Journal of Neuromorphic Intelligence

REVIEW ARTICLE

VOLUME 01 | 8 MAY 2024

Transition Metal Dichalcogenides- Based Memristors for Neuromorphic Electronics

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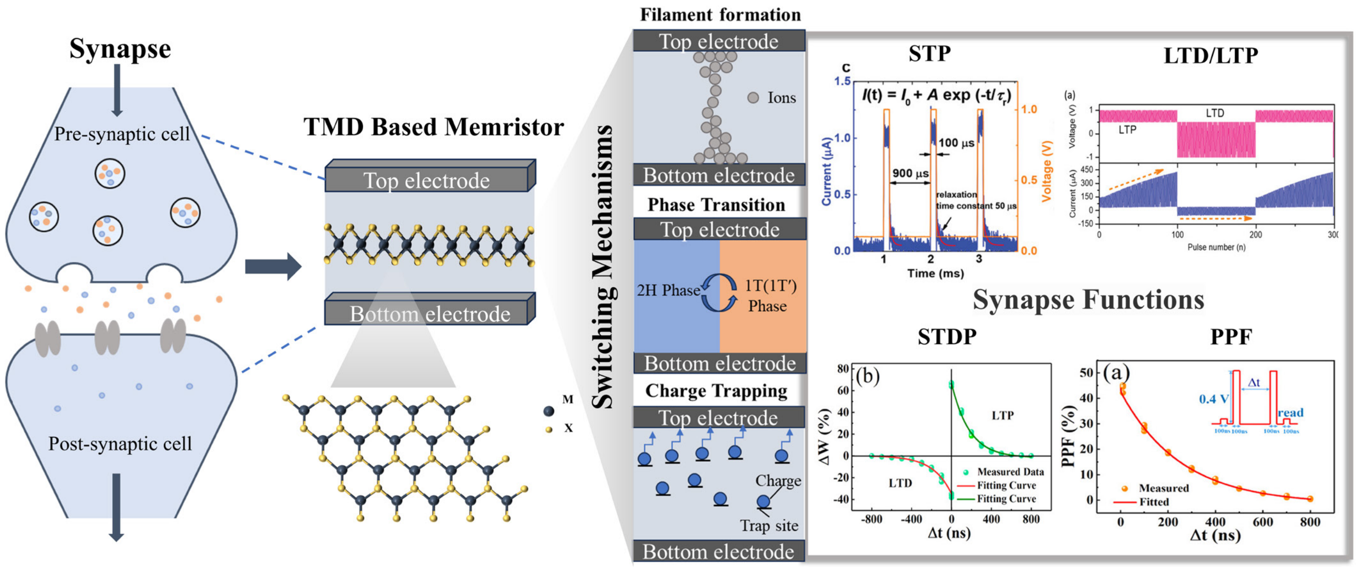

Received: 20 April 2024

Accepted: 28 April 2024

Published: 8 May 2024

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ABSTRACT

Due to the rapid advancement of neuromorphic computing, novel hardware is required to achieve high energy efficiency and performance. Memristors, a new electronic device, have gained significant attention due to their excellent non-volatile properties. These days, scientists are starting to focus on TMD materials as proper materials for making memristors to achieve neuromorphic computing due to their atomically thin nature, higher carrier mobility, weak vdW force between layers, bandgap tunability, and ability to possess multiple polymorphs. This article summarizes the latest developments in TMD material-based memristors as artificial synapses, focusing on three common mechanisms: filament formation, phase transition, and charge-trapping. This paper highlights how TMD materials contribute to achieving memristor's RS behavior, emulating biological synapses, and advancing neuromorphic electronics. Furthermore, the current challenges and future directions in this research area are discussed, indicating the potential for breakthroughs in neuromorphic computing.

Keywords: ***TMD, Memristors, Artificial synapse, Neuromorphic electronics***

1. INTRODUCTION

Neuromorphic computing represents an emerging and promising field of research that seeks to emulate the brain's distributed architecture. This approach aims to design systems capable of executing highly integrated functions with significantly reduced power consumption, trying to overcome the limitations inherent in traditional von Neumann computing architectures in which the computing and memory units are separated[1] The human brain is comprised of approximately 100 billion neurons. Each neuron communicates with an average of 1 thousand to 10 thousand other neurons by synapses, collectively generating the brain's complex and rich neural circuits responsible for transmitting and processing information[2] Synapses are critical connections between neurons, acting as fundamental units for learning and computation, enabling the efficient and concurrent storage and processing of information (Figure 1 provides a concise description of neurons and the function of synapses.). Consequently, the fabrication of artificial synapses capable of exhibiting synaptic behaviors constitutes a milestone in constructing artificial neural networks (ANNs) to realize neuromorphic computing. In 2008, the DARPA SyNAPSE program[3] sparked scientists' interest in synaptic electronics. From then on, research efforts on this topic have experienced a rapid expansion, from designing analog circuits4, 5 to beginning to focusing on using one electronic device to model the structure of single biological synapses.[6, 7] Several architectures have been introduced to achieve synapse function, such as floating-gate transistors[8-10], ferroelectric-gate transistors11, phase-change memories[12, 13] atomic switches,[14] and memristors,[15-18] utilizing changes in electrical properties to simulate the connection strength of biological synapses. Within the mentioned architectures, memristors are defined as two-terminal nonlinear dynamic electronic devices that exhibit nonvolatile resistive switching (RS) behaviors. Memristors usually have a sandwiched structure with a top electrode, an RS layer, and a bottom electrode. They are considered highly promising for constructing artificial neural networks, facilitating advancements in neuromorphic computing. This potential is primarily due to their inherent capability to retain the memory of their electrical state even after power disconnection. Their non-volatile properties enable a single, simple structure memristor to mimic synaptic functions, potentially enhancing the integration density of neuromorphic devices.[19] For several years, reports have highlighted the capability of single memristors to replicate synaptic functions. These include synaptic short-term plasticity (STP), long-term plasticity (LTP), short-term depression (STD), long-term depression (LTD), spike-rate-dependent plasticity (SRDP), spike-time-dependent plasticity (STDP), paired-pulse depression (PPD), and paired-pulse facilitation (PPF)[20-26]. Lately, Transition Metal Dichalcogenides (TMD) materials have been extensively used to create memristor-based artificial synapses that are both energy-efficient and high-performing. 2D materials possess the advantageous characteristic of being atomically thin, which allows many outstanding and unique qualities, such as flexibility,[28-30] electrical tunability,[31,14] low working voltage,[32, 33] large ON/OFF ratio,[34] and low power consumption.[35, 36] 2D materials can be simply split into individual layers due to the weak van der Waals (vdW) force interactions among adjacent layers[37] This property is particularly useful in the field of memristors. Remarkably, the TMD (with the general formula MX2, M represents a transition metal, such as Mo, W, Ta; X represents a chalcogen, such as S, Se, or Te. It usually performs like an X-M-X structure.) possesses multiple polymorphs,[38, 39] allowing for unique structural phase transitions. Also, these materials have layered crystal structures that result in highly anisotropic electrical and ionic transport properties.[40, 41] These characteristics enable them to achieve reliable and rapid-switching multilevel states with high capabilities.[42]

This article presents a comprehensive study of the latest developments in TMDs Material-Based Memristors with vertical and planar architectures used as artificial synapses. We first briefly introduce neuromorphic computing and the development of artificial synapses. Second, the working principles and advancements of TMDs-memristor-based artificial synapses, are classified into three common switching mechanisms, including filament formation memristors, phase transition memristors, and charge trapping memristors. Furthermore, how memristors achieve artificial synapses is discussed. Finally, the remaining challenge and perspective for TMDs-memristor-based artificial synapses and the future advancement

2. MEMRISTOR SWTICHING MECHANISMS BASED ON VARIOUS TMD MATERIALS

The two-terminal memristor can retain its resistance state even in the absence of power, akin to synapses' ability to sustain the str-ength of their connections between neurons. This concept can potentially create artificial synapses for constructing artificial neural networks (ANNs). TMD materials have been widely used to develop memristors with fast switching speeds, low working voltage, high electrical endurance, and low operation power[34, 44, 45] to improve neuromorphic computing efficiency. Based on the primary characteristics of I-V curves, electronic devices can be classified into three primary categories: bipolar, unipolar, and threshold (Figure 2a-c). Non-volatile bipolar memory switching is polarity-dependent, which means that the switching direction (transition between the high (HRS) and low

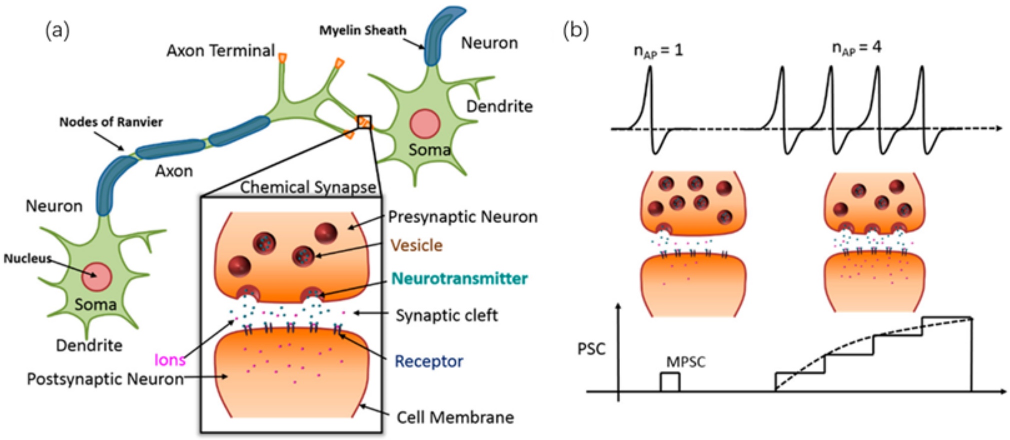

Figure 1 Bio-synapse description. (a) Neuron and synapse architecture. A biological synapse is a specialized connection between nerve cells that transmits information by releasing and receiving neurotransmitters. (b) Action potential (AP)-induced presynaptic neurotransmitter release. Vesicle exocytosis releases neurotransmitters proportionally to presynaptic AP firing frequency, which causes postsynaptic current (PSC). Adapted with permission. [27] Copyright 2017, American Chemical Society.

resistance states (LRS)) is determined by the polarity of the applied electric field. Figure 3c illustrates a typical example of bipolar switching using metal filaments formation mechanism. For non-volatile unipolar memory switching, both SET and RESET processes can occur under the same voltage polarity. The magnitude of the current or voltage controls the switching between HRS and LRS. Unlike non-volatile unipolar/bipolar switching, LRS in the threshold switching cycle can be changed back to HRS by applying a voltage below a specific threshold, which shows volatile properties. This paper discusses three common mechanisms of memristor, including filament formation, phase transition, and charge trapping. Furthermore, it examines how memristors achieve artificial synapses, such as simulating STP/STD, LTP/LTD, STDP, SRDP, PPD, and PPF.

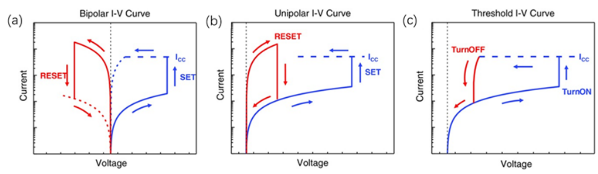

Figure 2. The schematic non-volatile memory I–V curve in (a)Bipolar switching mode and (b) Unipolar switching mode. The **S**chematic volatile I–V curve in (c) Threshold switching mode. Reproduced with permission. 46 Copyright 2017, Wiley-VCH.

Table 1: Sumary of TMDs-memristor based artifical synapses

Switching mechanisms	Structure	Set Voltage	STP STD	LTP, LTD	STDP	SRDP	PPF, PPD	Endurance cycle	ON/OFF Ratio	Retention	Ref
Metal Filaments Formation	Vertical Cu/ MoS_2/Au	0.25V	-	√	√	-	-	20	7	$1.8x10^4$s	[33]
	Vertical Ag/ MoS_2/Ag	0.18V	√	√	-	√	√	100	10^9	10^5s	[28]
	Vertical Ag/ ZrO_2/WS_2/Pt	0.16V	-	√	√	-	√	10^9	-	$4x10^4$s	[48]
Vacancy Filaments Formation	Vertical ITO/Al_2O_3/ $PdSe_2$/Al_2O_3/ TaN	0.8	√	√	-	√	√	800	10	-	[50]
Phase transition	Planar Au-Li_xMoS_2- Au	6V	-	-	-	-	-	$4x10^4$	100	$7.5x10^3$s	[56]
Charge trapping	Vertical Ag/ZnO/ WS_2/Al	1V	√	√	√	-	√	-	300	300s	[21]
	Vertical Pd/WS_2/Pt	0.6V	√	√	√	-	√	75	10^3	$1.8x10^4$s	[60]

2.1 FILAMENT FORMATION

2.1.1 METAL FILAMENT FORMATION

The mechanism of metal filament formation, based on cation migration, leads to a typical nonvolatile bipolar RS. The generation of the LRS and HRS of the memristor is caused by the creation and destruction of metal conductive filaments (CF) connecting the top electrode (TE) and bottom electrode (BE) within the RS layer. The RS characteristics are achieved through an electrochemical redox reaction of the active electrode under the influence of an electric field. Taking Cu electrode as an example, the atom of anode Cu are oxidized

$$Cu \rightarrow Cu^{2+} + 2e^{-1}$$

Then, the electric field moves Cu2+ cations through the RS layer to the cathode. Cu^{2+} then reduces with cathode electrons:

$$Cu^{2+} + 2e^{-1} \rightarrow Cu$$

Form the CF within the RS layer. Resistance will drop rapidly when the CF grows and connects the TE and BE, resulting in SET. Conversely, by applying an opposite bias voltage to the anode, the CF will progressively diminish by an oxidation reaction, increasing the resistance and leading to a RESET. Consequently, this mechanism based on the migration of cations is also known as the electrochemical metallization mechanism (ECM) .[47] The atomic-scale thinness and high carrier mobility characteristics of TMDs position them as up-and-coming candidates for the resistive switching (RS) layer in ECM memory devices. The atomic thinness significantly decreases the distance between electrodes, resulting in reduced switching voltages. Xu et al.[33] proposed a vertical MoS2 memristor as an artificial synapse with a working voltage approaching 100mV. The memristor exhibits a typical sandwich structure, with Cu as the TE and Au as the BE, with bilayer MoS2 in between (Figure 3a). Figure 3b displays the schematic of atomic-scale Cu filaments within the RS layer. The radius change of the conductive filament is related to the RS. The I-V curve (Figure 3c) clearly shows the polarity-dependent non-volatile bipolar memory switching, and the working voltage is around 100-200mV. Cu's lower migration barrier and diffusion activation energy in MoS2 than sulfur vacancies account for the low switching voltage. . What's more, this is the first time to report STDP (Figure 3d) in a vertical 2D material-based memristor. It helps achieve efficient ANNs and learn the Hebbian learning mechanism through electronics. Also, utilizing switching voltages very close to the action potentials of real neurons (around -70mV to +30mV) presents a potential avenue for novel direct interactions with mammalian neural systems. Furthermore, the employment of TMD materials in printed memristors has garnered significant attention, owing to their solution processability and flexibility. Feng et al.[28] reported a fully aerosol-jet-printed vertical cross-bar structure Ag/MoS2/Ag memristor with 0.18V switching voltage (Figure 3e). Moving silver ions across the gaps between the printed MoS2 flakes allows for fast percolation and CF formation, leading to RS. The metal filaments were confirmed on the surface of the printed MoS2 by c-AFM with different sizes and shapes (Figure 3f). This memristor could emulate both short-term (Figure 3g) and long-term memory (Figure 3h), demonstrating its promise as an artificial synapse in building efficient artificial neural networks. Furthermore, considering the human brain as a complex three-dimensional structure, developing low working voltage memristors through 2D ink printing techniques presents a promising approach for achieving hetero-integration of diverse components in three dimensions. A high endurance vertical Ag/ZrO_2/WS2/Pt memristor(Figure 3i) was reported by Yan et al. [48] Ag ions immigrate faster in WS2 than in ZrO_2, restricting the fracture and reconstruction of the metal filament in the memristor, decreasing the randomness of the CF. (Figure 3g). After 10^9 endurance cycles, the resistance of RHS and LHS remains steady in the memristor, showing excellent performance in endurance. Propose a solution for improving memristor endurance for neuromorphic electronics. Furthermore, the device successfully simulates the synaptic plasticity, including PPF (Figure 3k). Also, it achieves LTP/LTD, STDP (Figure 3l) based on the unsymmetrical Hebbian regulation, showing the ability to build reliable artificial neural system.

2.1.2 VACCANCY FILAMENT FORMATION

The process of producing non-metal filaments, which relies on the migration of anions, results in a typical nonvolatile memory. In TE and BE applications, inert electrodes or oxides such as platinum (Pt) and indium tin oxide (ITO) are often used. The RS mechanism in vacancy CF formation is associated with the movement, buildup, and reorganization of oxygen vacancies (V_{OS}), creating metal oxides with changing compositions and phases. The chemical formula can be represented as

$$O \rightleftharpoons V_o^{2+} + O^{2-}$$

TMD materials such as MoS_2 have ion barrier properties. Also, their low thermal conductivity could increase the temperature at RESET for the memristor, promoting the redox reactions[49] A vertical ITO/Al_2O_3/$PdSe_2$/Al_2O_3/TaN memristor based on vacancy CFs, which could achieve both bipolar and threshold switching and show excellent synaptic properties, is reported by Chandreswar et al[50] The $PdSe_2$ layer is incorporated to augment redox reactions during the RS process. The vacancy CF in this device is created through the motion and dispersion of oxygen vacancies and ions. Two ways of adjusting the CFs have been described in this paper: changing the amplitude and width, or frequency, of the input pulse. The dynamic excitatory postsynaptic current (EPSC) is detected under varying pulse amplitudes. Increasing the pulse amplitude highly improves the speed and strength of filament formation, resulting in a significant change in the EPSC value. This phenomenon establishes a solid basis for imitating biological synapses. The shift from STP to LTP can also be observed. Furthermore, the device can replicate short-term memory (STM) due to its ability to perform threshold switching at 800 uA, facilitated by the volatile nature of CFs produced within the switching interface under low current compliance. The device also verified that the duration of memory retention increased as the frequency increased. Weak CFs are formed at low pulse frequencies, and the device demonstrates STP properties. The robust CFs are formed at high pulse frequencies and have LTP characteristics. The PPF, PPD, and SRDP could also be simulated under this condition. Provide essential information for using the PdSe2-based memristor to fabricate an artificial synapse.

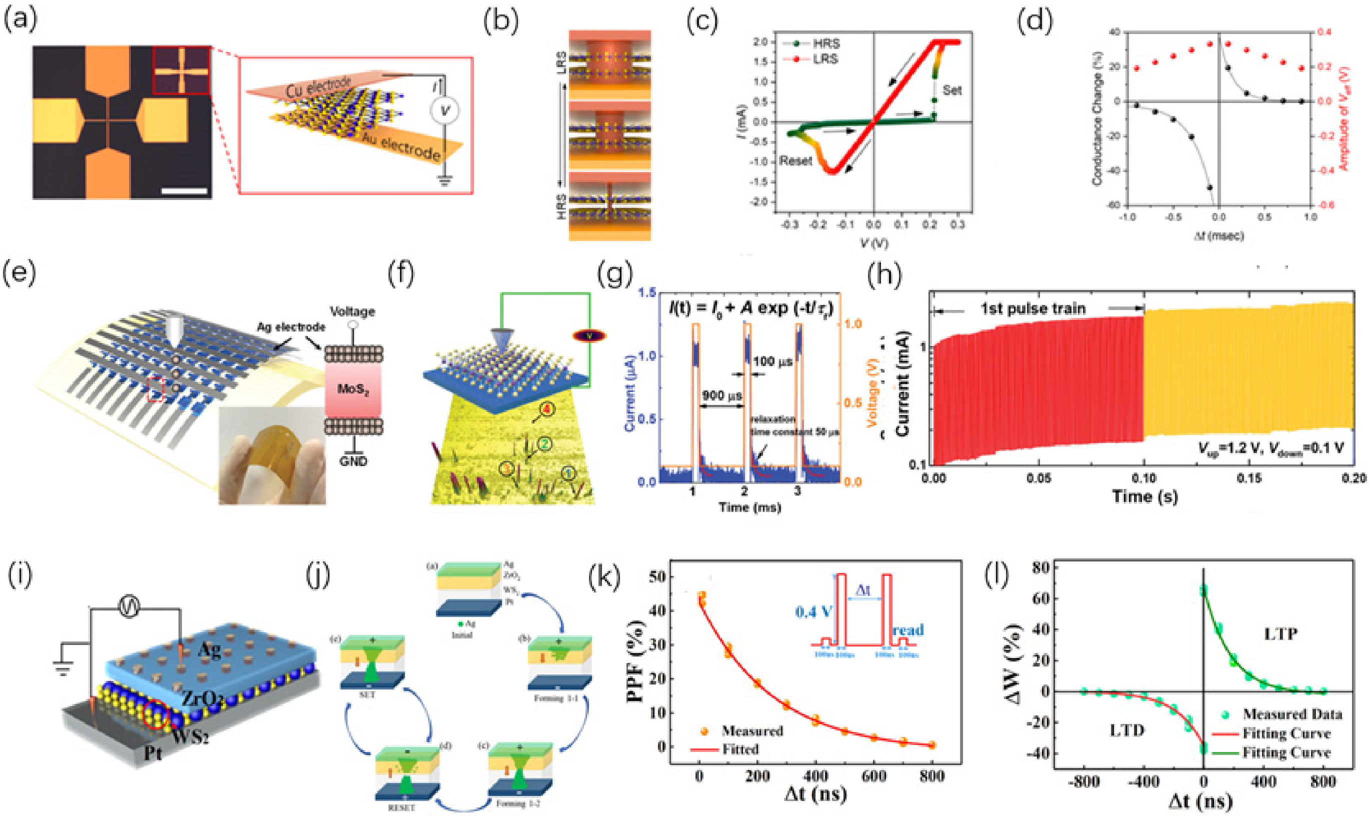

Figure 3: Metal filament formation TMD-based memristor. (a) Schematic of vertical Cu/MoS_2/Au memristor. (b) Schematic of Cu filaments within the RS layer. (c) The I-V curve of the vertical Cu/MoS_2/Au memristor shows typical polarity-dependent non-volatile bipolar memory switching; the set voltage is 0.25V, and the reset voltage is 0.15V. (d) STDP characteristics for Cu/MoS_2/Au memristor show the relationship between conductance change (%) and time (msec). (e) Schematic of vertical cross-bar structure Ag/MoS_2/Ag memristor. (f) c-AFM figure for printed MoS_2. (g) The relaxation characteristics show STP when the pulse width is 100 μs and the pulse period is 900 μs. (h) The pulse train exhibits progressive LTP with a pulse width of 0.5 ms and a Vup value of 1.2 V. (i) Schematic of vertical Ag/ZrO_2/WS_2/Pt memristor. (j) Schematic of RS mechanism of vertical Ag/ZrO_2/WS_2/Pt memristor. (k) PPF characteristics for Ag/ZrO_2/WS_2/Pt memristor. (l) STDP characteristics for Ag/ZrO_2/WS_2/Pt memristor. LTD and LTP in the figure show the negative correlation between Δt and ΔW. a–d) Adapted with permission.33 Copyright 2019, American Chemical Society. e–h) Reproduced with permission.28 Copyright 2019, The Authors, published by Wiley-VCH. Reprinted/adapted from ref.28. This is an open access article distributed under the terms of the Creative Commons CC BY license. https://creativecommons.org/licenses/by/4.0/. i-l) Adapted with permission.48 Copyright 2019, American Chemical Society.

2.2 PHASE TRANSITION

As mentioned before, TMD materials possess multiple polymorphs. Their physical properties could be changed due to the phase transition under the influence of external stimulation such as chemical,[51] thermal annealing,[52] laser,[53] mechanical.[54] Take MoS_2 as an example, MoS_2 can transition from a semiconductor to a metal by undergoing a change from its trigonal prismatic 2H phase to its octahedral 1T(1T′) phase.[55] Also, TMD materials allow different structural phases (such as 1T, 1T′, 2H phase) within a single TMD film, allowing for the creation of devices that rely exclusively on the specific configurations of these phases to determine their functionality. Zhu et al.[56] first report a kind of planar Au-Li_xMoS_2-Au memristor that could reversibly achieve 2H–1T′ phase transitions by the concentration of ions (Li+). The device is intercalated with a high-density ion (Li+) solution. The phase transitions between 2H and 1T′ within Li_xMoS_2 are controlled by the concentration of ions (Li+). They controlled the ion concentration by adding negative (positive) voltage to electrode A, which attracts (repels) Li+ ions and lowers (increases) the ion (Li+) concentration near electrode B. When the density of ions (Li+) increases, the 2H phase changes to the 1T′ phase. Conversely, the 2H phase reverses to the 1T′ phase. The device shows typical non-volatile memristive effects. Furthermore, Zhu and co-workers simulate synaptic competition and cooperation in building artificial neural networks by controlling the movement of Li+ in the fabricated network structure. Multi-terminal inputs activate neighboring synapses, which can be achieved based on the interplay between ionic and electronic coupling. This result demonstrates the potential for manipulating the phase of TMD materials using field-driven ionic processes to create artificial neural networks. Hou et al.[54] present a study on a vertical multilayer MoTe2 memristor based on the phase transition mechanism. They utilize process-induced strain engineering to achieve 2H(semiconducting)–1T′(semimetallic) phase transitions. The fabricated memristor has excellent performance with low switching voltage (90 mV), high on/off ratios (108), and long retention (over 10^5s), which shows potential for achieving neuromorphic computing with low power.

2.3 CHARGE TRAPPING

The atomic-scale thickness of TMD materials renders them highly vulnerable to inherent defects. These defects could sometimes provide a stable trap effect with significant promise in non-volatile memory[57] In TMD material-based memristors, charge trapping and de-trapping occur due to defects and dangling bonds within a charge-trapping interfacial layer, which can trap charge carriers (electrons or holes)[58] Electrons or holes are introduced into the RS layer by voltage and can trap at defect sites. The charge trapping mechanism reduces device resistance by adsorbing metal atoms or ions on surface defects or inserting them into semiconducting TMD materials. Using this mechanism, we could achieve synaptic functions such as LTP/ LTD, PPF, and STDP. Deshmukh et al[59] report a Al/CPB QDs/MoS2/FTO memristor (Figure 4a). The addition of CPB Quantum dots (QDs) has significantly enhanced the RS performance of the memristor. The observed phenomenon can be attributed to the transfer of charges across CPB QDs and MoS_2 under high-voltage conditions, which results in a transition of the conductive mechanism from ohmic conduction to space charge-limited conduction. Kumar et al[21] report a vertical Ag/ZnO/WS2/Al memristor((Figure 4b) with high spatial/temporal performance. The I-V curve (Figure 4c) shows the polarity-dependent non-volatile bipolar switching. XPS results demonstrate that the gradient of oxygen vacancies from the ZnO region to the WS_2 region induces the process of charge trapping and de-trapping (Figure 4d). Also, the vertical Ag/ZnO/WS2/Al memristor shows great potential for mimicking bio-synapses. LTP and LTD are found in the device by applying 100 programmed positive/negative voltage pulses. As the quantity of positive voltage pulses escalates, the current progressively increases, and upon the application of a negative voltage pulse, it demonstrates a suppressive impact (Figure 4e). At the same time, STDP is emulated by the device (Figure 4f), consistent with the asymmetric Hebbian rule. Yan et al.[60] proposed a vertical Pd/WS2/Pt memristor with electron hopping between sulfur (Vs) and tungsten vacancies(Vw) to imitate synaptic functions. (Figure 4g). The device exhibits a phenomenon where increasing the pulse interval results in a lesser increase in the EPSC amplitude, indicating a PPF-like behavior. (Figure 4h). The STDP characteristics could also be simulated (Figure 4i). Moreover, according to density functional theory, the defect states of Vw and Vs display deep and confined characteristics, resulting in decreased leakage current and power consumption.

This paper presents an excellent opportunity to optimize neuromorphic computing efficiency.

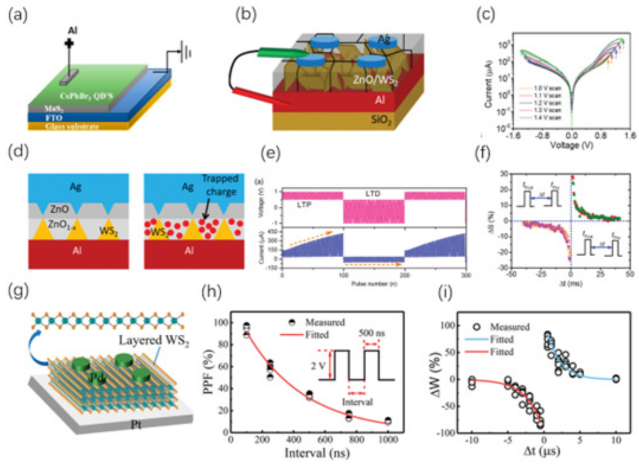

Figure 4. Charge trapping TMD-based memristor. (a) Schematic of vertical Al/CPB QDs/MoS_2/FTO memristor. (b) Schematic of vertical Ag/ZnO/WS_2/Al memristor. (c) I-V curve of the Ag/ZnO/WS_2/Al memristor shows non-volatile bipolar switching. (d) Schematic of the Ag/ZnO/WS_2/Al memristor based on XPS measurements. (e) Under 100 programmed positive/negative voltage pulse, the device shows synaptic functions: LTP/LTD. (f) Experimental results show STDP phenomenon. (g) Schematic of vertical Pd/WS_2/Pt memristor. (h) The relationship between the PPF ratio and pulse interval. (i) The STDP response is illustrated by changes in device weight with Δt variation. a) Reprinted with permission Copyright 2023, American Chemical Society. b-f) Reproduced with permission. [21] Copyright 2019, Wiley-VCH. g-i) Reproduced with permission.[60] Copyright 2019, Wiley-VCH.

3. CHALLANGES AND PERSPECTIVE

Upon reviewing the latest advancements in TMD material-based memristors employed as artificial synapses, it becomes evident that scientists have made significant efforts to leverage the distinctive characteristics of TMD materials. These include their atomically thin nature, higher carrier mobility, weak vdW force between layers, bandgap tunability, and ability to possess multiple polymorphs. These characteristics allow TMD material to be a potential material for many RS mechanisms, such as filament formation, phase changing and charge trapping. The objective is to achieve low working voltage, high endurance, a high switching ratio, and low operation power in memristors. Furthermore, these characteristics of TMD materials provide the memristor-based synapses potential to simulate intricate ion transport among adjacent neurons accurately and achieve synaptic functions, such as STP/STD, LTP/LTD, STDP, SRDP, PPD, and PPF, even more efficiently. Although scientists have researched TMD material memristor-based artificial synapses in neuromorphic computing for decades, challenges remain to improve reliability in the RS process. For example, the defects in the TMD material layer would impact the formation and rupture of the metallic CF. Furthermore, controlling redox reactions near the CF in TMD materials is challenging. These may lead to unpredictable RS behavior, making it challenging to achieve consistent device performance. Also, large-scale memristor arrays have been rarely reported. Current large-scale manufacturing methods for TMD materials, such as chemical vapor deposition (CVD) and solution-based processes, each exhibit inherent flaws, highlighting the challenges in integration and future industrialization. For future neuromorphic computing, using TMD material-memristor-based artificial synapses with numerous connections can create intricate circuits that emulate the human brain, facilitating complex and deep learning. Moreover, the photosensitivity exhibited by TMD materials presents a promising avenue for achieving associative learning. Based on human sensory systems, including vision, hearing, touch, taste, smell, to create devices that can respond to different external stimuli. Establishing an efficient and intelligent information perception system incorporating biological reality can facilitate the development of more intelligent bio-inspired robots

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ACKNOWLEDGMENTS

The authors are grateful for the funding support from Texas A&M University.

COMPETING INTERESTS

The authors declare no competing interests.