Memristor based on α - In_2Se_3 for emulating biological synaptic plasticity and learning behavior

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ABSTRACT Nowadays, memristors are extremely similar to biological synapses and can achieve many basic functions of biological synapses, making them become a new generation of research hotspots for brain-like neurocomputing. In this work, we prepare a memristor based on two-dimensional α - In_2Se_3 nanosheets, which exhibits excellent electrical properties, faster switching speeds, and continuous tunability of device conduction. Meanwhile, most basic bio-synapse functions can be implemented faithfully, such as short-term memory (STM), long-term memory (LTM), four different types of spike-timing-dependent plasticity (STDP), and paired-pulse facilitation (PPF). More importantly, we systematically study three effective methods to achieve LTM, in which the reinforcement learning can be faithfully simulated according to the Ebbinghaus forgetting curve. Therefore, we believe this work will promote the development of learning functions for brain-like computing and artificial intelligence.

Keywords: memristors, biological synapse, learning behaviors, 2D In_2Se_3

INTRODUCTION

Due to the limitations of the Von Neumann architecture, the traditional storage-separated computing approach can no longer meet the fast-growing demand for big data computing [1–3]. Recently, people have started to turn their attention to storage-integrated bio-neural computing due to that there are 10^{11} neurons and 10^{15} synapses in the human brain, which can integrate complex cognitive functions such as perception, calculation, logic, language, vision, and memory [4–6]. Hence, the implementation of biological neuron and synaptic bionics functions for artificial intelligence (AI) will break the bottleneck of traditional computing methods and promote the development of scientific computing technology. Fortunately, emerging memristors with continuous tunability of conduction have become a new generation of research hotspots due to their ability to effectively mimic biological neuro-synapses [7–9].

It is well known that the act of learning is one of the important behaviors of human psychology and one of the main means to train neural networks. In biological neural networks, the pre-

synaptic membrane releases neurotransmitters (Na^+ , K^+ , and Ca^{2+}) to stimulate the post-synaptic membrane for increasing or depression functions, and different concentrations of ions in the synaptic gap affect the strength of synaptic excitation/depression, that is, the synaptic weights change accordingly [10–12]. Ingeniously, the conductance of the memristor can be positively and negatively modulated to mimic weight changes for biological learning functions, such as short-term plasticity (STP), long-term potentiation (LTP), long-term depression (LTD) behavior, short-term memory (STM), long-term memory (LTM), spike-timing-dependent plasticity (STDP), and paired-pulse facilitation (PPF). However, new bio-synapse functions in memristor are requested to be further developed for AI, for example, reinforcement learning which has a very important role in enhancing memory ability and achieving targeted learning effects.

In recent years, two-dimensional (2D) materials are widely used as functional layers and structural materials in various fields, like transistors, batteries, catalysts, and nanodevices, due to their unique advantages, such as excellent electrical properties, structural stability, and unique layered structure [13–19]. Especially, 2D material-based memristors with small size, low power consumption, and fast switching speed can further extend their advantages and show good prospects for applications [20–25]. For example, Chen *et al.* [26] reported a high-density memory array based on hexagonal boron nitride as a resistive switching material, and constructed an artificial neural network for image recognition. Yan *et al.* [27] reported a low-power neurosynaptic device based on 2D WS_2 . Zhu *et al.* [28] reported a memristor device based on 2D transition-metal dichalcogenides (TMDS) for implementing synaptic competition and cooperation effects in a bionic artificial neural network. All these have proved that memristors based on 2D materials have great development prospects in the future.

In this work, we investigated the performance of a memristor based on α - In_2Se_3 thin films. We have spin-coated α - In_2Se_3 on the indium tin oxide (ITO) substrate and deposited Ag electrode as the top electrode to form a device structure of Ag/ α - In_2Se_3 /ITO. This memristor exhibits typical bipolar resistive switching behavior, excellent endurance, faster switching speeds (16.4 ns for SET process and 18.0 ns for RESET process), and continuous

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tunability under alternating-current (AC) pulse stimulation. The memristor also realizes four different kinds of STDP learning rules. Furthermore, we systematically investigated three effective learning methods (supervised learning, interest-based learning/memory behavior, and reinforcement learning); among them, the reinforcement learning behavior was successfully simulated, laying the foundation for further brain-like computing and AI applications.

EXPERIMENTAL SECTION

Fabrication

The sandwich structure of Ag/ α -In₂Se₃/ITO device was prepared by two methods: magnetic sputtering and spin coating. The α -In₂Se₃ nanosheet dispersions were prepared at a concentration of 1 mg mL⁻¹, with thicknesses of 1–10 layers and lateral dimensions of 0.05–1 μ m. At first, the α -In₂Se₃ nanosheet dispersions were spin-coated onto the ITO substrates and dried at room temperature, and the process was repeated three times to form three α -In₂Se₃ films, respectively. Then, with the help of a shaded mask of 90 μ m diameter, the top electrode of Ag was sputtered using direct-current (DC) reactive magnetron.

Characterization

The basic crystal structure of the material was tested using a

transmission electron microscope (TEM). The X-ray photoelectron spectroscopy (XPS) measurements were performed on an Escalab 250Xi using a monochromat Al-K α source at 1486.6 eV. Electrical characterization measurements of the device, including DC and pulsed electrical measurements, were performed in the atmosphere using a semiconductor characterization system (Keithley 4200SCS). The pulses were emitted using a Agilent 33250A signal generator and the pulse waveforms were captured with an oscilloscope (LeCroy62MXs-B). During the measurement, the Ag electrode was connected to the positive terminal and the ITO electrode was connected to ground.

RESULTS AND DISCUSSION

We used TEM to elaborate the basic microstructure morphology of α -In₂Se₃ nanosheets, which are distributed in sheets, as shown in Fig. 1a. The high-resolution TEM (HRTEM) image of the material is depicted in Fig. 1b, where the lattice spacing is 0.35 nm. The selected area electron diffraction (SAED) pattern in the inset is consistent with Ref. [29]. This material has good crystallinity and obvious layered structure, as shown in Fig. S1. XPS can reveal the chemical composition and chemical valence state of α -In₂Se₃. The peaks of 452.4 and 444.9 eV correspond to In 3d_{5/2} and In 3d_{3/2}, respectively (Fig. S2a), while the peaks of 54.1 and 55.0 eV correspond to Se 3d_{5/2} and Se 3d_{3/2} (Fig. S2b),

Figure 1 (a) The TEM image of α -In₂Se₃ nanosheets. (b) HRTEM of α -In₂Se₃ nanosheets in (a). The inset is SAED pattern. (c) Schematic of the Ag/ α -In₂Se₃/ITO device structure. (d) Typical *I-V* curve in linear coordinates. (e, f) Distributions of the SET/RESET voltage for a single device: (e) SET voltage; (f) RESET voltage. The red line is the result of Gaussian fitting. (g) The SET response time of this memristor. (h) RESET response time. The blue line denotes the input pulse, and the output conducting current in logarithmic scale is represented by orange shapes. (i) Statistical distribution of the endurance of the device.

respectively, in agreement with Ref. [30]. Then, we construct the Ag/ α -In₂Se₃/ITO device with ITO as the substrate, and the device structure is shown in Fig. 1c. Electrical tests were performed by applying a forward voltage to the top electrode and the bottom electrode. A typical current-voltage (*I-V*) curve, as shown in Fig. 1d, demonstrates that the device has apparent bipolar behavior [31]. Among them, the switch from the high-resistance state to the low-resistance state is the SET behavior with the threshold voltage of about 0.90 V. This device is pulled back from the low-resistance state to the high-resistance state (RESET behavior) by applying the -0.44 V reverse voltage. We further elaborate the conduction mechanism of the device (Fig. S3). In the high-resistance state region (the green dashed area), we split the fit into three parts with slopes of 1.02, 2.06, and 4.08. The fit results are more consistent with the trap-controlled space charge-limited current (SCLC) [32]. The low-resistance state (red region) shows the typical ohmic conduction behavior with a slope of 1.04, close to 1, which is in accord with the resistive switching mechanism based on the formation and rupture of Ag conducting filaments. In order to further prove the switching mechanism of the Ag conductive filament formation and rupture process of the device, we supplemented the research on the effect of temperature on the low-resistance state of the device. We read the resistance value at different temperatures with 500 mV, and found that as the temperature increases, the resistance value also increases, showing typical metal characteristics, as shown in Fig. S4. This also indicates the formation of Ag conductive filaments in the low-resistance state. And the relationship between the temperature and resistance can be expressed as: $R(T) = R_0[1 + \alpha(T - T_0)]$, where R_0 is the resistance at the initial temperature of T_0 , and α is a constant. The R/R_0 resistivity has a good linear relationship with temperature, with a slope of 3.3×10^{-3} , which is very close to the value of silver nanowires reported in previous studies [3].

To check the distribution of the device switching voltage, the Gaussian fitted histograms of the single device's switching voltages are statistically analyzed according to the *I-V* characteristics as shown in Fig. 1e, f [33]. It can be seen that the threshold voltages of the device are around 0.96 and -0.50 V. On this basis, we measured the *I-V* characteristics of five different devices and performed statistical and Gaussian fitting of their switching voltages, as shown in Fig. S5. The switching voltage distribution is relatively concentrated, which greatly improves the control accuracy of the programming voltage in the circuit and promotes the practical application of α -In₂Se₃. The SET and RESET response times of the device can be found to be 16.4 ns (Fig. 1g), and 18.0 ns (Fig. 1h), respectively, exhibiting fast switching speeds [34]. In addition, we measured the device in the pulse mode (Fig. S6) and found that it can repeat for 1×10^6 cycles, indicating its excellent endurance, as shown in Fig. 1i. Moreover, the distributions of high- and low-resistance states of the device are more concentrated, mainly in 10^6 and $10^3 \Omega$, respectively (Fig. S7). Also, we calculated the SET and RESET powers of the device as 4.96×10^{-6} and 2.45×10^{-5} W, respectively, indicating that the device possesses low power dissipation properties (Fig. S8). Notably, the device retains its original characteristics after one year of placement, demonstrating excellent ambient stability, as shown in Fig. S9. We compared the device performance with previously reported devices in the literature, demonstrating the good electrical properties of this device, as shown in Tables S1 and S2. Besides, the device can be

continuously tunable under AC pulse stimulation by changing the pulse parameters, which proves that it has a good synaptic bionic function, as shown in Fig. S10 [35,36].

To further demonstrate the continuous tunability of the conductance, we measured the device by defining individual pulse groups applied to the device (Fig. S11). Initially, we applied five positive pulses (pulse amplitude from 1 to 2 V, and from 2 to 1 V, Fig. S11a) and five negative pulse sets to stimulate this device (pulse amplitude from -1 to -2 V, and from -2 to -1 V, Fig. S11b). From Fig. S11c, d, it can be found that the conductance can be continuously changed, and the sandwich structure of the device can be viewed as a pre- and post-synaptic neuron and synapse, with Ag ions as neurotransmitters to simulate the change of biological synaptic weights, thereby simulating the function of biological neurons [37]. Further, biological neurological memory is divided into STM and LTM, while STM can form LTM by repeated stimulation, as shown in the model diagram illustrated (Fig. 2a) [38,39]. To demonstrate this function, we compiled pulse waveforms with a pulse amplitude of 1.5 V and a pulse width and pulse interval of 500 ns. Firstly, we applied 25 cycles of continuous pulses, and the current increased with increasing pulse stimulation, but decayed significantly after the thirteenth pulse stimulation, which can be explained by the spontaneous decay of the conducting filament. This process is called STM behavior, as shown in Fig. 2b. After that, we increased the pulse amplitude (Fig. 2c), and found that after five pulses, the current reached saturation, which is the LTM behavior in biological neurological memory. Therefore, this result proves that the device can realize the transition from STM to LTM behavior and effectively simulate biological synapses. We also systematically studied and simulated four different STDP learning rules to lay the foundation for the application of 2D material memristors to neural networks [37,40–42]. By applying the pulse waveform as shown in Fig. S12a, we found that as the pulse interval time increased, synaptic plasticity was almost close to 0. We calculated ξ values and fitted them according to Equation S1 to obtain the Hebbian STDP learning rule which is fully consistent with biological neural learning (Fig. 2d). Different types of pulses are imposed (Fig. S12b–d) to implement the anti-Hebbian STDP learning rule (Fig. 2e), symmetrical STDP (Fig. 2f) and visual STDP (Fig. 2g), respectively, laying the foundation for biological neural information processing. In addition, our device also effectively implements the PPF bio-synaptic function, as shown in Fig. S13 [43].

To further investigate the biological neural function, we simulated three learning behaviors in human psychology, including supervised learning, interest-based learning/memory behavior, and reinforcement learning [44–46]. We applied a pulse sequence with an amplitude of 2 V to the device as the initial voltage, and set the supervision voltage (V_s) in the range of 0.002–0.30 V, as shown in Fig. S14a. When the supervision intensity is milder ($V_s = 0.002$ V), the device conductance decays rapidly to the initial value (Fig. S14b). When the supervision voltage is enhanced to 0.01, and 0.10 V, the conductance holding time increases to 5 and 7 μ s, respectively. Then the conductance decays to the initial value again (Fig. S14c, d). We further increased the supervisory voltage to 0.20 V, at which point the conductance showed an increased retention time, but still decayed substantially, as shown in Fig. S14e. When the supervision voltage is enhanced to 0.30 V, the conductance always

Figure 2 (a) Schematic sketching map of the brain memory in Atkinson and Shiffrin's model. (b) Current response after 25 consecutive positive pulses with an amplitude of 1.5 V, STM behavior. (c) Current response after 25 consecutive positive pulses with an amplitude of 3 V, LTM behavior. (d) Hebbian STDP learning rule. (e) Anti-Hebbian STDP learning rule. (f) Symmetrical STDP learning rule. (g) Visual STDP learning rule.

remains in a stable state without significant decay (Fig. S14f). This result shows that when the intensity of supervision increases, the learning effect of biology is better. To further investigate this supervised learning behavior of the device, we applied a pulse with a supervised voltage of 0.30 V to the device, and then gradually withdrew the supervised voltage to 0.002 V to observe the conductance change of the device. Firstly, we set the time of withdrawal of supervision as 5 μs , and found that the conductance decreased significantly at each withdrawal of supervision, until the supervision voltage dropped to 0.18 V, when the conductance completely dropped to the initial state, as shown in Fig. 3a; secondly, when we shorten the time to 3 μs , the conductance had a slight instability at each withdrawal of supervision. When the withdrawn voltage reached 0.10 V, the conductance dropped to the initial state (Fig. 3b); Finally, when the supervised withdrawal time is 1 μs , there is almost no obvious change in conductance, indicating that the learning at this time is not disturbed, as shown in Fig. 3c. It can be seen

more clearly through the histogram that when the withdrawal voltage time is 1 μs , the high conductivity is not affected. When the withdrawal voltage time is 3 or 5 μs , the conductivity is relatively unstable (Fig. 3d). This demonstrates that strong supervision enables LTM of the device, effectively simulating human supervised learning behavior.

Interest-based learning/memory is another one of the important behavioral patterns of biological neural learning [44]. As we all know, learning efficiency for being interested in things is much better than uninterested learning, and the strength of learning interest often affects the time of learning or memory. In this study, we regarded different reading voltages as the strength of learning interest, and stimulated the device by a pulse sequence with an amplitude of 2 V to form conductive filaments. The reading voltage (0.3–0.5 V) placed during the pulse sequence represents the learning interest intensity (Interest voltage, V_1), which is used to stabilize the conductive filament. And the reading voltage (0.1 V) applied after 10 μs is the recall

Figure 3 Supervised behavior. (a–c) Supervised behavior is withdrawn to different levels for relaxation times of 1, 3, and 5 μs . (d) Conductivity states at different withdrawal voltages in different relaxation times.

Figure 4 Interest-based learning/memory behavior. (a) Test pulse sequences with different reading voltages representing different levels of interest. (b) 0.3 V represents the low interest. (c) 0.4 V represents the moderate interest. (d) 0.5 V represents the high interest.

ability, as shown in Fig. 4a. It can be found that when the interest voltage is 0.3 V, the learning interest is the lowest, and it has been forgotten after 10 μs , as shown in Fig. 4b. To further enhance the learning interest, we set the interest voltage to 0.4 V. At this time, after a 10 μs interval, the device could recall time up to 150 μs (Fig. 4c). Finally, when the interest voltage is increased to 0.5 V, the learning interest reaches the peak and the recall capacity is the strongest, which can reach 2000 μs , as shown in Fig. 4d. This proves that the strength of learning interest affects

the biological recall ability, and confirms that the device has biological learning performance.

The Ebbinghaus forgetting curve illustrates the law of forgetting information in the human brain, which shows that forgetting occurs after learning and is initially faster; over time, forgetting slows down [46]. And reinforcement learning is one of the most important tools to achieve LTM effectively. Reinforcement learning does not require any data to be given in advance, but rather stimulates learning by giving rewards

(punishments). Here, we applied ten sequential pulses of 2 V amplitude and 500 ns pulse width as the learning process, then sequentially gave five stimulus pulses as rewards and mainly investigated the effect of different times on the learning effect. Firstly, we set the stimulus intervals Δt to 5, 4, 3, 2, and 1 μs , respectively ($\Delta t_1 > \Delta t_2 > \Delta t_3 > \Delta t_4 > \Delta t_5$), as shown in Fig. 5a. When the initial stimulus is given after 5 μs , the conductance is found to decrease substantially, indicating a significant decline in learning memory at this time; while when the stimulus is given for a gradually shorter time, the conductance increases again, indicating that the stimulus is given all the time to strengthen the memory to a certain extent, as shown in Fig. 5b. Secondly, we set the stimulation interval time to a fixed value, $\Delta t_1 = \Delta t_2 = \Delta t_3 = \Delta t_4 = \Delta t_5 = 3 \mu\text{s}$ (Fig. 5c). At this time, the conductance decreased slightly when the stimulus is given for the first time, indicating that there is no significant decrease in memory and further stimulation is performed and the conductance is found to be steadily increasing, as shown in Fig. 5d.

Finally, we further shorten the time of the first stimulation, making $\Delta t_1 = 1 \mu\text{s} < \Delta t_2 = 2 \mu\text{s} < \Delta t_3 = 3 \mu\text{s} < \Delta t_4 = 4 \mu\text{s} < \Delta t_5 = 5 \mu\text{s}$ (Fig. 5e), and find that the conductance value increases significantly when the first stimulation time is shortened compared with the previous one. With the increase of stimulation time, the conductance value gradually rises and stabilizes, indicating that the learning memory at this time has been transformed into LTM, as shown in Fig. 5f. This result shows that reinforcement learning can achieve the transformation from STM to LTM and effectively enhance the learning effect. In addition, we compared the synaptic bionic functions of other devices in recent years to demonstrate the unique learning behavior of our devices as shown in Table S3.

CONCLUSIONS

In summary, we prepared a memristor using the 2D $\alpha\text{-In}_2\text{Se}_3$ as the functional layer. The device exhibits excellent properties, such as electrical properties, a relatively concentrated threshold

Figure 5 Reinforcement learning. (a) $\Delta t_1 = 5 \mu\text{s} > \Delta t_2 = 4 \mu\text{s} > \Delta t_3 = 3 \mu\text{s} > \Delta t_4 = 2 \mu\text{s} > \Delta t_5 = 1 \mu\text{s}$. (b) Corresponding to (a) conductance change. (c) $\Delta t_1 = \Delta t_2 = \Delta t_3 = \Delta t_4 = \Delta t_5 = 3 \mu\text{s}$. (d) Corresponding to (c) conductance change. (e) $\Delta t_1 = 1 \mu\text{s} < \Delta t_2 = 2 \mu\text{s} < \Delta t_3 = 3 \mu\text{s} < \Delta t_4 = 4 \mu\text{s} < \Delta t_5 = 5 \mu\text{s}$. (f) Corresponding to (e) conductance change.

voltage, faster switching speeds (16.4 and 18.0 ns), good endurance, and continuous tunability under AC pulse stimulation. The device can successfully implement LTM in the nano-second pulse time range and simulate the biological synaptic behavior, like STDP and PPF. Notably, this study successfully simulated important human psychological behaviors, such as supervised learning, interest-based learning/memory behavior, and reinforcement learning. Among them, the successful simulation of reinforcement learning in a single device provides a new way to further implement brain-like computers and artificial intelligence.

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Author contributions Yan X proposed the idea, integrated interpretation and revised the paper. Zhao Y completed the performance test of the device and drafted the manuscript. Pei Y fabricated the device and assisted in writing the manuscript. Zhang Z revised the paper. Li X, Wang J, Yan L and He H helped with characterization analysis. Zhou Z, Zhao J, and Chen J coordinated the work. All authors commented on the final paper.

Conflict of interest The authors declare that they have no conflict of interest.

Supplementary information Details of the simulation and supporting data are available in the online version of this paper.

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基于二维 α - In_2Se_3 忆阻器的突触可塑性和学习行为

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摘要 忆阻器与生物突触极为相似, 可以实现生物突触的基本功能, 使其成为了新一代类脑神经计算的研究热点. 在这项工作中, 我们制造了基于二维 α - In_2Se_3 材料的忆阻器件, 其表现出了优异的电学性能、较快的开关速度(16.4和18.0 ns) 以及器件电导的连续可调性. 同时, 大多数基本的生物突触功能得以实现, 如短时记忆(STM)、长时记忆(LTM)、四种不同类型的尖峰时间依赖可塑性(STDP)和双脉冲易化行为(PPF). 更重要的是, 我们系统性地研究了三种实现长时记忆的有效方法, 其中, 根据艾宾浩斯遗忘曲线成功地模拟了强化学习功能. 这项工作将促进类脑神经计算以及人工智能在学习方面的研究发展.