2D materials and van der Waals heterojunctions for neuromorphic computing

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Abstract
Neuromorphic computing systems employing artificial synapses and neurons are expected to overcome the limitations of the present von Neumann computing architecture in terms of efficiency and bandwidth limits. Traditional neuromorphic devices have used 3D bulk materials, and thus, the resulting device size is difficult to be further scaled down for high density integration, which is required for highly integrated parallel computing. The emergence of two-dimensional (2D) materials offers a promising solution, as evidenced by the surge of reported 2D materials functioning as neuromorphic devices for next-generation computing. In this review, we summarize the 2D materials and their heterostructures to be used for neuromorphic computing devices, which could be classified by the working mechanism and device geometry. Then, we survey neuromorphic device arrays and their applications including artificial visual, tactile, and auditory functions. Finally, we discuss the current challenges of 2D materials to achieve practical neuromorphic devices, providing a perspective on the improved device performance, and integration level of the system. This will deepen our understanding of 2D materials and their heterojunctions and provide a guide to design highly performing memristors. At the same time, the challenges encountered in the industry are discussed, which provides a guide for the development direction of memristors.

1. Introduction
Most modern computational systems are based on the von Neumann architecture, which plays a profound influence on the science and technology associated with our daily lives [1–8]. According to the traditional von Neumann architecture, the central processing unit and the data memory part are physically separated and communicate via the bus. The separated architecture prevents a high integration density of devices, and the data transportation in the bus leads to the high energy consumption of up to 60% of the total computing energy [9–11]. Moreover, the explosive growth of the data makes it further out of balance with the data bus size [3, 12–17]. In contrast with the von Neumann system, the neuromorphic computing system implements the memory and computations simultaneously in a brain-like style, which could overcome the bottleneck of the von Neumann architecture and provide a promising prospect for large data computing applications such as sound location, color and pattern recognition, and so on. Like biological systems, artificial synapses and neurons are the basic blocks of neuromorphic computing networks [18–21]. The function of each synapse is measured by ‘synaptic weight’, which corresponds to the number and size of neurotransmitter vesicles released by nerve stimulation signals in the process of signal transmission between neurons in the biological system [22–24]. The characteristics of nerve stimulation signals corresponding to different synaptic functions are...
simulated by changing the shape, frequency, and duration of the applied pulse voltage, which produces proper changes in the conductive state of the synaptic device. The resistance state emulates the plasticity of a nerve synapse, consisting of short-term plasticity (STP) and long-term plasticity (LTP), which constitutes the basis of cellular learning [25–27].

The aforementioned artificial synapse and neurons are mostly implemented on non-volatile resistive switching (NVRS), also known as the memristive effect [28], in which an external electric field is in charge of switching the resistance states of a two-terminal device. There are several kinds of approaches to realizing NVRS including conductive filaments (CFs) [29–43], oxygen vacancies migration [44–54], phase change [55–63], and Joule heating effect [2, 64, 65]. The switching mechanism varies by the channel material used in the memristor.

Traditional random accessible memory has been based on CMOS. However, as the scale of the device channel approaches several nanometers, quantum effects start to dominate the transport [66, 67]. Thus, new materials and device architectures are required to realize ideal memristors. Among many candidates for memristors, 2D layered materials have been widely investigated owing to their outstanding physical properties, such as electrical tunability, flexibility, low switching power, and hetero-integration compatibility [30, 68–72]. The van der Waals (vdW) heterostructures with 2D materials have drawn extensive interest because of their large surface-to-volume ratios [73–75], dangling-bond-free surfaces [73, 76–78], and highly gate-tunable bandgaps [79–82]. Owing to the atomically flat surface, atomic defects, and structural disorders of 2D layered materials and the dangling-bond-free vdW interfaces in vertical heterostructures, unprecedented device operation such as low operation energy and fast switching speed can be achieved [83–85], which is promising for next-generation memristors [86–90].

Here, we made a table to compare 2D material-based devices with other conventional 3D material-based devices, including oxides, organic materials, and perovskite materials, as shown in table 1. Compared with oxide materials, 2D materials demonstrate flexibility, lower threshold voltage, shorter response time, and broader work temperature ranges in device operation. Although the device performances of organic material-based devices are close to those of 2D materials-based devices, organic material-based devices cannot operate at high temperatures due to the lack of their temperature stability. As for perovskite material-based devices, their temperature ranges are narrower than those of 2D material-based devices, and the perovskite materials are unstable in ambient conditions.

In this review, we mainly focus on 2D materials and their vdW heterojunction-based neuromorphic computing devices, following a brief introduction to the fabrication approaches of the 2D materials and heterojunctions. Then, we discuss the memristors classified by the number of their terminal electrodes and various mechanisms for resistance switching. We will extend the discussion to integrated neuromorphic device arrays and their applications. The schematic diagram of main contents of this review is shown in figure 1. Accordingly, this review provides a brief outlook of the current challenges and prospects for the development of 2D materials and their heterostructures and device applications in the future.

2. Preparation of 2D materials and vdW heterostructures

The extraordinary physical properties of 2D materials, such as atomically thin geometry and chemical inertness, have attracted the attention of diverse research communities. Each atomic layer of 2D materials can be stacked with the adjacent layers via weak vdW interactions, not limited by the lattice mismatch and fabrication processing. The advantages of 2D materials realize feasible and transferable vdW heterostructures [109]. The fabrication methods of 2D materials and their vdW heterostructures have been extensively studied, as summarized below.

2.1. Preparation of 2D materials

Various preparation methods for 2D materials have been developed since the discovery of graphene in 2004: mechanical exfoliation [110–113], chemical exfoliation and intercalation [114, 115], and chemical vapor deposition (CVD) [116–122].

Mechanical exfoliation is the most convenient method to obtain 2D monolayers in the laboratory [123–125]. The quality, purity, and cleanness of the 2D materials prepared by the mechanical exfoliation method are found to be the best among 2D materials by various fabrication methods. However, producing large-area monolayers by the mechanical exfoliation is difficult [126]; the mechanical exfoliation method is not scalable and time-consuming, which makes it difficult for the systematic control of the thickness and lateral dimension of the exfoliated flakes.

Chemical methods including chemical exfoliation and intercalation could boost the production yield of atomically thin flakes. However, the quality of the 2D materials prepared by the chemical methods is low; external ions or compounds are inevitably involved in the chemical exfoliation/intercalation processes. In
Table 1. Comparison between devices based on 2D materials and other materials.

| Material       | On/off ratio | Endurance (cycles) | Retention time (V<sub>set</sub>/V<sub>reset</sub>) | Operation time (set/reset) | Power consumption | Temperature | References |
|----------------|--------------|--------------------|-----------------------------------------------|----------------------------|------------------|-------------|------------|
| 2D-materials   |              |                    |                                               |                            |                  |             |            |
| MoS<sub>2</sub> | 10<sup>7</sup> | DC > 100 bending 1000 | >11 h 0.08–0.3 V 40–70 ns 4.5 fJ |                          |                  | RT         | [91]       |
| MoS<sub>2</sub> | 10<sup>3</sup> | DC 20 10<sup>4</sup> s | 3.0 V/–2.0 V 0 °C–120 °C 4.5 fJ |                          |                  | RT         | [92]       |
| WS<sub>2</sub>  | 10<sup>3</sup> | Pulse > 10<sup>4</sup> s | >25 h 1.6 V/–1.5 V 0 °C–120 °C 4.5 fJ |                          |                  | RT         | [93]       |
| h-BN/Gr/h-BN   | 10<sup>3</sup> | Pulse 10<sup>6</sup> s | — — 290–450 K 4.5 fJ |                          |                  |            | [90]       |
| CuSe          | 10<sup>2</sup> | DC 300 10<sup>4</sup> s | ±0.4 V 10 μs 11.4/0.95 μW 80–420 K 4.5 fJ |                          |                  |            | [94]       |
| Oxide materials|              |                    |                                               |                            |                  |             |            |
| ZrO<sub>2</sub>; Cu | 10<sup>3</sup> | — 10<sup>4</sup> s | >3.6 V/0.8–1.5 V 50 ns/100 ns 290–450 K 4.5 fJ |                          |                  | RT         | [95]       |
| HfLaO         | 10<sup>3</sup> | DC > 10 000 10<sup>4</sup> s | 0.25 V/–1.81 V 10 ns 290–450 K 4.5 fJ |                          |                  | RT         | [96]       |
| TiO<sub>x</sub> | ~7.5%       | Pulse > 200        | — 3 V/–3.5 V 100 μs/100 μs 290–450 K 4.5 fJ |                          |                  | RT         | [97]       |
| TiO<sub>2</sub>; Ag | —            | —                   | — 0.4–0.8 V 290–450 K 4.5 fJ |                          |                  | RT         | [98]       |
| Ta<sub>2</sub>O<sub>5</sub> | 360         | AC 10<sup>2</sup>  | — 2.0 V/–2.0 V 290–450 K 4.5 fJ |                          |                  | RT         | [99]       |
| Organic materials |            |                    |                                               |                            |                  |             |            |
| PEI           | 10<sup>3</sup> | DC > 100 pulse > 10<sup>3</sup> | 10<sup>4</sup> s 0.5 V/–0.2 V 290–450 K 4.5 fJ |                          |                  | RT         | [100]      |
| pV3D3         | 10<sup>6</sup> | Pulse 10<sup>3</sup> s | — 2.6 V/0.5 V 290–450 K 4.5 fJ |                          |                  | RT         | [101]      |
| PTPA          | 10<sup>6</sup> | DC 10 10<sup>6</sup> s | — –0.8 V/–0.5 V 290–450 K 4.5 fJ |                          |                  | RT         | [102]      |
| [Ru(L)<sub>3</sub>](PF<sub>6</sub>)<sub>2</sub> | 10<sup>3</sup> | Pulse 10<sup>6</sup> s | — 0.52 V/–0.35 V 290–450 K 4.5 fJ |                          |                  | RT         | [103]      |
| PEO           | 10<sup>6</sup> | DC 10 000          | — 0.45–0.7 V/–0.1–0.2 V 290–450 K 4.5 fJ |                          |                  | RT         | [104]      |
| Perovskite materials |          |                    |                                               |                            |                  |             |            |
| BiFeO<sub>3</sub> | > 10 000    | Pulse 2000         | 68 h –2.5––3 V/–1.5 V 290–450 K 4.5 fJ |                          |                  | RT         | [105]      |
| BiFeO<sub>3</sub> | >10         | —                  | — — 290–450 K 4.5 fJ |                          |                  | RT         | [105]      |
| BaTiO<sub>3</sub> | 10<sup>3</sup> | Pulse 10<sup>4</sup> s | ~4 V/–7 V 600 ns 290–450 K 4.5 fJ |                          |                  | RT         | [106]      |
| PdZr<sub>6</sub>Ti<sub>6</sub>Co<sub>4</sub>O<sub>19</sub> | 10<sup>3</sup> | Pulse 10<sup>4</sup> s | ~4 V/–7 V 600 ns 290–450 K 4.5 fJ |                          |                  | RT         | [107]      |
| LaAlO<sub>3</sub> | 200         | Pulse > 2000       | >10<sup>4</sup> s 25 000 ns/15 ns 290–450 K 4.5 fJ |                          |                  | RT         | [108]      |

<sup>a</sup>Here, A:B means B doped A; RT: room temperature.
addition, the thickness, morphology, and size of the flakes prepared by the chemical methods are difficult to be controlled.

On the other hand, due to its excellent controllability and scalability, the CVD method has attracted extensive attention [116–122], and is considered to be a promising way to prepare high-quality and large-area monolayers of 2D materials [127]. CVD techniques are often classified by the reaction type, source, and pressure: low-pressure CVD (LPCVD), atmospheric pressure CVD (APCVD), plasma enhanced CVD (PECVD), and metal–organic CVD (MOCVD). Traditionally, CVD methods have been limited because the growth of 2D materials is not controllable in terms of the dimension and location of 2D materials on the substrate [128]. But the CVD methods have been improved as follows; LPCVD can reduce the generation of particulates generation by quickly pumping out the contaminants [128–131]. APCVD can reduce manufacturing costs, increasing the yield of the fabrication of 2D materials with appreciable quality [120, 132, 133]. PECVD is simple, low-cost, and scalable, enabling the deposition of 2D materials films up to wafer scale [134–137]. MOCVD is an emerging method for 2D materials, which can grow uniform and high-performance 2D materials [138–140].

2.2. Preparation of 2D vdW heterostructures

Methods for preparing vdW heterostructures are classified by chemical (e.g., CVD) and physical (e.g., artificial transfer method) ones. VdW heterostructures can be directly synthesized by CVD methods with highly uniform geometry, but the control of interlayer twist angle and location is hard to be achieved [141].

The dry/wet transfer assembly techniques provide a versatile platform for the preparation of 2D vdW heterostructures with the control [141]. The common dry transfer techniques include poly-dimethyl siloxane (PDMS) exfoliation method [142–144], thermoplastic sacrificial layer method [145, 146], vdW pick-up method [147, 148], wafer bonding method [149], fully dry polymethylmethacrylate transfer method [150], covalent-like quasi-bonding (CLQB) transfer method [111, 151–153], and Al2O3 substrate assisted transfer method [154], etc. The wet transfer techniques include the chemical etching method [155–160], electrochemical bubbling transfer [161–165], adjustable wettability-assisted transfer (AWAT) [166, 167], and capillary-force-assisted clean-stamp transfer [168–171], etc.

Some transfer methods such as the PDMS exfoliation method [142–144] and thermoplastic sacrificial layer method [145, 146] rely on the use of polymer layers, so substantial residues may remain on the 2D materials transferred using such polymers. The critical feature of the vdW pick-up method is the repeatability of picking up the flakes to create target stacks without any contact between the active interfaces of 2D materials and the polymer [148]. Similar to the vdW pick-up method, the wafer bonding method also has a similar benefit [149]. In addition, a gold-assisted exfoliation method underpins a universal and efficient route to produce large-area monolayers [111, 151, 152, 172–174]. Inspired by the method assisted by the Au adhesion layer with CLQB, an Al2O3-assisted exfoliation method has been developed [154]. During the process of the dry transfer technique, 2D materials are not directly exposed to water, effectively avoiding the contamination of the interface of the vdW heterostructures by unwanted chemical residues [175].

In the case of wet transfer, the chemical etching method is to detach 2D materials grown on metal substrates (e.g., copper) by the CVD technique, which produces high-quality vdW heterostructures on a large scale [156, 157, 159, 160]. However, completely dissolved metals cannot be reused in the chemical process.
The electrochemical bubbling transfer method directly exfoliates the samples from the metal, which is cost-saving without losing the metal substrate, but the bubbles may crack the 2D materials [161–165]. The AWAT method is a modified wet transfer process, which achieves uniform and wrinkle-less 2D materials on arbitrary substrates [166, 167]. Capillary-force-assisted clean-stamp transfer is also a simple and clean stamp transfer method; recently, higher capillary forces stimulate further investigation of this transfer technique for the fabrication of high-quality 2D heterostructures [168–171].

3. 2D memristor-based neuromorphic devices

The human brain is a neural network composed of neurons whose axons and dendrites are connected via synapses. The major functions of the neurons are to transmit, store, and process information, enabling humans to conduct various psychological activities that dominate and control most behaviors of humans. Synapses are the input and output channels of the nerve signals.

The evolution of strength (connectivity) of synaptic connection between neurons is called synaptic weight [176, 177]. Synaptic plasticity can be classified into two major types: short-term synaptic plasticity (STSP) and long-term synaptic plasticity (LTSP) by the timescales of the changes. LTSP consists of long-term enhancement (or long-term potentiation, LTP) and long-term inhibition (or long-term depression, LTD), which last for a long time (typically hours or longer). STSP consists of short-term potentiation (STP) and short-term-depression functions. We note other types of synaptic plasticity such as spike-time-dependent plasticity (STDP), spike-rate-dependent plasticity, spike-amplitude-dependent plasticity, and paired-pulse facilitation (PPF) [72, 178].

To mimic the human brain, hardware neuromorphic devices such as artificial neurons and artificial synapses are critical. We mentioned that memristors based on traditional CMOS devices have been widely studied with stable performance and a mature preparation process. But it is difficult to reduce the dimension of the memristor array with CMOS devices; numerous transistors are required to realize the memristor arrays. Thus, 2D material-based neuromorphic devices can be used to achieve novel technology applications that cannot be implemented by traditional oxide materials, such as low-power consumption, in-sensor resistance-switching devices, 3D integration, scalability, etc [16, 67, 71, 73, 83, 179–183]. Depending on the number of terminals, these devices can be classified into two-terminal devices, three-terminal devices, and multi-terminals devices. In the following section, we introduce the recent progress on 2D materials and their heterojunction-based neuromorphic devices, considering various underlying mechanisms of the switching devices. We collected information from several two-terminal devices and three-terminal/multi-terminal devices, consisting of 2D materials. The devices’ structures, on/off ratios, retention time, and other important device characteristics are summarized in two tables shown after sections 3.1 and 3.3. Then we made a comparison between 2D material memristor and some random access memory (RAM) devices in commercial, as shown in table 4. The details will be introduced as below.

3.1. Two-terminal devices

As the name indicates, these kinds of devices with two terminals, drain and source electrodes, emulate the characteristics of biological synapses and neurons. By using 2D materials as the active channel material, various objects such as atoms, ions, holes, and defects in the 2D materials take part in the resistance switching by applied bias voltages. Two-terminal devices can be divided by their distinct underlying working mechanisms, including atom switches/metal ion migration, anion/defects migration, charge trapping/detrapping, phase change, ferroelectric, joule heating effect, etc.

Memristors operating by atom switches/metal-ion-migration are often designed by a sandwich structure of metal/2D material/metal in a plane or vertical direction. In this kind of memristors, a 2D material usually plays a role as a dielectric layer with poor electrical conductivity, and the active metal (chosen as the top electrode) is electrochemically active, compared with the bottom electrode [184–186].

Drifted by external bias, the atoms are dissociated from the metal electrode; free atoms or ions can be diffused into the 2D material, forming CFs or conductive bridges between the drain and source electrodes, which is also called electrochemical metallization (ECM) [186–189]. The ECM results in a SET transition from high resistive states (HRS) to low resistive states (LRS) and generates a hysteresis in $I–V$ characteristics.

The metallic filaments (or CFs) can be either fractured spontaneously via an external bias or recovered via a reversed bias: SET and RESET processes between LRS and HRS. Although many CF phenomena based on traditional oxidized materials have been reported, we note that traditional oxidized materials are difficult to be integrated into 3D because the characteristic dimension of neuromorphic devices with traditional oxide materials requires larger than 5 nm, according to the device dimensional scaling rule first proposed by Dennard et al [67, 190–192].
While many memristors based on 2D materials or their heterostructures have been reported, transition metal di-chalcogenides (TMDs) have gained a lot of interest among numerous 2D materials. Hao et al formed a volatile bridge in Ag/MoS$_2$/TiW memristors (figure 2(a)) and got an on/off current ratio of up to $\sim 10^5$. The possible resistance switching schematic diagram is shown in figure 2(b). In high-resolution SEM, an Ag filament could be seen clearly, shown in figure 2(c) [193]. Dev et al mimicked the leaky-integrate-and-fire model with a $\sim 10^9$ on/off current ratio by using Au/MoS$_2$/Ag threshold switching memristors (TSM) (figures 2(d) and (e)). The work demonstrated $5 \times 10^6$ pulsed cycles with the device, which represents good reliability (figure 2(f)), and realizes an artificial neuron system by connecting the Au/MoS$_2$/Ag TSM to an external RC circuit. They achieved a stable spike output [194]. Shi et al reported an electronic synapse with a structure of Au/Ti/h-BN/Au, shown in figure 2(g). In the synaptic device, a CAFM probe was used as one terminal. After a forming process, the synaptic device exhibits stable NVRS (figure 2(h)). The conductive nanofilaments are clearly observed in the cross-sectional TEM (figure 2(i)). When applying a sequence of voltage pulses to the synaptic device, synaptic potentiation could be demonstrated (figure 2(i)) [30]. Moreover, Ge and his collaborators reported the observation of NVRS in monolayer TMDs sandwiched by metal electrodes in 2017 [70], which is contradictory to the conventional thought that nonvolatile switching cannot be scaled to sub-nanometer owing to the exceeding leakage current [195–197].

Depending on the mobile ion species, resistive switching can occur by valence change mechanism (VCM). The VCM is based on anion migration and redistribution, leading to the change of conductance with a valance state change [186]. Traditionally, metal oxides have been placed between two metal electrodes, which makes it easy for the formation of oxygen vacancies. In the SET process, the migration of oxygen anions and vacancies leads to the formation of CFs while the recombination of the oxygen anions and vacancies ruptures the CFs.
Figure 3. (a) Schematic diagram of the structure of memristive device with vertically aligned MoS$_2$ layers. (b) and (c) Endurance data for 100 manual DC switching cycles performed in ambient conditions before vacuum (b), and in vacuum (c). The absence of the resistance switching effect in a vacuum indicates a water adsorbent-driven process and OH$^-$ migration. Reproduced from [205]. CC BY 4.0. (d) Dual–$I–V$ characteristics of set process by positive and negative sweep with compliance of 500 $\mu$A. (e) and (f) Cross-sectional schematic of the device in (e) LRS and (f) HRS, where green balls represent anions, maybe oxygen vacancy or hydroxyl groups, further experiments are needed to confirm this. [207] John Wiley & Sons. © 2021 Wiley-VCH GmbH.

(i.e., the RESET process) [186, 198]. Besides oxygen anions (O$^{2-}$) [186, 199–204], hydroxy ions (OH$^-$) [205], halide ions such as chloride ion (Cl$^-$), bromide ions (Br$^-$) [186], iodide ions (I$^-$) [186], nitride, and sulfurs (S$^{2-}$) [206] could be used to form the CFs in the insulating materials (metal oxides).

Belete et al reported a NVRS in a vertical heterostructure of silicon, vertically aligned MoS$_2$, and chrome/gold metal electrodes, as shown in figure 3(a). When the MoS$_2$ is exposed to air (figure 3(b)), water molecules in ambiance could be introduced to the MoS$_2$-based memristor; the MoS$_2$ works as a catalyst to split water molecules and generate hydroxyl ions. The experimental results and analytical simulations suggest that an electric field can drive the movement of OH$^-$ ions and modify the energy barrier at the interface of MoS$_2$ [205]. Xiong et al reported an anion conductor-based memristor using layered double hydroxide (LDH) [M$_{2x+1}$M$_{3x-1}$(OH)$_2$]$^{2x-}$·[A$^{n-}$]$_m$H$_2$O as the active material, where M represents metal cations, and A represents anions. LDHs are considered to be candidates for the OH$^-$ ion conductors due to their high concentration of hydroxyl groups covalently bonded within the 2D brucite layer. Xiong et al also reported a dual resistive switching behavior in the MgAl LDH-based memristor, as shown in figure 3(d), utilizing the formation of anion CFs [207]. In summary, the CFs formed in 2D materials can reduce the scale of neuromorphic devices to the atomic level, which represents their potential for an ultrahigh density of data storage for neuromorphic applications.

Numerous atomic defects and dangling bonds are present in the 2D materials. When the metal atoms/ions are adsorbed on surface defects or inserted into a semiconducting 2D material, the conductance of the 2D material is largely modified without any microstructural change. Thus, we could get memristive devices with charge trapping/detrapping processes. Under a certain (reverse) bias voltage, metal atoms are trapped (detrapped) in (from) inherent defects or interlayer space, leading to an increase (decrease) in the conductance of the 2D materials.

Ge et al in 2020, proposed a model to explain the resistive switching phenomenon in a metal/2D material/metal crossbar structure, called as dissociation–diffusion–adsorption (DDA) model (figures 4(a)–(c)). In the model, free atoms/ions are adsorbed in inherent vacancies of 2D materials and bonded with neighboring atoms, causing a conductive point that drives a conductance increase. They calculated the energy barrier for the diffusion and adsorption process, as shown in figures 4(d) and (e). Following scanning tunneling microscope (STM) experiments supported the model (figure 4(f)). Then, they investigated and added transition metal sulfides (MS$_2$, M = Re, Sn), transition metal selenides (MSe$_2$, M = Re, Sn, Pt), a transition metal telluride (MoTe$_2$), a TMD heterostructure (WS$_2$/MoS$_2$), and an insulator (h-BN) into the category for charge trapping/detrapping [197].
Materials with different crystalline structures demonstrate significantly different optical and electronic properties. Using the phase change between different crystalline structures, we could achieve resistance switching. This kind of memristor with two-terminal electrodes is a phase-change memory memristor, also called a phase change random access memory (PCRAM) device, using the conductance modification during the phase change of materials [208].

Since the phase change in traditional oxide materials is driven thermally, there appears accumulation of 'waste' heat in the channel [208]. However, the highly anisotropic electrical and ionic transport characteristics...
and the large interlayer space of the 2D materials achieve the phase change without waste heat. The ions are inserted into the oxide layer by an electric field and make the phase transition, which is different from the anion migration for forming CFs [207]. For example, the energy consumption per unit volume of the electrostatically driven phase transition in monolayer MoTe2 at room temperature is 9% of the thermally driven phase transition in Ge2Sb2Te5 [209] according to the density functional theory-based calculations.

There have been continuous demands to develop new materials to realize phase change memristors. Jiao et al. reported that through geometrical shrinkage and interfacial confinement, thin Sb film with a thickness of 4 nm can be used to make a phase change memristor via phase transition between the amorphous and crystalline phase. The device demonstrated iterative RESET and cumulative SET operations, with a low resistance drift and low noise [208]. TMDC materials, such as MoS2, MoTe2 [210], and so on, have been demonstrated with their inherent different phases such as 1H, 1T, distorted 1T, 2H, and 3R. Zhu et al. reported reliable reversible memristive behaviors with layered MoS2 films via electric field-driven Li+ ions redistribution, which generates reversible local 2H (semiconducting)–1T’ (metallic) phase transition. The high in-plane diffusivity of Li+ ions allows efficient ionic coupling of multiple MoS2 devices and provides a mechanism to implement synaptic competition and synaptic cooperation effects in bio-inspired artificial neural networks, as shown in figures 5(a) and (b).

Fu and his co-workers reported photo-induced phase change in 2D nanosheets (NS)/0D quantum dots (QDs) MoS2 structures, realizing dynamic resistive memory. The device can be modulated by local QD excitation and shows the resistive switching in an electric field, as shown in figures 5(c)–(e). The Raman spectra demonstrate reversible photo-induced phase transition from 2H to 1T’ phase [56, 211]. Furthermore, the phase change, controlled by the charges and temperature, generates tunable periodic oscillations with a chaotic
behavior similar to what occurs in a biological neuron network \cite{211}, as reported by Panin et al. They claimed the potential of their devices for further artificial neural networks.

Non-centrosymmetric materials have attracted research interests owing to their polarizations and related applications. Ferroelectricity possesses a spontaneous electric polarization that can be controlled by an applied external electric field \cite{213}, making it promising for non-volatile RAM. Ferroelectric RAM (FeRAM) provides a fast read and write access compared with dynamic RAM, the most commonly used type in digital computers. The commercial FeRAM devices are mostly based on the ferroelectric effect of a lead zirconium titanium (PZT) material. Under a certain electric bias voltage, the central atom is driven to move and finally stays at a stable state (i.e., non-volatile); a reverse bias can recover the state, which can be detected by an external circuit.

However, the scale of current devices based on traditional ferroelectric materials prevents a high density of integration; new 2D ferroelectric materials are required. Unlike 3D oxidized materials, the competence of 2D layered materials to retain ferroelectric properties originates from their weak interlayer coupling, which stabilizes individual layers from out-of-plane perturbations \cite{72}.

TMDs, such as In$_2$Se$_3$ \cite{214, 215}, SnS \cite{216}, SnSe \cite{216} and so on, and other materials such as CuInP$_2$S$_6$ \cite{217}, demonstrate ferroelectricity. Wang et al reported a ferroelectric semiconductor field effect transistor (FeSFET) using ferroelectric and semiconductor characteristics of $\alpha$-In$_2$Se$_3$. The switching characteristics of the device demonstrated its potential as an artificial synapse. The structure schematics of the device and material are shown in figure 6(a). Figure 6(b) shows different ferroelectric states and band diagrams corresponding to the different states. When a negative voltage is applied to the back gate (BG), its polarization direction is affected by the electric field and turns to downward near the interface between $\alpha$-In$_2$Se$_3$ and Al$_2$O$_3$. In the lower surface of $\alpha$-In$_2$Se$_3$, positively polarized charges are accumulated, resulting in downward band bending. The charge accumulation on the bottom surface of the channel greatly increases the carrier density and channel current and realizes the set process. However, when a positive voltage is applied to the BG, the polarization direction is opposite and negative polarization charges are accumulated, resulting in upward band

Figure 7. (a) and (b) Artificial synaptic device by Joule heating. Schematic diagrams of two-terminal devices based on monolayer MoS$_2$ without (a) and with (b) Joule heating. The violet and yellow spikes represent an input spike and a transmitted EPSC, respectively. (c) Joule heating-driven conductance ($G$) facilitation with multiple voltage sweeps. (d) Continuously increased synaptic strength (50 continuous synaptic strengths) by a series of pulses with an amplitude, width, and time interval of 25 V, 50 ms, and 50 ms, respectively. The auto reset process was measured with a small voltage of 0.1 V. (e) PPF and PPD indexes after two consecutive pulses as a function of the inter-pulse interval. Reprinted with permission from \cite{2}. Copyright (2018) American Chemical Society. (f) Schematic of the Pd/WS$_2$/Pt device. (g) Typical $I$–$V$ curve in linear coordinates. The inset shows the electroforming curve. (h) ON switching using a 13 ns voltage pulse. \cite{44} John Wiley & Sons. © 2019 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim.
| Materials          | Structure                             | Mechanism             | On/off ratio | Threshold voltage (V) | Retention (s) | Endurance | References |
|-------------------|---------------------------------------|-----------------------|--------------|------------------------|---------------|-----------|------------|
| WS₂               | Pd/WS₂/Pt                             | Joule heating effect  |              | ~0.6(set) ~0.2(reset)   | 1.8 × 10⁴     |           | [44]       |
| MoS₂              | Au/MoS₂/Au                            | Joule heating effect  |              |                        |               |           | [2]        |
| h-BN/graphene/h-BN| Au/h-BN/graphene/h-BN/Ag              | CF                    | 10⁴          |                        | 10⁵           | 10⁴       | [90]       |
| MoS₂              | Ag/MoS₂                               | CF                    | 10⁵          | 0.35–0.4               |               |           | [193]      |
| MoS₂              | Au/MoS₂/Ag                            | CF                    | 10⁵          | ~0.7 V                 |               |           | [194]      |
| MXene             | Cu/MXene/Cu/SiO₂/Si                   | CF                    | >50          |                        | 10⁶           | 0.35–0.4  | [221]      |
| BP/ZnO            | BP/ZnO                                | Anion migration       | 10⁹          |                        | 5             |           | [222]      |
| LDH               | Ti/Au/[Mₓ⁺⁺Mₓ⁺⁺⁺(OH)ₓ⁺⁺⁺⁺⁺·Aₓ⁻⁻⁻⁻⁻⁻·nH₂O/Gr | Anion migration       | 10⁹          | 2.7(set)–8.2(reset)    | 2.7(set)–8.2(reset)–8.6(reset) | [207]      |
| MoS₂              | Si/MoS₂/Cr                            | Ion migration         |              |                        |               |           | [205]      |
| MoS₂/WO₃          | Cr/Au/MoS₂/WO₃                        | Ion migration         | 60×          |                        |               |           | [206]      |
| MoS₂              | Au/Li/MoS₂/Au                         | Phase change          | ~10³         |                        | 7000          | 10⁴       | [212]      |
| MoS₂              | MoS₂ NS/QD structure                  | Phase change          | ~2           | 4 V                    |               |           | [56]       |
| In₂Se₃            | In₂Se₃/Au                            | Ferroelectric         | 10³          |                        |               |           | [214]      |
| CrI₃              | h-BN/Gr/GrI₃/Gr/h-BN                  | Ferroelectric         | ~10²         | 2.94                   |               |           | [213]      |
| SnS               | Pt/SnS/Pt                             | Ferroelectric         | 20           | 4/–4                   | 10³           |           | [216]      |
| SnS               | Cr/Au/SnS/Au/Cr                       | Charge trapping/detrapping | ~1.2       |                        |               |           | [223]      |
| MoS₂/ h-BN/ graphene | MoS₂/h-BN/ graphene                   | Charge trapping/detrapping | 10⁵         | 10                     | 4.5 × 10⁴     | 10⁴       | [224]      |
bending. Thus, the charge depletion occurs on the bottom surface of the channel, which realizes the reset process [218].

The inevitable interface traps at the interface of 2D semiconductor/ferroelectric materials suppress the ferroelectric characteristic, so a self-assembly monolayer material, three-aminopropyltriethoxysilane (APTES) was proposed to work as an interfacial passivation layer at MoS$_2$ and Hf$_{0.5}$Zr$_{0.5}$O$_2$ (HZO) to minimize the influence of the traps (figures 6(c) and (d)) [219]. Hernandez-Martin et al demonstrated the interplay between electrochemical and ferroelectric degrees of freedom at the interface using the dynamic formation of an oxygen vacancy profile in a ferroelectric tunnel junction [220]. Most 2D ferroelectric materials are semiconductors with an appropriate bandgap, so ferroelectric field-effect-transistor devices can be made without using an additional semiconductor channel layer, which can improve the integration density of devices. Despite the advantages of 2D ferroelectric materials, there is still a long way to achieve high-performance 2D ferroelectric material-based memristive devices due to the difficulty of large area production and the low spontaneous polarization value.

When an electric current flows through a conductor or a semiconductor channel, some heat is generated and released, which is called Joule heating. Modulating the conductivity of the channel material, the amount of heat and its release can be controlled. Distinct from all former working mechanisms, the change of conductance driven by the Joule heating effect demonstrates a mono-stable conductance threshold switching: once a bias
Figure 9. (a) The schematics of band diagrams of the metal/dielectric layer/semiconductor device in OFF/ON state, respectively. (b) The schematics of the charge trapping/detrapping process of the MoS₂ transistor in OFF/ON state, respectively. The red and blue arrows show the migration direction of electrons. Reprinted with permission from [231]. Copyright (2017) American Chemical Society. (c) Schematic of the sr-SiNx memory in a FET configuration. The inset shows the transfer characteristics of the MoTe₂ channel on the sr-SiNx substrate, sweeping in forward and backward directions, where the inset shows the corresponding transfer curve on the logarithm scale. The bias $V_{sd} = 0.5 \text{ V}$. (d) Schematic of the operational mechanism of the sr-SiNx memory. The rectangles in the pink and gray colors represent the sr-SiNx substrate and the highly p-doped Si, respectively. The ellipses in the sr-SiNx substrate denote the traps. The blue circles represent the holes that move between the sr-SiNx substrate and the p-doped Si under different gates. Reprinted with permission from [232]. Copyright (2021) American Chemical Society.

Figure 10. (a) A schematic diagram of the neuromorphic transistor with PVA proton conductor IL gate. (b) Transfer curve of the MoS₂ transistor with a fixed bias of $V_{DS} = 0.1 \text{ V}$, in which a hysteresis window is shown clearly. (c) A pair of presynaptic spikes and a triggered EPSC are shown versus time, representing the PPF phenomenon. [239] John Wiley & Sons. © 2017 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim. (d) Structure of the dual-gate MoS₂ IL (Li⁺) neuristor. (e) STP and (f) LTP of the MoS₂ neuristor. Gray lines indicate the stimulus applied to the TG (ionic gate). Reprinted with permission from [240]. Copyright (2019), American Chemical Society.

Voltage lower than a certain threshold is applied, the heat is immediately released, and the conductance of the channel materials quickly gets recovered [178].

Sun et al reported synaptic computation based on Joule heating and versatile doping induced metal–insulator transition in a scalable monolayer-molybdenum disulfide (MoS₂) device with biologically comparable energy consumption ($\sim 10 \text{ fJ}$) (shown in figures 7(a) and (b)) [2]. With the increase in temperature, the conductance increases simultaneously, supporting the synaptic computation. A circuit with tunable
excitatory and inhibitory synaptic devices demonstrated a key function of synapses such as set/auto reset, PPF, and PPD, as shown in figures 7(d) and (e).

Yan et al. reported a 2D layered WS$_2$ NS-based memristor with vacancy-induced synaptic behaviors (figures 7(f)–(h)) [44]. The electric current generated heat, leading to the generation of the vacancies of S and W atoms; electrons move by hopping through the vacancies. The device exhibited low-power consumption of the order of femto-Joules ($10^{-15}$ J). Furthermore, it successfully emulated biological synaptic functions, such as learning and memory operations under PPF, STDP, excitation and inhibition under positive and negative pulse-stimulus trains, and STP to LTP transition.

Here, we made a table summarizing two-terminal devices based on 2D materials such as TMDs, MXene, h-BN, graphene, LDM, and so on, as shown in table 2. The device structures, mechanisms, on/off ratios, threshold voltages, retention times, and endurance of the various devices are shown in table 2. In table 2, CF devices tend to have higher on/off ratios, lower threshold voltages while anion migration devices need higher threshold voltages for similar on/off ratios (compared with CF devices). The on/off ratios of ion migration, phase change, and ferroelectric devices are lower with higher threshold voltages than those of CF and anion migration devices. Charge trapping/detrapping devices require the highest threshold voltages of $\sim 10$ V while their performances are good with a high on/off ratio of up to $\sim 10^6$ and a long retention time of up to $10^4$ s.
| # of terminals | Materials | Structure            | Control gate/FG/BG | Mechanism                        | On/off current ratio | Power consumption | Threshold voltage (V) | Retention (s) | Endurance | References |
|---------------|-----------|----------------------|--------------------|----------------------------------|----------------------|-------------------|----------------------|---------------|-----------|------------|
| 3             | MoS$_2$   | v-MoS$_2$/Gr/SiO$_2$/Si | —/—/Si            | VdW structure                    | $\sim 10^6$          | —                 | —                   | —             | —         | [228]      |
| 3             | MoS$_2$   | Al$_2$O$_3$/PTCDAMoS$_2$/SiO$_2$/Gr/++-Si | Electrical/optical spike | VdW structure | — | — | — | — | — | [227] |
| 3             | MoS$_2$/h-BN/Gr | MoS$_2$/h-BN/Gr | Light | VdW structure | $\sim 10^6$ | $\sim 2$ nJ | $\sim 3$ V | $3.6 \times 10^4$ | $>10^4$ | [224] |
| 3             | MoTe$_2$  | MoTe$_2$/Gr/SiO$_2$/Si | —/—/Si            | Charge trapping/detrapping       | $4 \times 10^3$      | $\sim 2nJ$        | $3.6 \times 10^4$ | $>10^4$ | $>570$   | [232] |
| 3             | MoS$_2$   | IL/MoS$_2$/SiO$_2$/Si | Proton-PVA         | IL control                       | $>10^4$               | 1.5 V             | —                   | —             | —         | [239]      |
| 3             | WSe$_2$   | WSe$_2$/LiNbO$_3$ (LNO) | Metal            | Ferroelectric                    | 898.4                | 6–9 V             | 6–9 V               | —             | —         | [243]      |
| 3             | MoS$_2$   | MoS$_2$/APTES passivation layer/Hf$_0.5$Zr$_0.5$O$_2$/p-Si | p-Si | Ferroelectric | $<10$ V | $<10$ V | — | — | — | [219] |
| 3             | WSe$_2$   | WO$_{3-x}$/WSe$_2$/Gr/SiO$_2$/p$^{++}$/Si | SiO$_2$/p$^{++}$/Si | Oxygen migration                  | $10^3$               | 0.2–0.5 V          | $<10^3$ | $<10^3$ | [244] |
| 3             | MoS$_2$   | Au/HF/Hf$_0.5$Si$_0.5$/Sapphire | Au | Diffusion of double sulfur vacancy | $10^3$ | $<10^3$ | — | — | — | [245] |
| 4             | WSe$_2$/WO$_3$ | WSe$_2$/WSe$_{3-x}$/O/WO$_3$ SiO$_2$/p-Si | Light/—/p-Si      | VdW structure                    | $\sim 10^7$          | 2.7 pF            | —                   | 100           | —         | [226]      |
| 4             | MoS$_2$   | h-BN/MoS$_2$/h-BN/Gr/SiO$_2$/Si | —/Au/Gr           | Charge trapping/detrapping       | $\sim 300$           | $\sim 7.3$fF     | $<10$ V            | $>10^3$       | —         | [183]      |
| 4             | 2D MoO$_3$ | IL/MoO$_3$/SiO$_2$/Si | Li$^+$/IL/Si      | IL control                       | $10^4$               | —                | —                   | —             | —         | [236]      |
| 4             | MoS$_2$   | IL/MoS$_2$/SiO$_2$/n$^+$-Si | Li$^+$/IL/n$^+$-Si | IL control                       | $3000$               | —                | —                   | —             | —         | [240]      |
3.2. Three-terminal devices

In contrast to two-terminal devices, three-terminal synaptic devices can achieve both stimuli transmission and the learning process. Therefore, they can provide a new component for synaptic devices and potentially for complex neural network circuits. In the three-terminal geometry, a gate electrode is introduced to mimic the presynaptic processes or external stimulus, acting as a signal modulator. 2D materials and source and drain electrodes mimic the post-neuron, while channel currents can imitate the synaptic weight plasticity in real-time. There are three subtypes of three-terminal devices: vdW heterostructures, charge trapping/detrapping, and ionic gating, which would be introduced in detail below.

2D vdW heterostructures have drawn great interest due to their appealing optoelectronic, mechanical, and electronic properties. In a modified operation, more charge trap sites can be generated to serve as hopping sites for charge carriers; then, the channel formed in the 2D vdW heterostructure and the energy band alignment is substantially changed, leading to an increase in conductance.

In 2015, monolayer MoS2 was used to feature NVRS in a lateral device [225]. In 2020, He et al reported a multi-gate memristor with a lateral heterostructure of 2D WSe2 and WO3, as shown in figures 8(a) and (b). Between the WSe2 and WO3 parts, there is an intermediate transition layer, a mixture of WSe2−x and WO3−y. Protons are injected into or removed from the intermediate layer, generating the memristive behavior. Using the device structure, the authors emulated synaptic functions such as post-synaptic current, STP, and LTP, with highly linearity, symmetry, and ultra-low energy consumption of ~20.7 pJ per spike [226].

Wang et al demonstrated the first multi-functional synaptic transistor based on a fully 2D inorganic/organic, MoS2/polyethylene-3,4,9,10-tetracarboxylic dianhydride (PTCDA) hybrid heterojunction. Via the optical/electrical modulation of the interface band structure, the synaptic inhibition and excitation could be switched with the minimum inhibition of 3% and the maximum excitation of 50% [227]. However, the lateral structure lacks the integration density that is achievable in two-terminal vertical devices. Accordingly, a vertical device structure is ideal for high-density system integration [197].

Kalita et al reported the integrate-and-fire response of an artificial neuron based on the volatile threshold switching behavior of a vertical-MoS2/graphene vdW heterostructure. They successfully emulated the typical behaviors of biological neurons such as all or nothing spiking, the threshold-driven spiking of the action potential, the post-firing refractory period, and strength modulated frequency response [228].

To overcome the limits of the capacity for device miniaturization and to increase circuit complexity, Tran et al reported a two-terminal multibit memory via MoS2/h-BN/graphene vdW heterostructure. An electric field can modulate the energy band structure, leading to the transition between on/off states, as shown in figures 8(c) and (d) [224]. He et al demonstrated a novel multi-terminal device based on the 2D vdW heterostructure with a dual-gate, which exhibited admirable multi-level NVRS (figures 8(e) and (f)). Applying different external voltage biases between the source and drain electrodes, they realized cooperative programming and erasing process [183].

In summary, three-terminal memristive devices based on vdW heterostructures demonstrate their potential to extend the range of memristors with promising applications such as image sensing, logic gates, and synaptic devices for neuromorphic computing to meet the power saving, high efficiency, and parallel computing.

Distinct from the two-terminal charge trapping/detrapping devices, there is a selective dielectric layer placed between the charge donor and the charge reservoir (localized defects or trapping layers). When a bias voltage is applied on the control gate, charges are trapped/detrapped from the charge reservoir without back-flow and charge neutralization, generating the switching between LRS and HRS [229, 230]. In the studies, the charge donor is often chosen as a photosensitive material so that, upon light irradiation, numerous hole/electron pairs are formed.

Arnold et al mimicked neurotransmitter release dynamics, and LTP in chemical synapses, by using hysteresis engineering in a BG MoS2 FET. The band diagram shown in figure 9(a) gives a clear energy perspective of the switching process. The resistive switching process by the electrons trapped/de-trapped into/from the MoS2 layer is shown in figure 9(b). Applying a negative voltage to the p-doped Si leads electrons to be detrapped from the semiconductor, resulting in the off-state. When applying a positive voltage, electrons are trapped into the semiconductor, resulting in the on-state [231]. Xiang et al reported a memristor based on silicon-rich silicon nitride (sr-SiNx) as the charge trapping layer owing to its large trap density and high trapping efficiency (figures 9(c) and (d)). They further demonstrated a 2D TMDs-based artificial synaptic array on the sr-SiNx substrate in the three-terminal FET configuration. The device array exhibited a minimal device-to-device (5.3%) and cycle-to-cycle (1.5%) variability, high analog on/off ratio, and conductance update linearity [232].

The charge trapping/detrapping devices discussed above show their potential for next-generation neuromorphic computing systems. They introduced that a 2D dielectric layer can prevent the reverse electric field generated by the accumulated charges and help the anions to migrate, realizing the linearity and higher saturation values of the conductance.
Table 4. Comparison between the 2D material devices and several commercial RAMs.

| Operating mechanisms                        | Merits                                      | Demerits                                                   |
|---------------------------------------------|---------------------------------------------|------------------------------------------------------------|
| Resistive RAM/RRAM                          | Non-volatile                                | Unclear mechanism                                          |
|                                             | Fast speed and high density                 | Large fluctuation                                          |
|                                             | High reliability                            | Insufficient reliability                                   |
|                                             | Multi-value storage                         | Integration problems                                       |
|                                             |                                             | Low read speed in the integrated array                     |
| Magnetic RAM/MRAM (take spin-transfer torque MRAM as an example) | Non-volatile                                | Barrier layer damage and data storage reliability caused by high current density |
|                                             | Fast speed                                  | Non-ideal circuit design                                    |
|                                             | High reliability                            |                                                            |
|                                             | Near-infinite erasure times                 |                                                            |
| Phase change RAM/PCRAM                      | Low production cost and high scalability    | Low endurance                                              |
|                                             | Large storage capacity                      | Low data-retention reliability                              |
| Ferroelectric RAM/FeRAM                     | Non-volatile                                | Destructive read out-FeRAM:                                |
|                                             | Fast speed and high density                 | Fatigue failure                                            |
|                                             | Low power consumption                       | Non-destructive read out-FeRAM:                            |
|                                             | Radiation-proof                            | Laboratory research stage                                   |
| 2D-material RAM                              | Non-volatile                                | Wafer-scale synthesis of high-quality and uniform 2D materials |
|                                             | Smaller sizes                               | Integration difficulty of high-density neural networks     |
|                                             | Faster speed and higher density             |                                                            |
|                                             | Lower power consumption                     |                                                            |
|                                             | Compatibility with traditional CMOS devices |                                                            |
| CF and anion migration                       | High on/off ratio                           | Stochastic path of CFs, especially for low current operation (by unstable paths) |
|                                             | Low threshold voltage                       | Structure change led by ion-migration                       |
|                                             | Low power consumption                       |                                                            |
| Charge trapping/detrapping                  | Utilizing inherent defects in 2D materials  | Affected by environmental conditions greatly                |
|                                             | No damage to channel materials              |                                                            |
| Phase change                                | Inherent different states                   | High cost                                                   |
|                                             | Low energy consumption                      | High heating effect and power consumption                   |
|                                             |                                            | Faulty circuit design                                       |
| Ferroelectric                               | Electric-field-driven polarization transformation | High threshold voltage                                    |
|                                             | Fast read and write access                  |                                                            |

(continued on next page)
Besides the three-terminal memristive devices discussed above, ionic liquid (IL) gating control has attracted wide attention from researchers due to its powerful charge control ability via an IL control gate. Researchers have discovered that, in addition to the effect of net charge under IL gate voltage particularly in oxides, there are often complicated ion insertion/extraction processes [233–238].

The introduction of an IL gate leads to the formation of a double electric layer in the IL, inducing charge carriers electrostatically and/or inserting ions into and out of the 2D material's lattice. The charge and ion dynamics produce considerable changes in the electronic, optical and magnetic properties of the material, and can even modify the lattice crystal structure as well.

In 2017, Jiang et al for the first time, proposed the proof-of-principle neuromorphic devices with multiple presynaptic inputs based on a polyvinyl alcohol (PVA) proton-conducting electrolyte, laterally coupled, 2D MoS2 electric-double-layer transistor, as shown in figures 10(a)–(c). They successfully emulated excitatory post-synaptic current (EPSC), PPF, dynamic filter, spatiotemporal signal dendritic integration, and spike logic. Bao et al demonstrated a MoS2 neuristor with a dual-gate transistor structure. An ionic top gate (TG)/electronic BG was designed to control the migration of ions/electrons, as shown in figures 10(d)–(f). When the BG (electronic gate) is grounded, the MoS2 neuristor behaves as a synaptic transistor, demonstrating STP, LTP, PPF, and a potential/depression process with good linearity and symmetry [240]. Furthermore, the IL gating control memristors could be used to achieve artificial modulation of many novel physical phenomena, such as metal–insulator phase transition, magnetic phase transition, superconducting transition, and so on.

### 3.3. Multi-terminal devices

Although two-terminal and three-terminal devices have demonstrated great potential for basic neural functions, synapses in the human brain outnumber neurons by more than a thousandfold, which implies that multi-terminal devices are needed to realize complex functions such as heterosynaptic plasticity. In the human brain, multi-synapses interact and work together. To mimic the multi-synapse interactions, Zhu et al reported a multi-terminal neuromorphic device, as shown in figure 11(a). The synaptic behaviors are modulated by the change of Li⁺ concentration, driven by a bias voltage. Comparing the two devices with the two configurations (figure 11(c)), we learn that the device with a higher concentration of Li⁺ ions has a better conductance response, revealing that Li⁺ ions take part in the conductance switching process [212].

Sangwan et al experimentally realized a multi-terminal hybrid memristor and transistor (memtransistor) using polycrystalline monolayer MoS2 [241]. The device demonstrated great switching properties and acted as a LRS–HRS memtransistor. The LRS and HRS resistances change by a factor of about 10⁴. The 2D planar geometry of the MoS2 memtransistor realizes multi-terminal neural circuits. In a six-terminal memtransistor, the conductance between any two of the four inner electrodes can be modulated by high-bias pulses applied to the two outer electrodes while the inner electrodes are disconnected. The six-terminal device demonstrated LTP/LTD, and indirect STDP.

Xie et al mimicked spatiotemporally-processed visual neurons by a coplanar multigate 2D MoS2 transistor with PVA electrolytes as laterally coupled gate dielectrics experimentally (figure 11(d)). The multigate array was regarded as the receptive field of a visual cortex cell, whereas the EPSC amplitude was measured as an activity of the spatial cortical cell, respectively. Based on the device, they realized some fundamental neuromorphic behaviors such as EPSC and paired-pulse facilitation successfully, as shown in figures 11(e)–(h), providing a promising approach to the current system design for artificial visual recognition [242].

Here, we made a table summarizing three-terminal/multi-terminal devices based on 2D materials such as TMDs, MXene, h-BN, graphene, LDM, and so on, as shown in table 3. The device structures, mechanisms, on/off ratios, power consumption, threshold voltages, retention times, and endurance of the various devices are shown in table 3. The on/off ratios vary from 10² to 10⁶, and power consumption reaches several nJ and even pJ. The threshold voltages of three-terminal/multi-terminal devices are generally higher than two-terminal devices.

| Operating mechanisms | Merits | Demerits |
|----------------------|--------|----------|
| Joule heating effect | Low threshold voltage | Heat-driven resistive switching |
| IL gating            | Better electron control ability | Integration difficulty of large-area IL gating |

Possible electrochemical reactions

| Possible electrochemical reactions |
|-----------------------------------|
| 1. Li⁺ + e⁻ → Li₀ |
| 2. Li⁺ + e⁻ → Li⁺⁺ |

Table 4. Continued.
Figure 12. (a) Schematic picture of the Ag/h-BN/graphene/h-BN/Au in the crossbar memory array architecture. (b) SEM images of 12 × 12 crossbar Ag/h-BN/graphene/h-BN memory array/Au. Scale bars: 5 µm for the SEM images. (c) I–V characteristics of a single memory cell. Reproduced from [90]. CC BY 4.0. (d) Optical microscope images of metal/h-BN/metal memristive crossbar arrays fabricated on two-inch wafers. (e) Optical of six 10 × 10 metal/h-BN/metal memristor crossbar arrays. Scale bars, 50 µm. (f) I–V cycling characteristics of a single Au/h-BN/Au memristor. (g) I–V cycling characteristics of a single Ag/h-BN/Ag memristor. Reproduced from [254], with permission from Springer Nature. (h) Schematic of a single WSe2 photodiode. (i) Macroscopic image of the photodiode array. Scale bar, 15 µm. (j) Illustration of the ANN photodiode array and circuit diagram of a single pixel in the photodiode array. (k) Schematics of the classifier and the autoencoder. Reproduced from [255], with permission from Springer Nature.

Given so many different mature RAM devices in commercial, we made a comparison between 2D material-based devices and other RAM devices. We summarized their merits and demerits in table 4. We can use 2D materials to make the device size smaller, overcoming the energy consumption problems with a high on/off ratio, and so on. Despite great advances in the neuromorphic devices based on 2D materials, there remain some problems to be solved.

4. Neuromorphic device array

Since the neural network of our brain is composed of 10¹¹ neurons and 10¹⁵ synapses, and its structure and function are complex, simulating the biological brain requires memristors integrated with a high density. The crossbar array consists of perpendicular top and bottom metal lines, named word lines and bit lines. Memristors can be integrated at the intersections of the word and bit lines.

The crossbar array can conduct parallel operations for matrix multiplication and addition, which can accelerate neural network computation. Therefore, the neural network based on memristors provides a solution for the construction of a new computing architecture that combines storage and computing [246–248]. The oxides-based memristive array has been widely reported [30, 249–252]. With the development of ultra-large-scale integration process technology, the size of silicon-based electronic devices has entered the nanometer scale, which is expected to encounter quantum effects that are different from classical devices [253].

With the development of large-scale integration process technology, the size of silicon-based electronic devices has reached nanometer scale. In the geometry, quantum phenomena such as short channel effect or tunneling (TL) current, which do not occur in large-scale devices, newly appear. Sun et al reported a 12 × 12 self-selective memory array constructed by stacking h-BN and graphene layers into a vertical structure of h-BN/graphene/h-BN between silver (Ag) and gold (Au) electrodes in a crossbar array structure, as shown in...
Figure 13. (a) The human optic nerve system is realized by integrating with the h-BN/WSe2 photodetector and h-BN/WSe2 synaptic device. (b) The simplified electrical circuit for the optic-neural synaptic device. (c) The recognition rate of the optic-neural network and neural network at 600 epochs. Reproduced from [19]. CC BY 4.0.

In addition, Chen et al. reported a wafer-scale integration of high-density memristive crossbar arrays for artificial neural networks using 2D h-BN. The arrays exhibit a high yield (98%), low cycle-to-cycle variability (1.53%), and low device-to-device variability (5.74%). Memristive crossbar arrays with Au/h-BN/Au and Ag/h-BN/Ag structures were fabricated by CVD large-area growth of multilayer h-BN and transferred to two-inch SiO2/Si wafers, as shown in figures 12(d) and (e). Figures 12(f) and (g) corresponds to the electrical tests of Au/h-BN/Au and Ag/h-BN/Ag, respectively [254].

Despite great achievements in memristor-based electrical crossbar arrays, the conversion of optical signals to electrical signals remains a bottleneck. Therefore, Mennel et al. designed a 2D photodiode array, where WSe2 plays a role as the photoactive material (figures 12(h) and (i)). Figure 12(j) schematically illustrates the basic layout of the image sensor and the corresponding circuit diagram. They implemented two types of ANNs: a classifier and an autoencoder, as shown in figure 12(k), which realizes the perception-storage-calculation of optical signals. Furthermore, the device could realize positive and negative photoconductivity. The optical signals can be used for training in the neural network, avoiding the digital-to-analog conversion of the signal, and greatly improving efficiency [255].

5. Applications of neuromorphic devices

Most of our brain’s knowledge of the surrounding environments comes from the sensory organs: eyes, ears, nose, tongue, and skin. Therefore, emulating human’s visual, auditory, tactile, and olfactory nerves is fascinating and attracts extensive research. Brain-inspired neuromorphic computing is considered as the most promising solution to provide effective emulation of the functionality of the human brain via the integration of artificial neuron and synaptic device components. Due to their remarkable electronic, optical, mechanical, and thermal properties, 2D materials provide an ideal platform to develop diverse functionalities for brain-inspired neuromorphic devices. Here, we introduce recently reported applications of neuromorphic devices based on 2D materials and their heterostructures.

5.1. Artificial visual

As one of the most important organs for acquiring information for human beings, biological vision has attracted many scientists to mimic the visual functions of the human brain through machine vision to realize ‘seeing’ and real-time recognition and information storage. Visual perception grasps more than 80% of the information in the process of human interaction with the surrounding environment [174, 256–259].

Recently, Sun et al. proposed a new multi-dimensional photoelectric fusion memristive device based on 2D layered tin sulfide (SnS). The device demonstrated reservoir computing by combining the sensory functions and successfully made such in-sensor reservoir computing electronic devices used in language symbol recognition and learning. Even with extremely similar interference items, a 91% recognition rate can be achieved for complex language systems. This research work overcomes the technical bottleneck of physically separated
sensors and reservoir computing calculations and greatly reduces the learning system complexity and operating costs. The new device deals with the urgent requirements of the explosive growth of big data processing in the Internet of Things era and provides a technological breakthrough for more effective machine learning and brain-like computing [223].

Similarly, Seo et al used a vdW heterostructure (h-BN/WSe2) to realize an optic-neural synaptic device with synaptic and optical-sensing functions, as shown in figure 13(a). The device demonstrated diverse synaptic dynamics with various light illumination conditions (red ($\lambda = 655$ nm), green ($\lambda = 532$ nm), and blue ($\lambda = 405$ nm)), and preserved its synaptic plasticity. Figure 13(b) shows the simplified electrical circuit for the optic-neural synaptic device. Simultaneously, the colored and color-mixed pattern recognition capability of the human visual system was emulated by an optic neural network, which achieved an >90% (figure 13(c)) recognition rate for the color-pattern recognition task, which is an analog to a color-blindness test [19].

5.2. Artificial tactile

Touch is another sensory way, one of the main ways for humans to coordinate and interact with surrounding environments. Sense of touch can help humans to evaluate the properties of objects, such as size, shape, texture, temperature, etc; ‘touch’ could transmit various sensory information such as pressure, vibration, pain, and temperature to the central nervous system, helping humans to perceive the surrounding environment and avoid potential harm. The human touch system consists of receptors, transmitters, and synapses that act as a medium for the perception of external mechanical stimuli and the transmission/processing of sensory signals [133, 260–264].

Chen et al reported a piezotronic graphene artificial sensory synapse by integrating a piezoelectric nanogenerator (PENG) with an ionic gel–gated transistor demonstrated (figure 14). Figure 14(a) shows the tactile receptors in human skin by which the received mechanical stimulus can be converted into presynaptic potentials. The presynaptic potentials are transmitted to the central nervous system through neurons and synapses. Figure 14(b) shows the schematic illustration of the proposed piezotronic artificial sensory synapse and the corresponding circuit diagram shown in figure 14(c).

The working mechanism of the device is to trigger the PENG to generate an induced electric field through mechanical strain, which causes the uneven distribution of ions in the IL, resulting in the change of the conductance in the graphene channel. In addition, they demonstrated the LTP and LTD of artificial synaptic plasticity by the tension and compression strain pulses applied to the PENG [265] (figures 14(d) and (e)).

5.3. Artificial auditory

Generally, the ear, as an auditory organ, receives the sound from all directions, converts the sound signals into neural signals, and transmits them to the nerve system in the brain, which is finally processed and understood.
by the brain. The functional goal of the auditory system is to detect and extract information from pressure waves in the surrounding medium, usually air or water. Sound waves with different amplitudes, frequencies, and compositions are produced by motion or collisions, and they primarily tell the perceiver what is happening in the environment [266–272].

Sun et al designed a synaptic device with the structure of Au–MoS$_2$–Au, realizing a key function for the most precise temporal computation in the human brain, sound localization, through detecting binaural time difference by suppressing sound intensity or frequency-dependent synaptic connections, as shown in figures 15(a) and (b). Synaptic weights could be modified by the metal–insulator transition in a scalable monolayer-molybdenum disulfide (MoS$_2$) induced by Joule heating and versatile doping. Furthermore, the energy consumption ($\sim$10 fJ) is comparable to biological (figure 15(c)). Figure 15(d) illustrates the confounding effect of the cue of interaural level difference (ILD) without our synaptic computation or inhibitory synaptic device [2]. This work demonstrates a breakthrough in neuromorphic computing in mimicking the complex and accurate information processing in the human brain.

Recently, Seo et al demonstrated a vdW-hybrid synaptic device with linear and symmetric conductance update characteristics and realized their applications in acoustic pattern recognition (figure 15(e)). Through the training and inference simulation, a high recognition rate of 93.8% was achieved by using the vdW-hybrid synaptic device, as shown in figure 15(f). The vdW-hybrid synaptic device holds great promise in highly accurate neuromorphic computing [272].
6. Challenges and outlook

In summary, recent advances in understanding the microscopic working mechanisms and applications of neuromorphic devices are reviewed in this manuscript. We summarized several methods for preparing 2D materials and heterojunctions. Then, we discussed the emerging neuromorphic devices. Depending on the number of terminals, the device could be divided into three kinds: two-terminal, three-terminal, and multi-terminal. For each kind, we clarify the underlying resistance switching mechanisms. After that, we introduced the neuromorphic device array and its applications on artificial visual, artificial tactile, and artificial auditory.

While great progress has been made so far, we are still far from system-level, brain-inspired neural computing that enables practical artificial intelligence with 2D materials. Some technical challenges remain to be addressed before the realization of commercial applications as follows.

(a) Wafer-scale synthesis of high-quality and uniform 2D materials; large-area and high-quality 2D crystals are the basis for the development of next-generation electro-optical devices. The synthesis of wafer-scale 2D materials is a critical step for industrial applications. At present, the CVD method is still considered to be the most effective technique, which can be industrially produced and relatively simple to operate. Besides CVD, there are several other methods to synthesize TMDCs. Such as pulsed laser deposition, atomic layer deposition ALD (PEALD), and molecular beam epitaxy.

(b) The compatibility with traditional CMOS fabrications; integrating devices with different functions in one chip significantly improves the performance of the circuit and reduces the cost. Solving the CMOS process compatibility problem of neuromorphic computing devices will enable large-scale integration of brain-like devices for commercial applications.

(c) Device development with low power consumption and high stability; the energy consumption of synaptic devices plays a critical role in the realization of neuromorphic computing. We need to further improve the device efficiency by reducing the contact resistance of 2D materials. As the metal electrode deposited on the 2D material will partially damage the covalent bonds in the atomic lattice and may introduce the Fermi level pinning and form Schottky contacts, the contact resistance increases.

(d) The high-density integration serving neural networks; the current integration density is much lower than the human brain containing $10^{11}$ neurons and $10^{15}$ synapses, so we need to develop high-density/high-capacity 3D stacked device arrays to address the challenge. Low-integration-density integrated circuits that take advantage of the advanced properties of 2D materials have remained an issue, and several companies have begun to commercialize them. In the future, high-density integrated circuits impose more stringent requirements in terms of yield, variability, reliability, and stability, thus requiring lower defect densities at 2D materials and their interfaces with other materials.

Therefore, for future computing, we should first prepare proper 2D materials that can achieve high-performance devices through a precise physical design. Secondly, designing new device structures to overcome the limitations of the von Neumann bottleneck is critical. Finally, integrating reliable device functions is required for system-level applications. All the efforts should be comprehensively promoted in all aspects of the developments: materials design, device fabrication, and circuit integration. Then, we will be able to realize brain-like computing systems for real-world applications.

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Data availability statement

All data that support the findings of this study are included within the article (and any supplementary files).
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