Emerging Optical In-Memory Computing Sensor Synapses Based on Low-Dimensional Nanomaterials for Neuromorphic Networks

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1. Introduction

Current digital computers are expected to operate more like the human brain and emulate bionic abilities like massive parallelism, self-learning behavior, self-cognition progress, and self-adaptation operation. Exploring the emerging hardware devices or systems that can incorporate storage and computing has developed the “in-memory computing (IMC)” techniques.[1–5] The IMC techniques can be divided into two main kinds: off-chip IMC and in-chip IMC techniques.[1–7] The off-chip IMC technique focuses on hardware configuration, which can enhance the read and write rate by implanting the calculator chip or the logic programming unit into the memory,[1–4] while the in-chip IMC technique attaches more attention to the software optimization of the memory. The optimized algorithm with the weight update function is inserted into the memory and makes the memory algorithmic, which allows the computer to mimic the human brain more closely in both function and form.

1.1. Artificial IMC Synapses

Due to neurons (≈10^{11}) and synapses (≈10^{15}), the human brain can handle massive information in ultrashort time with extremely low energy consumption.[8–14] It is critical and meaningful to investigate the imitation of biological synapses by emerging electronic devices with the IMC technique due to the requirement of lower energy consumption, higher data storage density, and faster operating speed.[8,10,15–20] The biomimetic device with similar advantages has been named an “artificial synaptic device.” The input information induced by consecutive...
stimuli is stored in the functional layers of these devices as a form of electric signal and processed by continuous synaptic weight updates, which indicates that it is feasible and promising to explore allowing the computing capability to electronic devices with the long-term memorizing behaviors by IMC techniques. Current IMC synaptic devices (three-terminal transistor, two-terminal memristor, and atomic switch) represented by the popular nonvolatile memory (NVM) are significantly affected by the purely electronic control, which has an important influence on electrical input/output, delay of resistance–capacitance (RC) interconnection, and circuit crosstalk.\[8–11,19,21–27]\]

1.2. Artificial Optical In-Memory Computing Sensor Synapses

Biologically, the normal life of human beings cannot be separated from vision, and the content of vision depends on the light reflected on the surface of things. Evidence has suggested that humans receive the most external information through the visual perception system.\[28–30]\] The investigation of optogenetics of biological neuroscience also promoted the integration of light and synaptic devices, which resulted in optical synapses.\[8–10,31]\] Compared with IMC synaptic devices that only rely on electrical control, optical synapses could realize the interconversion between optical and electrical signals, which was positively influenced by the integration of the optical neuron network system.\[31–34]\] Besides, the external optical stimuli for the optical synapses could emulate the light hit into the human eyes, which could develop the concept of “in-memory computing sensor (IMCS).” IMCS synaptic devices can be considered as evolutionary IMC devices, and IMCS devices rely more on the control of bionic stimuli (mostly optical stimuli for now).\[1–4,35]\] Compared with IMC devices, IMCS devices can demonstrate a more comprehensive biomimetic significance, which indicates a perfect integration of bionic perception, processing, and memory.\[1–3,5,8,14,32,33]\] Optical IMCS synapses also exhibited advantages with low circuit crosstalk, high operation speed, large bandwidth, and ignorable RC delay. In addition, optical IMCS synapses revealed the utilization potential in the application of signal processing, image recognition, and visual sensory.\[8,14,32,33]\] These properties more closely mimic the information exchange between the human retina and synapses. Therefore, it is inevitable to investigate the functional materials utilized in the devices for exploring the potential of optical IMCS synapses. For now, a wide variety of materials have been investigated with their application potential on optical IMCS synapses, including perovskites, metal oxides, organics, and low-dimensional nanomaterials.\[19,21,36]\] Perovskites have shown high electro-optic conversion efficiency, metal oxides have exhibited high carrier mobility and excellent transparency, while organics also demonstrated advantages like low-temperature manufacturing and degradability.\[19,21,36]\] These properties have made these conventional materials become popular candidates during the research of artificial optical synapses. Compared with traditional materials, emerging low-dimensional nanomaterials such as CsPbBr\(_3\) quantum dot (QD)\[31\] InAs nanowire (NW)\[37\] and molybdenum disulfide (MoS\(_2\)) nanosheet\[14\] have received incredible growing interest due to their unique advantages like nonlinear optical character, high optical-sensitivity efficiency, and wide bandgap in the field of photovoltaic conversion.\[8–11,19,21,36]\] Figure 1 exhibits significant milestones highlighted in the historical evolution of optical IMCS synapses with low-dimensional materials.\[38–50]\] Besides, other properties including ease of preparation, low leakage current, and feasibility of bandgap modulation also make low-dimensional nanomaterials attractive and popular to researchers of microelectronic devices.\[9,10]\] For now, some researchers like Kim,\[10\] Pi,\[9\] Zhao,\[8\] and Pan\[51\] have provided their overall

![Figure 1](https://www.advancedsciencenews.com/)

**Figure 1.** Significant milestones are highlighted in the historical evolution of optical IMCS synapses with low-dimensional materials.
2. Biological and Biomimetic Characteristics of Neuron Synapses

The primary purpose of exploration in biomimetic synaptic devices is to emulate the synaptic function of biological synapses. Therefore, it is necessary to provide a detailed discussion on the connection between biological synapses and biomimetic synaptic devices. In this section, we focus on the characteristics of biological synapses and biomimetic synaptic devices first. Then related reports about optical IMCS synapses based on low-dimensional materials are included.

2.1. Biological Synapse

In the biological neuron system, two primary transmission modalities, electrical and chemical, of synapses have been considered to provide the physiological basis to neural activities, which derived electrical synapses (the left part of Figure 2a) and chemical synapses (the right part of Figure 2a). For the electrical synapse, the transmission of nerve signals is realized in the form of electrical transmission, and the transmission channel is a low-resistance gap junction between pre- and postsynaptic membranes. Electrical synapses are mainly found in the nervous tissues of invertebrates and lower vertebrates. Compared with the electrical synapse, the chemical synapse relies on the terminal of the presynaptic neuron to release special chemicals (neurotransmitters) as media for transmitting information to affect the postsynaptic neuron, which is closely related to the high-level neural activities, such as memorizing and learning.

2.2. Biomimetic Synaptic Behaviors of the Synaptic Device

At present, most of the research on biomimetic synaptic devices focuses on the imitation of chemical synapses. The function variation of the biological chemical synapse is determined by the synaptic weight, which is the fundamental presence of neural connection strength. For the biomimetic synaptic device, this synaptic weight can be obtained with specific resistance or conductance, and the change of this synaptic weight indicates the synaptic plasticity. When the external spiking was applied upon the synaptic device, the biomimetic synaptic performance is always evaluated by the change of the postsynaptic current (PSC), which may be inhibitory (IPSC) or excitatory (EPSC). With various modulation techniques of the PSC, different characteristics of the synaptic plasticity can be obtained, including classic STP, LTP, and STDP. As illustrated in Figure 2b–g, some popular and emerging forms of synaptic plasticity have been demonstrated, including STP, LTP, STDP, SNPD, and SRDP. Biological synaptic plasticity is responsible for neural activities like memorizing, learning, recognition, and logical thinking. Therefore, it is significant to investigate the synaptic plasticity of biomimetic synaptic devices.

STP and LTP have been accepted as the typical response of external stimuli. STP is associated with the short-term response and recognition of external information, which can transit to LTP after repeated rehearsal and training processes. Based on the fundamental behaviors of STP, LTP is related to long-term learning and memorizing. In general, STP behaviors are always reflected by paired-pulse facilitation (PPF) and paired-pulse depression (PPD). PPF or PPD is demonstrated by the response of the synaptic device to two consecutive external stimuli. After the first stimuli, the synaptic device may exhibit an increased (decreased) current to respond to the second stimuli, which results in PPF (PPD). The PPF index is always determined by the current ratio of EPSC or IPSC. The relationship between the PPF/PPD index and the time interval (Δt) of two consecutive stimuli is always used to evaluate STP. With the increment of Δt, the PPF index always decays following the equation

\[ \text{PPF index} = C_1 \cdot \exp \left( -\frac{\Delta t}{\tau_f} \right) + C_2 \cdot \exp \left( -\frac{\Delta t}{\tau_r} \right) \]
was associated with the long-term escape of photo-derived electrons in the 2D polymer layer. In addition, they also utilized the PPF index to evaluate the PPF characteristic. When the time interval between the first and second photonic pulse was 50 ms, the maximum value of the PPF index was being obtained with 1.63.\[59\] For LTP, the long-term potentiation and depression are always under investigation, which corresponds to the enhancement and inhibition of the connection strength between two adjacent neurons.\[9,10,14\] As illustrated in Figure 2d, Crawford et al. fabricated a photonic synaptic device based on organic–inorganic halide perovskite QDs (PQDs) and multiwall carbon nanotube (MWCNT), which demonstrated the long-term potentiation under the 405 nm all-optical stimuli and the long-term depression with the consecutive electrical pulses.\[63\] After that, they weakened the connection strength with higher efficiency by increasing the pulse width of electrical pulses, which indicated that it was feasible to adjust the learning process of neuromorphic computing by modulating various parameters of pulses.\[63\]

For STP and LTP measurement of a single biomimetic synaptic device, the external stimuli are usually applied onto one terminal (the presynaptic terminal or the postsynaptic terminal).\[8,9,28\] However, it is more meaningful to investigate the interaction relationship between pre- and postneurons in the artificial neuromorphic system, which indicates the significance of research on STDP, SRDP, and SNDP.\[59–62\]

Based on the Hebbian learning rule, STDP is a dependency relationship between time interval (\(\Delta t\)) of pre-/postsynaptic spikes and synaptic weight change (\(\Delta w\)) induced by chronological difference of pre-/postsynaptic spikes.\[8,9,28\]

Different chronological orders of pre-/postsynaptic spikes result in the potentiation or depression of the synaptic weight, which indicates symmetric...
and asymmetric STDP. Asymmetric STDP based on a competitive characteristic of the Hebbian learning rule can result in sequence learning and influence future events. At the same time, symmetric STDP is usually related to associative learning and enhances the synaptic weight during the learning process. Unlike STDP, SRDP is presented by modulation based on various firing frequencies of pulses. The potentiation and depression responses of SRDP are triggered by high and low firing frequency, respectively. SNDP is also under exploration, which is associated with the modulation method based on spiking numbers. As illustrated in Figure 2e, a memristive synaptic device with the structure of reduced graphene oxide (RGO)/GO and N-doped carbon QD (GO-NCQD)/graphene was proposed by Liu et al. This device demonstrated the STDP performance with the UV light and electrical spiking. When $\Delta t < 0$, the presynaptic spiking arrived earlier than the postsynaptic spiking, the long-term potentiation performance was obtained. Reversely, the long-term depression performance was observed when $\Delta t > 0$. In addition, Liu et al. found that EPSC of RGO/GO-NCQD/graphene synaptic device increased with higher spiking frequency, which verified SRDP characteristic in Figure 2f. The synaptic learning behavior based on SRDP demonstrated the experience-dependent property. The same spiking sequence could trigger either depression or potentiation of the synaptic weight, which was determined by the stimuli history. Figure 2g shows SNDP performance of a synaptic device fabricated with low-dimensional SnO$_2$ nanoparticles (NPs) and CsPbCl$_3$ perovskite, which was proposed by Lin et al. With the increase of photonic spiking number, the device exhibited larger PSC under each photonic stimuli with fixed amplitude.

3. Categories of Optical IMCS Synapses

For now, most biomimetic synaptic devices have been proved to be quite dependent on the stimulation induced by external electrical impulses. In contrast, the research on optical IMCS synapses is more in line with the demand for bionic devices. In general, categories of optical IMCS synapses are associated with device structure, device function, and utilized materials. In this section, the primary effort of the investigation is put onto optical IMCS synapses based on low-dimensional materials, and a detailed discussion on the operation mechanisms of these optical IMCS synapses will be presented in the next section.

3.1. Structure and Function Categories

As illustrated in Figure 3, based on the structure variation, optical IMCS synapses can be divided into two- and three-terminal devices, which take vertical-structure memory and planar-structure transistors as typical representatives, respectively. Advantages like the easy fabrication process of planar-structure devices and excellent scalability of vertical-structure devices indicated the developing tendency of integration with high density and power consumption with low energy. Apart from the structure category, based on the various functions of optical stimuli, optical IMCS synapses can be divided into all-optical-stimuli, optical-assisted, and optical-output devices. All-optical-stimuli IMCS synapses can operate with only light input and mimic artificial synaptic behaviors with the modulation of all-optical stimulations. Optical-assisted IMCS synapses always work with the effect of both external optical stimuli and electrical stimuli, while optical-output IMCS synapses are associated with the conversion between the electrical signal (input) and the optical signal (output).}

3.2. Materials Categories

As illustrated in Figure 3, various materials have been investigated in the field of optical IMCS synapses, including metal oxides (MO), perovskites, organics, and low-dimensional (0D, 1D, and 2D) materials. As listed in Table 1, various optical IMCS synapses fabricated with different materials are demonstrated with their related performance. Compared with pure electrical synaptic devices, the sensitivity of the devices to light and electricity should be considered at the same time when studying optical IMCS synapses. Therefore, the materials used in a single optical IMCS synapse are often not single. High carrier mobility, excellent transparency, and light-induced ionization of oxygen vacancy indicated the feasibility of MO materials as functional layers. Easy low-temperature preparation, simple solution-processed preparation, and diversity of hybridization also make perovskite, organic, and low-dimensional (0D, 1D, and 2D) materials more popular. In addition, low-dimensional materials always play the role of enhancing and improving the performance of optical IMCS synapses. In this section, optical IMCS synapses fabricated with various low-dimensional materials will be discussed first. Some published work has demonstrated that the device function could be driven by only one
kind of low-dimensional material (0D PQD, or 1D CNT or 2D graphene). Some other work also indicated the excellent or enhanced electrical and artificial synaptic performance of devices fabricated with hybrid low-dimensional materials, which included hybrid groups like “polymer QD + graphene” and “PQD + CNT.” In most cases, these low-dimensional materials are always utilized to fabricate functional layers of optical IMCS synapses, including dielectric layers and semiconductor layers. The functions of two-terminal optical IMCS synapses are always influenced by the dielectric layers, while the dielectric and semiconductor layers always influence the three-terminal optical IMCS synapses simultaneously.

### 3.3. Optical IMCS Synapses with 0D Materials

0D materials consist of a few particles (atoms or molecules) stacked together and the size of the particles is on the order of nanometers. It is very significant for particles of such small size to observe various quantum effects (quantum confinement effect, quantum tunneling effect, and quantum interference effect). The common 0D materials are NP, QD, ultrathin powder, atomic cluster, and nanocluster. Pi et al. also proposed an optical IMCS synapse with a near-infrared QD light-emitting diode (NIR QLED) incorporated with the poly(3-hexylthiophene) (P3HT) and P3HT was used as the hole transport layer (HTL) due to its high hole mobility of $1.0 \times 10^{-3}$ cm$^2$V$^{-1}$s$^{-1}$. Besides, due to the nonabsorbing behavior of NIR light, it was significant for P3HT to balance the injection process of carriers instead of deterioration of light extraction. With the solution-processed spin-coating process, device 1 was obtained with the structure of Ag/ZnO/Si-QD/P3HT/PEDOT:PSS/ITO firs. The electron-transport layer was the ZnO layer. Holes from the highest occupied molecular orbital (HOMO) of PEDOT/P3HT layers and electrons from the ZnO layer were injected into the Si-QD layer and resulted in electroluminescence (EL) behavior due to the radiative combination. The device performance was influenced by the thickness of the P3HT layer, which was associated with the concentration of the P3HT precursor solution. They obtained the best performance of device 1 with the P3HT concentration of 15 mg mL$^{-1}$. To improve the performance of the device, they inserted a PFN (a conjugated polymer containing amine group polyfluorene) layer as an interlayer between the P3HT and Si-QD layers of device 2 to weaken the leakage of electrons from the Si-QD layer to the anode Ag layer. Compared with the lowest unoccupied molecular orbital (LUMO) of P3HT, the higher LUMO was observed in PFN, which had a more positive influence in preventing the leakage of electrons from the Si-QD layer. This device showed the EL synaptic behaviors with the spiking of electrical pulses, the NIR light was emitted by the electrical stimuli, and the decreasing tendency of the optical output power was observed in the range from 1 to 10$^{5}$ ms. Then two continuous electrical pulses triggered the typical PPF behavior, which indicated the STP performance of this enhanced synaptic device.

| Materials | Materials category | Thickness of active layer [nm] | Optical stimuli intensity [μW cm$^{-2}$] | Electrical stimuli intensity [V] | Minimum power consumption [μW] | Ref. |
|-----------|--------------------|-------------------------------|------------------------------------------|---------------------------------|---------------------------------|-----|
| ZTO       | MO                 | ≈5                            | 10                                       | 10                              | 2.5                            | [67]|
| IZO       | MO                 | ≈7                            | 0.6                                       | 10                              | –                              | [55]|
| In$_2$O$_3$/ZnO | MO             | ≈150                          | 0.4-4.0                                     | 2                               | 0.2                            | [68]|
| Al$_2$O$_3$ | MO                 | ≈75                           | 18.23                                     | 1.8-4.2                         | –                              | [69]|
| MAPbI$_3$ | Perovskite         | ≈250                          | 1 μW                                       | 1-4                             | 1.0                           | [70]|
| CuPc/p-6P | Organic            | ≈30                           | 0.1-1.0                                    | 10                              | 0.7                            | [71]|
| PQT-12    | Organic            | ≈10                           | 0.015-0.75                                 | 10                              | –                              | [72]|
| P3HT      | Organic            | –                             | 30 μW                                      | 10                              | –                              | [73]|
| P3HT-MWCNT| 0D/1D              | –                             | 125 μW                                     | –                               | 6.09                            | [63]|
| GO-QCQD   | 0D/2D              | ≈100                          | 20 μW                                     | 2                               | –                              | [60]|
| QLED      | 0D                 | ≈50                           | Output                                     | 4                               | 1.2                            | [74]|
| G-PQD     | 0D/2D              | ≈20                           | 190 nW–1.1 μW cm$^{-2}$                     | 10                              | 36.75                          | [75]|
| CdS/MWCNT | 1D                 | –                             | –                                         | –                               | –                              | [76]|
| SWCNT/HfO$_2$ | 1D                 | ≈50                           | 0.34 μW                                     | 2.5                             | –                              | [77]|
| p(VDF-TrFE)/SWCNT | 1D       | –                             | –                                         | 1                               | –                              | [78]|
| InAs NW   | 1D                 | ≈3                            | <400 μW                                    | –                               | –                              | [37]|
| Graphene/MoS$_2$ | 2D        | ≈40                           | 3.5–11.5 μW cm$^{-2}$                       | –                               | –                              | [79]|
| WSe$_2$   | 2D                 | ≈5                            | $1.25 \times 10^{-3}$ μW cm$^{-2}$         | 4                               | 75                            | [80]|
| IGZO/GO   | 2D                 | –                             | 2000 μW                                    | –                               | –                              | [81]|

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Table 1. Electrical performance of artificial optical IMCS synapses fabricated with various materials.
Han et al. provided their research on optical IMCS synaptic flash memory with the solution-processed CsPbBr$_3$ QDs and the CsPbBr$_3$-QD/semiconductor heterostructure provided the foundation of optical programming and electrical erasing.[31] The 365 nm-wavelength light was employed as an optical pulse (0.153 mW cm$^{-2}$, 30 s) to trigger the programming behavior, while the $-50$ V/1 s electrical pulse was used to induce the erasing operation of the device. As illustrated in Figure 4, the programming and erasing performance were associated with the capture and release of trapped electrons in the CsPbBr$_3$-QD layer. In addition, the Au/pentacene/PMMA (polymethyl methacrylate)/CsPbBr$_3$-QDs/SiO$_2$/Si device showed the high-level EPSC under the optical stimuli and indicated the obvious change of the synaptic weight, which suggested the existence of STP and LTP. During the optical modulation process with various intensities, wavelength, and time intervals, the transformation from volatile memory to nonvolatile memory was obtained. The retention curves after removing consecutive optical pulses were plot and the decay tendency of current followed the Kohlrausch equation, which was described as the “Forgetting Function” in the field of psychology.[83,90-95] The linear performance of the relaxation time with optical stimuli in the plot verified the transition from STP to LTP. The performance of STP and LTP could be observed in Figure 4f,g.[31]

3.4. Optical IMCS Synapses with 1D Materials

1D materials refer to materials with nanometer scale in dimensional direction and macroscopic scales in length, such as NT, nanorod (NR), NW, nanofiber, and nanoribbon; electrons only freely move in the form of straight lines in a nanometer-scale direction.[10,82] Ren et al. reported their discussion on the optical synaptic plasticity of a transistor with InAs NW.[37] The high field-effect mobility of up to 1448 cm$^2$ V$^{-1}$ s$^{-1}$ in the device resulted in a long mean free path of thermal electrons, which make it easier for the trapping process of thermal electrons and these thermal electrons had enough time to arrive traps in the photogating layer (PGL) before the occurrence of the thermalization process. Photoinduced photons with high energy were trapped in the PGL region close to InAs NW. Then the electron density was reduced by the combination process that occurred between electrons and thermal holes, which resulted in the decrease of the leakage current. During their experiment, the device demonstrated no optoelectronic response to optical stimuli with extremely high-power intensity. Therefore, optical stimuli lower than 400 W cm$^{-2}$ were utilized. STP and LTP response were triggered by optical pulse with various power intensity, while the transition from STP to LTP was associated with the increment of optical power intensity, which indicated
the cooperativity of the modulation process for artificial synaptic plasticity. When a 200 W cm$^{-2}$ optical pulse was applied to the device, it took more than 50 s for the synapse weight to slowly recover to a stable state lower than the original current. When the optical pulse intensity was lower than the threshold voltage ($V_{TH}$), although a small number of electrons were blocked, the conduction in the channel is not significantly affected and the STP performance could be triggered. With the continuous increase of optical pulse intensity, the trap barrier blocked more trapped electrons due to the intensity of the optical pulse being higher than the $V_{TH}$, which resulted in the LTP performance.\[37\]

Cui et al. proposed an optical neuromorphic synaptic thin film transistor (TFT) based on a single-walled carbon nanotube (SWCNT).\[77\] The n-doped Si layer acted as an optical-drive gate and the HfO$_x$ layer was used as a semiconductor layer. After depositing the 50 nm HfO$_x$ layer with the atomic layer deposition method, the inks of polymer-sorted SWCNT were printed into the channels. When the external optical stimuli were applied to the device, the photogate effect occurred in the n-doped Si layer trapped photoinduced holes, and then trapped photoinduced holes were released from positions where they were captured. The gradually increasing number of optical pulses caused more photoinduced holes to accumulate in the channel. When the balance between photoinduced holes and released holes was broken, an obvious increment of the drain current could be observed, which resulted in the optoelectronic response of this device. As illustrated in Figure 5b,c,e,f, the drain current could be modulated by the trapped carriers in this SWCNT interface and optical voltage triggered by external optical stimuli, which indicated that the device could exhibit some bionic synaptic functions of neuromorphic devices. After the optical spike from a 940 nm-wavelength pulsed light (0.25 mW cm$^{-2}$), the EPSC abruptly increased to 5.12 nA at first and then decayed to 4.58 nA gradually during the following 100 s, the current signal decreased by

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**Figure 5.** a) Schematic view of the optical synaptic TFT with SWCNT. Photogating effect with the carrier drift of the SWCNT device spiked the first pulse cycle b) in darkness without optical stimuli and c) with optical stimuli. d) SEM image of the SWCNT thin film in the channel. Photogating effect with the carrier drift of the SWCNT device spiked the second pulse cycle e) in darkness without optical stimuli and f) with optical stimuli. g) Attenuation response of the SWCNT device after optical stimuli with different wavelengths. h) SNDP characteristic of the SWCNT device stimulated by optical stimuli with various frequencies. Reproduced with permission.\[77\] Copyright 2019, ACS.
12%. A similar gradual decay was also triggered by the optical stimuli from a 520 nm light (0.25 mW cm\(^{-2}\)) and the current decayed by 22%, which exhibited the LTP performance of this Au/Ti/HfO\(_x\)-SWCNT/Si device. With the external optical stimuli, photoinduced carriers were trapped in interfaces of HfO\(_x\) and SWCNT with a fast velocity, and these trapped carriers were slowly released, which resulted in the gradual decay of EPSC. When researchers increased the power intensity of the optical pulses up to 80 mW cm\(^{-2}\), the amplitude of EPSC increased around 3500 times and then decayed by 90%, which demonstrated that the memory behavior of the printed neuromorphic TFT could be enhanced with the increment of external optical stimuli.[77]

3.5. Optical IMCS Synapses with 2D Materials

Compared with 0D materials like QD and 1D materials like NW, recently, 2D materials have received more interest in the research field of artificial neurons and synaptic devices.[8,9,21] In 2004, Konstantin Novoselov and AndreGeim discovered the famous “graphene” with the single-layer 2D honeycomb lattice structure formed by the close packing of sp\(^2\)-hybrid connected carbon atoms.[19,36] 2D materials, including GO, hexagonal boron nitride (h-BN), transition metal dichalcogenides (TMDC), and black phosphorus (BP), represented by graphene have been widely accepted in various fields like field-effect transistor (FET), diode, optoelectronic detector, catalyst, and TFT.[36] For neuromorphic devices, 2D materials always exist with the forms of thin film or interface (such as nanosheet and nanoflake), and the depth of interface and the thickness of thin film was measured at the nanometer scale. With the high carrier mobility and high electrical conductivity of 2D materials, two-terminal synaptic devices like low-power synaptic RRAM (resistive random access memory) can obtain faster operation speed and lower delay time compared with 0D materials like QD and 1D materials like NW, which was associated with the trapping and detrapping of carriers in 2D materials.[34] In conclusion, three-terminal synaptic devices like synaptic transistors can be enhanced with the integration density and be fabricated with lower economic cost due to the ease of solution-processed preparation and the structure of atomic-level thickness of 2D materials.[21] Wang et al. proposed a mecha-electro-optical artificial synapse comprising a triboelectric nanogenerator (TENG) and a transistor fabricated with 2D heterostructure (graphene (Gr)/MoS\(_2\)).[79] The optoelectronic transistor was driven by the triboelectric potential generated by TENG, which had an influence in modulating synaptic behaviors. The Gr/MoS\(_2\) heterostructure was made up of multilayer MoS\(_2\) and monolayer graphene, which were fabricated by mechanical exfoliation and chemical vapor deposition (CVD) methods, respectively. The triboelectric potential generated by the TENG integrated into the gate terminal acted as the gate voltage of the Gr/MoS\(_2\) transistor. With the 525 nm-wavelength light emitted from the green LED, the drain current (\(I_\text{D}\)) decreased with the enhancement of the optical power intensity when the gate voltage (\(V_\text{G}\)) was lower than \(V_\text{TH}\). However, the charge exchange was suppressed by the MoS\(_2\) of the heterostructure; no change in \(I_\text{D}\) could be observed no matter how the light intensity changed when \(V_\text{G} > V_\text{TH}\), which was different from the current change of devices with only graphene or MoS\(_2\) materials. In addition, the optical responsibility of the device was enhanced with the decrease of \(V_\text{G}\) due to the electron-induced modulation on the Fermi level of graphene and states of the electron in MoS\(_2\), which suggested the feasibility of the modulation process of the optical responsibility and the adjustment of the synaptic weight. With the increment of the optical stimuli power intensity from 0.73 to 13.5 mW cm\(^{-2}\), more photoinduced carriers were generated and then increased the resistivity of the graphene, which contributed to that the PSC decayed by 38.4%. After removing the optical stimuli, a slight reduction of the current was observed, which indicated the long-term depression of LTP.[79] Ni et al. also explored the optical synaptic behaviors of devices fabricated with other 2D materials beyond graphene and related materials (GRMs).[80] The device operated based on the synergistic effect between silicon nanocrystals (Si-NCs) with the photonic absorption ability on strong broadband and 2D tungsten diselenide (WSe\(_2\)) with strong charge transport characteristics. Compared with 2D materials, Si-NCs doped with boron elements showed better optical absorption to NIR light and the device could operate under optical stimuli with low power intensity (1.25 × 10\(^{-3}\) μW cm\(^{-2}\)). As one of the materials with the lowest thermal conductivity, WSe\(_2\) hindered the thermal conduction between the device and the external environment, which could effectively reduce the energy consumption (∼75 fJ). They mainly discussed the relationship between the change of EPSC and the duration time of optical spiking. Photonic absorption was induced in the band tail of the B-doping Si-NCs and then optical-induced holes were transferred from Si-NCs to WSe\(_2\). This absorption characteristic resulted in EPSC and the EPSC continuously increased with the increment of optical pulse duration time until the current reached the saturation state, which indicated the effective modulation of the synaptic weight of the Si-NCs/WSe\(_2\) optoelectronic transistor. The STP performance represented by the PPF response was triggered by two consecutive optical pulses (1.25 × 10\(^{-3}\) μW cm\(^{-2}\), 40 ms). With the increment of optical pulse number and the optical spiking frequency, the transition from STP to LTP was obtained with extremely low energy consumption.[80]

Park et al. proposed an optical synapse that could update the synaptic weight under the optical stimuli with high frequency and low power intensity, which was associated with the trapping process of electrons in the GO layer and charge-transporting process of electrons in the indium–gallium–zinc oxide (IGZO) layer.[81] A large amount of electron–hole pairs were generated in the IGZO channel by the optical stimuli, which made it easier for electrons to be trapped or detrapped in the GO nanosheet layer. Due to the existence of the long alkyl chain demonstrated in Figure 6b, electrons were detrapped in the GO layer by photoinduced holes located on the valence band (VB) of the IGZO layer with the effect of the negative optical pulses, which resulted in the increment of the conductance. During the modulation process of the synaptic plasticity, the electrical voltage bias with 5 V was applied to the device for 500 ms at first and the initial current decreased to ∼1 nA, which was attributed to enough electrons in the GO layer. With the pure optical pulse (2000 W cm\(^{-2}\), 50 ms), a slight increment of the EPSC from 1.2 to 5.9 nA was obtained and then decayed back to the initial current state with a fast velocity, which exhibited no retention property and indicated the biomimetic STP performance. However, an obvious increment of the EPSC from 7.9 to
187.1 nA was observed with the combination spiking from electrical (1 V/50 ms) and optical (2000 W cm$^{-2}$, 50 ms) stimuli. At last, the amplitude of the EPSC was maintained at around 20 nA after the decay, which suggested the successful transition from STP to LTP.\[81\]

### 3.6. Optical IMCS Synapses with Hybrid Low-Dimensional Materials

In order to obtain better artificial synaptic characteristics, some researchers have investigated employing various low-dimensional materials in a single optoelectronic device.\[60,62\] Zhang et al. emulated the bionic synaptic behavior with a flexible optical IMCS synapse fabricated with the structure of pyrenyl graphdiyne (Pyr-GDY)/graphene/PbS (poly (butylene succinate))-QD heterostructure.\[38\] As illustrated in Figure 7d, based on the combining stimuli from optical can electrical pulses, the bidirectional photoresponse behavior of the device was obtained with the photogating effect controlled by Pyr-GDY and PbS-QD layers. Pyr-GDY and PbS-QD layers were responsible for trapping electrons and the conductance channel was the graphene layer between Pyr-GDY and PbS-QD layers. The conductance of the graphene layer could be effectively modulated by trapped carriers in Pyr-GDY and PbS-QD layers, which indicated the updating process of the synaptic weight. The inhibitory performance (depression) was determined by the Pyr-GDY layer, while the PbS-QD layer was in charge of the excitatory behavior (potentiation).\[38\]

Thomas et al. successfully obtained the growth of 0D PQDs on 1D MWCNTs in Figure 8a, which enhanced the optoelectronic responsibility of the optical IMCS synapse.\[63\] Strong photosensitivity of PQDs and excellent electrical performance of MWCNTs realized the photonic memorizing behavior with the low optical power intensity of 125 μW cm$^{-2}$ and this optical IMCS synapse operated without $V_G$. In order to complete the modulation of the synaptic weight, the PQD-MWCNT device was stimulated by optical and electrical pulses. The optical stimuli triggered the bionic synaptic potentiation response, while the electrical stimuli induced the bionic synaptic depression performance, which was obtained with the form of current change. Under the optical spike (125 μW cm$^{-2}$, 0.375 s), electrons were trapped in the PQDs/MWCNTs interface, resulting in the potentiation response with the enhanced current. When the negative electrical pulses (1 V/0.1 s) were applied, the release process of charges occurred at previous trapping sites, which resulted in the depression response.\[63\]
3.7. Optical IMCS Synapses with Non-Low-Dimensional Materials

Apart from the low-dimensional materials mentioned above, other classic materials like oxides, perovskites, and organics have also been used to fabricate functional layers of IMCS synapses.\[6,7,62,96–98\] Kim et al. reported the optical synaptic characteristic of the transparent device with the structure of InO$_x$/ZnO/FTO.\[68\] Due to the formation of type-II InO$_x$/ZnO$_x$ heterojunction, the device demonstrated a loop-opening characteristic with the positive voltage, which was resulted from charge trapping/detrapping. Their transparent device could be triggered by UV illumination and showed artificial synaptic responses of PPF, STP, and LTP. Besides, behaviors of optical potentiation and electrical habituation were also implemented, which explained the type-II band alignment between ZnO and InO$_x$.\[68\] Lu et al. demonstrated a two-terminal memristor device with the spin-coated MAPbI$_3$ perovskite thin film.\[62\] The light illumination was used to control the generation and annihilation process of iodine vacancies by increasing the formation energy of iodine vacancies, which emulated the function of calcium ions in biological synapses. They also controlled the bionic memorizing and forgetting behaviors by modifying light intensity and wavelength. The Ag/MAPbI$_3$/Ag device exhibited the long-term potentiation response in darkness, while the long-term depression response could be observed with the optical pulses. Lin et al. also proposed an optical synapse based on CsPbCl$_3$ perovskite materials, which emulated the retina-like PPF, STP, and LTP with the spiking of UV illumination.\[62\] The synaptic performance was attributed to the carrier trapping and detrapping in the CsPbCl$_3$ interface. Moreover, the ability to detect deep red light without changes in synaptic behavior indicated the potential for dual-mode operation.\[62\] Huang et al. showed an organic field-effect transistor (OFET) with organic materials of PAN (polyacrylonitrile) as the gate dielectric and C8-BTBT (2,7-dioctyl[1]benzothieno[3,2-b][1]benzothiophene) as the organic semiconductor.\[97\] The artificial synaptic characteristics of EPSC, PPF, and bionic learning were all attributed to the charge trapping effect near or between the C8-BTBT/PAN interface instead of photosensitivity induced by organic semiconductors. Besides, an array comprising 20 OFETs was established to emulate the bionic memorizing and forgetting behaviors without the silicon substrate.\[97\] Huang et al. reported a photonic transistor based on PQT-12 (poly(3,3-didodecylquaterthiophene)) and...
CsPbBr$_3$. As one of the typical hole-transport materials, PQT-12 showed a high bandgap (~2.27 eV) when its maximum UV–vis absorption peak at about 480 nm. Huang’s device showed enhanced the separation efficiency of optical-induced charges, which improved the optical responsivity of the transistor. Based on this high-efficient photoresponsivity, optical synaptic behaviors like EPSC, IPSC, PPF, and STP were obtained.

4. Mechanism of Optical IMCS Synapses with Low-Dimensional Materials

Although the diversity of device structures or device functions, the current research tendency has indicated that the artificial synaptic behaviors of the most optical IMCS synapses were determined or influenced by the operation mechanisms based on defects and carrier traps/traps in the semiconductor structure, such as the ionization of oxygen vacancy, the capture of defect trap, the release of photoinduced carriers, and potential-well capture triggered by heterojunction. These operation mechanisms are closely associated with various properties of utilized materials. Therefore, the following investigation in this section will focus on mechanism categories of optical IMCS synapses with low-dimensional materials (including 0D, 1D, and 2D materials).

4.1. Trapping/Releasing Process of Photoinduced Carriers

For low-dimensional materials used in the functional layers of optical IMCS synapses, the basic optoelectronic characteristics and derived artificial IMCS synaptic behaviors were associated with the trapping/releasing process for photoinduced carriers. In general, the trapping/releasing process of photoinduced carriers based on low-dimensional materials was mainly influenced by main factors like the heterostructure of functional layers and the defect of functional layers. The effect induced by the heterostructure could be produced by the contact of two low-dimensional materials, or by the contact of low-dimensional materials and non-low-dimensional materials.

Figure 8. a) Schematic view of the optical IMCS synapse with PQD-MWCNT materials. b) TEM image of the hybrid PQD-MWCNT. c) Fabrication process of the hybrid PQD-MWCNT materials. d) PPF effect of the PQD-MWCNT device with two consecutive pulsed stimuli. e) Long-term potentiation and depression performance of the PQD-MWCNT device with 40 consecutive optical pulses and 80 consecutive electrical pulses. Reproduced with permission.[63] Copyright 2020, Wiley-VCH.
As illustrated in Figure 4, Han et al. investigated the optical IMCS synapse based on 0D CsPbBr3 QD, the heterostructure functional layers included a pentacene layer as the semiconductor layer, a PMMA layer as the passivation layer, and a 0D CsPbBr3-QD layer as the floating gate. The photonic programming operation generated plenty of carriers into the functional layer and then resulted in the photogenerated current. The escape phenomenon of photoinduced holes occurred from the CsPbBr3-QD layer to the pentacene layer and photoinduced electrons left in the conduction band (CB) of the CsPbBr3-QD layer. The captured electrons played a role of an internal conductor with a faster speed. Trapped carriers could retain a long-lifetime potential well after removing the optical stimuli. This mechanism was verified by the in situ Kelvin probe force microscopy (KPFM) test conducted on the surface potential change of functional layers, as illustrated in Figure 9a. In Figure 9b, optical stimuli with fixed intensity and different wavelengths were applied to the pentacene thin film, and the obvious increment of its surface potential was observed with the decrease of the light wavelengths (Figure 9c). Due to the escape behavior of photoinduced holes in the CsPbBr3-QD layer, the remaining photoinduced electrons in the CB increased the surface potential. In addition, in Figure 9d, the CsPbBr3-QD thin film was electrically scanned and holes/electrons were horizontally injected into the film, which was used to evaluate the trapping ability of holes/electrons. As illustrated in Figure 9e, injected holes vanished in the CsPbBr3-QD thin film with a quicker speed and indicated the higher drift rate of holes, which proved that electrons were deeply trapped and supported the optical synaptic behaviors.

Zhou et al. reported the optical IMCS synapse with a heterojunction based on 2D MoS2 and organic perylene-3,4,9,10-tetracarboxylic dihydride (PTCDA); the synaptic plasticity was modulated by the carriers drift at the hybrid heterojunction interface, as illustrated in Figure 10. The discussion on the mechanism included five phases, which were I, II, III, IV, and V phases illustrated in Figure 10d. Figure 10b,c have demonstrated the typical artificial synaptic behaviors of IPSC and EPSC. In phase I of Figure 10d, a dynamic equilibrium of carriers in the MoS2/PTCDA heterojunction was obtained with the baseline voltage (VCG) at −17 V. When the VCG changed from −17 V to −12 V, on the one hand, electrons accumulation occurred in the MoS2 channel with this positive change of VCG and then resulted in the higher current. On the other hand, in phase II of Figure 10e, due to the weakening performance of the gate electric field and the bending of the energy band, some electrons drifted to the PTCDA layer. After removing the VCG, electrons in the MoS2 layer depleted because these electrons had drifted to the PTCDA layer and then resulted in the current decrease in Figure 10b. However, in phase III, the diffusion process of electrons was slow due to the low ion mobility, and the current returned back to the reference state, which indicated the IPSC behavior in Figure 10b. Therefore, when the negative change (−17 V to −20 V) of VCG was applied to the device, the reversed behavior, EPSC, could be obtained due to the reversed drift of photoinduced carriers and electrons in the 2D MoS2/PTCDA heterojunction (phases IV and V in Figure 10f).

Compared with the trapping/releasing process of photoinduced carriers influenced by the heterostructure, the trapping/releasing process of photoinduced carriers influenced by the defect could be more observed on optical IMCS synapses with 0D and 1D low-dimensional materials. As illustrated in Figure 11a, Yang et al. proposed their optical IMCS synapse with a spin-coated ≈300 nm functional layer based on 0D

Figure 9. a) Schematic view of the KPFM test for the surface potential. b) Surface potential change of the pentacene thin film induced by the optical stimuli with the fixed intensity and various wavelengths. c) Distribution results of the relationship between the surface potential change and various light wavelengths. d) Injection of holes/electrons into the CsPbBr3-QD thin film by electrical scanning. e) In situ records of trapped charge at 0, 1, 2, and 3 h with the scale bar were 1 μm. f) Distribution results of surface potential variation from (e). Reproduced with permission. Copyright 2018, Wiley-VCH.
B-doped Si-NCs. Measurements of electron paramagnetic resonance (EPR) and scanning tunneling spectroscopy (STS) were used to investigate the operation mechanism and related characterization results could be observed in Figure 11c,d.

The signal of 2.006 in Figure 11c indicated the indeed existence of a large number of surface dangling bonds (SDBs) on the Si-NCs and the EPR measurement estimated the density of SDBs was about 3 \times 10^{14} \text{ cm}^{-2}, which indicated the successful...
enhancement of the boron doping.\cite{109} As a typical producer of defect-induced trap states, SDBs had a significant influence on the drift of photoinduced carriers, and this theory was supported by the following STS measurement in Figure 11d. In-gap states were found near CB, which indicated that energy levels could be introduced to the position under the CB by SDBs of B-doped Si-NCs.\cite{111} In addition, the extension of the band tail from VB was observed with the STS measurement, which made the reduced bandgap of \( \approx 0.52 \) eV and verified the photonic absorption ability of B-doped Si-NCs from the UV region to the NIR region.\cite{111} As the illustration in Figure 11e, when photons were absorbed by the B-doped Si-NCs, the excitation operation drove electrons to drift from VB to CB and a great deal of photoinduced holes enhanced the hopping behaviors of neighboring holes in the Si-NCs thin film, which resulted in the increment of EPSC of the device. During this process, partial electrons were trapped by SDBs defect in the Si-NCs thin film. These trapped electrons were released to CB with the effect of the thermal fluctuation and then recombined with photoinduced holes mentioned before, which verified the synaptic plasticity.\cite{112,113}

Pilarczyk et al. demonstrated the optical IMCS synaptic behaviors based on 1D MWCNTs as the functional layer with a hybridization structure.\cite{76} The optoelectronic performance and artificial synaptic behaviors of the device resulted from the trapping/releasing process of photoinduced electrons due to the existence of surface defects located in the MWCNT semiconductor layer.\cite{76} Apart from the research on typical 1D materials like CNT, Sun et al. reported optical IMCS synapse behaviors based on 1D SnO\(_2\) NW, which was triggered by the deep-ultraviolet (DUV) signals.\cite{110} A combination effect between internal defects of the 1D SnO\(_2\) thin film and its surface states resulted in the change of photoconductivity of the device. Because of the strong sensitivity to the DUV radiation, surface acceptors of SnO\(_2\) NWs trapped photoinduced holes, and the trapping process required a long relaxation time. During this trapping process, the electron density was continuously increased by these trapped holes, and then the photoconductivity was enhanced obviously.\cite{76}

### 4.2. Effect of Ferroelectric Polarization

So far, more and more attention has been attached to both electrical-control IMC and optical IMCS synapses based on low-dimensional materials with the effect of ferroelectric polarization, especially for ferroelectric materials based on 2D van der Waals system such as PbZr\(_{0.2}\)Ti\(_{0.8}\)O\(_3\) (PZT), Bi\(_2\)O\(_2\)Se, and \( \alpha \)-In\(_2\)S\(_3\).\cite{114–119} However, no one has summarized the ferroelectric polarization mechanism of optical IMCS synapses with low-dimensional materials. In this section, systematic discussions on the ferroelectric polarization mechanism of 2D materials optical IMCS synapses will be investigated for the first time.

The ferroelectric polarization represented that the electric polarization of the ferroelectric materials could be transferred or switched among multiple states (the number of states could be two or more).\cite{118,119} The ideal Clausius–Mosotti (CM) model also pointed that the definition of polarization was the average dipole moment density.\cite{118,119} Effective polarization (\( \Delta P \)) was described as an integral function of the charge flow, as illustrated in the following equations

\[
P_{\text{unit}} = \frac{1}{V_{\text{unit}}} \int_{V_{\text{unit}}} \rho(r) dr
\]

\[
\Delta P_{\text{unit}} = \int \frac{dt}{V_{\text{unit}}} \int_{V_{\text{unit}}} j(r, t) dr
\]

where \( V_{\text{unit}} \) is the desired unit cell, \( \rho(r) \) is the charge density, and \( j(r, t) \) is the flow of charge. This definition indicated that the quantified polarization values might be influenced by the measurement techniques. This theory was the foundation of the 2D ferroelectric polarization materials and also indicated their quantum characteristics.\cite{117,119} Compared with conventional ferroelectric materials represented by perovskite-type oxides, the most outstanding advantage of 2D ferroelectric materials was the regulation of directions of electric field and depolarization field.\cite{119} The spontaneous polarization of 2D ferroelectric materials resulted from the asymmetric distribution of the atomic layers perpendicular to the 2D plane, the direction of polarization can be reversed by clever multiaimatic motions.\cite{119} As there was no inversion symmetry in the 2D plane, the spontaneous polarization occurred in the direction of in-plane, and the in-plane polarization direction was also associated with the out-plane polarization.\cite{115,117–119} Therefore, the coupling modulation of the electric field direction and the polarization direction could be obtained, which was helpful to the transition process from semiconductor characteristics to metal-like characteristics.\cite{119}

Zheng et al. proposed an optical IMCS synapse based on 2D ferroelectric Bi\(_2\)O\(_2\)Se materials, as illustrated in Figure 12a.\cite{115} The electrical measurement had pointed out the \( n \)-type semiconductor characteristic of the 2D ferroelectric Bi\(_2\)O\(_2\)Se thin film and the device showed a resistive ratio of 900%, which indicated that the nonvolatile modulation between the electric field and resistance could be obtained. As the illustrations in Figure 12b,c, when the external electric field was applied onto the gate terminal, polarized charges occurred on the top surface of the \( \text{Pb(Mg}_{0.1}{Nb}_{0.2})_{3}\)O\(_3\)–PbTiO\(_3\) (PMN-PT) layer and influenced the carrier motion in the Bi\(_2\)O\(_2\)Se layer, which affected the electron density of the 2D ferroelectric Bi\(_2\)O\(_2\)Se thin film. Besides, the pyroelectric effect induced by PMN-PT monocrystals also influenced the ferroelectric polarization of Bi\(_2\)O\(_2\)Se materials. In Figure 12d, negative polarized charges on the top surface of the PMN-PT layer trapped positive charges from the Bi\(_2\)O\(_2\)Se layer. The 980 nm IR light was chosen as the input optical stimuli in Figure 12a, which increased the temperature of the device and weakened the remnent polarization intensity. Trapped positive charges were released to the Bi\(_2\)O\(_2\)Se layer, which enhanced its hole density and then reduced the conductance. The reversed phenomenon was demonstrated in Figure 12e. The changed conductance influenced by the pyroelectric effect and the ferroelectric polarization had a great impact on the following synaptic weight update.\cite{115}
Figure 13c–e, the generation and accumulation of photoinduced carriers resulted in the increment of the channel conductance when the optical pulse was added to the channel. Besides, the effect of ferroelectric polarization also occurred along with the direction of the electric field, and the motion of the electron was accelerated. After being removed from the photonic environment, the channel conductance demonstrated a depression state due to the reversed polarization in ferroelectric domains. Hence, this optical IMCS synapse with the 2D ferroelectric α-In_{2}Se_{3} materials showed a longer attenuation time with the optical stimuli instead of the pure electric stimuli, which indicated the potential of the long-term synaptic plasticity.\[118\] Interestingly, due to the different synaptic responses to various light stimuli, the 2D α-In_{2}Se_{3} IMCS synapse emulated biomimetic color recognition. The visible purple light with the 450 nm wavelength induced the rapid saturation of the synaptic weight update, while the 980 nm IR laser almost resulted in no photoinduced current in the device. Only the invisible 808 nm light triggered bionic synaptic potentiation current.\[118\]

Apart from 2D materials with their build-in effect of ferroelectric polarization, some other work also demonstrated optical IMCS synapses based on the combination of 2D materials without ferroelectric characteristics and ferroelectric non-low-dimensional materials.\[114,117\] Gruverman et al. reported an optical IMCS synapse fabricated by ferroelectric PbZr_{0.2}Ti_{0.8}O_{3} (PZT) and 2D WS_{2} materials.\[114\] The conductance states of the WS_{2} channel were controlled by the external electrical and optical stimuli, which were associated with the tunable ferroelectric polarization of the PZT layer. The polarization of the PZT layer resulted in the accumulation and depletion of charges in the WS_{2} channel, which influenced the carrier density of WS_{2} materials and then affected the degree of polarization compensation. With the change degree of the ferroelectric polarization effect, the excitatory or inhibitory currents resulted in the potentiated and depressed responses of the optical IMCS synapse.\[114\] In addition, Park et al. showed their research on an optical IMCS synapse based on 2D MoS_{2} and ferroelectric Hf_{0.5}Zr_{0.5}O_{2} (HZO) materials.\[117\] Between the 2D MoS_{2} channel and the HZO layer, a 3-aminopropyltriethoxysilane (APTES) passivation layer was inserted to control the number of interface traps. With the external stimuli applied to the gate, the ferroelectric polarization direction of the HZO was influenced, which resulted in the corresponding accumulation or depletion of electrons in the channel. Then different synaptic behaviors were demonstrated with the potentiated and depressed updates of synaptic weight.\[114\]

4.3. Ionized and Deionized Processes of Oxygen Vacancies

In most cases, mechanisms based on the trapping/releasing process of photoinduced carriers and ferroelectric polarization effect were investigated on three-terminal optical IMCS synapses. However, for partial two- and three-terminal optical IMCS synapses with semiconductive oxide materials, although low-dimensional materials were included for functional layers, their all-in-one IMCS performance was considered to be...
associated with the ionized and deionized process of oxygen vacancies induced by external optical stimuli.[35,120,121] The photoinduced ionization resulted in the neutral oxygen vacancies with a positive charge due to the persistent photoconductivity (PPC) characteristic of the oxide semiconductor and these ionized vacancies transferred back to the neutralized state after removing the optical stimuli.[35,120–122] The ionized and deionized processes could be represented by the following equations:[35]

$$V_0^\circ + O_{WB}^- + 2h^+ \rightarrow V_{O}^{2+} + 2e^- \quad (4)$$

$$\frac{V_{O}^{2+}}{O_1} + 2e^- + E_a \rightarrow V_O^\circ \quad (5)$$

where $V_0^\circ$ is the insulating oxygen vacancy without charge, $O_{WB}^-$ is the bound oxygen, $h^+$ is the hole with a positive charge, $O_1$ is the interstitial oxygen, $e^-$ is the electron with a negative charge, and $E_a$ is the activation energy.

Mahata et al. reported a two-terminal optical IMCS synapse with 0D TiN-NPs embedded in the HfAlO switching layer.[121] With the optoelectric field brought by the optical stimuli, rupture and formation processes of conductive filament (CF)-based ionized oxygen vacancies were limited by 0D TiN-NPs. Ionization phenomena of oxygen vacancies occurred near the interface between HfAlO and TiN-NPs; partial oxygen vacancies with a positive charge formed CFs to connect the top and bottom electrodes of the device, which enhanced the current and modulated the conductance state. Reversely, the potentiated conductance state could be modulated by the rupture of CFs. Hence, the change of CFs with ionization and deionization processes based on oxygen vacancies could realize the modulation of synaptic weight.[121] Duan et al. also investigated the optical IMSC performance of a three-terminal transistor with perovskite (PVK) NPs and IGZO materials.[122] Due to the built-in electric field induced by optical stimuli, photoinduced electron–hole pairs were separated. Compared with the n-type IGZO, the p-type CsPbBr$_3$-NPs had a deeper Fermi level ($E_F$), which promoted electrons in IGZO transfer to CsPbBr$_3$-NPs and then $E_F$ of IGZO drifted in a downward direction near the interface of

Figure 13. a) Schematic view of the optical IMCS synapse based on 2D ferroelectric \(\alpha\)-In$_2$Se$_3$ materials. b) The emulation process of the bionic retina with various optical stimuli. c–e) The structures of band diagrams and f–h) corresponding PSC currents are induced by optical stimuli with different wavelengths. Reproduced with permission.[118] Copyright 2021, Wiley-VCH.
IGZO/CsPbBr3-NPs. The downward-moving $E_f$ could result in the ionization of oxygen vacancies, which induced the PPC characteristic. More ionized oxygen vacancies could enhance the synaptic plasticity of the optical IMCS synapse, which could complete the synaptic modulation.[122]

4.4. Nonvolatile Resistive Switching Induced by Optical Stimuli

For some two- and three-terminals IMCS synaptic memristors fabricated with low-dimensional materials, the artificial synaptic behaviors were controlled by the nonvolatile resistive switching (RS) behaviors, which the optical stimuli could trigger.[123–128] Furthermore, these nonvolatile RS behaviors were attributed to different physical mechanisms of materials, such as optical-mediated Schottky barrier, optical-induced formation/rupture of CFs, and photogate effect. Lourembam et al. developed an RRAM device with wurtzite ZnO NRs deposited on perovskite Nb-doped SrTiO$_3$ (NSTO) thin film, which formed a Schottky barrier.[123] Optical-induced electrons moved to the anode with the UV illumination, while holes moved to the ZnO/NSTO interface and then combined with electrons in the space charge region. Due to the Schottky barrier of the ZnO/NSTO interface, ionized oxygen vacancies moved to the interface. Captured holes and ionized oxygen vacancies had large effective mass and low mobility, which resulted in the PPC characteristic and further RS performance.[123] Wang et al. reported a memristor comprising the Ag/MoSe$_2$/Bi$_2$Se$_3$/PMMA/ITO hybrid layer that could modulate the formation/rupture process of CFs.[124] With the 790 NM NIR illumination as the input spiking, optical-generated electrons were trapped by the MoSe$_2$/Bi$_2$Se$_3$ hybrid layer, while untrapped holes made Ag clusters turn into Ag ions (Ag$^+$). Gradual accumulated Ag$^+$ resulted in the optical-induced annihilation of the Ag CFs and then reduced the operation current of the device.[124] Lv et al. proposed RRAM devices with carbon dots (CDs)-silk protein and CsPbBr$_3$ QDs, respectively, which proved that the photogate effect could affect the feasibility of optical-tunable memory characteristics.[125,127] Optical-generated electron–hole pairs at the interface were separated as electrons and holes, which were driven to anode and cathode, respectively. Optical-generated carriers were trapped in the QDs. The following photogate effect resulted in the faster formation of the conductive path, which reduced the energy consumption of the RS process.[125,127] Although some researchers mentioned above did not show their investigation results of synaptic behaviors, their memristors still had great potential as candidates of artificial synapses due to their common multistate conductance characteristics and excellent optical-induced nonvolatile RS performance.

5. Neuromorphic Applications of Optical IMCS Synapses

Biologically, the external optical stimuli act on the neurons in the form of perceptible nerve impulses. With the complex visual neuron network system, the external information is transmitted to the visual cortex of the human brain and then explained with various characteristics such as wavelength, color, intensity, and frequency.[8,14,52,129–132] Different external stimuli resulted in...
cognition and memorizing variation, which is associated with the neural function in the biological system.\textsuperscript{[51,131–138]} The application research on optical IMCS synapses is always inseparable from the emulation of artificial neural performance such as memorizing and forgetting, which is related to the synaptic short-term memory (STM) and long-term memory (LTM).\textsuperscript{[97,112,139–144]} The foundation of STM and LTM is synaptic plasticity (STP and LTP) with excellent stability and adjustability. The STM is always triggered by short-term external stimuli with low power intensity and few spiking cycles. With the repeated and frequent stimuli (rehearsal), the STM can be transmitted to LTM before the occurrence of the forgetting behavior.\textsuperscript{[10,11,106,107,112,129,140]} The transition process can be compared to the learning behavior, as illustrated in Figure 14a. In contrast, compared with artificial synaptic devices, biological synapses demonstrate the sensory memory (SM) before the STM, which is associated with the stimuli perception of the human sensory organs.\textsuperscript{[8,9,58]} The SM process of the synaptic device can be explained by the reception of external pulses. For now, some applications on optical IMCS synapses such as pattern recognition, image memorizing, logic operation, and filtering focus on two-terminal memory and three-terminal transistor have received more and more attention, and most of them emulated the biomimetic visual perception and forgetting/learning behaviors.\textsuperscript{[13,19,37]} In this section, application research of optical artificial synaptic devices with low-dimensional materials is under discussion, which includes pattern/image recognition, mathematical calculation, logic operation, and filtering.

5.1. Pattern/Image Recognition

As one of the most popular applications of artificial synaptic devices, pattern/image recognition has been investigated since the occurrence of artificial synapses.\textsuperscript{[8,10,32–34]} The biomimetic pattern/image recognition was supported by the operation principle of the biological visual system.\textsuperscript{[31,36]} Optical receptors on the surface of the retina generated excitatory signals after receiving external optical stimuli. These signals were processed and then transmitted through a neural network comprising of neurons and synapses in the retina, and finally transmitted to the cerebral cortex through optic nerve fibers.\textsuperscript{[58,130,145,146]} Based on this supported biological theory, Wang et al. emulated the pattern recognition process with the mechanophotonic synapse fabricated with 2D h-BN/WSe\(_2\) heterostructure.\textsuperscript{[79]} In Wang’s device, the TENG was integrated into the gate terminal of the transistor.\textsuperscript{[79]} Therefore, the synaptic weight of the optical synaptic transistor was updated by the mechanical displacement of the TENG and the optical stimuli of the transistor. Before starting the recognition process, the long-term potentiation and depression were measured and some key parameters were obtained. The updating process of the synaptic weight was determined by four vital parameters, including the maximum value of the conductance (\(G_{\text{MAX}}\)), the minimum value of the conductance (\(G_{\text{MIN}}\)), the nonlinearity of the potentiation curve (\(NL_p\)), and the nonlinearity of the depression curve (\(NL_d\)). The whole recognition process of the artificial neuron network (ANN) was supported by the supervision learning of the Mixed National Institute of Standards and Technology database (MNIST) database. The structure of ANN was composed of input, hidden, and output layers. The hidden layer with 100 neurons was the connection of the input layer with 784 input neurons and the output layer with 10 output neurons. The input layer emulated the biological optical receptors, the hidden layer simulated the intermediate neuron cells, and the output layer was compared to the ganglia cells. The input source was an image with 28 × 28 pixels and the output results were Arabic numbers from 0 to 9. Number 7 was chosen as the target number. With the combined modulation of mechanical displacement and optical stimuli, the obvious increment of updated synaptic weight was obtained and number 7 was recognized, which indicated the feasibility of dual-model modulation (mechanical displacement of TENG and optical stimuli of the transistor).\textsuperscript{[79]} Park et al. demonstrated their research on the high-efficient recognition process of mixed colorful patterns based on an optical-neural synaptic device fabricated with 2D h-BN/WSe\(_2\) materials.\textsuperscript{[131]} As illustrated in Figure 14b, a conventional neural network (NN) system and an optical neural network (ONN) system were built to emulate the color-blindness test, which could complete the recognition process of the target number from the numerical matrix with complex mixed color. For the conventional NN, the input layer of the conventional NN was comprised of the color-filtering neuron array, which could emulate the operation mode of cone cells in the retina. However, the synaptic connection in the neuron array could not respond to the external optical stimuli, which indicated that the recognition progress was emulated in the dark environment. By contrast, the synaptic connection in ONN had the function of responding to the optical stimuli, and the array was built by 784 (28 × 28) biomimetic cone cells. All cone cells in the input layer of the network had different responses to the wavelength of visible RGB (red/green/blue) light. Each cone cell in the input layer was connected to six classification neurons in the output layer. Due to the wavelength variation of the RGB light, the NN and ONN were triggered by the different input signals with various voltage biases (1 V for R light, 0.5 V for G light, 0.3 V for B light), and the input source was the MNIST database. Numbers 1 and 4 were chosen as target numbers and six different datasets comprising 600 images were prepared for the training process. During each training epoch, 600 different images were used in the test process of the network. When at the 50th training epochs, the ONN system exhibited high recognition accuracy of 90%, while the accuracy of the conventional NN system was lower than 40%. In addition, the ONN system maintained this stable recognition accuracy until end of the training. During the whole training process, further optimization of the synaptic weight was obtained to be suitable for the recognition process of numbers with mixed RGB color. Their h-BN/WSe\(_2\) synaptic device demonstrated the strongest response to the B light, which resulted in the richer synaptic weight for blue patterns and indicated the successful recognition of numbers with RGB lights. The ONN system exhibited a high recognition accuracy of 98% after 600 training epochs.\textsuperscript{[131]}

Park et al. also researched the recognition process on digital handwriting images with an ANN, which was performed based on synaptic behavior of the IGZO-alkylated/GO device.\textsuperscript{[89]} As illustrated in Figure 15, with vital parameters like \(G_{\text{MAX}}/G_{\text{MIN}}, NL_p,\) and \(NL_d,\) the recognition process was simulated with...
Figure 15. (a) Schematic view of the ANN recognition process with an SLP structure comprising of 785 input signals (784 input neurons and 1 bias neuron) and 10 output signals. (b) Recognition result of number 6 with different weight status. (c) Change of recognition rates due to the various function relationships between electrical stimuli and the number of learning phases. (d) Change of recognition rates due to the various function relationships between electrical stimuli (black)/mixed stimuli (red) and the number of learning phases. (e) Change of recognition rates due to the various function relationships between electrical stimuli (black)/mixed stimuli (red) and the number of weight states. Reproduced with permission.[81] Copyright 2018, Wiley-VCH.

Figure 16. (a) Schematic view of the pattern recognition process with an MLP ANN system. (b) Schematic view of the synaptic weight defined as conductance difference of two optical synapses. (c) Input image data with various noise ratios from 0% to 90%. (d) Recognition accuracy of the device at the flat, bending, and folding states. (e) Recognition accuracy of the device in different noise environments. (f) Change of recognition accuracy for the device with the effect of noise pixel proportion. Reproduced with permission.[38] Copyright 2021, ACS.
a single-layer perceptron (SLP) structure comprising 785 input signals (784 input neurons and 1 bias neuron) and 10 output signals. Image data in the MNIST dataset were sent to the input layer, the output \( y \) was decided by the following equation

\[
y = \frac{I}{V \times W}
\]

(6)

where \( I \) was the current vector, \( V \) was the input vector, and \( W \) was the synaptic weight matrix. \( I \) was transferred to the output vector \( y \) through a sigmoid activation function \( y = f(I) \). Then the updated synaptic weight \( \Delta W \) was obtained by evaluating the difference value between the output value (\( y \)) of \( I \) and the label value (\( k \)) of the input image. After 60,000 training epochs, with mixed spikes in Figure 15b, number 6 was recognized successfully with the recognition accuracy at 73%.\(^{[81]}\)

Zhang et al. completed the pattern recognition process with a multilayer perceptron (MLP) ANN system, which had an input layer comprising 784 input neurons, a hidden layer comprising of 300 hidden neurons, and an output layer comprising of 10 input neurons, as illustrated in Figure 16.\(^{[38]}\) Compared with other recognition processes, the training under noise environment was investigated; related results could be observed in Figure 16e,f. With optical synaptic behaviors of their Pyr-GDY/graphene/PbS-QD device, the highest was up to 98.1%. Excellent average recognition rates were also obtained when the device was at the flat (90.8%), bending (88.9%), and folding (89.5%) states. Based on the high-accuracy recognition process, they also emulated real-time pattern detection with a sensing–memorizing–processing system, as illustrated in Figure 17.\(^{[38]}\) At the initial state, all devices in the conductance map showed low conductance, which was defined as \( G_0 \) in Figure 17c. After 150 consecutive optical pulses (150 mW cm\(^{-2}\), 20 ms, 980 nm) were applied onto the map, the letter “G” was obtained in Figure 17d, which indicated the in situ memory. Based on this in situ memorizing performance, the input letter “O” was chosen as the reference pattern, which resulted in the nonvolatile updated conductance. After measuring the synapse array with conductance change, a matrix \( M \) comprising of 7 \(	imes\) 6 pixels was obtained. With more training by optical stimuli, unknown images were updated with a further matrix \( R \). Unknown images with numbers “1” and “2” represented letters “S” and “O,” which were corresponding to matrixes “\( R_1 \)” and “\( R_2 \)” respectively. With continuous optical training, the output variation \( V \) was defined as

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**Figure 17.** a) Schematic view of the sensing-memorizing-processing system with optical stimuli. b) Variation among letter “O,” number “1” (S), and number “2” (O). Conductance map of pixels with the structure of 7 \(	imes\) 6 at c) initial state, d) after 150 consecutive optical pulses, and e) after 1000 s at the ending of optical stimuli. Reproduced with permission.\(^{[38]}\) Copyright 2021, ACS.
The difference value of 7 from 10 was obtained by calculating the quantity of optical pulse number required to the current increment from EPSCm to EPSCn. Based on the substance of the addition and subtraction process, the processes of multiplication and division were obtained in Figure 18d,f.\textsuperscript{[147]}

5.3. Logic Operation

Yang et al. also investigated the synaptic application of the device based on 0D Si-NC.\textsuperscript{[152]} Besides, they proposed a device with optical output and emulated the logic functions of “AND,” “OR,” “NAND,” and “NOR” gates. The input signals were determined by electrical pulses $E_{1}\mu$ and $E_{2}\mu$; the output signal of the AND-OR gate in Figure 19a was represented by the total optical output power ($I_{\text{out}}$) of the synaptic devices. For the AND gate, input signals of a low voltage of 0 V and a high voltage with 6 V were defined as “0” and “1,” respectively. The output signal was defined as “0” with the $I_{\text{out}}$ lower than the threshold power of 1 $\mu$W while the output “1” indicated that the optical output power was higher than 1 $\mu$W. For the OR gate, input signals “0” and “1” were a low voltage of 0 V and a high voltage with 8 V; the evaluation criteria of output was the same as AND gate. Results of the AND-OR logic operation were obtained in Figure 19b. $I_{\text{out}}$ of “00,” “01,” “10,” and “11” were 0.13, 1.68, 1.68, and 3.54 $\mu$W, respectively. Apart from the AND-OR gate, Yang et al. also emulated the NAND-NOR gate. Compared with the AND-OR gate, the most significant difference was that the output of the NAND-NOR gate was the output resistance ($R_{\text{out}}$) and the threshold value was 0.1 $\Omega$ (Figure 19c). For the NAND gate, 6 V and 0 V voltage signals were selected as input “1” and “0.” 0 V and 8 V voltage signals were chosen as input “1” and “0” for the NOR.
gate. As illustrated in Figure 19d, during the logic operation of the NAND-NOR gate, $R_{out}$ values of 0.07, 0.16, 0.16, and 15.1 $\Omega$ were corresponding to the output signals of “0,” “1,” “1,” and “1,” which was induced by input signals of “11,” “10,” “01,” and “00.”

5.4. Filtering

In biological neural networks, the filtering function of synapses is responsible for screening the information passing through the synapses. Information within the target frequency will be allowed...
to transmit through synapses while the strength of information at other frequencies will be weakened. The conductance change of biomimetic synaptic devices indicated that these synaptic devices also have similar characteristics. For some optical IMCS synapses, the high-pass (low-pass) filtering function was always triggered by optical stimuli with high (low) frequency. Pi et al. have demonstrated the filtering function of their optical IMCS synapse fabricated with 0D Si-NC. At first, they investigated the effect of the number and frequency of optical pulses on EPSC, as illustrated in Figure 20a,b. With the increment of optical pulse number, an increased tendency of the EPSC could be obtained and the decay time of the EPSC was longer, which was called SNDP. A test cycle with 50 consecutive optical pulses was selected as the input source and the EPSC increased with the change of frequency (2.0, 2.9, 4.0, 6.7, and 14.3 Hz), which was called SRDP. The ratio between the maximum value of the EPSC triggered by the 50th optical pulse ($A_{50}$) and the maximum value of the EPSC triggered by the first optical pulse ($A_{1}$) was defined as the optical gain ($A_{50}/A_{1}$). This optical gain grew from 2 to 6 with the frequency increasing from 2.0 to 14.3 Hz, which indicated the dependency of the optical gain on the pulse frequency. The cutoff frequency ($f_{1}$) of this optical IMCS synapse was evaluated as 4.8 Hz with the fitting operation between the optical gain and a high-pass filter function, which indicated the great potential of the device as a high-pass filter (Figure 20c). The $f_{1}$ was defined as a threshold frequency, the signal with a frequency higher than this threshold frequency could pass the filter while the signal with a frequency lower than this threshold frequency was attenuated. They took an original image illustrated in Figure 20c as the filter target, and Figure 20d,e shows results of the image filtered by various frequencies. With the 4.8 Hz filtering frequency used in the filtering process, the image was sharpened effectively.

Cui et al. also evaluated the filtering characteristics of the optical IMCS synapse based on SWCNT. The EPSC of the device was triggered by various stimuli cycles of optical stimuli with different frequencies and each cycle contained 30 consecutive optical pulses. As illustrated in Figure 21a, the increment of EPSC was observed with the increment of spiking time. In the range from 0.5 to 10 Hz; a higher EPSC was observed with a lower frequency at the same time. There was no change in the EPSC with the optical stimuli at 10 Hz while the EPSC increased about 100 times when the optical stimuli frequency was 0.5 Hz. They considered 5 Hz as the threshold frequency due to no obvious variation occurred during each stimuli cycle when the frequency was higher than 5 Hz, which resulted in those carriers like photoinduced carriers and trapped carriers could not be released. The optical gain of each stimuli cycle was defined as $|A_{g}/A_{f}|$. A decay tendency of the optical gain with the increase of the optical stimuli frequency could be observed in Figure 21b.

6. Conclusions and Outlook

In this review, the definition and demonstration of optical and other artificial synaptic characteristics such as STP, LTP, STDP, SNDP, and SRDP have been introduced. Based on the relationship between biological neuromorphic systems and biomimetic artificial neuron systems, optical IMCS synapses have been investigated in terms of function and materials categories and then we mainly provided a comprehensive summary of optical synapses with low-dimensional materials. Many excellent electrical performances and optical-induced synaptic behaviors of optical IMCS synapses fabricated by 0D (PQDs and silicon nanocrystals), 1D (InAs NWs and CNTs), and 2D (graphene and MoS$_2$) have been discussed with their operating mechanism. Finally, neuromorphic applications such as pattern/image recognition, mathematic calculation/logic operation, and filtering of optical synapses based on low-dimensional materials have been under discussion.

At present, various factors influence the performance of optical IMCS synapses in terms of structure, size, energy consumption, materials categories, and input source, which results in that there is no general standard to define the performance of optical IMCS synapses. For a fully optical-control synaptic device, the inhibitory and excitatory of PSC is completely affected by the power intensity, optical frequency, and wavelength of input optical stimuli. Photoinduced carriers in the functional layers determined the optical-electronic response of the device. For an optical IMCS synapse with a three-terminal structure, the inhibitory response of PSC is almost controlled by electrical stimuli and the operation power is always in the range from several
tens of nanowatts to several microwatts. Compared with the power level in the biological neuron system from 1 to 100 fJ, the artificial neuron network requires extra energy consumption during the operation process. Low-dimensional materials with high optical sensitivity, nanoscale structure, and high electrical conductivity enhance the feasibility of performance optimization and energy consumption reduction. In addition, properties like manageable transport rates of charge carrier, relatively high trap density, and atomic-level thickness structure are conducive to the implementation of 0D, 1D, and 2D materials in further research on artificial neuromorphic computing.

The final goal of researching optical IMCS synapses is to realize the high integration of the neuromorphic system in hardware while ensuring excellent device performance and reducing device energy consumption, which indicated the tendency of shrinking the size of optical IMCS synapses. The nanoscale structure of low-dimensional materials makes them become extremely competitive candidates in the future. In the field of optoelectronic devices fabricated with silicon-based materials, feasible and relative-mature experiences have been obtained when investigating computing operations based on von Neuman architecture. Related research and investigation similar to this review will positively affect the exploration of information interconnection between optical and electrical signals, which also reveals that brain-inspired optical IMCS synapses applied in neuromorphic systems will also benefit from these advanced experiences. In addition to sensing external optical stimuli, future optical IMCS synapses may integrate optical-sensing, neuromorphic computing, and nonvolatile memory to better adapt to artificial intelligence in the Internet of Things (IoT) environment.

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Conflict of Interest

The authors declare no conflict of interest.

Keywords

artificial intelligence, in-memory computing sensors, low-dimensional nanomaterials, neuromorphic networks, optical synapses
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