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Review

Two-dimensional materials for synaptic electronics and neuromorphic systems

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ABSTRACT

Synapses in biology provide a variety of functions for the neural system. Artificial synaptic electronics that mimic the biological neuron functions are basic building blocks and developing novel artificial synapses is essential for neuromorphic computation. Inspired by the unique features of biological synapses that the basic connection components of the nervous system and the parallelism, low power consumption, fault tolerance, self-learning and robustness of biological neural systems, artificial synaptic electronics and neuromorphic systems have the potential to overcome the traditional von Neumann bottleneck and create a new paradigm for dealing with complex problems such as pattern recognition, image classification, decision making and associative learning. Nowadays, two-dimensional (2D) materials have drawn great attention in simulating synaptic dynamic plasticity and neuromorphic computing with their unique properties. Here we describe the basic concepts of bio-synaptic plasticity and learning, the 2D materials library and its preparation. We review recent advances in synaptic electronics and artificial neuromorphic systems based on 2D materials and provide our perspective in utilizing 2D materials to implement synaptic electronics and neuromorphic systems in hardware.

1. Introduction

In 1945, von Neumann first proposed the concept of "storage program" and the binary principle [1]. Later, the electronic computer systems designed with this concept and principle were collectively referred to as the "von Neumann-type structure" computer. The von Neumann architecture processor uses the same memory and is transmitted over the same bus [2]. Although the von Neumann architecture has achieved unprecedented success in dealing with many problems, it is not perfect to separate the central processing unit (CPU) from the memory. The traffic between the CPU and the memory (data transfer rate) is quite small compared to the memory capacity. In modern computers, the traffic is very small compared to the CPU efficiency. In some case (when the CPU needs to execute some simple instructions on huge data), data traffic becomes a very serious limitation of overall efficiency. The CPU will be idle when the data is input or output, which leads to the so-called von Neumann bottleneck [3]. In addition, the power consumption and efficiency of modern computers cannot be well balanced when dealing with complex system problems. The significant difference between modern computers and the human brain lies in the form of operation of memory and processing. Inspired by the biological nervous system, Alan Turing proposed a new type of computing system in 1948: a machine consisting of artificial neurons (i.e., simple processing units) that are connected together by modifiers between them. It can effectively solve the intensive computation problems limited by the von Neumann architecture with an acceptable energy consumption, owing to the inherent analog capability and efficient parallel connection [4]. In the era of big data and high-speed information, it is more urgent to solve problems such as pattern recognition, sound localization, unsupervised learning, association learning, adaptability, and energy efficiency etc. [5]. Consequently, neural network hardware architecture inspired by biological systems such as the human brain is being extensively explored by society.

Inspired by the unique features of biological synapses that the basic connection components of the nervous system and the parallelism, low power consumption, fault tolerance, self-learning and robustness of biological neural systems, artificial synaptic electronics and neuromorphic systems have the potential to overcome the traditional von Neumann bottleneck and create a new paradigm for dealing with big data and complex problems. Neuromorphic computation can be divided into software and hardware implementation: (1) software implementation (such as Google AlphaGo) mainly adopts von Neumann structure, which is still limited by high power consumption, low energy efficiency and integration density; (2) traditional hardware implementation methods mainly include static random access memory (SRAM),

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resistive change memory (RRAM), phase change memory (PRAM), etc. Neural synaptic electronics and neuromorphic computing network systems based on a variety of materials and mechanisms are in rapid development, including metal oxide (such as AlO_x [6–8], HfO_x [9–13], TiO_x [14–16], WO_x [17,18], TaO_x [19], etc.) RRAM [6,20–25], Ag or Cu based conductive bridge synapses [16,26–35], ferroelectric material based synapses [36–39], phase change (PC) material based synapses [40–46], magneto-resistive (MR) material based synapses [47–49], carbon nanotube (CNT) based synapses [50–53] and nanoparticles-organic molecular based synapses [54–57], etc. However, development is also subject to key factors such as insufficient formation mechanism, lack of reliability and low integration density. Traditional software and hardware strategies are faced with bottlenecks and limitations (high power consumption, reliability, low integration, mechanisms, etc.). A new material or structure based system is urgently needed to promote the development of neuromorphic/brain-like computing and meet the era of artificial intelligence big data. For hardware ways, researchers are always looking for new materials or architectures to implement neuromorphic networks/brain-like computing. The first thing that comes to mind is the two-dimensional (2D) materials that have received much attention: with atomic thickness (negligible volume), clean surfaces without dangling bonds, peculiar physical properties (metal-insulator transition), high integration density and possible low energy consumption operation. It is considered an important candidate for future memory and neuromorphic/brain-like computing technologies. More recently, 2D materials have drawn great attention in simulating synaptic dynamic plasticity and neuromorphic computing with their unique properties. The discovery of exfoliated few layers or monolayer graphene and its extraordinary characteristics have triggered great interest on the new kind of materials known as "2D materials" [58–60]. Graphene, possessing perfectly mechanical, electronic and optical properties, had sparked enormous interest to wide exploration. Nevertheless, it still has many drawbacks (for instance, pristine graphene has a zero bandgap, which is not suitable for electronic and optoelectronic applications [61]) that have yet to be overcome [62]. Meanwhile, an increasing number of research groups concentrate on other novel various 2D materials such as transition metal dichalcogenides (TMDCs, i.e., MoS_2, WSe_2), black phosphorus (BP), hexagonal boron nitride (h-BN). With its atomic-level thickness, 2D materials can effectively reduce the energy consumption caused by short-channel effects, achieve the equilibriums between consumption and efficiency, and can scale down the device size to further increase integration, as close as possible to the human brain. 2D materials exhibit great compatibility in tuning the electronic features, which make it viable by altering the number of layers in a given material to realize the band-gap engineering [63,64], and which make it possible to achieve different resistance or conductance states with low power consumption, the key characteristic of synaptic plasticity emulation [65]. Various 2D materials have a rich band structure, including insulators, semiconductors, semi-metals, metals and superconductors, indicating their versatility. 2D semiconductors have high carrier concentration and mobility, which is a good alternative for Si-based channel materials and offers the possibility to achieve synaptic plasticity and neuromorphic computation through carrier dynamics either internal or at the interface. In general, 2D materials open up a whole new path for unprecedented parallel, energy-efficient, fault-tolerant synaptic electronics and hardware design neuromorphic computing systems.

Our review will center on the processes in neural synaptic devices and neuromorphic computing systems based on two-dimensional materials. First, we will give a brief introduction of synaptic plasticity and learning behavior. Secondly, we present the main 2D material library and preparation process. Subsequently, synaptic devices and artificial neuromorphic network systems based on various 2D materials will be discussed. At last, we summarize the opportunities and challenges faced by 2D-based synaptic devices and neuromorphic computing systems, and foresee a glorious future of the hardware implementation of the brain-like neural architecture.

2. Synaptic plasticity and Hebbian learning

Two different types of synapses: excitatory and inhibitory synapses, which depend on the type of neurotransmitter receptor on the synaptic membrane. Excitatory synapses correspond to excitatory post-synaptic potentials (EPSP), and postsynaptic neurons produce an action potential. Inhibitory synapses correspond to the production of an inhibitory post-synaptic potential (IPSP), and postsynaptic neurons do not reach the threshold for the formation of action potentials. In neuroscience, synaptic plasticity refers to the connection between nerve cells, that is, synapses, whose connection strength is adjustable. A characteristic or phenomenon in which the surface morphology and function of a synapse undergoes a lasting change [66]. Synapses will be strengthened and weakened as their activities are strengthened and weakened. Synaptic plasticity is considered to be an important neurochemical basis for memory and learning. Synaptic plasticity can occur for a variety of reasons, such as changes in the number of neurotransmitters released in synapses, and the efficiency of cellular responses to neurotransmitters [67].

Synaptic plasticity mainly includes short-term synaptic plasticity and long-term synaptic plasticity. Short-term plasticity (STP) mainly includes facilitation, inhibition, and potentiation [68]. Paired-pulse depression (PPD)/facilitation (PPF) is a dynamic reduction/enhancement of neurotransmitter release, which is considered to be the key to transmitting information in biological synapses, that is, the inhibition/promoting effect of presynaptic-induced IPSC/EPSC weakens or even disappears with the increase of the interval time (Δt) between two consecutive pulse signals. For small Δt (tens to thousands of milliseconds), the inhibitory synapse further inhibits the magnitude A of the postsynaptic current, resulting in $A_2 < A_1$, and is enhanced by excitatory synapses, i.e., $A_2 > A_1$, corresponding to PPD/PPF. The following double exponential decay function can be used for fitting [69]

$$A_2/A_1 = C_1 \times \exp(-\Delta t/t_1) + C_2 \times \exp(-\Delta t/t_2) + C_0, \quad (1)$$

Δt is the time interval between pairs of consecutive pulse signals, C_1 and C_2 are the initial magnitudes of the fast and slow phases, the relaxation times of the fast and slow phases are represented by t_1 and t_2, respectively. Short-term synaptic plasticity is an important manifestation of synaptic plasticity and plays an important role in achieving the normal function of the nervous system. The short-term plasticity of synapses can enhance the certainty of synaptic transmission, regulate the balance between excitation and inhibition of the cerebral cortex, form the temporal and spatial characteristics of neural activity, and form and regulate the synchronous oscillation of the cortical thalamic network [70]. In addition, synaptic short-term plasticity may also be involved in the realization of advanced functions of the nervous system such as attention, priming effects, sleep rhythm and learning and memory.

Long-term plasticity (LTP), including long-term potentiation and long-term inhibition, has been recognized as the biological basis for the level of cellular learning and memory activity. Long-term potentiation is a persistent enhancement that occurs in the transmission of two neurons due to the simultaneous stimulation of two neurons [71]. This is one of several phenomena associated with synaptic plasticity, the ability of synapses to change intensity. Since memory is thought to be encoded by changes in synaptic strength, long-term potentiation is widely regarded as one of the main molecular mechanisms that underlie learning and memory [72]. Besides, long-term potentiation has several features, including input specificity, relevance, synergy, and persistence. Otherwise, long-term depression (LTD), also known as long-term inhibition, long-term inhibition, refers to the inhibition behavior of nerve synapses for several hours to several days [73]. Strong synaptic stimuli (Cerebellum Purkinje cells) or long-term weak synaptic stimuli (hippocampus) can lead to LTD. Long-term inhibition is usually thought to be caused by changes in post-synaptic receptor density, but changes in the release of presynaptic neurotransmitters may also have an effect. The LTD of the cerebellum is assumed to play an important role in the learning of motor nerves, and the LTD of the hippocampus is thought to have a special effect on clearing past memories [72]. The effect of LTP is long-lasting and can last for a few minutes or even months, which is the fundamental difference between it and other synaptic plasticity. Nowadays, artificial neural networks mainly use the learning and memory function of nerve cells. In the construction of neural networks, the application of the theory of synaptic plasticity is indispensable.

The Hebbian theory proposed by Donald Olding Hebb in 1949 describes the basic principle of synaptic plasticity, that is, the continuous repetitive stimulation of presynaptic neurons to postsynaptic neurons can lead to an increase in synaptic transmission efficiency [74]. This theory is often summed up as "cells that fire together, wire together" [75,76], which can be used to explain "associative learning", in which the nerves are passed through the stimulation of the element increases the synaptic strength between the neurons. However, Hebb emphasized that the neuron "A" must make a certain contribution to the excitation of the neuron "B", i.e., the excitation of the neuron "A" must precede the neuron "B" and not simultaneously. This part of the study in Hebbian theory, later called spike-timing-dependent plasticity (STDP), suggests that synaptic plasticity requires a certain time delay [77]. STDP is an indicator of the development of nervous system activity, particularly in terms of long-term potentiation and LTD. The process of synaptic connection weight (ΔW) varies with the relative time (t_{post}–t_{pre}) of pre- and post-synaptic pulse spikes. The presynaptic pulse precedes the post-synaptic pulse for a period of time ($t_{post}-t_{pre} > 0$), and the synaptic connection is strengthened to form a long-term potentiation effect. Otherwise ($t_{post}-t_{pre} < 0$), a LTD effect is formed typically. Fig. 1 shows the different forms of STDP in biological synapses. In addition to STDP, spike-rate-dependent plasticity (SRDP) is also a basic learning rule in LTP Hebbian theory [78], i.e., the process of synaptic weights varies with the frequency of the presynaptic spike signal transmission: a series of high frequency (20–100 Hz) presynaptic spike signals result in long-term potentiation, while low frequency (1–5 Hz) signals result in LTD typically [79]. Such a learning method is called Hebbian learning, and Hebbian learning has also become the biological basis of unsupervised learning.

Contemporarily, 2D materials have gained extensive attention and progress in simulating synaptic plasticity and neuromorphic computation due to their superior electrical and optical properties. In the following sections of this review, we will detail the synaptic devices and neural networks computing based on various 2D materials.

3. 2D material library and preparation

3.1. 2D material library

The library of 2D materials has been expanding rapidly since the first exfoliation of graphene. The versatility of 2D material, including conductors, semimetals, semiconductors with varying bandgaps and insulators, provides greatly flexibility of van der Waals stacks building, as shown in Fig. 2. Among these 2D material candidates, graphene, black phosphorus, h-BN, and TMDCs attract most scientific and engineering interest, due to the stable chemical, thermal and complementary properties. Because of the hexagonal rings of carbon atoms, graphene has a zero bandgap and remarkable high mobility of carries. Therefore, graphene presents as conductive 2D material and usually used as contact electrode [81,82] and assembled with 2D semiconductor forming van der Waals heterostructures. Graphene electrode can avoid the Fermi-level pinning effect of the Schottky-barrier [83,84], so as to reduce contact resistance [85,86]. The TMDC semiconductors have a varying bandgap in the range of 1–2 eV. For instance, MoS_2 has indirect bandgap of 1.4 eV in the bulk state, while it transits into direct bandgap of 1.8 eV at monolayers state [63]. Semiconducting TMDCs are promising candidates for channel in transistors, due to the free of dangling bonds, layers of atomic thickness and air-stable property. The h-BN is in sequence of hexagonal layers with alternating boron and nitrogen atoms. The h-BN has a wide bandgap (~6 eV) of insulator and is very compatible to mechanical transfer and chemical process, so that it always be used an encapsulation layers [87]. Besides, the h-BN layer has an atomically smooth surface to minimize the effect of defects, which is essential for gate dielectrics [88]. The BP is the most stable allotrope of phosphorus. It has a 0.33 eV direct bandgap in the bulk, and the bandgap can be expanded to more than 1.0 eV if black phosphorus is in monolayer form [89]. The bandgap of BP has filled the gap between that of TMDCs and graphene, which is highly important to 2D material class. BP typically exhibits as p-type characteristic, but the origin is unclear. Transistors have been researched with BP channels, exhibiting larger drive current compared to most of TMDCs based transistors.

3.2. 2D material preparation

The preparation methods of 2D materials mainly include the following: exfoliation, epitaxial growth, chemical vapour deposition (CVD) growth and hydrothermal growth. Each method has its own advantages and disadvantages, and is suitable for device preparation requirements of different scales. The micromechanical exfoliation method relies on the 2D layered material interlayer van der Waals force to strip a monolayer or multilayer film of graphene [90–92], MoS_2 [93–95], WS_2 [96], WSe_2 [92,94,97], h-BN

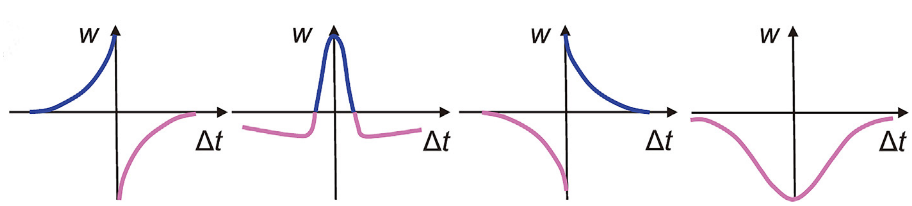

Fig. 1. (Color online) Different forms of STDP in biological synapses, where the change in synaptic weight depends on the stimulation time interval.

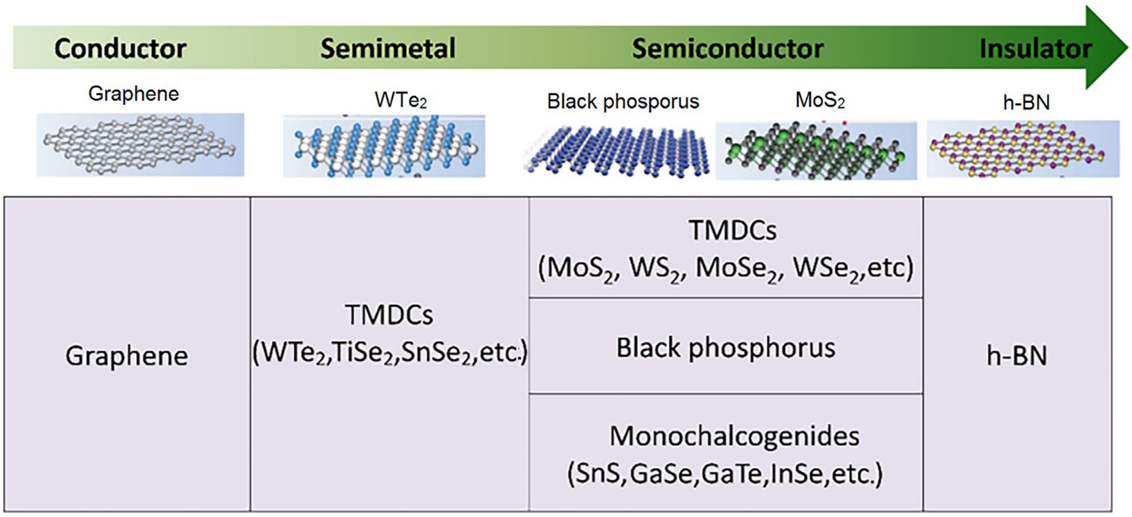

Fig. 2. (Color online) The library of 2D materials, covering conductors, semimetals, semiconductors and insulators [80].

[91,92,95,98], BP [99,100] etc. from a bulk crystal to prepare various electronic and optoelectronic devices. The biggest advantage of micromechanical exfoliation is that it is easy to operate. However, his limitations are non-extendability, the uniform area of the stripped film is small (about 5–20 μm). Another method for scalable growth of 2D materials is epitaxial growth, that is molecular beam epitaxy (MBE) or van der Waals epitaxy. Because the surface of the 2D material has no dangling bonds, it can be grown on the surface of the layered material and the inert substrate by interlayer van der Waals interaction without requiring lattice matching, which is different from traditional epitaxial growth [101]. However, if the substrate and the overlapping layer lattices do not match, the grown film may be polycrystalline or have more dislocation defects [102]. The most common method for growing 2D films today is CVD. The film domain grown by CVD has good electrical optical properties and room temperature mobility [103], but the presence of grain boundaries degrades the mobility of the film material [104]. CVD can grow a variety of different TMDCs, including MoS_2 [105], $MoSe_2$ [106,107], WS_2 [108,109], WSe_2 [110,111], ReS_2 [112,113], $ReSe_2$ [114], $MoTe_2$ [115], WTe_2 [116], etc. In addition, it can also be used to grow various novel 2D layered heterostructures [117,118], but the electrical properties of large-area films need to be further improved. Recently, hydrothermal growth has been used to synthesize 2D materials. It is reported that the monolayers MoS_2 and $MoSe_2$ with no restacking tendencies are synthesized based on the reaction of ammonium molybdate with sulfur and selenium in a solution of hydrazine hydrate in 2001 [119]. Inspired by the synthesis of MoS_2, hydrothermal methods have expanded to the growth of other layered 2D materials, including group IV and V TMDCs [120] and h-BN [121,122] etc.

4. Synaptic electronics and artificial neuromorphic network systems implemented by 2D materials

Imitating the classification of resistance switching mechanism by Waser [24], according to the physicochemical mechanism of dominant synaptic behavior, synaptic electronics and artificial neural network systems based on 2D materials can be divided into electrostatic/electron ion synapses, electrochemical metallization/conductive bridge synapses, oxidation reductive/chemical valence synapses, phase change synapses and thermochemical synapses, etc. A detailed description of each type of synaptic electronics and neural network will be given below.

4.1. Electrostatic/electronic(ionic) neural synapses and neuromorphic network

4.1.1. Transistor-based electrostatic/electronic ion synaptic devices and networks

There are some impurities and defect states in the dielectric material. When the impurity levels and defect levels in the forbidden band are occupied and released by electrons/ions, the electron/ions transport ability in the 2D material channel is affected, forming synaptic plasticity. Recently, a polycrystalline single-layer

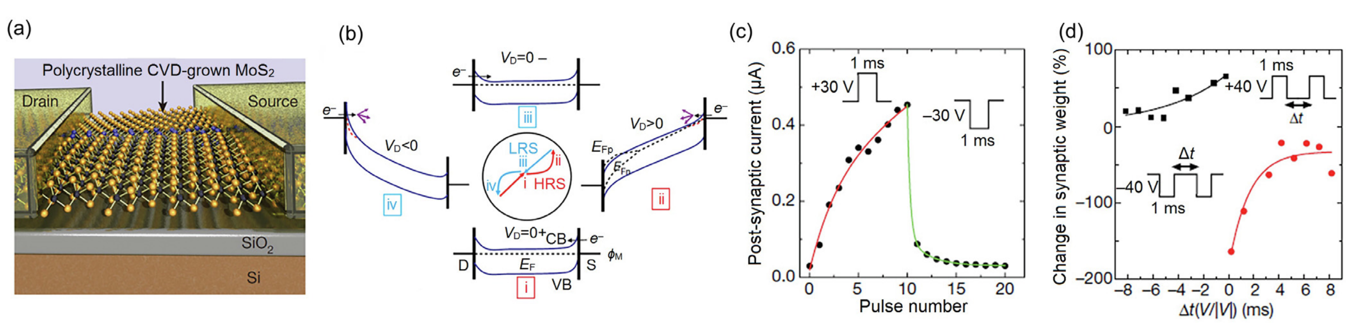

Fig. 3. (Color online) Six-terminal MoS_2 memtransistors with gate-tunable synaptic function and complex neuromorphic learning. (a) Schematic of the MoS_2 memtransistor. (b) Schematic of the energy band corresponding to four different states of MoS_2 memtransistor. The arrow represents the defect migration. (c) The post-synaptic current as a function of the pulse number, showing long-term potentiation and inhibition of +30 and −30 V pulses. (d) The change in synaptic weight as a function of the paired pulses time interval.

MoS_2 multi-terminal hybrid memtransistor based on the bias-induced MoS_2 defect moving resistance switching mechanism has been demonstrated by Sangwan et al. [123]. They realized traditional long-term potentiation/depression neuro-learning behavior and six-terminal MoS_2 memtransistors with gate-tunable synaptic function (Fig. 3), demonstrating to the community that integrating memristors and transistors into a multi-terminal device enables complex neuromorphic learning and defect dynamics of 2D materials. Prior to this, Arnold et al. [124] have reported that hysteresis-based MoS_2 transistors are used to mimic the release of neurotransmitters in synapses. Arnold et al. accurately captured the randomness and excitatory or inhibitory properties of neurotransmitter release in experiments, and also mimicked and successfully modulated the basis of human brain memory formation, namely long-term potentiation phenomenon. Synaptic and neuromorphic networks based on static or electronic ions have been widely reported in recent years, including 2D MoS_2 neural transistors with basic synaptic function made by poly(vinyl alcohol) electrolyte in Jiang et al.'s work [125]. Poly(vinyl alcohol) electrolyte-based 2D MoS_2 electric double-layer transistor simulates spatiotemporal processing of visual neurons [126], synaptic transistors based on bias modulated ultra-thin MoS_2 of Wang et al.'s work [127]. MoS_2 memristive transistor with optical short-term plasticity at room temperature, based on 1T-phase quantum dot superlattice and 2H-phase single layer single crystal [128], three (electron, ionic, and photoactive) modes of the MoS_2 neural device were used to simulate intracellular ion dynamics and neurotransmitter release in chemical synapses, and simulations of classical conditional experiments were successfully performed as shown in Fig. 4 [129].

In addition to MoS_2, there are reports of electrostatic/electronic (ionic) synapses and neuromorphic networks based on other two-dimensional materials, including twisted graphene dynamic synapse demonstrated by Ren and co-workers [130], and achieved both synaptic excitatory and inhibitory behaviors owing to the ambipolar conductance of graphene. Tian et al. [131] introduced an field-effect transistor (FET) mode of Al/AlO_x/graphene stack for simulating synaptic plasticity and neuromorphic computing. In addition, Boolean logic computing, temporal and spatial dendritic integration and principle-proven visual networks for simulating Lobula Giant Motion Detector (LGMD) neurons have been implemented by Wan et al. [132] using metal oxide/graphene oxide hybrid transistors as shown in Fig. 5. In the same year, they also demonstrated the orientation tuning characteristics through a transistor coupled with proton conductive graphene oxide [133]. Obviously, optical stimulation has more flexible and adjustable synaptic plasticity than electrical stimulation, such as graphene-carbon nanotube hybrid phototransistor reported by Qin et al [134]. For the first time, Tian et al. [135] realized an anisotropic synaptic device based on PO_x/BP heterojunction, which achieved synaptic behaviors through charge transfer between heterostructure interfaces and proved a simplified multi-synaptic network. Besides, Zhu et al. [136] reported an ion-gated synaptic transistor based on WSe_2 and phosphorus trichalcogenides with a variety of short-term and long-term plasticity and power consumption as low as 30 fJ (per spike). Moreover, Tian et al. [137] simulated the dynamic reconfigurable excitatory and inhibitory synaptic behavior using the tunable electronic properties of black phosphorus and tin selenide heterostructures. The heterojunction between BP and SnSe produces tunable rectifying electrical properties, and

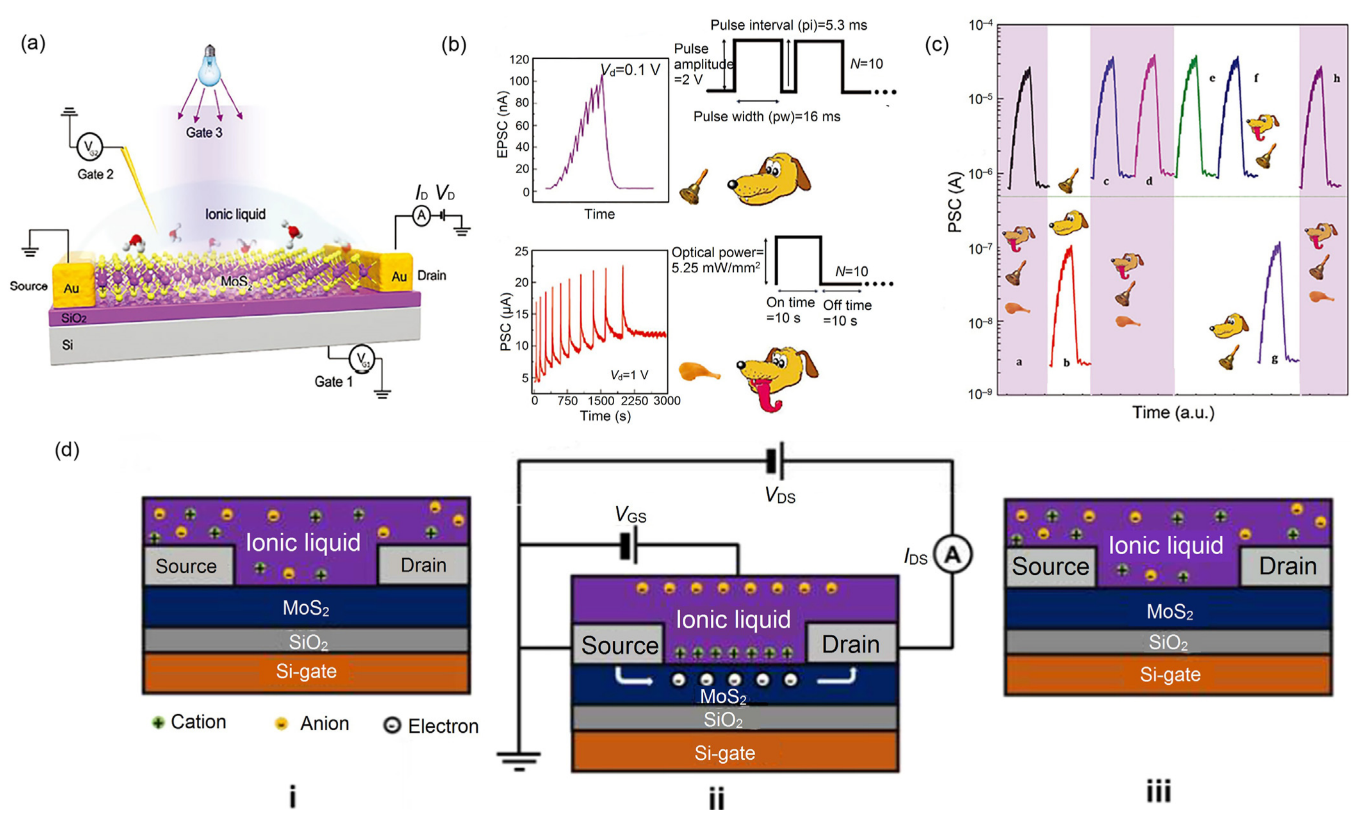

Fig. 4. (Color online) Electro-iono-photoactive MoS_2 neuristors. (a) Three modes (electron, ion electron and photoactive) of the MoS_2 neural device. (b) Electrical pulse as conditional stimulus and optical pulse as unconditional stimulation simulated classical conditional experiment. (c) Realize the simulation of Pavlov's dog classical conditional experiment. (d) Working mechanism of MoS_2 electric-double-layer transistor in ionotronic-mode.

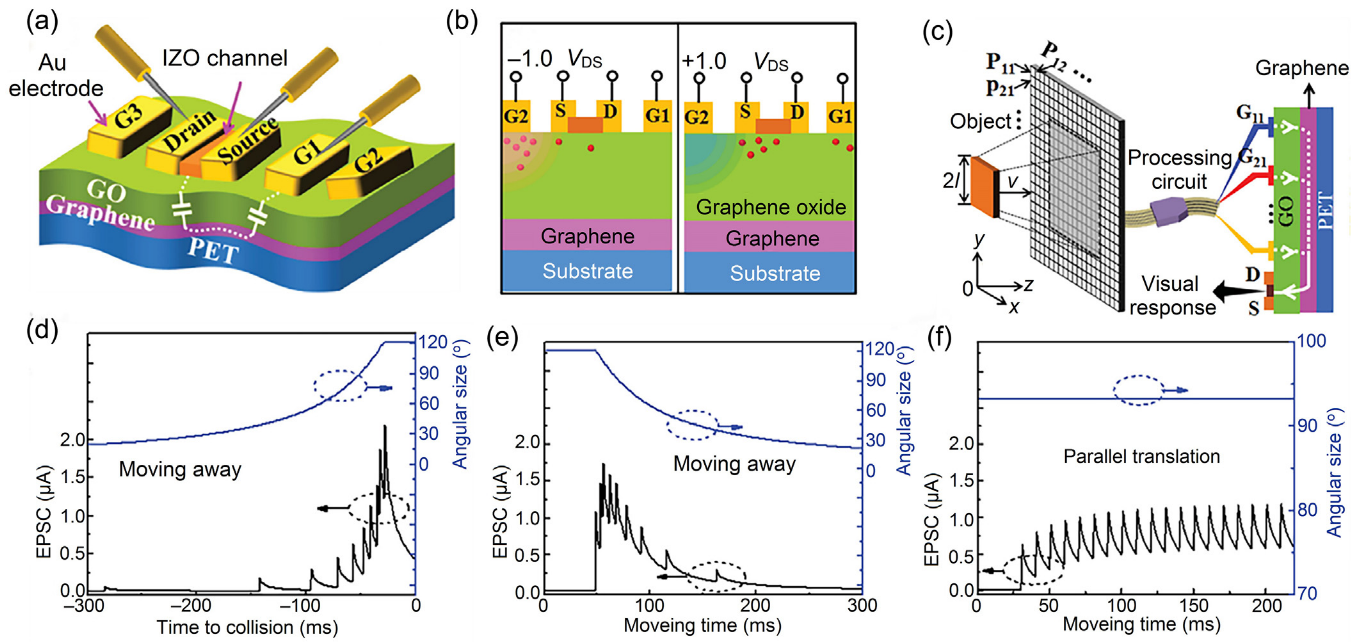

Fig. 5. (Color online) Metal oxide/graphene oxide hybrid neuromorphic transistors for imitating the LGMD neuron. (a) Schematic of a tri-gate flexible neuromorphic transistor. (b) Schematic diagram of proton distribution at different VGS. (c) Principle-proven visual networks for simulating LGMD by metal oxide/graphene oxide hybrid transistors. (d–f) The visual system records the EPSC output in response to the object moving parallel/ away/toward the array of photoreceptor, respectively.

charge transfer between the naturally oxidized BP layer and the channel BP layer results in synaptic plasticity.

4.1.2. RRAM-based electrostatic/electronic ion synaptic devices and networks

In addition to transistor-based electrostatic/electronic ion synaptic devices and networks, there are reports of synapses and neuromorphic networks based on RRAM structures, including a graphene electrochemical synapse that conductance is reversibly modulated by Li ions by Xiong and co-workers [138]. Li et al. [139] studied the switching characteristics of mechanically printed MoS_2 memristors and found that device interconnection scheme can be used to simulate ionic dynamic in biologic neural networks. Tian et al. [140] used 2D perovskite to demonstrate that ultralow current (pA) ion migration and diffusion are used to simulate artificial synapses, which is important for achieving low consumption neuromorphic networks. Besides, they constructed a Markov chain algorithm in the native oxide 2D multilayer tin selenide single device by using oxygen ions transport [141].

4.2. Electrochemical metallization/conductive bridge (ECM/CB) synapses and neuromorphic network

Electrochemical metallization or conductive bridge synapses, mainly proposed for solid electrolyte-based synapses, usually require special electrode materials, one side is an electrochemically active electrode (such as Ag, Cu, Ni, etc.), and the other side is an auxiliary electrode, which is often composed of an electrochemically inert metal material (Pt, W, etc.). The reason for such synaptic plasticity is that the oxidation, migration and reduction processes of cations (Ag^+, Cu^{2+}, Ni^{2+}, etc.) lead to the formation and fracture of metallic conductive filaments/conductive bridges in solid electrolytes, changing the current state of the 2D material channel. Newly, Lanza and co-workers [142] use 2D h-BN instead of traditional transition metal oxides as conversion layer materials, and have volatile and non-volatile resistance conversion characteristics, corresponding to STP and LTP of electrical synapses as shown in Fig. 6. Subsequently, filling the boron vacancies in h-BN based on adjacent electrode metal ions to form a conductive bridge, resulting in resistance transition behavior and electrical synaptic function also proved by them [143]. And this structure can be either volatile or non-volatile, and can be used to simulate brain-like synaptic properties with both STP and LTP again. It is worth mentioning that each time the resistance conversion power consumption is as low as 0.1 fW and the conversion time is less than 10 ns, which shows its potential application in low power consumption and high efficiency neuromorphic computation. The same year, Yan et al. [144] used the coexisting tunneling and ECM effect to achieve bidirectional progressive resistance modulation through $Ag/Zr_{0.5}Hf_{0.5}O_2$:graphene oxide quantum dots/Ag structure, in which 0.6 V amplitude and 30 ns duration can achieve effective modulation, indicating its potential for low power consumption and fast conversion applications in neuromorphic systems.

4.3. Redox/valence change synapses and neuromorphic network

Redox/chemical valence-type synapses are similar to ECM/CB synapses and are related to the electrochemical and electromigration processes of ions, but ECM is based on electrochemical reactions of easily oxidizable metal ions (Ag^+, Cu^{2+}, Ni^{2+}, etc.), while valence change is a redox reaction based on the movement of protons or oxygen-related defect vacancies present in the dielectric materials, causing changes in the chemical valence of the elements in the 2D materials dielectric or channel. Lately, the memristive structure based on quasi-2D α-MoO_3 for simulating biological synapses was proposed by Yang et al. [145] and demonstrated typical synaptic behaviors such as excitatory postsynaptic currents, synaptic weight suppression and enhancement, PPF, and the transition from STP to LTP (Fig. 7). As the gate voltage increases, some protons overcome the barrier and diffuse into the α-MoO_3, where this redox behavior makes the valence of Mo^{6+} to become Mo^{5+} with the formation of new chemical bonds ((Mo–O)–H). The applied positive gate electric field causes protons to move into the α-MoO_3 channel and into the lattice, resulting in enhanced channel conductance.

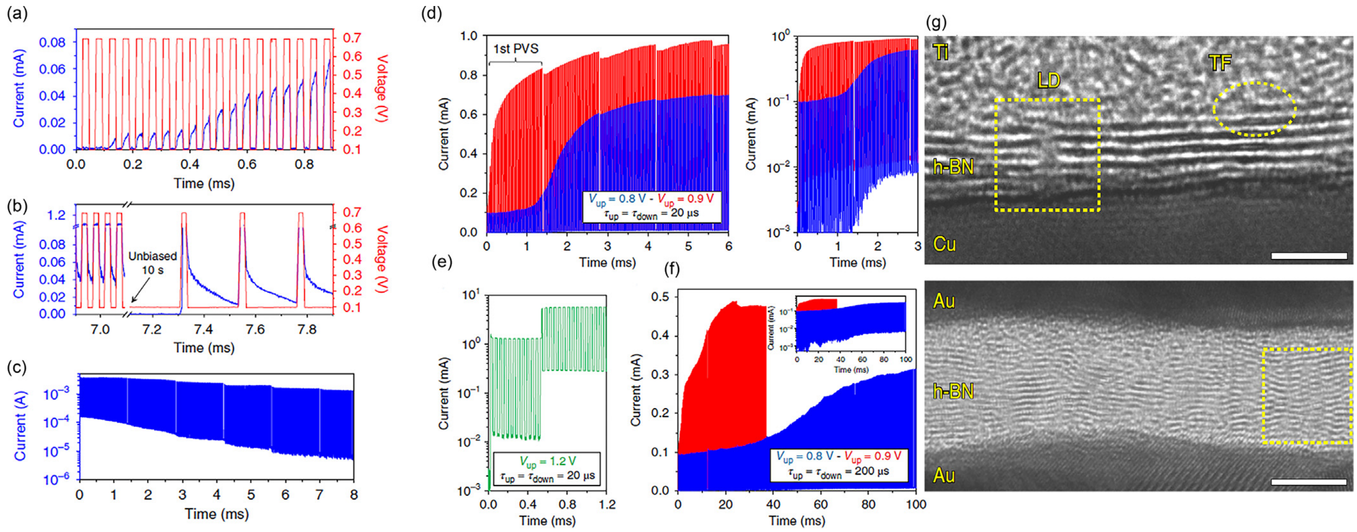

Fig. 6. (Color online) (a–c) Dynamic response of Au/Ti/h-BN/Cu synapses. (a) Applying a series of PVS pulses to achieve synaptic enhancement (PPF). (b) Changes in synaptic current relaxation over time after application of the PVS pulse sequence. (c) By applying PVS, the current signal is gradually reset, which proves the existence of non-volatile RS. (d–f) Dynamic response of Au/Ti/h-BN/Au synapses. (d) The current in multilayer h-BN synapse after two sequences of PVS, showing progressive synaptic potentiation. The right figure shows an enlarged view of current. (e) Applying the PVS sequences, first showing a sudden increase, then an additional current increase of the non-volatile RS. (f) Two PVS sequences show synaptic potentiation with a pulse period of $\tau_{up} = \tau_{down}$ = 200 µs. (g) Cross-sectional transmission electron microscope (TEM) images of Au/Ti/h-BN/Cu and Au/h-BN/Au synapses, which show the mechanism of resistance transition behavior and electrical synaptic function: filling the boron vacancies in h-BN based on adjacent electrode metal ions to form a conductive bridge.

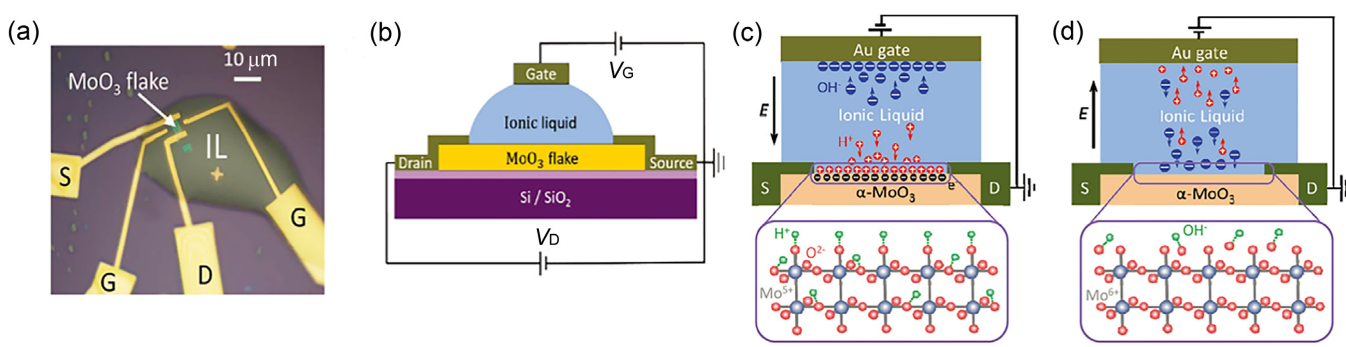

Fig. 7. (Color online) Quasi-2D α-MoO_3 based neuromorphic transistors for simulating biological synapses. (a) Optical micrograph of quasi-2D α-MoO_3 artificial synapses. (b) Schematic diagram of quasi-2D α-MoO_3 memristive structure. (c) Schematic diagram of the movement of protons and hydroxyl groups in the memristive structure under positive gate voltage. (d) Corresponds to a schematic diagram at a negative gate voltage.

4.4. Phase change (PC) synapses and neuromorphic network

Phase change materials have been rapidly developed in the past few decades since they were developed for non-volatile memory in 1968 [146]. Phase change materials typically have the advantages of scalability, durability, reliability, multi-level programming resistance and low device variation, etc. [147–149]. The phase change material is usually a chalcogenide that can be converted from an amorphous phase to a crystalline phase (SET, low resistance state) by a medium amplitude and longer voltage pulse (µs), while switching to the amorphous phase (RESET, high resistance state), a voltage pulse with a higher amplitude and a very short pulse width (ns) is often used [40]. Recently, phase-change 2D materials have been extensively investigated for synaptic devices and neuromorphic computation. And Huh et al. [150] developed a new class of electronic devices for simulating biological synaptic behavior using phase-engineered tungsten oxide, WSe_2 and graphene 2D heterostructures, and called synaptic barrister as shown in Fig. 8. In addition to some basic synaptic characteristics, the structure can also realize the gate-controllable memristive switch behavior through the electrostatic regulation barrier height in the heterostructure. And the adjustable programming voltage is 0.2–0.5 V. It is worth noting that the adjustment of the degree and tuning rate of synaptic weights can be achieved by gate electrostatic regulation, regardless of the programming pulses of the source and drain. All of these properties ultimately enable the enhancement and transformation of synaptic plasticity, similar to the neuromodulation of synapses in biological neural networks. And Zhu et al. [151] report the electric field controls the migration of Li^+ ions in MoS_2 to achieve reversible modulation of the 2H-1T′ phase, in which the increase/decrease in local Li^+ ion concentration leads to phase transition, and observed the biologically similar synaptic competition and synaptic cooperation behavior.

4.5. Thermochemical/Joule heating synapses and neuromorphic network

For resistive random access memories (RRAM), when the electric field drives oxygen ion reaction and migration dominates, it exhibits a bipolar resistance change trend, that is, SET and RESET operations must be performed under opposite voltage polarities. When the Joule heating effect dominates, the device exhibits a

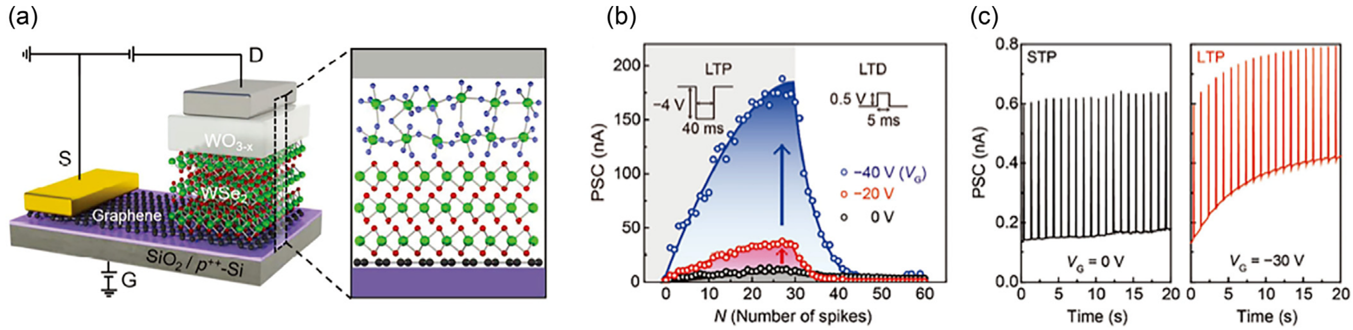

Fig. 8. (Color online) Phase-engineered synaptic barristers for emulating synaptic behaviors. (a) Schematic of phase-engineered WO_{3-x}/WSe_2/graphene synaptic barrister. (b) Characteristics of LTP and LTD with spike number. (c) Implementation of STP and LTP.

monostable resistance threshold switching. When the voltage is less than a certain threshold, the conductive filament spontaneously fuses under the action of Joule heat, and the device returns to a high resistance state, that is, in the case of power failure, the device's low resistance state cannot be maintained. This phenomenon of electric field drive competing with Joule heat drive is also present in 2D layered material-based synaptic devices. A single-layer MoS_2 device based on Joule heating effect and electrostatic doping induced synaptic computation with biocompatibility power consumption (about 10 fJ) has been demonstrated by Sun et al. (Fig. 9) [152]. Moreover, the peripheral circuit with adjustable excitability and suppression synapses implements a sound localization function: detecting an interaural time difference by inhibitory sound intensity or spike frequency dependent synaptic plasticity. It opens up a new approach to synaptic computing in neuromorphic applications that overcomes the drawbacks of traditional CMOS neuromorphic devices, i.e., non-scalability and high energy consumption.

5. Perspective and summary

Recently, relying on the unique advantages and strengths of 2D materials, the use of 2D materials to realize the hardware unit of the neuromorphic computing system has attracted more and more interest. Although 2D materials have made great progress in the application of synaptic devices and neuromorphic systems, there is still a long way to go to implement brain-like computing. In the current study, the application of two-dimensional materials is mainly focused on simulating biological synaptic plasticity, and the current neuromorphic system based on 2D materials contains only one or several synapses or neurons, and the system integration is still at a relatively low level. As a consequence, in order to achieve neuromorphic calculations, large-scale integrated artificial neural units are required, in which each neuron can be interconnected and regulated by artificial synapses, corresponding to the connection and action of neurons in the biological nervous system through synapses, which is a huge challenge for artificial neuromorphic devices. In addition, the reliability of synaptic devices in the construction of artificial neuromorphic network systems is worth exploring, which is often overlooked. Taking into account the current major limitations, relying on the novel advanced 2D material growth technology, the growth of large area, uniform, thickness controllable and high-performance 2D material films used to improve the integration of artificial neural devices, is conducive to the realization of ultra-high density neuromorphic networks with multiple artificial synapses and further promoting the development of brain-like computation.

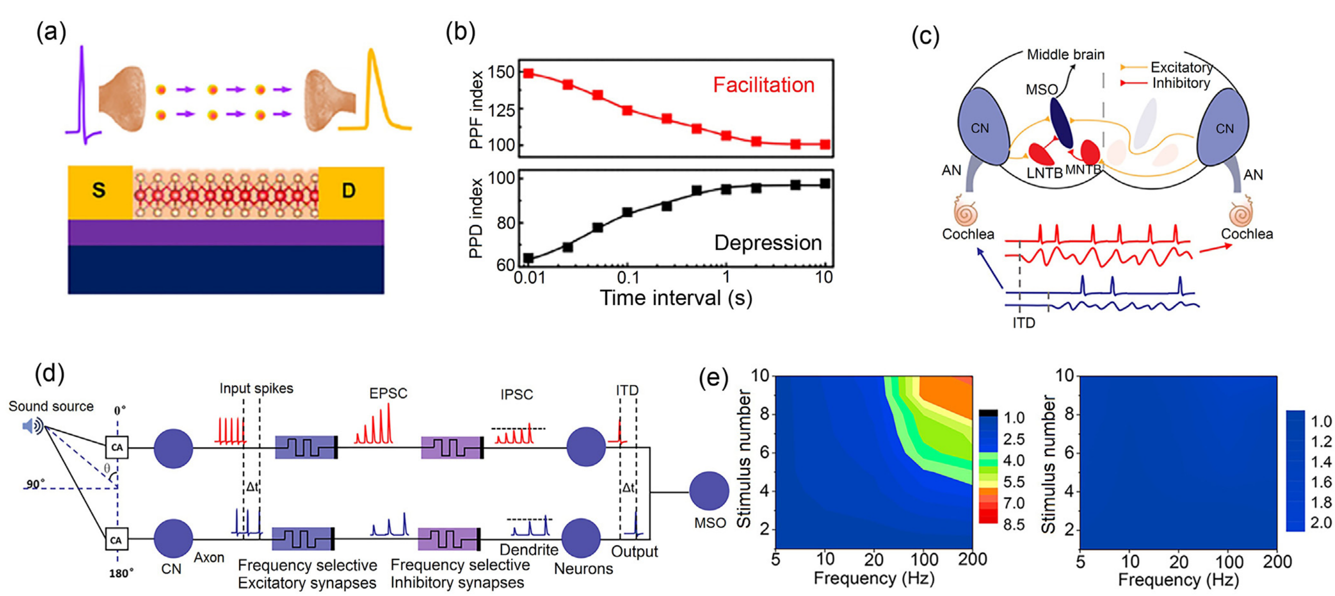

Fig. 9. (Color online) Monolayer MoS_2 devices with Joule heating effect and electrostatic doping induced synaptic computing. (a) Schematic of monolayer MoS_2 synaptic transistor based on Joule heating effect. (b) PPF index of facilitation and PPD index of depression. (c) Graphical ITD and ILD sound localization. (d) Working mechanism of synaptic computing based on ITD sound localization. (e) The frequency- and number-dependent output of the MSO with/without synaptic computation in sound localization.

Conflict of interest

The authors declare that they have no conflict of interest.

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Author contributions

Shuiyuan Wang and Peng Zhou proposed and revised the review framework. Shuiyuan Wang collected and analyzed the related work reported in recent years. All authors discussed the review and contributed to the preparation of the manuscript. Shuiyuan Wang wrote the manuscript. Peng Zhou gave guidance and polished the manuscript.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scib.2019.01.016.

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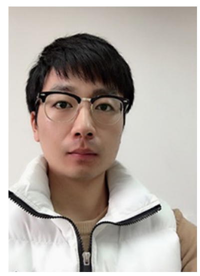

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