Coexistence of Digital and Analog Resistive Switching Behaviours in Ag/CuAlO$_2$/TiO$_2$/p$^{++}$-Si Memristor

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Abstract. The digital resistive switching is suitable for the applications of information storage and logical operation, while the analog resistive switching is required in the neuromorphic computing system. This paper reports the stressed bias voltage-dependent digital and analog resistive switching behaviours coexisted in Ag/CuAlO$_2$/TiO$_2$/p$^{++}$-Si memristor devices. At high biased voltage, the device has demonstrated bipolar resistance switching functions with a resistance ratio over $10^7$ and reliable durability. Moreover, in the low voltage sweeping region, the device showed potentiation and depression characteristics. It is suggested that the bipolar resistive switching may be due to the local migration of Ag and oxygen ions within the dielectric layers. This new memory structure with digital and analog resistive switching is expected to reduce to decrease the manufacturing complexity of the electronic circuit containing digital/analog memristors.

1. Introduction

Resistance switching (RS) has been demonstrated in two-terminal devices (known as memristors) for various applications, such as nonvolatile memory (NVM), and neuromorphic computing [1][2]. The manifestation of resistive switching behaviour is closely related to the modulation of internal states which is influenced by an externally applied stimulus. In general, applications of memristor-based on two distinguishable resistance states known as digital type and analog type of switching [3]. An abrupt change in resistance in digital circuits, which is characterized by a high on/off ratio, can cause two distinguishable states, indicating the possibility of nonvolatile data storage and logic operations. Digital type of resistance change mostly has been found in filamentary memristors [4]. The switching mechanism of the memristor can be ascribed to the growth and disruption of conductive filaments (CFs) between the electrodes. In filamentary memristor, the formation of conductive filaments (CFs) is attributed by forming a conduction path between electrode by the migration of oxygen or active metal ions (Ag$^+$, Cu$^{2+}$, Ni$^{2+}$, Al$^{3+}$, In$^{3+}$, etc.). In such kind of memristors, SET and RESET can be achieved by growth and disruption of conductive filament.

In general, an Off/On conductivity ratio greater than 10 is sufficient to store data with small read errors. Numerous studies show that the higher on/off ratio (greater than $10^3$) offers a wide memory window that enables the real applications of memristor devices [5]. Unlike digital switching, the gradual change (analog type) in resistance is controlled by the charge or electric flux through the memristor. The analog type of change in resistance is similar to a gradual change in resistance state in biological synapses, which is responsible for learning behaviour and memory in the brain [6]. Unlike
to abrupt change in resistance, the gradual change has been found in filamentary memristors [7]. Both digital and analog switching behaviours are not found in a single memristor. However, if an abrupt and gradual change in resistance is realized in a memristive structure reduces the complex circuitry. It can be seen that in most of the memristor device, by controlling experimental conditions, digital and analog switching behaviours can be realized, including Pt/BiFeO$_3$/Pt [8] and Ag/SiO$_x$:Ag/TiO$_x$/p$^{++}$-Si [9].

This article demonstrates the relationship between digital and analog behaviour as in the memristor of Ag/CuAlO$_2$/TiO$_2$/p$^{++}$-Si structure. Compared to the structure of Ag/CuAlO$_2$/p$^{++}$-Si, the gradual resistance changes in the Ag/CuAlO$_2$/TiO$_2$/p$^{++}$-Si memristive structure. Here, owing to a higher dielectric constant (~90 with less bandgap ~3 eV) of TiO$_2$ thin film leading to the distribution of the applied electric field locally and more applied voltage could be concentrated on CuAlO$_2$ layer. It can prevent excessive injection of metal ions and the growth of filaments through the memristive layer, which will improve performance (e.g. increase uniformity and reliability).

2. Experimental Detail
A thin layer of CuAlO$_2$ (200 nm) and then a TiO$_2$ (10 nm) are deposited on p$^{++}$-Si substrates, simultaneously using CuAlO$_2$ and TiO$_2$ and Ar sputtering gas (0.5 Pa working pressure). Using DC sputtering and lithography process, an Ag thin layer (300 nm) was then deposited as top electrode (TE) to fabricate the Ag/CuAlO$_2$/TiO$_2$/p$^{++}$-Si devices. The entire deposition process took place at room temperature. The current-voltage (I-V) characteristics are measured using a power source and semiconductor analyzer Keithley 2636B.

Figure 1. Schematic structure of Ag/CuAlO$_2$/TiO$_2$/p$^{++}$-Si memristor.

3. Results and Discussion
The schematic structure of the Ag/CuAlO$_2$/TiO$_2$/p$^{++}$-Si memristor is shown in figure 1. Figure 2(a) shows the I-V curve when DC-bias sweeps from 0 V to 6.0 V and from 0 V → -3.0 V are applied with a compliance current (I$_{cc}$) of 1.0 mA is applied to avoid the current overshoot. The I-V curve reveals a similar hysteresis phenomenon similar to the first found memristor [10]. When TE is positively biased, the memristor achieves a high resistance state (HRS) to a low resistance state (LRS), called "SET" state. In contrast, when TE is negatively biased, the device achieves its original state (i.e., HRS) called "RESET" state. In order to test the endurance performance, the device undergoes to $10^3$ times, as shown in figure 2(b), demonstrating uniform switching behaviour. Here the abrupt changes in resistance during SET and RESET operations indicate a typical digital switching.
Figure 2. (a) Bipolar switching behaviour of Ag/CuAlO$_2$/TiO$_2$/p$^{++}$-Si structure and (b) endurance-cycling performance test for $10^5$ cycles; (c) The current response and retention characteristics obtained at pulse mode operation.

Moreover, for a potential nonvolatile memory application, ‘WRITE/READ/ERASE’ by SET and RESET at pulse mode operation needs to be demonstrated. The resistance states are measured after each SET and RESET stimulus of +5 V (for SET) or -3.5 V (for RESET) alternately. Figure 2(c) shows the current response subject to SET and RESET pulse voltages, and the memristor is switched between Low and high states, alternately. Figure 2(d) indicates that the LRS and HRS are stable over $10^4$ times of SET/RESET under a pulse-type bias, representing an excellent uniformity of the switching behaviour.

In order to understand the switching mechanisms involved during the set and the reset processes, the electrons transport behaviour is taken account. The positive and negative region of the I-V curve in figure 2(a), are plotted in figure 3(a) and figure 3(b) on a logarithmic scale. From the fitting I-V curve in linear scale, in LRS and HRS the charge transport behaviour can be explained by space charge limited conduction (SCLC) mechanism [11]. The linearly fitted I-V curve consists of three regions, such as the Ohmic (I/V), the Child’s law (I/V$^2$) and the sharp. It can be seen at low-voltage ~ 0.35 V. The curve shows linear behaviour indicating the Ohmic conductance resulting from the thermally generated free electrons. Whereas, when voltage is increased further from 0.35 V to 6 V, a sharp increase in current is observed with slope ~ 2.8 V indicating the SLCL mechanism. When further high voltage bias is applied, the current increases abruptly, which might be explained by electrochemical metallization of Ag atoms and formation of Ag-CFs across the electrodes [12]. Unlike HRS, at LRS the linearly fitted curve shows Ohmic (I/V) behaviour with a slope of 1.3 indicating the formation Ag-CFs at LRS.
Figure 3. I-V curve of the Ag/CuAlO$_2$/TiO$_2$/p$^{++}$-Si structure plotted with corresponding linearly fitted curves at (a) positive and (b) portions; (c) A schematic diagram of a physical model is drawn as in pristine device, (d) LRS state and (e) HRS state of the memristor.

As compared to the single-layer CuAlO$_2$-based device, in Ag/CuAlO$_2$/TiO$_2$/p$^{++}$-Si structure, where the CuAlO$_2$ thin film possesses higher bandgap (~9.0 eV) than TiO$_2$ (~3 eV) and a higher dielectric constant (~90). It results in uniform distribution of electric field across the dielectric layer between the electrode. Figure 3(c) shows that electric field drives the Ag ions interfacial layer from the Ag TE to p$^{++}$-Si BE, and Ag$^+$ ions reduced in Ag atoms near the surface of BE resulting in the formation of Ag-CFs from TE to BE. This Ag-CFs are tending to maintain their original shape till TE is biased negatively. Upon negatively biased voltage, Ag-CFs are partly broken due to the joule-heating effect [13], and the device achieves its original state.

Very surprisingly, under a consecutive voltage sweep of 0 V → +3.0 V → 0 V and 0 V → -3.0 V → 0 V respectively, the current increases or decreases apparently, which can be seen in figure 4(a) and figure 4(b). This kind of resistance change is analogous to an analog switching type, which differs from a digital switching one that shows an steep increase in current achieving SET and RESET states under 0 → 6.0 V → -3.0 V → 0 V. As discussed above, when Ag filaments are formed, a digital type of switching could be realized while the top and bottom electrodes are entirely bridged. It is noted that in the case of repeated bias voltage sweeping, +3.0 V is not sufficient to create a conducting Ag filament bridge across the electrodes. Notably, it is observed the during biased I-V following the where the former one is left, indicating the analog type of memristive behaviour in our device. When the TE is biased negatively, the I-V curve exhibits SLCL behaviour at HRS and Ohmic behaviour at LRS similar to the I-V curve at positive region. During the negative bias, the conductance of the device is gradually decreases which might be due to the increase in the effective gap between electrodes and the lateral size of CFs decreases as well. Figure 4(a) and 4(b) show that when the repeated negative bias is applied, the gradual decrease in current flow. These phenomena signify analog switching behaviour.
Figure 4. Analog switching characteristics of Ag/CuAlO$_2$/TiO$_2$/p$^{++}$-Si Memristor. (a) Gradual SET and (b) RESET process using consecutive positive and negative sweep; (c) Gradual SET and RESET processes of the identical repeated pulse.

The same behavior is observed when using a repeated pulse. As shown in figure 4(c), when a $+2.5$ V identical pulse is applied successively, the current read at $+0.2$ V rises slowly, which is called potentiation [14]. While using a repeated negative pulse of $-2.5$ V decreases the current flow, which is called depression. Such incremental increase and decrease are called potentiation and depression, respectively, which are beneficial in artificial synapse applications.

4. Conclusion
In summary, we fabricated the Ag/CuAlO$_2$/TiO$_2$/p$^{++}$-Si memristor by RF sputtering method. Bipolar digital switching is observed after the electrical-forming operation. This can be explained on the basis of the formation and rupture of the leading pathway by formation and rupture of Ag filament. Prior to electroforming, the device has an analog switching that can be realized by local migration of Ag ions and oxygen under the electric field. The digital and analog switching behavior in a single device of Ag/CuAlO$_2$/TiO$_2$/p$^{++}$-Si structure will provide a new way to design a system of consciousness and memory in the future.

5. Acknowledgements
This work was supported by the National Natural Science Foundation of China (Grant No. 61421002).

6. References
[1] Zhu X, Lee S H and Lu W D 2019 Nanionic Resistive-Switching Devices Adv. Electron. Mater. 5
[2] Zhu J, Zhang T, Yang Y and Huang R 2020 A comprehensive review on emerging artificial neuromorphic devices Appl. Phys. Rev. 7
[3] Lv F cheng, Yang R and Guo X 2017 Analog and digital Reset processes observed in Pt/CuO/Pt memristive devices Solid State Ionics 303 161–6
[4] Abbas Y, Jeon Y R, Sokolov A S, Kim S, Ku B and Choi C 2018 Compliance-Free, Digital SET and Analog RESET Synaptic Characteristics of Sub-Tantalum Oxide Based Neuromorphic Device Sci. Rep. 8
[5] Wang Y, Liu Q, Long S, Wang W, Wang Q, Zhang M, Zhang S, Li Y, Zuo Q, Yang J and Liu M 2010 Investigation of resistive switching in Cu-doped HfO$_2$ thin film for multilevel nonvolatile memory applications Nanotechnology 21 045202
[6] Wu W, Wu H, Gao B, Deng N, Yu S and Qian H 2017 Improving Analog Switching in HfOx-Based Resistive Memory with a Thermal Enhanced Layer IEEE Electron Device Lett. 38 1019–22
[7] Yoo E, Lyu M, Yun J-H, Kang C, Choi Y and Wang L 2016 Bifunctional resistive switching behavior in an organolead halide perovskite based Ag/CH$_3$NH$_3$PbI$_3$Cl$_2$/FTO structure J. Mater. Chem. C 4 7824–30
[8] Shi T, Yang R and Guo X 2016 Coexistence of analog and digital resistive switching in
BiFeO3-based memristive devices *Solid State Ionics* **296** 114–9

[9] Ilyas N, Li D, Li C, Jiang X, Jiang Y and Li W 2020 Analog switching and artificial synaptic behavior of Ag/SiO$_x$:Ag/TiO$_x$/p$^+$.Si memristor device *Nanoscale Res. Lett.* **15** 30

[10] Strukov D B, Snider G S, Stewart D R and Williams R S 2008 The missing memristor found *Nature* **453** 80–3

[11] Yang Y C, Pan F, Liu Q, Liu M and Zeng F 2009 Fully Room-Temperature-Fabricated Nonvolatile Resistive Memory for Ultrafast and High-Density Memory Application *Nano Lett.* **9** 1636–43

[12] Pan R, Li J, Zhuge F, Zhu L, Liang L, Zhang H, Gao J, Cao H, Fu B and Li K 2016 Synaptic devices based on purely electronic memristors *Appl. Phys. Lett.* **108** 013504

[13] Liu Q, Sun J, Lv H, Long S, Yin K, Wan N, Li Y, Sun L and Liu M 2012 Real-time observation on dynamic growth/dissolution of conductive filaments in oxide-electrolyte-based ReRAM *Adv. Mater.* **24** 1844–9

[14] Tong G, Malenka R C and Nicoll R A 1996 Long-term potentiation in cultures of single hippocampal granule cells: A presynaptic form of plasticity *Neuron* **16** 1147–57