biological synapses. Reproduced with permission. Copyright 2019, The Royal Society of Chemistry.
on the device immediately after the electrical spikes, simulating the taking of emetine after the drinking of alcohol. As the treatment proceeded, the PSC of the device decreased, leading to a transition from craving to aversion for alcohol. After the treatment, the PSC of the device remained at a rather low level (aversion state) even if a series of electrical spikes were used in the end.

4.2. Logic Functions

A biological brain is capable of performing complex computations in a parallel or hierarchical manner with the help of the neural network. As the basic elements of the computation, logic functions are desired for synaptic devices. For optoelectronic synaptic devices, several logical functions such as AND, NAND, OR, and NOR have been emulated with optical or electrical stimuli. Precise time- and information-encoded inputs are typically needed when realizing the logical functions. Zhao et al. realized the logical functions by connecting two EL synaptic devices (Figure 10a). The electrical spikes on the two devices were the inputs. When realizing the AND and OR gates (NAND and NOR gates), the total optical power (\(L_{\text{out}}\)) (total resistance \(R_{\text{out}}\)) of the devices was defined as the output with a threshold value. The output signal was defined as “1” when its value was higher than the threshold. Otherwise, the output signal was defined as “0.” With designed electrical inputs, the synaptic devices could enable the logical functions of AND/OR (Figure 10b) and NAND/NOR (Figure 10c). The logic functions of AND and OR were also realized using a synaptic SiNC phototransistor. Optical spikes denoted by \(L_{1\text{in}}\) and \(L_{2\text{in}}\) were regarded as the input signals, whereas the amplitude of the IPSC \((E_{\text{PSC}})\) of the device was used as the output signal. Electrical spikes on the gate were used to switch the logical functions between AND and OR, as schematically shown in Figure 10d. For the AND operation, the electrical spikes with a positive voltage were synchronously applied with the optical spikes. In this case, \(E_{\text{PSC}}\) was larger than the threshold value only when \(L_{1\text{in}}\) and \(L_{2\text{in}}\) were simultaneously used (Figure 10e). For the OR operation, the electrical spikes with a negative voltage were used together with the optical spikes. Now \(E_{\text{PSC}}\) was larger than the threshold value when either \(L_{1\text{in}}\) or \(L_{2\text{in}}\) was used (Figure 10f). The realization of logical functions is based on the difference between the output and threshold current or resistances. It is crucial for devices having a clear and well-controlled distinction of the output and threshold conductance.

4.3. Filtering

In biological neural systems, synaptic filtering can impact information transmission from via a synapse in a positive way. Signals with certain frequency range are passed, whereas the others are smoothed to a lower level. The filtering function of synaptic devices is closely related to the nonlinearity of conductance change. For high-pass (low-pass) filtering, the synaptic devices show clear conductance change only stimulated by high- (low-) frequency signals. The emulation of high-pass filtering and low-pass filtering has been demonstrated using optoelectronic synaptic devices. MoS\(_2\)-based synaptic phototransistor to realize the synaptic filtering. For this device, high-frequency optical inputs (20–50 Hz) led to synaptic potentiation, whereas low-frequency optical inputs (2–17 Hz) gave rise to synaptic depression. The absolute value of the change of the current was defined as the synaptic weight. As shown in Figure 10g, the synaptic weight decreased (increased) with the increase in the stimulus frequency in the depression (potentiation) region. Hence, the low-pass (high-pass) filtering could be realized in the depression (potentiation) region (Figure 10h). In addition, Shao et al. demonstrated the low-pass filtering with a synaptic transistor based on CNTs. The device exhibited higher gain when stimulated by optical pulses with lower frequency (Figure 10i).

4.4. Neuromorphic Visual Systems

It has been proved that humans receive nearly 80% of information from the external environment through the sense of vision. The visual pathway is composed of the retina circuitry, optic nerves, and visual cortex. The retina circuitry first senses the optical signals and triggers neural events. The optical information then passes through the optic nerves and reaches the visual cortex for visual recognition and perception. To emulate the human vision, artificial visual systems should integrate detecting, memorizing, and processing. Conventional artificial visual systems are integrated with image sensors, memory, and processing units. The systems normally have the issue of complex circuitry with high energy consumption. To address this issue, optoelectronic synaptic devices have been recently used to build neuromorphic visual systems. To build a neuromorphic visual system, synaptic devices are expected to have broadband response, high photosensitivity and intensity adaptation and follow learning rules.

The retina is a light-sensitive layer with the functions of light detection, adaptation, and image preprocessing. The retina is composed of photoreceptor cells, bipolar cells, and ganglion cells. The photoreceptor cells sense light and translate the light signal into the changes of the neuron membrane potential. The potential changes are then transmitted into the ganglion cells through the bipolar cells and finally work on the visual cortex in the brain. Synapses are the connections between these neurons. Chen et al. designed a hybrid optoelectronic device based on bulk Si and a HfO\(_2\) film. This device realized STDP, indicating its potential for the simulation of a retina. Image preprocessing such as contrast enhancement in a retina could improve the accuracy and quality of transmitted information. Zhou et al. realized contrast enhancement of images using synaptic devices with the structure of Pd/MoO\(_3\)/ITO. The 3 × 5 device array was repeatedly trained with a four-grayscale input image (image (1) of Figure 11a) for 100 times. The input signals were optical signals and the four grayscales of input images corresponded to four different light intensities (right panel of Figure 11a). The output image (image (2) of Figure 11a) was based on the output current of the array. After training, the differences in output currents according to the different light intensities are enlarged. This resulted in an output image with contrast higher than the input image. The device also showed faster decay when stimulated by optical signals with lower light intensity (right panel of...
Figure 11a). After 1 min, the output currents corresponding to a lower light intensity decay faster than those corresponding to a higher light intensity. Thus, the contrast of the output image was further enhanced (image (3) of Figure 11a). In this way, the device array can effectively extract the main feature, and it can be applied at the front of a neural network to improve the subsequent processing tasks.

Image memory is an essential intermediate between perception and thinking. Optoelectronic synaptic devices can also be used as the image memory of neuromorphic visual systems. Dai et al.\cite{102} achieved image memory using a T-shaped synaptic device array (Figure 11b). The array was stimulated 20 times with an input optical image encoded by different light intensities, resulting in a well-recognized current contrast image. The times of the optical stimuli represented the learning times. After the learning process, the array exhibited a dynamic forgetting process. The output image was longlasting with memory time over 30 s, demonstrating the potential of optoelectronic synaptic devices as the image memory devices.

As an application of the neuromorphic visual systems, pattern recognition may be executed using ANNs. Synaptic devices are commonly used to form a crossbar array of synaptic weight layer in ANNs. Although the hardware implementation of the ANNs using optoelectronic synaptic devices requires further investigation, the pattern recognition can be simulated with algorithms nowadays.\cite{113-115} The feasibility of pattern recognition is usually validated using the Modified National Institute of Standards and Technology (MNIST) handwritten digit images. The MNIST handwritten digit images with a resolution of 28 × 28 pixels are widely used to train ANNs. The acquired recognition rate (recognition accuracy) is used to evaluate the learning capability of ANNs. Ham et al.\cite{55} demonstrated a single-layer ANN based on synaptic devices with the structure of Ag/perovskite/ITO for the MNIST pattern recognition. As shown in Figure 11c, the ANN consisted of 784 input neurons and ten output neurons with 7840 synaptic weights. The ten output neurons corresponded to numbers from "0" to "9." As the training process (i.e., learning process) proceeded, the synaptic weights were adjusted and updated according to a learning algorithm based on the long-term potentiation and long-term depression of the synaptic devices. In this case, the ANN was exposed to light, a rapid recognition accuracy of 82.7% was achieved after 2000 learning phases (Figure 11d). Compared with the case without light exposure, the light-assisted ANN could achieve a similar accuracy at a lower power.

5. Perspectives

All kinds of optoelectronic synaptic devices are shown in Table 1. Various materials such as oxide semiconductor films, 2D-layered semiconductors, organic semiconductor films, CNTs, semiconductor nanocrystals (NCs), and QDs have been used to fabricate optoelectronic synaptic devices.\cite{139-141} Despite the difference in the working mechanism of these devices, they usually adopt the structures of memristors and transistors.\cite{142-144} The wavelength of the optical stimulation has spanned from the UV to NIR, signifying the broadband spectral response of optoelectronic synaptic devices. For the optically emitting synaptic devices, it is worth extending the wavelength beyond 1 μm to better facilitate optical transmission in an integrated system.\cite{145} It is clearly advantageous that synaptic devices could accept both optical and electrical stimulations. The synergy of optical and electrical stimulations leads to added freedom for the simulation of synaptic
Table 1. Compilation of optoelectronic synaptic devices.

| Typea) | Active materials | Structure | Dimension (D or W × L) | Wavelength [nm] | Optical stimulation | Electrical stimulation | Synaptic functionalities | Energy consumption | Ref. |
|--------|------------------|-----------|------------------------|-----------------|---------------------|-----------------------|------------------------|-------------------|-----|
| **Optically stimulated** | ISGZ, ISG, ISG, ISG | Two terminal | 180 × 70 μm² | 380–385 | ✓ | ✓ | STP/LTP | – | [82] |
| IGZO | Two terminal | 100 μm | 250 | ✓ | ✓ | STP/LTP | – | [90] |
| **Optically stimulated** | Bottom gate | 30 × 100 μm² | 470 | ✓ | ✓ | STP/LTP | ≈0.35 nJ | [101] |
| **Optically stimulated** | ISG lateral gate | 80 × 1600 μm² | 275/365/405 | ✓ | ✓ | STP/LTP/STDP | – | [92] |
| **Optically stimulated** | ISG lateral gate | 80 × 1600 μm² | 365 | ✓ | ✓ | STP/LTP/STDP | ≈0.4 nJ | [116] |
| PEDOT/SnO₂/IGZO | Bottom gate | 500 × 800 μm² | 450 | ✓ | ✓ | STP/LTP | ≈1 nW | [88] |
| IGZO | Two terminal | 10 × 100 μm² | 254 | ✓ | ✓ | STP/LTP | – | [90] |
| **Optically stimulated** | Bottom gate | 20 × 40 μm² | 365 | ✓ | ✓ | STP/LTP | ≈13 pJ | [91] |
| **Optically stimulated** | Two terminal | 100 μm | 310 | ✓ | ✓ | STP/LTP | – | [96] |
| **Optically stimulated** | Multigate | 10 × 20 μm² | 445 | ✓ | ✓ | STP/LTP/STDP | – | [126] |
| **Optically stimulated** | lateral gate | 405 | ✓ | ✓ | STP/LTP | – | [98] |
| **Optically stimulated** | ISG/chitosan Bottom gate | 80 × 1000 μm² | 405 | ✓ | ✓ | STP/LTP | – | [109] |
| **Optically stimulated** | ISG/chitosan Bottom gate | 80 × 1000 μm² | 405 | ✓ | ✓ | STP/LTP | – | [103] |
| **Optically stimulated** | ISG NT | Bottom gate | D = 387 nm | 532 | ✓ | ✓ | STP/LTP/STDP | – | [97] |
| **Optically stimulated** | ISG/alkylatedgraphene oxide | Lateral gate | 20 × 1000 μm² | 520/940/1310 | ✓ | ✓ | LTP | – | [132] |
| **Optically stimulated** | Bottom gate | 10 × 10 μm² | 405 | ✓ | ✓ | STP/LTP | – | [137] |
| **Optically stimulated** | Bottom gate | 10 × 120 μm² | 405 | ✓ | ✓ | STP/LTP/STDP | – | [14] |
| **Optically stimulated** | Bottom gate | 30 × 90 μm² | 405/532 | ✓ | ✓ | STP/LTP/STDP | ≈5 nJ | [58] |
| **Optically stimulated** | Bottom gate | D = 387 nm | 532 | ✓ | ✓ | STP/LTP/STDP | – | [112] |
| **Optically stimulated** | Bottom gate | 50 × 1000 μm² | 365/520/660 | ✓ | ✓ | STP/LTP/STDP | – | [138] |
| **Optically stimulated** | Bottom gate | 10 × 120 μm² | 405 | ✓ | ✓ | STP/LTP/STDP | – | [113] |
| **Optically stimulated** | Bottom gate | 20 × 40 μm² | 365 | ✓ | ✓ | STP/LTP | – | [111] |
| **Optically stimulated** | Bottom gate | 30 μm × 1 mm | 500 | ✓ | ✓ | STP/LTP | – | [111] |
| **Optically stimulated** | Bottom gate | 2 × 5.3 μm² | 532 | ✓ | ✓ | STP/LTP | – | [114] |
| **Optically stimulated** | Bottom gate | 50 × 1000 μm² | 365 | ✓ | ✓ | STP/LTP/STDP | – | [117] |
| **Optically stimulated** | Bottom gate | 50 × 1000 μm² | 365 | ✓ | ✓ | STP/LTP | – | [117] |
| **Optically stimulated** | Bottom gate | 20 × 20 μm² | 405 | ✓ | ✓ | STP/LTP/STDP | – | [117] |
| **Optically stimulated** | Bottom gate | 100 μm | 310 | ✓ | ✓ | STP/LTP | – | [97] |
| **Optically stimulated** | Bottom gate | 250 μm | 365 | ✓ | ✓ | STP/LTP | – | [117] |
| **Optically stimulated** | Bottom gate | 300 × 1 mm | 500 | ✓ | ✓ | STP/LTP/STDP | – | [117] |
| **Optically stimulated** | Bottom gate | 405/532 | ✓ | ✓ | STP/LTP | – | [117] |
| **Optically stimulated** | Bottom gate | 50 × 1000 μm² | 365 | ✓ | ✓ | STP/LTP/STDP | – | [117] |
| **Optically stimulated** | Bottom gate | 20 × 20 μm² | 405 | ✓ | ✓ | STP/LTP | – | [117] |
| **Optically stimulated** | Bottom gate | 100 μm | 310 | ✓ | ✓ | STP/LTP | – | [97] |
| **Optically stimulated** | Bottom gate | 250 μm | 365 | ✓ | ✓ | STP/LTP | – | [117] |
| **Optically assisted** | Bottom gate | 300 × 1 mm | 500 | ✓ | ✓ | STP/LTP | – | [117] |
| **Optically assisted** | Bottom gate | 405/532 | ✓ | ✓ | STP/LTP | – | [117] |

a)Dimensions: diameter (D) or width × length (W × L). Energy consumption is estimated using Equation (4).

5.1. Energy Consumption

Synaptic devices should be integrated to form a synapse unit, which is coupled with an axon unit and a dendrite unit in an ANN for neuromorphic computing. While the energy consumption of the ANN is not solely dependent on...
the synapse unit, the energy consumption of synaptic devices plays an important role in the realization of neuromorphic computing with low energy consumption.\textsuperscript{[148]}

For optoelectronic synaptic devices, different methods have been used to calculate energy consumption.\textsuperscript{[149]} In the first method, researchers use the formula of

\[
dE = S \times P \times dt
\]

where \( S \) is the area of the device, \( P \) is the power density of the optical spike with the duration of \( t \).\textsuperscript{[95,113]} This means that the energy consumption is the energy of the optical spike. The second method relies on

\[
dE = V \times I \times dt
\]

where \( V \) (I) is the voltage (current) of the device, \( t \) is the duration of the optical spike.\textsuperscript{[109,112]} In this case, the energy consumption is the energy of the electrical response of an optoelectronic synaptic device. Alternatively, Equation (3) and (4) is combined to calculate the energy consumption. It seems more reasonable to take the energy of the electrical response as the energy consumption and use the energy of the optical spike to evaluate the sensitivity of the optoelectronic synaptic device.\textsuperscript{[149]}

Up to now, few optoelectronic synaptic devices consume extremely low energy, which should be comparable with the energy consumed by a biological synapse (\( \approx 10^{-15} \text{fJ} \)). It has been shown that the reduction of the device size and the decrease in the duration of the optical spike are effective means to lower the energy consumption.\textsuperscript{[60,112,113]} Moreover, novel device structures with photovoltaic effect can be used to enable optoelectronic synaptic devices to work without electrical power supply.\textsuperscript{[150]} This may be the ultimate goal of optically stimulated synaptic devices in the context of minimizing the energy consumption.

5.2. Variation

There is no standard evaluation on the variation of optoelectronic synaptic devices so far. This may be largely due to the fact that optoelectronic synaptic devices have not been well incorporated into ANNs. For traditional optoelectronic devices, spatial (device-to-device) and temporal (cycle-to-cycle) uniformities are critical to their applications. In principle, however, the uniformity of optoelectronic synaptic devices is not crucial as neuromorphic computing has certain tolerance on variation at the level of an integrated system.\textsuperscript{[151]} Researches on the robustness of different electronic neural networks showed that a task could be successfully conducted even with a relatively obvious variation (30%) in a neuromorphic system.\textsuperscript{[152,153]}

Nevertheless, cycle-to-cycle tests with up to thousands of cycles for optoelectronic synaptic devices have demonstrated relatively small temporal variation (e.g., 4.4%).\textsuperscript{[54,55,102]} Device-to-device tests with up to tens of devices have shown that the spatial variation of optoelectronic synaptic devices may also be not very large (e.g., 8.8%).\textsuperscript{[54,117]} These previous tests have signified the controllability of the variation of optoelectronic synaptic devices, which renders freedom during the incorporation of optoelectronic synaptic devices into ANNs.

5.3. Exact Simulation

A synapse is either excitatory or inhibitory in a biological neural network.\textsuperscript{[154]} If one wants to exactly mimic a biological neural network for the fabrication of neuromorphic computers, he/she needs to separately make excitatory synaptic devices and inhibitory synaptic devices. In the point of view of compact integration, however, it is advantageous for a synaptic device to have both EPSC and IPSC. This also helps the facile realization of other important synaptic functionalities such as STDP.

In addition, we know that LTP refers to the consolidated connection of a synapse in a biological neural network. During the process of the consolidation, the LTP is signified by the significant increase in PSC. After the consolidation process, the PSC that is either excitatory or inhibitory actually disappears. However, the consolidation-induced physical changes (e.g., AMPA receptors are incorporated into the synaptic cleft) of the synapse remain.\textsuperscript{[154]} It is these physical changes that are responsible for the LTM in a biological neural system. Therefore, we need to consider how to introduce physical changes in a synaptic device under optical stimulation if we want to obtain the LTP by exactly mimicking a biological neural system. However, we notice that researchers are more actively working toward neuromorphic computing by combining what has been known for a biological neural network with the technologies of current mainstream computing based on CPUs and memory. Hence, concerns such as the retention time of the LTP may arise in the point of view of memory devices. The retention time of the LTP now ranges from a few seconds to a few hours and even longer in the literature.\textsuperscript{[55,61,99,126]} It is worth mentioning that requirements of the retention of synaptic devices depend on the applications.\textsuperscript{[15]} For neural networks that are trained online, the weights in such networks are updated rapidly and the retention performance of synaptic devices is less stringent. On the contrary, the offline training neural networks require good long-term retention because the longstanding transformation in synaptic connection is critical for memory and learning in such neural networks. Hence, optoelectronic synaptic devices with short-term retention\textsuperscript{[53,113]} and those with long-term retention\textsuperscript{[60,117]} are both useful for ANNs.

6. Conclusions

In this Review, we have introduced the state-of-the-art optoelectronic synaptic devices. A series of important biological synaptic functions such as STP, LTP, and STDP have been simulated using optoelectronic synaptic devices. Various materials and related hybrid structures have been used to fabricate optoelectronic devices. It has been clear that materials science and engineering are critical to the realization of high-performance optoelectronic synaptic devices. On one hand, conventional technologically important materials such as Si and oxide semiconductors need to be explored for their new application in optoelectronic synaptic devices. On the other hand, emerging materials such as 2D-layered materials and perovskite deserve investigation, enabling their excellent optical and electrical properties to be exploited for optoelectronic synaptic devices.
As optoelectronic synaptic devices are eventually used in ANNs, their fabrication requires to facilitate optoelectronic integration. In the current early stage of the development of optoelectronic synaptic devices, the device size is often rather large. This is obviously not good for the integration of optoelectronic devices. Hence, nanofabrication should be more seriously used for optoelectronic synaptic devices in the future.

In addition, we notice that great experiences on optoelectronic integration have been gained in other fields such as Si-based optoelectronic integration for the computing based on the von Neuman architecture.\[146\] By taking advantage of these experiences in the device design, researchers should effectively improve photoelectric conversion and the transmission of optical and electrical signals in ANNs based on optoelectronic synaptic devices. In such a context, Si-based optoelectronic synaptic devices may hold great promise.\[157\] Meanwhile, synaptic devices should form a synaptic unit, which is coupled with an axon unit and a dendrite unit to form an ANN for neuromorphic computing. The pursuit of large-scale deployment of neuromorphic computing clearly depends on the balanced experiences in the device design, researchers should effectively improve photoelectric conversion and the transmission of optical and electrical signals in ANNs based on optoelectronic synaptic devices. In such a context, Si-based optoelectronic synaptic devices may hold great promise.\[157\] Meanwhile, synaptic devices should form a synaptic unit, which is coupled with an axon unit and a dendrite unit to form an ANN for neuromorphic computing. The pursuit of large-scale deployment of neuromorphic computing clearly depends on the balanced development of each unit. Up to now synaptic devices have been more extensively studied than devices in the axon and dendrite units, which desire more research efforts in the future.

When the response of optoelectronic synaptic devices to external stimulation is deemed as sensing, these devices are actually able to integrate sensing, memory, and computing. This is highly desired by the burgeoning artificial intelligence (AI) and Internet of Things (IoTs). Therefore, research on optoelectronic synaptic devices is quite exciting in the intelligent era. Continuous efforts on the development of optoelectronic synaptic devices should significantly contribute to the large-scale deployment of neuromorphic computing in the future.

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Conflict of Interest

The authors declare no conflict of interest.

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