Memtransistors Based on Non-Layered $\text{In}_2\text{S}_3$ Two-Dimensional Thin Films With Optical-Modulated Multilevel Resistance States and Gate-Tunable Artificial Synaptic Plasticity

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ABSTRACT

Memtransistor, a hybrid structure that integrates the function of memristor and transistor, is a promising device prototype for the realization of complex neuromorphic learning owing to its diverse functionality and additional flexibility in emulating synaptic behaviors. Memtransistor of two-dimensional (2D) chalcogenide materials have received many interests as it has distinctive memristive mechanism quite different from conventional oxide memristors. Here, we report a memtransistor based on the two-dimensional thin films (2DTFs) of non-layered $\beta$-In$_2$S$_3$. The In$_2$S$_3$ 2DTFs grown by physical vapor deposition method have microscopically visible grain boundaries (GBs) formed by the stacking and interconnecting of 2D In$_2$S$_3$ flakes. The memtransistors of In$_2$S$_3$ 2DTFs show tunable bipolar resistive states with resistance ratio up to $10^5$, endurance over 200 cycles, and a retention time of $10^4$ s. Illumination of laser light from visible and near-infrared are able to induce intermediate resistance states in memtransistors, enabling optical-modulated multilevel memory storage. Also, the memtransistors are able to emulate the synaptic function of long-term potentiation (LTP) and long-term depression (LTD) with tunable synaptic weight in response to presynaptic stimuli of drain/gate pulses. Interestingly, the plasticity of LTP and LTD behavior can be switched in a highly tunable manner by simply varying the gate voltages. The diverse optoelectronic properties and controllable functionality of memtransistors based on the emerging 2D In$_2$S$_3$ offer a useful guide to potential application in electronic memory and artificial synapses.

INDEX TERMS

Memristors, field effect transistors, thin film devices, electric resistance.

I. INTRODUCTION

Human brain, the most efficient computational entity, consists of $10^{11}$ neurons interconnected by $10^{15}$ synapses, forming a sophisticated and complex nervous system. The neuron-to-neuron connection strength is regulated by synapses through the short-term plasticity (STP) and long-term potentiation (LTP), synaptic learning rule, dendrite integration and neural arithmetic [1]. Inspired by human brain, neuromorphic computing at physical level provides a promising solution to overcome the long-standing bottleneck of energy efficiency and processing speed in conventional von-Neumann computing architecture [2], [3]. Artificial synapses in neuromorphic computing system have been widely investigated in a class of basic circuit element called memristor [4]–[6]. Memristor-based device can keep track of the past resistance state through which it has experienced. The associate editor coordinating the review of this manuscript and approving it for publication was Anisul Haque.
decades, such as resistive memristor [8], [9], phase change memory [10], organic memristor [11], ferroelectric memristor [12], manganite memristor [13], resonant-tunneling diode memristor [14]. In recent years, 2D materials attract intensive research interests since it has demonstrated excellent memristive properties and device performance in artificial synapse applications [15]–[20]. Particularly, Sangwan et al. revealed a brand-new memristive mechanism based on the migration of sulfur vacancy defects mediated by grain boundaries (GBs) [19], followed by a demonstration of device’s scalability and diverse functionality using bottom-gate multiterminal memristive transistor (memtransistor) based on CVD-grown polycrystalline monolayer MoS$_2$ [20]. In this regard, one may anticipate grain-boundary mediated memristive phenomena can be found in some other 2D materials (especially layered transition metal dichalcogenides). However, relevant reports have not appeared in material other than MoS$_2$ [19]–[21]. Based on the underlying physics of this phenomena, a kind of 2D material building block, namely 2D van der Waals thin films (vdWTFs) [22], may be a potential candidate. The vdWTFs are fabricated by assembling 2D nanosheets into continuous thin films in which neighboring sheets interact via van der Waals forces and form natural GBs in interfaces. The fabrication strategy of vdWTFs not only ensure optimum charge transport properties in large-area thin film, but also produce moderate transport barrier across neighboring 2D nanosheets that would give rise to the possible memristive phenomena. An attempt to uncover the memristive properties and synaptic device application for the emerging vdWTFs and their analogue is greatly worthy.

$\beta$-In$_2$S$_3$ is an emerging 2D nonlayered material with medium bandgap and unique defective structure [23]. Our previous studies have revealed the unique thickness-dependent optical properties of 2D $\beta$-In$_2$S$_3$ and the ultrasensitive broad-range optical response of In$_2$S$_3$ based van der Waals heterostructure [24], [25]. In this letter, a high-quality $\beta$-In$_2$S$_3$ analogue of van der Waals thin films was grown through physical vapor deposition (PVD) method. Similar to the structure of vdWTFs, the as-grown thin films are assembled by the stacking and interconnecting of 2D In$_2$S$_3$ flakes, thus forming rich microscopically-visual GBs on surface. It is noteworthy that 2D vdWTFs refer to nanosheet-assembled thin films of layered materials in which neighboring sheets interact via weak van der Waals forces. But for non-layered material, an atom has strong covalent bonds in three dimensions. Therefore, in spite of the very similar construction, we referred our continuous thin films of non-layered $\beta$-In$_2$S$_3$ as 2D thin films (2DFTFs) rather than vdWTFs, to avoid confusion. The memtransistors based on In$_2$S$_3$ 2DFTFs exhibit stable resistive states with switching ratio up to $10^6$ and retention time over $10^4$ s. The memtransistors exhibit multilevel resistance states and resistive switching ratio under the illumination of 405, 532, 635 and 808 nm laser light. Also, the devices are able to emulate the synaptic plasticity of long-term potentiation (LTP) with tunable potentiation strength depending on the amplitude and width of the input-pulses. Three behaviors of the synaptic plasticity, plasticity “off”, LTP and long-term depression (LTD), as well as their synaptic weight, can be effectively controlled by changing the gate voltage, offering diverse and controllable functionality in mimicking biological plasticity of synapses.

**II. EXPERIMENT**

The In$_2$S$_3$ 2DFTFs were grown on mica substrate through epitaxy by physical vapor deposition (PVD). As illustrated in Fig. 1(a), 20 mg In$_2$S$_3$ (99.999%, Alfa) powder was placed in the center of the furnace, while the precleaned mica substrates on a ceramic boat was placed at the downstream end of the furnace, 10 cm away from the In$_2$S$_3$ powder. The system was purged by Ar (99.999%) with 200 sccm flow rate for 1 h. Then the furnace was ramped to a temperature of 980 °C for 10–20 min under an ambient pressure and a constant Ar flow rate of 50 sccm. After the growth, the samples were naturally cooled down to room temperature in Ar atmosphere. Finally, the In$_2$S$_3$ thin films were transferred from mica to SiO$_2$ (300nm)/Si substrates and 50-nm-thick Au as electrode was deposited on In$_2$S$_3$ through standard lithography process and thermal evaporation, detail of which can be found in our previous work [25], [26].

![FIGURE 1.](image-url)
The morphology of the resulting products was analyzed with a scanning electron microscope (SEM, Hitachi, S-3400N). The thickness and surface topology were measured using an atomic force microscope (Bruker, FastScan). Raman spectroscopy was conducted using a confocal Raman microscope (ZOLIX, Finder Vista) at an excitation wavelength of 532 nm. X-ray diffraction (XRD) of $\beta$-In$_2$S$_3$ was performed with a Bruker D8 Advance X-ray diffractometer (Co Kα, $\lambda = 0.178897$ nm) equipped with a LynxEye detector in a Bragg-Brentano configuration. The chemical composition analyses of samples were conducted by x-ray photoelectron spectroscopy (Thermo Fisher, Escalab 250Xi).

The electrical characteristics of the devices were studied on a four-probe table (Semishare SM-4, China) combined with the 2614B and 4200 source meters (KEITHLEY, America).

III. RESULTS

A. MATERIALS AND DEVICES

The as-grown In$_2$S$_3$ is in the form of substrate-scale continuous thin films, as shown on SiO$_2$(300 nm)/Si substrate in Fig. 1(b) after transferred from mica. A back-gate field effect transistor (FET) configuration using the In$_2$S$_3$ 2DTFs as channel material is illustrated in Fig. 1(c). The FET is fabricated by depositing Au as source and drain electrode through standard lithography process and thermal evaporation. The 500 $\mu$m thick phosphorus-doped Si substrate with resistivity of 0.001-0.005 $\Omega \cdot$ cm is used as the gate electrode. As can be seen in Fig. 1(d)-(g), In$_2$S$_3$ 2DTFs with different thicknesses exhibit very different morphology, from which the grow mechanism in various stage is discussed. On the early stage, In$_2$S$_3$ nucleates and epitaxially grows in lateral direct under appropriate growing condition, forming triangle flakes on mica substrate (Fig. 1(d)). As the growth continues, triangle flakes cover the whole substrate and new flakes start to grow on the existent one (Fig. 1(e)). Then, the 2D flakes grow and merge together, gradually forming a continuous and smooth film (Fig. 1(f)). This growing mechanism suggests that the In$_2$S$_3$ thin films are assembled by the stacking and interconnecting of the 2D flakes. Also, by choosing samples with appropriate thickness, we are able to balance the requirement of GBs and surface smoothness for the realization of memristor device. As shown in the high-magnification optical image of Fig. 1(g), a 60 nm thick In$_2$S$_3$ 2DTFs with vivid stacking and interconnecting among In$_2$S$_3$ flakes were used for the device fabrication. The flakes with lateral length of 10−50 $\mu$m contacts mutually to form visible physical boundaries. The images of scanning electron microscope (SEM) in Fig. 1(h) can see more identified superficial GBs as it focuses on the material surface. As visualized in Fig. 1(i) and 1(j), there are many clear boundaries of In$_2$S$_3$ flakes cross the channel area between two Au electrodes, which corresponds to the so-called bridge-GB memristor [19]. The AFM image of a 60 nm-thick In$_2$S$_3$ films in Fig. 1(k) and its height profile in Fig. 1(l) further reveal the layered structure on its edge and a vertical stacking of thin nanosheets. The stacking In$_2$S$_3$ nanosheets have typical thickness ranged from several nanometres to tens of nanometres, suggesting that the thin films are lamellar. Optical characterization might be an effective approach to distinguish 2D material from its bulk counterpart. However, experimental results of our previous work [24] and an earlier paper [23] have both demonstrated that Raman peak position of $\beta$-In$_2$S$_3$ does not shift as its thickness reduces from bulk to few-layer. Also, there are no experimental evidences so far that the bandgap energy of $\beta$-In$_2$S$_3$ is thickness-dependent. Therefore, it might be difficult to identify the 2D structure of In$_2$S$_3$ thin films via optical approach.

Rather than using the colloidal chemical synthesis process reported in the fabrication of vdWTFs [22], we for the first time adopt the PVD growth approach to achieve the analogue of vdWTFs. The stacking of 2D materials by epitaxy growth provides much stronger plane-to-plane contacts between neighbouring nanosheets with an efficient reduction of dangling bonds at interfaces. In the meantime, while maintaining superior charge transport within the film, the surface of In$_2$S$_3$ 2DTFs preserves eminent GBs that can possibly induce memristive phenomena.

The Raman spectra in Fig. 2(a) exhibits several characteristic Raman peaks located at 115.2, 135.6, 161.8, 246.8, 308.1, 327.1 and 367.6 cm$^{-1}$, which correspond to Raman active modes $E_g$ and $B_{2g}$ [24]. As shown in Fig. 2(b), the very sharp four peaks of X-ray diffraction (XRD) spectra located at 14.3°, 28.8°, 43.7° and 59.5° suggest the very high quality of the tetragonal In$_2$S$_3$ (PDF#73-1366). Fig. 2(c) exhibits X-ray photoelectron spectroscopy (XPS) spectra of In$_{3d}$ and S$_{2p}$, for the In$_2$S$_3$ 2DTFs. The binding energies for In$_{3d5/2}$ and In$_{3d3/2}$ are 443.2 and 450.7 eV with a $\Delta$E of 7.5 eV, while those of S$_{2p3/2}$ and S$_{2p1/2}$ are 160.5 and 161.7 eV with a $\Delta$E of 1.2 eV, which agrees well with results from the previously reported $\beta$-In$_2$S$_3$ [27], [28]. The above characterizations reveal the pure tetragonal phase of $\beta$-In$_2$S$_3$ and agree well with the results of 2D $\beta$-In$_2$S$_3$ flakes synthesized by similar PVD approach in our previous study [24].

B. ELECTRICAL PROPERTIES

The memtransistors based on In$_2$S$_3$ 2DTFs were under the test of electrical hysteresis to switch their resistance states. A closed-loop sweeping of $I_{ds}$-$V_{ds}$ at $V_g = 0$ under vacuum is plotted in Fig. 3. The memtransistors in vacuum shown a largely separated high-resistance state (HRS) and low-resistance state (LRS) after dual sweep, indicating that the hysteresis is the intrinsic property of In$_2$S$_3$ vdWTFs rather than from the absorption of O$_2$ and H$_2$O in the air by vacancies of sulfur atoms [29]. In addition, the FETs fabricated on single In$_2$S$_3$ flake (Fig. 3(b) and its inset) recorded a weak hysteresis in vacuum with resistance switching ratio about four orders of magnitude smaller than that of In$_2$S$_3$ 2DTFs in Fig. 3(a), suggesting that the hysteresis might related to the rich GBs of the 2D flakes. As exhibited in Fig. 3(a), the electroformed memtransistor in the positive bias sweep shows an HRS at $V_{gs} = 0$ V that changes to an LRS at high bias after an abrupt increase in current. In the negative
bias sweep, the device is at an LRS initially and then changes to HRS, corresponding to a reset process. The HRS and LRS of the memtransistors can be effectively modulated by changing the sweep range of $V_{ds}$, as illustrated by the $R_{HRS}/R_{LRS}$ at $V_{ds} = 5$ V in Fig. 3(c). The switching ratio changes from 3 for $\pm 6$ V to $5.6 \times 10^3$ for $\pm 20$ V, showing very wide tunable range of the resistive states. Fig. 3(d) and (e) shows the gate-tunable output and transfer characteristics of the In$_2$S$_3$ memtransistors, respectively. The current $I_{ds}$ can be effectively adjusted by the gate voltage under the entire forward and reverse scanned $V_{ds}$. It is apparent that the current amplitude increases with the increase of positive gate voltage, suggesting an n-type semiconductor behavior. The In$_2$S$_3$ device exhibits a calculated carrier mobility of 0.073 cm$^2$/V·s and an on/off ratio of $10^4$. The results above clearly suggest that the In$_2$S$_3$ device can function as memristor and FETs simultaneously. Therefore, it is reasonable to term our device as memtransistors while keeping the controversy in mind.

In spite of this, the notion of memristor has been extended to a more general one recently. Leon Chua, in his paper “If it’s pinched it’s a memristor” [32], extended the notion of memristor to all two-terminal resistive devices that show a hysteresis loop pinched at the origin. Serrano-Gotarredona et al. defined the memristor as a “two-terminal electronic device which is similar to a resistor, but whose resistance changes dynamically as the device is being used.” [33]. In most of the literature, the concept of “memristor” is widely accepted as passive electrical component whose output is dependent on a system-specific state variable. In this paper, we follow the extended notion of Chua and Serrano-Gotarredona and still term our device as memtransistors while keeping the controversy in mind.

The gate-tunable memristive properties of the memtransistors in ambient conditions were measured using continuously dual sweeping of $V_{ds}$ at $\pm 15$ V under different gate bias. As shown in Fig. 4(a), the window of bipolar resistance at $V_{ds}$ become larger when increases the gate voltage from $-40$ to $40$ V, resulting in a wide gate-tunable range of resistive states. As can be seen in Fig. 4(b), the switching ratio changes from 0.4 at $-40$ V to 523 at $40$ V. To investigate
the endurance of the resistance states in ambient conditions, a typical device of memtransistors with smaller switching ratio was under continuously dual sweeping of $V_{ds}$ at ±15 V for 200 cycles. The results in Fig. 4(c) show repeatable characteristic and robust resistance states in $I_{ds}$ curve during cycling. The resistance of LRS in Fig. 4(d) increases slightly after 80 cycles. However, the change of switching ratio (inset of Fig. 4(d)) is still within one order of magnitude after 200 cycles, suggesting good endurance of the memtransistors. Retention characteristics of HRS and LRS values were measured for the same device with a reading bias of 5 V to demonstrate the electrical reliability of devices (Fig. 4(e)). Despite the apparent fluctuation, persistent HRS and LRS with switching ratio larger than $10^2$ (inset of Fig. 4(e)) was maintained even after $10^4$ s. Switching ratio does not decrease from its initial value but even increases to almost $10^3$ as the measure time goes on. All in all, the performance of our device is competitive in terms of high resistive switching ratio, good electrical stability, and long endurance.

The presented hysteresis properties of our device are similar to those of GB-mediated memristor using single-layer MoS$_2$ [19]. The nonlayered In$_2$S$_3$ is well known for its rich energy states and surface states introduced by the high-density vacancy sites [34], [35]. For 2D In$_2$S$_3$ nanosheet, the surface is abundant with dangling bonds that mostly formed by vacancies. These vacancies in β-In$_2$S$_3$ include tetrahedrally coordinated S vacancies, octahedrally coordinated In vacancies, In interstitial on the position of a tetrahedral structural vacancy, and In vacancies in both octahedral and tetrahedral positions, which have been theoretically predicted and experimentally investigated in previous studies [24], [34], [35]. The results of gate-tunable properties of the memtransistors (presented and discussed later) suggest the migration of positive charge defect on the surface of In$_2$S$_3$ 2DTFs. As S vacancies act as donor, the large change of resistance in In$_2$S$_3$ 2DTFs may originate from the GB-mediated migration of S vacancies. The external electric field applied between source and drain provides a large driving force for the transport of positive charge S vacancies, while the GBs provide physical interfaces that render potential barrier to regulate the migration of S vacancies. The accumulation and depletion of S vacancies on the edge of GBs is tuned by $V_{ds}$ (or $V_g$) and the potential height of GBs, thus changing the resistance of the area between GBs and electrodes.

**C. OPTICAL MODULATION ON RESISTANCE STATES**

Previous studies of β-In$_2$S$_3$ single crystal have reported a bandgap about 1.94 eV and a number of vacancy-related energy states near the band edges [24], [34], resulting in ultrasensitive optical response to light from visible and near-infrared [23], [25]. In this regard, we conducted a test of resistance states in response to the illumination of laser light in 405, 532, 635 and 808 nm. As exhibited in Fig. 5(a), the current of LRS and HRS all increases remarkably after the illumination of 635 nm light at power of 1.2 μW/mm$^2$. However, the current of HRS increases much larger than that of LRS (Fig. 5(b)), resulting in a change of switching ratio of the bipolar resistance before and after the illumination. It is found that the gradual increase of laser power can induce monotonous decrease of the switching ratio, as shown in Fig. 5(c). In the case of 635 nm laser, power from 0.2 to 7.4 μW/mm$^2$ induces seven switching levels. Notes that if
the light power is tuned more finely, more intermediate resistance state and switching levels can be obtained. This result indicates that optical stimuli can be employed as the extra terminal of the In$_2$S$_3$ memtransistor devices to ensure a variation margin of multiple storage levels.

Illuminations of laser light of other wavelengths were also used to examine the optical modulation on resistance states. As the optical energy of 808 nm is smaller than the bandgap of $\beta$-In$_2$S$_3$, the response to the near-infrared light originates from defects transition below the bandgap, which exhibit inferior photoresponse compared to visible light [23]. Therefore, as shown for the case of 808 nm in Fig. 6(a), light power up to 955 $\mu$W/mm$^2$ is used to induce resistance change, but results in fewer intermediate states and steps of switching ratio (Fig. 6(b)) compared to the case of 635 nm. On the other hand, light of 532 and 405 nm have higher photoresponsivity than 635 nm due to their larger optical energy [23], [25]. As a result, illuminations of these two wavelengths give rise to dramatically increased photocurrent (Fig. 6(d)) that in turn greatly suppress the switching ratio (Fig. 6(d)). Very low power of 1~2 $\mu$W/mm$^2$ already leads to switching ratio smaller than 10, making the fine optical modulation difficult. Thereby we can only obtain three and two storage levels in our experiment for 532 and 405 nm laser, respectively. The tunability from other wavelengths were not as effective as that from 635 nm, which might suggest that the choice of illuminating light with appropriate photoresponse is crucial in achieving optical-induced multilevel storage. For the In$_2$S$_3$ memtransistor in our work, 635 nm light is proved to be more suitable for the use of optical modulation in terms of the appropriate optical energy and photoresponsivity.

**D. TUNABLE ARTIFICIAL SYNAPTIC PLASTICITY**

In a biological synapse system (Fig. 7(a)), the release of neurotransmitter is caused by the arrival of action potentials fired by a presynaptic neuron, and then a signal is transmitted as a synaptic potential [7]. The memtransistors of In$_2$S$_3$ 2DTFs is used to emulate the synaptic plasticity of the biological synapses. As shown in Fig. 7(b), voltage pulse of $V_{ds}$ or $V_g$ is applied as presynaptic input while the current of $I_{ds}$ is recorded as postsynaptic current.

We at first demonstrate the tunability of postsynaptic current by changing the pulse $V_{ds}$. The time-dependent plasticity is found to be highly dependent on the width (W) and amplitude (A) of the input $V_{ds}$ pulses, as demonstrated in Fig. 7(c) and 7(d). When using electrical $V_{ds}$ pulses with a pulse period of 50 ms and a positive amplitude of 6 V, a duty cycle of 0.1 (pulse width of 5 ms) is insufficient to induce any observable change in output postsynaptic current. Starting from a threshold duty cycle of 0.2, $I_{ds}$ exhibits small increases and thereafter enhancement of $I_{ds}$ ($\Delta I_{ds}$) becomes more significant with the increase of duty cycle,
corresponding to gradual reduction of device resistance. This is analogue to the transition from “off” states to potentiated states in biological synapses, exhibiting the LTP behavior. Also, the device has an amplitude threshold for input $V_{ds}$ pulses to induce LTP. Time-dependent postsynaptic current exhibits almost no change if pulse amplitude is 1 V, but obvious enhancement when amplitude becomes larger than 2 V. Also, larger pulse amplitude results in a faster potentiation. On the other hand, LTD behavior can be mimicked using negative $V_{ds}$ pulses, which also exhibits effective tunability. As demonstrated in Fig. 7 (e) and (f). Using pulse period of 50 ms, duty cycle as small as 0.004 (200 $\mu$s) can induce a decrease of the postsynaptic current and faster depression can be achieved when increasing the duty cycle. The negative $V_{ds}$ pulses also have threshold amplitude of 1 V to induce the obvious decreasing of the postsynaptic current.

Fig. 7 (g) gives the response of postsynaptic current to a consecutive pulse sequence with varying pulse amplitude. The postsynaptic current continues to increase with positive $V_{ds}$ pulses, and increases more rapidly when larger pulse amplitude is applied. It is important to persist the change of the resistive state for a long period of time, so that we evaluate this property by measuring the conductance in response to the change of positive $V_{ds}$ pulses, as seen in Fig. 7(h). It can be seen that the conductance increases continuously under the stimulation of $V_{ds}$ pulse. After the pulse is removed, the conductance decays to about 70% of the maximum value and become stable thereafter. It clearly indicates that permanent transition to higher-conduction states is observed with repeated application of stimuli pulses. The above device performances successfully mimick the memorization events in biological nervous system, where the persistent increase in the strength of synaptic connection is achieved through repeated stimulation with sufficient frequency by action potentials [1].

The gate terminal in memtransistors gives another degree of freedom for synaptic modulation. Gate voltage exhibits effective control on the states of synaptic plasticity. As demonstrated in the case of positive $V_{gs}$ pulse amplitude of 6 V in Fig. 8(a), the $V_{gs} = -20$ and $-40$ V give rise to no obvious change in the time-dependent $I_{ds}$, representing the “off” states of synaptic plasticity. However, the device can transit from potentiated states (LTP) at $V_{gs} = 0$ V to depressed states (LTD) at $V_{gs} = 40$ V. In the case of $V_{gs} = 20$ V, the $I_{ds}$ decreases rapidly and then increases as time goes on, representing a transitional state between potentiated state and depressed state. The gate-dependent synaptic behavior can be seen clearly from the change of absolute value of $I_{ds}$.
in Fig. 8(b), where $I_{ds}$ was studied by using two back-to-back paired $V_{ds}$ pulses. In the first paired pulse, the gate voltage $V_g$ is set to 40 V and changed to 0 V in the second paired pulse. For the positive $V_{ds}$ pulse, the $I_{ds}$ switches from decrease to increase when the $V_g$ changes from 40 V to 0 V, corresponding to the transition from LTD to LTP behavior. It is obvious that the potentiation or depression responses are much faster at $V_g = 40$ V than that at 0 V, implying that gate voltage may not only tune the synaptic behavior but also the synaptic weight. However, for the negative $V_{ds}$ pulse, the $I_{ds}$ slightly decreases for both $V_g$ of 40V and 0 V, exhibiting no notable transition from LTD to LTP. Therefore, we focus our investigation to the case of positive $V_{ds}$. To verify the influence of $V_g$ on the synaptic weight of the device, we use the pulse gate voltage $V_g$ as input to update the postsynaptic current $I_{ds}$. As expected, the variation of amplitude of pulse $V_g$ changes the postsynaptic current effectively, as demonstrated in Fig. 8(c). Amplitude variation from $-10$ V to 60 V results in $10^3$ current difference, which gives plenty of room for synaptic weight modulation. As shown in Fig. 8(d), multiple step levels of postsynaptic current are acquired when gradually increase the pulse amplitude of $V_g$. Overall, the memtransistors of In$_2$S$_3$ 2DTFs as artificial synapse can exhibit diverse functionality by proper setting of the gate voltage. Specifically, gate voltage can modulate the behavior of synaptic plasticity in a highly controllable manner.

The gate tunable properties of the In$_2$S$_3$ memtransistors may shed light on the explanation of the potentiated and depressed postsynaptic current. Since the $I_{ds}$ does not change much at large negative $V_g$ (e.g. $-40$ V) for various $V_{ds}$, the potentiation and depression of $I_{ds}$ should be related to migration of positive charge defects. In$_2$S$_3$ is an n-type semiconductor [23]–[25]. Thus, when applying negative gate voltage, majority carrier of electron drifts to the surface of 2DTFs and recombine with positive charge defects (S vacancies), making the migration of defects invalid in changing the local resistance. On the other hand, when applying positive gate voltages, electrons are drained away from the surface of 2DTFs, where positive charge defects are able to accumulate through the potential barrier constructed by GBs. In such way, gate bias is able to tune the concentration of the effective defects and in turn change the channel resistance in a controllable manner.

**IV. CONCLUSIONS**

Based on the two-dimensional continuous thin films of non-layered $\beta$-In$_2$S$_3$ with microscopically visual superficial GBs, we have realized the memtransistors of In$_2$S$_3$ 2DTFs with resistance ratio up to $10^5$ and retention time over $10^4$ s. Laser light from visible and near-infrared was introduced to modulate resistance states of memtransistors. 635 nm light, which have optical energy near the bandgap of $\beta$-In$_2$S$_3$, is effective in inducing many intermediate resistance states for multilevel storage. The LTP and LTD behavior of biological synaptic plasticity can be realized by using the memtransistors of In$_2$S$_3$ 2DTFs as artificial synapses. $I_{ds}$ as postsynaptic can be highly tuned by using pulse drain/gate voltage as input stimuli. The behavior of the artificial synapses, including depressed and potentiated states, can be switched with tunable synaptic weight by the changing of gate bias. The realization of memtransistors based on In$_2$S$_3$ 2DTFs and their diverse and controllable functionality reveal their great potential in neurocomputing applications.

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