Anatomy of high-uniform unidirectional volatile switching behavior in SiO2/TiO2-based selection device

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ABSTRACT

Unidirectional volatile switching behavior is a typical resistive switching phenomenon in oxides, which has been proposed as attractive candidates for selection devices for high density memory and neuromorphic systems applications with cross-point arrays. However, the microscopic origin of the underlying mechanism is still unclear due to the lack of direct experimental evidence. Here, a high performance selection device with a low forming voltage, low OFF-state current, high nonlinearity, and excellent uniformity is proposed in SiO2/TiO2 stacked structure. The underlying nature of the unidirectional volatile switching behavior in the Pt/Ag/SiO2/TiO2/Ti selection device is revealed by using scanning electron microscope and energy-dispersive X-ray spectroscopy analysis in planar structure devices, which can be attributed to the unstable conductive filament (CF) consisting of Ag nanoparticles formed in the SiO2 layer, and the Schottky contact formed between the Ag CF and TiO2 interface. This work provides clearly experimental evidence to deepen understanding of the mechanism for unidirectional volatile switching behavior, which can provide a guide to further improve the device performances for high density memory and neuromorphic systems applications.

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1. Introduction

Resistive random access memory (RRAM) devices, based on resistive switching characteristics between a high resistance state (HRS) and a low resistance state (LRS) driven by voltage or current bias, have been intensively studied and considered to be promising candidates for both neuromorphic computing [1–6] and next-generation nonvolatile memory applications [7–11] due to their excellent scalability, low power consumption, low cost, high density 3D integration, excellent compatibility with the current complementary metal oxide semiconductor (CMOS) technology [1,7–13], and a close resemblance to the operating characteristics of bio-synapse [1,2,6,15]. These applications typically require a large cross-point array of RRAM devices [16–18]. However, the integration of cross-point array consisting of only RRAM cell suffers unavoidable cross-talk interference due to sneak path currents through neighboring unselected cells, which severely impede the proper operation of the array [17–22]. To solve the critical issue in cross-point array, numerous access devices with nonlinear response are usually connected to each RRAM cell of the array [23–27].

Recently, two-terminal thin-film-based volatile RRAM devices which feature high nonlinearity, fast switching speed, high endurance, short relaxation time, as well as 3D stacking capability [17,22,27–31] have been used as selector devices to suppress crosstalk effect in cross-point arrays [17,22,27,28]. On the other hand, they have also been proposed as attractive candidates in bio-inspired neuromorphic systems as short-term synapses [6,14,29] and integrate-and-fire neurons [30,31]. Although various types of volatile RRAM devices have been developed [27–38], the intrinsic mechanism of the volatile switching behavior is still unclear. Indeed, the intrinsic mechanism is particularly critical for optimizing device structure and improving device performance. Therefore, the revelation of the intrinsic mechanism of volatile switching behavior will provide important guidelines and shed new insights into the resistive switching study of volatile RRAM.
devices.

Generally, volatile resistive switching behaviors can be classified as two modes: bidirectional volatile switching (BS) [17,22,27,28] with symmetric switching characteristics and unidirectional volatile switching (US) [32] with asymmetric switching characteristics. In the BS mode, the volatile switching behavior has been attributed to the spontaneous rupture of the thin and unstable conductive filament (CF) within the switching layer as the external electrical bias is removed [17,32,33]. In the US mode, although thermally activated drift/diffusion switching models based on Monte Carlo simulations of Ag nanoparticle Brownian-like motion [34], Gibbs–Thomson effect, and interfacial energy minimization [17] have been proposed to quantitatively interpret the asymmetric unidirectional switching phenomenon, a detailed experimental study of the underlying mechanism for this phenomenon is still lacking, especially lacking of direct evidences to uncover why asymmetric unidirectional switching characteristics happen in the US mode.

In this work, we report an asymmetric unidirectional volatile switching selection device in SiO2/TiO2 bilayer stacked structure. The fabricated two-terminal Pt/Ag/SiO2/TiO2/Ti volatile selection device shows a low forming voltage (~1.5 V), high selectivity (~106), low OFF-state current (~10 pA), and uniform resistive switching. Using scanning electron microscope (SEM) and energy-dispersive X-ray spectroscopy (EDS) analysis, we provide unambiguous evidence to directly reveal the microscopic origin of the unidirectional volatile switching behavior. Obtaining these pieces of information not only help elucidate the underlying nature of the unidirectional volatile switching phenomenon, but also guide the design of volatile RRAM devices for cross-point memory arrays and neuromorphic systems applications.

2. Experimental

The unidirectional volatile selection device with Pt/Ag/SiO2/TiO2/Ti vertical structure was directly fabricated on SiO2/Si substrate. A 65-nm-thick Ti film was first deposited on the SiO2/Si substrate by electron beam evaporation to form the bottom electrode. Subsequently, a 40-nm-thick TiO2 film was deposited on the Ti bottom electrode by an atomic layer deposition (ALD) system at 250 °C substrate temperature. TiCl4 was used as the Ti precursor and H2O was used as the oxygen precursor. An atomic layer deposition cycle with 4 steps was adopted for this process. The 4 steps were TiCl4 reactant, N2 purge, H2O reactant, and N2 purge, respectively. Then, a 20-nm-thick SiO2 film was deposited on the TiO2 film by sputtering. Finally, 10/35 nm Pt/Ag top electrode (TE) with areas of 100 × 100 μm2 was deposited by electron beam evaporation followed by a lift-off photolithographic process. The thin Pt film acts as the part of the protective layer to avoid the oxidation of the Ag TE, and to prevent the probe tip from scratching the device surface during testing. For the Ag/SiO2/Ti and Ag/TiO2/Ti planar structure devices, 40-nm-thick SiO2 and TiO2 films were prepared on the SiO2/Si substrate by sputtering and ALD, respectively. After first photolithography, 80-nm-thick Ti layers were deposited on the SiO2 and TiO2 films by electron beam evaporation. Then, rectangle Ti electrode with area of 100 μm × 5 mm was released using a lift-off process. Subsequently, Ag electrode patterns composed of a 100 μm × 100 μm square and a 20 μm × 5 μm rectangle tip were generated by photolithography. Subsequently, 80-nm-thick Ag layers were deposited by electron beam evaporation and released by lift-off process. The gap between Ag and Ti electrodes ranges from 200 nm to 2 μm.

The electrical characteristics of all the fabricated devices were performed by using an Agilent B1500A semiconductor device analyzer at room temperature. During the measurements, the bias voltage was applied on the Ag electrode while the Ti electrode was grounded. TEM measurements were performed with a 200 kV high-resolution TEM (HR-TEM, JEOL JEM2100F). The TEM specimens were prepared using a dual-beam focused ion beam (FIB) system (Seiko 4050MS). The elemental distribution was determined by the EDS mapping. The morphology of CF in the device was detected by using a field-emission scanning electron microscopic (Apreo S).

3. Results and discussion

3.1. Structural and electrical properties

The schematic of the device structure is shown in Fig. 1a, and a cross-sectional bright-field transmission electron microscopy (TEM) image and relevant EDS mapping images of our fabricated device with Pt/Ag/SiO2/TiO2/Ti stacked structure on SiO2/Si substrate are shown in Fig. 1b and c, respectively. It is clear that the Pt/Ag/SiO2/TiO2/Ti multi-layer films are well stacked on SiO2/Si substrate with thicknesses of about 10/35/20/40/65 nm, respectively.

The as-deposited fresh devices are normally in the high resistive state (OFF-state), and their initial resistances are about 1013 Ω at a 0.3 V read voltage. To trigger repeatable resistive switching behavior in the fresh device, an electroforming process (switching the fresh device from the initial OFF-state to the low resistive state (ON-state)) is generally needed [33]. Because of the asymmetric structure of our devices, an electroforming process with positive voltage on the Ag TE is required, and a negative voltage applied to the TE could not lead to the electroforming process. Fig. 2a shows the typical current–voltage (I–V) curve of the Pt/Ag/SiO2/TiO2/Ti device with a high compliance current (Icomp) of 1 mA. When a Icomp of 1 mA is applied on the fresh device, a gradual increase of device current can be observed at a low voltage range, while a sudden increase of device current to the Icomp at a forming voltage of about 2.35 V, and the device switches to a low ON-state. Unfortunately, the ON-state obtained under the Icomp of 1 mA cannot be reset to a stable OFF-state again either in positive voltage or negative voltage, meaning that the device may be completely broken down due to the higher forming voltage when a higher Icomp is applied [39]. However, it is gratifying to note that repeatable volatile resistive switching characteristics can be obtained when lower Icomp are used. Fig. 2b shows the I–V curves of the Pt/Ag/SiO2/TiO2/Ti device with lower Icomp values varied from 0.1 μA to 100 μA. During the negative-voltage sweep, the current is extremely low, and no forming process can be observed, while forming clearly appears for a positive voltage. By sweeping the voltage from zero to a positive voltage, a gradual increase of device current can be observed. The device current reaches to Icomp at voltages about 1.5 V, and the device switches from the initial OFF-state to ON-state. When the voltage is swept from 2 V back to 0 V, the ON-state resistance of the device cannot be maintained and reverts spontaneously to the OFF-state as soon as the applied voltage is close to zero. These results reveal the unidirectional volatile switching behavior in the Pt/Ag/SiO2/TiO2/Ti device. More importantly, low leakage current in the pA range and nonlinearity as high as 3 × 106 at the read voltage (VREAD = 1.5 V) and half-readvoltage (1/2VREAD = 0.75 V) can be achieved by using the Icomp of 100 μA (Supporting Information Fig. S1).

In order to explore the origin of the volatile switching behavior in the Pt/Ag/SiO2/TiO2/Ti device, a kinetic Monte Carlo model was
Fig. 1. (a) Schematic of the unidirectional volatile selection device with two-terminal Pt/Ag/SiO$_2$/TiO$_2$/Ti stacked structure. (b) A cross-sectional bright-field TEM image of the unidirectional volatile selection device with Pt/Ag/SiO$_2$/TiO$_2$/Ti stacked structure on SiO$_2$/Si substrate. (c) EDS mapping images corresponding to the TEM image.

Fig. 2. (a) $I-V$ characteristic of the Pt/Ag/SiO$_2$/TiO$_2$/Ti device under a high compliance current of 1 mA. No stable resistive switching characteristic could be observed in the device with a higher $I_{\text{comp}}$ of 1 mA during the electroforming process. (b) Typical $I-V$ curves of the Pt/Ag/SiO$_2$/TiO$_2$/Ti device under different compliance current of 0.1 $\mu$A, 1 $\mu$A, 10 $\mu$A, and 100 $\mu$A. Unidirectional volatile switching behaviors were observed when different compliance currents from 0.1 $\mu$A to 100 $\mu$A were used during the electroforming process with positive voltage. (c) Simulated dynamic microstructure evolution of Ag CF in SiO$_2$/TiO$_2$ films.
constructed to investigate the microstructure evolution of metal Ag atoms/ions in SiO2/TiO2 films. To reflect stochastic nature in migration of Ag atoms/ions which induces instability of Ag conductive filament (CF), noise function, depicted by a Wiener process with parameters related to applied bias and electrical field, was added to model. The dynamics of the Ag ions are simulated by the following equation:

\[ \frac{d\mathbf{u}}{dt} = \nabla \cdot (\mathbf{v} n_{D} - D \nabla n_{D}) + \nabla \cdot D \nabla n_{D} dW_t \]

where \( n_{D}, \mathbf{v} \) are the concentration and displacement of the Ag ion, \( D \) and \( \mathbf{v} \) are diffusivity and drift velocity, respectively. The calculation of these parameters is the same as \([40,41]\). The equation is solved by Matlab. To obtain necessary temperature and electrical field profile, FreeFem ++ is utilised to solve the Fourier Equation.

Fig. 2c shows the simulated Ag CF microstructure evolution during the forming process in SiO2/TiO2 films. It is noted that there are some Ag atoms/ions within SiO2 film, but there are almost no Ag atoms/ions within TiO2 film when a voltage is supplied. This may be due to that a high Schottky barrier is formed between Ag and TiO2 interface \([42]\). The high Schottky barrier between Ag and TiO2 interface prevents Ag ions from moving forward into the TiO2 film. More importantly, it can be seen from Fig. 2c (1–3) that discontinuous CF channel consisting of discrete Ag nanoparticles with poor continuity is formed within the whole SiO2 layer when the voltage reaches 1.5V. However, when the voltage is swept back to 0V, the discontinuous CF channel is completely disrupted, as shown in Fig. 2c (4–6). Then, the device will be back to the OFF-state.

To further evaluate the uniformity of our fabricated selection device, we analyzed the statistical distributions of switching voltages and currents obtained by a direct current (DC) cycling test. Fig. 3 shows the cycle-to-cycle statistical distributions under the \( I_{\text{comp}} \) of 100 \( \mu \)A. As shown in Fig. 3a, the shape of the \( I-V \) curve is similar to that in the first sweep and only a little voltage and current variations are observed after \( 10^3 \) successive DC cycles. The statistical distributions of the current values at 1.5V and 0.75V during \( 10^3 \) successive switching cycles are shown in Fig. 3b. As shown in Fig. 3b, only small current variations and high nonlinearity of more than \( 10^6 \) are observed. Such a high selectivity could be used to suppress crosstalk effect in the cross-point array. The statistical data of the histograms for the turn-on voltage (\( V_{\text{ON}} \)) and turn-off voltage (\( V_{\text{OFF}} \)) are shown in Fig. 3c and d, along with the Gaussian fit of each histogram. Excellent uniformity with a negligible standard deviation is achieved with an ultralow \( \sigma \) value of 0.01 for \( V_{\text{ON}} \) and 0.02 for \( V_{\text{OFF}} \) (\( \sigma \) is aparmeter of the Gaussian distribution), indicating excellent uniformity is achieved in the Pt/Ag/SiO2/TiO2/Ti selection device. Fig. 3e shows the simulated microscopic changes and dynamic evolution process of Ag CF in both ON-state and OFF-state during the first (1st) cycle and the thousandth (1000th) cycle. It is found that the morphology of Ag CF of both states in the 1st cycle is similar to that in the 1000th cycle (image 1 versus image 3, image 2 versus image 4), and only a little variations are observed after 1000 successive switching cycles. Extrapolating from these results, the excellent uniformity of current values and voltage values in the Pt/Ag/SiO2/TiO2/Ti selection device can be attributed to the similar morphology of Ag CF in different switching cycles.

3.2. Resistive switching mechanism

In order to further elucidate the original nature of the unidirectional volatile switching characteristics in the Pt/Ag/SiO2/TiO2/Ti selection device, we fabricated two control samples with Ag/TiO2/Ti and Ag/SiO2/Ti structures, where the thicknesses of the TiO2 and SiO2 layers are the same as those in the Pt/Ag/SiO2/TiO2/Ti selection device. The \( I-V \) curves of the Ag/TiO2/Ti and Ag/SiO2/Ti structure devices are shown in Supporting Information Fig. S2 and Fig. S3, respectively. In the case of Ag/TiO2/Ti structure, the device exhibits nonlinear diode characteristics, as shown in Supporting Information Fig. S2. The rectifying \( I-V \) characteristic of the Ag/TiO2/Ti device can be attributed to the Ohmic contact at TiO2/Ti interface and the Schottky contact at the Ag/TiO2 interface \([18,42]\). As for the Ag/SiO2/Ti structure device, the \( I-V \) characteristics shown in Supporting Information Fig. S3 are performed under different \( I_{\text{comp}} \) of 1 \( \mu \)A, 10 \( \mu \)A, 100 \( \mu \)A, and 1 mA, respectively. From the \( I-V \) curves in Supporting Information Fig. S3 and Fig. 1, the Ag/SiO2/Ti device has the similar initial resistance as the Pt/Ag/SiO2/TiO2/Ti selection device (about \( 10^{11} \) \( \Omega \)). Therefore, the bulk resistance of the SiO2 layer is the main contributer for the Pt/Ag/SiO2/TiO2/Ti fresh device. However, different from the unidirectional volatile switching behavior obtained in the Pt/Ag/SiO2/TiO2/Ti selection device (Fig. 1), the ON-state of the Ag/SiO2/Ti device cannot be switched to a stable OFF-state after the forming process even at a very low \( I_{\text{comp}} \), as shown in Supporting Information Fig. S3.

There have been numerous studies that have shown the contact of bi-layer film plays an important role and has great influence on the resistive switching characteristics of RRAM devices \([43,44]\). To further clarify this, we then fabricated another control sample with the Ag/TiO2/SiO2/Ti bi-layer structure under the same process. The \( I-V \) curve of the control Ag/TiO2/SiO2/Ti device with \( I_{\text{comp}} \) of 100 \( \mu \)A is shown in Supporting Information Fig. S4. It is worth noting that, different from the Pt/Ag/SiO2/TiO2/Ti selection device, volatile switching characteristic cannot be achieved in the Ag/TiO2/SiO2/Ti structure device. Thus, it is indicated that the unidirectional volatile switching characteristics in the Pt/Ag/SiO2/TiO2/Ti selection device is not resulted from the contact of SiO2 and TiO2.

In recent years, planar structure RRAM devices based on an active electrode (e.g. Ag) and an inert electrode (e.g. Pt or W) are often used to study the underlying mechanism of resistive switching phenomenon due to easy observation on the change of Ag CF between two electrodes \([33,45,46]\). To further reveal the underlying mechanism of the unidirectional volatile switching behavior in the Pt/Ag/SiO2/TiO2/Ti selection device from the microscopic point of view, both SiO2-based and TiO2-based planar structure devices were fabricated. In order to ensure the electroforming operation successfully, an appropriate distance between Ag and Ti electrodes is needed. Thus, the gaps between the Ag and Ti electrodes of the fabricated planar structure devices were designed from 200 nm to 2 \( \mu \)m. The schematic of the planar structure device and typical SEM images of the as-fabricated SiO2-based planar devices with different gaps are illustrated in Fig. 4a. Fig. 4b shows an as-fabricated SiO2-based planar device with a gap of 850 nm. As shown in Fig. 4c, the as-fabricated planar device is in the OFF-state with a very low current. In order to trigger the electroforming process and further observe the change of Ag CF in the Ag/SiO2/Ti planar device, a 40 V constant voltage stress was applied.
on Ag electrode. As shown in Fig. 4d, the resistance of the planar device suddenly decreased to $I_{\text{comp}}$ after 35 s. However, the low resistance state was only maintained for a short time and returned to a high resistance again after 40 s. This result indicates that the CF formed in the Ag/SiO$_2$/Ti planar structure is unstable during the electroforming process, which leads to a spontaneous rupture. The corresponding SEM image after the electroforming process is shown in Fig. 4e. As can be seen from Fig. 4e, the Ag nanoparticle chain formed between the Ag and Ti electrodes is discontinuous, where a larger number of Ag nanoparticles are generated around Ag electrode and the nanoparticle chain extends towards Ti electrode. On the other hand, the Ag/SiO$_2$/Ti vertical structure device cannot be switched to a stable OFF-state after the electroforming process, as shown in Supporting Information Fig. S3, which may serve as another clue to claim the unstable CF within the Ag/SiO$_2$/Ti structure. The chemical compositions of the CF region in the initial-state and after the electroforming process were analyzed by using EDS analysis for further understanding the nature of the CF. As
shown in Fig. 5a, no Ag element signal is found in the initial-state in the SiO₂ film. However, a high Ag element signal was detected in the CF region (e.g. circular region labeled by “1” and “2”) after the electroforming process, as shown in Fig. 5b and c. The corresponding EDS elemental mapping images also confirmed that Ag element was present in region 1 and 2, as shown in Supporting Information Fig. S5. In addition, line profile analysis (Fig. 5d) along the red line from region 1 to region 3 in Fig. 4e, again revealing clear Ag element signal along the CF region. These results clearly indicate that Ag is the primary elemental component of the CF in the Ag/SiO₂/Ti device, which is consistent with the previous results obtained by SEM and in situ TEM [33,45].
Fig. 6a shows an as-fabricated TiO2-based planar device with a gap of about 1 μm. To explore electrical properties of the TiO2-based planar device, a first negative-voltage sweep (0V → -3V) followed by a positive-voltage sweep (0V → 10V) is applied on Ag electrode to program the fresh device. It is worth noting that the Ag/TiO2/Ti planar device exhibits rectifying $I - V$ characteristic, as shown in Fig. 6b, which is similar to the asymmetric $I - V$ characteristic observed in vertical structure device (Supporting Information Fig. S2). Compared with the SEM image of initial-state (before programming) shown in Fig. 6a, the SEM image observed after programming process reveals that no significant change is observed in the region between the two electrodes, as shown in Fig. 6c. On the other hand, the chemical compositions of the interelectrode region before and after programming were analyzed by EDS, as shown in Fig. 6d. It can be seen from Fig. 6d that the element signals are almost the same, and no Ag element signal is detected in the interelectrode region after programming. These results prove that it is difficult for Ag ions to migrate onto TiO2 film.

Based on the experimental results obtained above, it is clear that unstable Ag nanoparticles based CF is formed in Ag/SiO2/Ti structure device, movement of Ag ions in TiO2 film is difficult, and rectifying $I - V$ characteristic is observed in Ag/TiO2/Ti structure device because of the Schottky contact at the Ag/TiO2 interface. Therefore, the intrinsic mechanism of the unidirectional volatile switching behavior in our fabricated Pt/Ag/SiO2/TiO2/Ti selection device can be schematically summarized in Supporting Information Fig. S6. When a positive voltage is applied on the Ag electrode, Ag ions are generated and will migrate onto the Ti cathode. Because the migration of Ag ions in TiO2 film is difficult, unstable CF consisting of Ag nanoparticles is only able to form in the SiO2 layer rather than complete CF between the two electrodes, as illustrated in the ON-state of the Supporting Information Fig. S6. Once the Ag CF formed between the Ag electrode and SiO2/TiO2 interface, a Schottky contact is thus formed between the Ag CF and TiO2 interface, leading to the rectifying $I - V$ characteristic of the Pt/Ag/SiO2/TiO2/Ti selection device. When the voltage is swept back to 0V, the CF is completely disrupted due to the discontinuous CF channel consisting of discrete Ag nanoparticles, as illustrated in the OFF-state of the Supporting Information Fig. S6.

4. Conclusions

In summary, an asymmetric unidirectional volatile switching selection device with Pt/Ag/SiO2/TiO2/Ti structure was presented. Outstanding features such as a low forming voltage, low OFF-state current, high nonlinearity, and excellent uniformity were achieved.
Furthermore, the underlying mechanism of the unidirectional volatile switching behavior in the Pt/Ag/SiO₂/TiO₂/Ti selection device was revealed by simulation and experiments. The simulated analysis and SEM results from the planar structure devices directly prove that unstable CF consisting of Ag nanoparticles is only formed in the SiO₂ layer, and then a Schottky contact between the Ag CF and TiO₂ interface begins to appear, leading to the unidirectional volatile switching with rectifying $I-V$ characteristic in the Pt/Ag/SiO₂/TiO₂/Ti selection device. The underlying nature of the unidirectional volatile switching behavior illustrated in this work will provide a guide to further improve the device performances for numerous applications, such as high density memory and bio-inspired neuromorphic systems.

**Credit author statement**

Liping Fu: Conceptualization, Methodology, Measurement, Writing-Original draft preparation, Kuan Yang: Software, Simulation, Xiaoping Gao: Methodology, Experiment, Discussion, Xiaoliang Fan: Experiment, Visualization, Xiaojian Song: Experiment, Measurement, Zewei Wu: SEM, Visualization, Xiaolong Fan: Experiment, Discussion, Yingtao Li: Supervision, Conceptualization, Writing-Reviewing and Editing.

**Declaration of competing interest**

All authors declare that no conflict of interest exists.

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**Appendix A. Supplementary data**

Supplementary data to this article can be found online at https://doi.org/10.1016/j.mtadv.2021.100197.

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