Demonstration of Threshold Switching and Bipolar Resistive Switching in Ag/SnO$_x$/TiN Memory Device

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Abstract: In this work, we observed the duality of threshold switching and non-volatile memory switching of Ag/SnO$_x$/TiN memory devices by controlling the compliance current (CC) or pulse amplitude. The insulator thickness and chemical analysis of the device stack were confirmed by transmission electron microscope (TEM) images of the Ag/SnO$_x$/TiN stack and X-ray photoelectron spectroscopy (XPS) of the SnO$_x$ film. The threshold switching was achieved at low CC (50 µA), showing volatile resistive switching. Optimal CC (5 mA) for bipolar resistive switching conditions with a gradual transition was also found. An unstable low-resistance state (LRS) and negative-set behavior were observed at CCs of 1 mA and 30 mA, respectively. We also demonstrated the pulse operation for volatile switching, set, reset processes, and negative-set behaviors by controlling pulse amplitude and polarity. Finally, the potentiation and depression characteristics were mimicked by multiple pulses, and MNIST pattern recognition was calculated using a neural network, including the conductance update for a hardware-based neuromorphic system.

Keywords: memristor; threshold switching; resistive switching; metal oxides

1. Introduction

Resistive switching is behavior during which a dielectric abruptly or gradually changes its resistance state by the electric field [1–4]. When the state of the resistor is programmed and erased once by a pulse, the state can be maintained for a long time, and the state can be changed by programming and erasing several times [1–5]. The resistance change memory can be further subdivided by various physical effects as follows. In phase-change memory (PRAM) [6], phase-change materials such as GST are changed in a reversible manner by a metal heater. In magnetic memory (MRAM) [7], the resistance state is changed as the magnetization direction is changed by the electric field. In ferroelectricity memory (FeRAM) [8], the resistance state of the dielectric can be changed by electrical polarization under an electric field. In resistive memory (RRAM) [1–4], the resistance state can be tunable by the electric field or joule heating. Among resistance-based non-volatile memories, RRAM has advantages in terms of low power consumption, high-density integration, and CMOS compatibility [9–18]. Moreover, the resistive switching characteristics in RRAM can be varied depending on the various material systems [9–18]. Typically, in the case of the filamentary type, metal ions such as Ag or Cu [19–21], which are top electrodes, enter the dielectric to form a metal filament. In addition, oxygen vacancy formation in the dielectric can make a conducting filament by the oxygen movement under the electric field [22,23]. When metal oxides such as TaO$_x$ and HfO$_2$ are used for the resistive switching layer, very uniform and excellent endurance and retention properties have been reported [22,23]. However, when the current level is lowered to a microampere or less, the variation in the resistance value increases, and the retention characteristics deteriorate. However, the interface type uses all areas between the metal electrode and the dielectric...
for resistive switching. In this switching type, the operating current is greatly affected by the cell area [24,25]. It has a relatively good variation of resistance states, but it has disadvantages such as a long switching time and poor retention characteristics.

Recently, in non-volatile memory, the concept of in-memory processing was introduced, in which operations are performed in or near the memory rather than simply storing data in the memory by a process in which operation logic is integrated [26]. This suggests the possibility that RRAM can be used as artificial neural network hardware. RRAM can provide both long-term memory and short-term memory functions that are suitable for hardware-based neuromorphic systems [27–35]. Especially, oxide semiconductors such as In-Ga-Zn-O (IGZO) [36,37] and ZnO [38–40] provide gradual conductance modulation, whose property is favorable to implement the neuromorphic system as the synaptic device. The threshold switching that has volatile memory properties can be utilized by the neuron function in a neuromorphic function [41] and the memory switching with multi-levels is suitable for synaptic devices [42]. The threshold switching and memory switching are easily obtained by controlling the strength of the Ag filament when using Ag top electrodes in metal–insulator–metal (MIM) system memory [43].

In this work, we demonstrate the threshold resistive switching and non-volatile memory switching of an Ag/SnO$_x$/TiN device for short-term memory and long-term memory. The XPS analysis was conducted to investigate the chemical and material characteristics of SnO$_x$ film. In addition, the accurate thickness and amorphous solids of SnO$_x$ were detected by the TEM images. For electrical short-term and long-term memory characterization, DC sweep and pulse measurement was conducted. By adjusting CC, the optimal bipolar resistive switching condition was searched and threshold switching was confirmed at low CC (50 µA). Furthermore, the set process, reset process, and threshold switching were verified by pulse transient sequence. Finally, MLC operation was achieved by identical pulse conditions during the set and reset processes. The dual resistive functions from one AOS device are rarely realized, which will give more degrees of freedom to the resistive switching operation for the selector and neuromorphic device applications.

2. Materials and Methods

Ag/SnO$_x$/TiN devices were prepared through the following process. A 100-nm-thick TiN bottom electrode was deposited on a SiO$_2$/Si wafer by reactive sputtering. Ar (20 sccm) and N$_2$ (3 sccm) were used at pressure of 1 mTorr and power of 500 W for TiN film. SnO$_x$ film with a thickness of about 30 nm was deposited by DC magnetron sputtering in which an Sn target was used with Ar (10 sccm), O$_2$ (9 sccm) gases at DC power (95.5 W). A 100-nm-thick Ag top electrode was deposited by an e-beam evaporation system via a shadow mask containing a circular pattern with a diameter of 100 µm. A Keithley 4200-SCS semiconductor parameter analyzer (SPA) and a 4225-PMU pulse measurement unit in the probe station were used to measure electrical characteristics using DC sweep mode and transient characteristics. While a bias was applied to Ag, the top electrode and TiN bottom electrode were grounded.

3. Results and Discussion

Figure 1a shows the TEM image of Ag/SnO$_x$/TiN memory devices. The thickness of SnO$_x$ film is about 30 nm. Figure 1b,c show the XPS spectra of Sn 3d and O 1s from SnO$_x$ film. Here, since the SnO$_x$ film thickness is thicker than the detection range of one point of XPS, the information for pure SnO$_x$ film can be purely observed. The binding energy of the peak of Sn3d$_{5/2}$ is centered at 486.73 eV, which means the sputtered deposited SnO$_x$ film is non-stoichiometric (Figure 1b) [44]. The peak binding energy of O1s is located at 530.58 eV (Figure 1c).
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Figure 1. (a) TEM image of Ag/SnO$_2$/TiN stack. XPS spectra of (b) Sn 3d and (c) O 1s.

Next, we investigate the I-V characteristics with different CC. Figure 2a shows the threshold switching with low CC (50 μA). In the forward sweep, the current increases rapidly from HRS to LRS, but the current seems to be maintained during the backward sweep; however, in the subsequent forward sweep, it again follows HRS with a lower current level. This kind of switching is attributed to a weak filament in the dielectric that is formed during the set process. Next, we investigate the memory switching after forming process. Figure 2b shows the bipolar resistive switching with a CC of 1 mA. The LRS has a large fluctuation. This is mainly due to the current increment before the reset process in a negative bias. This indicates that a sufficiently high current is required for the reset process. Figure 2c shows the bipolar resistive switching after the forming process with a CC of 5 mA. Set process and reset process occur in a positive bias and negative bias, respectively. Note that the current is gradually increased and decreased. This property can be helpful to realize multilevel cells (MLCs). Moreover, the LRS current is so high and inevitable to negative-set behavior at a higher CC (30 mA) in Figure 2d. The negative-set behavior usually occurs because of further breakdown during the reset process. Here, one-time programming behavior can be used for write once read many (WORM) applications [45].

Figure 2. I-V characteristics of Ag/SnO$_2$/TiN device with different compliance current conditions. (a) 50 μA, (b) 1 mA, (c) 5 mA, (d) 30 mA.
Next, we verified the pulse’s volatile switching, set, reset, and negative-set processes for practical switching operation. Figure 3a shows the volatile process in which the read pulse, set pulse, and read pulse are successively applied on the device. The read voltage (0.3 V) is set low in order not to affect the state significantly. A pulse of voltage with a magnitude of 0.8 V is insufficient to proceed with the set process, and when the main pulse is turned off, it is observed that there is no significant difference in the current before and after the main pulse. However, when a set voltage of 2 V is applied, the current increases in Figure 3b. Moreover, it was confirmed that the current increased after the set pulse was applied by 0.2 V read voltage. Figure 3c shows the reset process by pulse. The current is decreased after a reset pulse of −1 V. However, when a negative pulse with a larger amplitude is applied, the current is rather increased in Figure 3d. This can be explained by the negative-set phenomenon as confirmed by DC sweep.

We identified the long-term potentiation (LTP) and depression (LTD) of the Ag/SnO$_2$/TiN device. This device could store the multi-level states, so it is suitable for the neuromorphic system. To demonstrate the adequacy of the synaptic device, a potentiation pulse of 0.9 V and depression pulse of −0.7 V was applied with reference switching in the I-V curves, which are for the set process at a positive voltage and reset process at a negative voltage. In order to see the changing conductivity with the pulse, the current was extracted by applying the read pulse of 0.1 V at each pulse. At this point, the pulse width was equally set up for 5 ms to avoid being affected by time.

As shown in Figure 4a, the conductance values are controllable with the pulse, and the regular potentiation residual is less than ±0.0001, and the depression regular residual is less than ±0.00015 in the logistic fitting. To determine whether it is appropriate for the neuromorphic system, the Modified National Institute of Standards and Technology (MNIST) was used to analyze the LTP and LTD values as synaptic weights to recognize the handwriting of different people. In Figure 4c, in the input layer, two-dimensional images of 60,000 training samples that have 28 × 28 pixels respectively are transformed to one-dimensional 784 vectors that correspond to input neurons and transmit to 128
neuron nodes (h1 ... h256) in the hidden layer. The hidden layer calculates the weights and the strength of the synapse between the input and output layer based on the conductance values and logistic fitting values measured. The output layer prints the 10 numbers (from 0 to 9) from these weights via the 10,000 test images. Each synaptic device consists of an excitatory device and an inhibitory device that correspond to the excitatory weight $G^+$ and inhibitory weight $G^-$, respectively. The weight of the device is defined as $W_{ij} = G^+ - G^-$. The inference scheme of this neural network system depends on the voltage. A binary MNIST image (white pixel) is a 1 V pulse voltage, and the background (black pixel) is a 0 V pulse voltage, and the crossbar array through WL \(_i\) is used. This configuration exhibits up to 80.7% accuracy in Figure 4b.

![Figure 4.](image)

**Figure 4.** (a) Conductance with the potentiation and depression pulse of the Ag/SnO/TiN device. (b) Recognition accuracy of the MNIST. (c) Schematic of three-layer neural network for the neuromorphic system.

4. Conclusions

In summary, we have characterized the threshold switching and bipolar resistive switching with non-volatile memory properties of a Ag/SnO\(_x\)/TiN memory device. Firstly, volatile switching and memory switching were controlled by CC in DC sweep mode. The volatile switching was observed due to the weak conducting filament formation at low CC (50 \(\mu\)A). However, stable and gradual resistive switching was achieved at 5 mA without negative-set behavior and unstable LRS. Next, the transient characteristics of threshold switching, set and reset process, and negative-set behaviors were conducted by pulse. Finally, biological potentiation and depression were emulated by the repetition of identical pulses and MNIST pattern recognition was evaluated in a neural network.
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