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Enlarged read window in the asymmetric ITO/HfO_x/TiN complementary resistive switch

H. Z. Zhang,¹ D. S. Ang,^{1,a)} Y. Zhou,¹ and X. P. Wang²

¹*School of Electrical and Electronic Engineering, Nanyang Technological University, Nanyang Avenue, Singapore 639798*

²*Institute of Microelectronics, A*STAR (Agency for Science, Technology and Research), Singapore 117685*

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The narrow read-window of most complementary resistive switches proposed to-date poses a significant challenge to array level implementation, as inherent variations in the set and reset voltages result in an unacceptably small read margin. In this work, we present the asymmetrical ITO/HfO_x/TiN complementary resistive switch, with a significantly enlarged positive read window of 1.6 V as compared to the much narrower window of 0.5 V of the symmetrical TiN/HfO_x/TiN switch. A read margin of 1.1 V is obtained after accounting for statistical variations, representing a significant improvement over the 0.1 V margin of the symmetrical counterpart. Analyses show that the enlarged read window may be ascribed to two important attributes: (1) the stronger affinity of ITO for oxygen, which leads to a reduced positive set voltage and (2) a larger work function of the ITO, resulting in an increase in the positive reset voltage. *Published by AIP Publishing.*
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The resistive random access memory (RRAM) has shown the potential in becoming the future universal memory, due to its fast switching time ($\sim$ ns), low switching energy ($\sim$ pJ), and high write endurance ($\sim 10^{12}$ cycles).^{1–8} For high-density memory integration, RRAM cells are usually deployed in the form of a crossbar array. However, a major issue with a crossbar array is the sneak-path current, a parasitic current which arises from neighboring RRAM cells programmed to the low-resistance state (LRS).⁹ A typical way to solve the sneak-path current is to connect each RRAM cell in series to a selector device, which serves to isolate the cell from its neighbours.^{10–13} To preserve the packing density and three-dimensional stacking advantages of the crossbar architecture, recent effort is mainly centered on oxide-based thin-film selectors^{14–16} having the same metal-insulator-metal structure as the RRAM cell. But for a selector to be effective, it must have a low off-state current and a highly non-linear, i.e., steep turn-on current-voltage curve. The attainment of both characteristics has remained a difficult challenge.¹⁷

On the other hand, complementary resistance switching (CRS)⁹ offers the promise of a selector-less crossbar RRAM array free of the sneak-path current problem. In this approach, two distinct high-resistance states (HRS) are used to represent the binary memory data, as opposed to the usual low- and high-resistance states. Only one of the two high-resistance states would switch to a low-resistance state during the read process, thus allowing them to be distinguished electrically. Because a selector is not needed, CRS enables a more straightforward realization of RRAM on flexible substrates.¹⁸ Since the introduction of the idea in 2010 based on two anti-serially connected cells,⁹ CRS has been reported in single RRAM cells with a bilayer oxide stack,^{19–22} and also in cells having a single oxide layer.^{23–26} However, a major

obstacle of CRS implementation lies in the generally small “read window,” i.e., the difference between the set and reset voltages (V_{set} and V_{reset}) for one of the high-resistance states.^{9,19–28} Coupled with inherent variations in V_{set} and V_{reset} , the resultant array-level read margin becomes unacceptably narrow. Attempt to increase the read window has been made through multi-layer stack engineering.¹⁸

A recent study has found that CRS in the TiN/HfO_x/TiN RRAM cell is due to an oxygen exchange between the vacancy-type conducting filament in HfO_x and the reactive TiN electrodes.^{23,29} The exchange enables the position of filament rupture to be alternated between the electrode interfaces, thus giving rise to a CRS behavior. On the one hand, this finding indicates that a cell with the same top and bottom electrodes would tend to have a small read window, due to a similar interaction between the filament and the electrodes.²³ On the other hand, it suggests the possibility of enlarging the read window through electrode interface engineering. In this work, we report the CRS characteristics of the ITO/HfO_x/TiN RRAM cell, with different top and bottom electrodes. A significant improvement in the positive read window to 1.6 V is obtained. The resultant read margin is 1.1 V after accounting for V_{set} and V_{reset} variations. On the other hand, the negative read window is comparable to that of the TiN/HfO_x/TiN. The results may be consistently ascribed to the difference in oxygen affinity and work function of the ITO and TiN electrodes.

Figure 1(a) shows a scanning electron micrograph of the asymmetric ITO/HfO_x/TiN crossbar device. Electron-beam lithography (EBL) was first performed on a pre-cleaned SiO₂/Si substrate, followed by DC reactive magnetron sputtering for TiN deposition (Ti:N ratio of 1.1:1) and lift-off to form the bottom electrode. Atomic layer deposition (ALD) of the HfO_x layer at 250 °C was then carried out with tetrakis(dimethylamino) hafnium as the precursor and H₂O as the oxidizing agent. After the second EBL step, ITO deposition

^{a)}E-mail: edsang@ntu.edu.sg

FIG. 1. (a) High-magnification scanning electron micrograph of the ITO/HfO_x/TiN crossbar resistive memory device. Dimensions of the TiN bottom electrode and ITO top electrode are as indicated. (b) High-resolution cross-sectional transmission electron microscopy of the ITO/HfO_x/TiN stack.

(via physical sputtering of a commercial ITO target with ~5 wt. % Sn in an argon-only ambient) followed by lift-off was carried out to complete the top electrode formation. High-resolution cross-sectional transmission electron microscopy of the test device revealed a 7 nm thick HfO_x [Fig. 1(b)]. The bright shade between the HfO_x and TiN may refer to an interfacial oxide formed by the oxidation of the exposed TiN surface during initial ALD cycles.²³ For comparison, symmetric TiN/HfO_x/TiN devices, only substituting the top ITO electrode by TiN, were also fabricated. To facilitate later discussion, the former will be referred as the ITO device, while the latter as the TiN device. DC voltage-sweep testing was performed at room temperature (27 °C) using a Keithley SCS4200 parameter analyzer. The voltage-bias was applied to the top ITO or TiN electrode with the bottom TiN electrode always grounded.

Figure 2 shows typical CRS behaviors of the ITO and TiN devices. Forming was carried out using a positive-voltage sweep (not shown) to create a conductive filament within the HfO_x layer so that the subsequent resistive switching effect could be observed. For the ITO device [Fig. 2(a)], a positive-voltage sweep triggers a high to low resistance (HRS1 → LRS) or set transition @ $V_{\text{set}}^+ \sim 0.2$ V. If the voltage sweep is stopped after this set transition, the device can only be reset by a negative-voltage sweep (@ $V_{\text{reset}}^- \sim -1$ V), i.e., it exhibits a typical positive set/negative reset bipolar switching behavior. However, if the positive-voltage sweep is further extended, a reset subsequently happens @ $V_{\text{reset}}^+ \sim 1.5$ V, bringing the device to a different high-resistance state (HRS2). Following this reset, the device can no longer be set again using a positive-voltage sweep. Instead, it can only be set by a negative-voltage sweep (@ $V_{\text{set}}^- \sim -0.4$ V) and reset by a positive-voltage sweep (@ $V_{\text{reset}}^+ \sim 1.5$ V), i.e., the device now operates in a complementary switching mode (negative set/positive reset). An extension of the negative-voltage sweep

yields a reset @ $V_{\text{reset}}^- \sim -1$ V to HRS1, after which the device is changed back to the positive set/negative reset mode. The device may be alternated between the two switching modes by extending the voltage sweep beyond the initial set transition (Fig. S1, [supplementary material](#)). A similar behavior is obtained for the TiN device [Fig. 2(b)]. An oxygen-exchange mechanism between the oxygen vacancies in the conducting filament formed in the HfO_x and the ITO or TiN electrode, adopted from our earlier study on the CRS behavior of the TiN/HfO_x/TiN device,²³ may also be used to explain the CRS behavior in the ITO device. A conducting filament, comprising oxygen vacancies, is generated in the HfO_x after forming.^{30–35} A negative-voltage sweep drives oxygen from the top ITO interface region to the adjacent vacancy sites in the filament, rupturing the filament and causing a reset to HRS1. A subsequent positive-voltage sweep reconnects the filament and causes a set by reversing the oxygen migration process. An extended positive-voltage sweep would draw oxygen from the bottom TiN interface region into the adjacent filament and in turn ruptures the filament there, resetting the device to HRS2 (see Fig. S2 in the [supplementary material](#) for a schematic illustration).

A comparison of the CRS characteristics of the ITO and TiN devices reveals two major differences. First, for most of the ITO devices tested, the resistance of HRS1 is significantly higher than that of HRS2 [cf. Fig. 2(a)]. The respective resistance distributions differ by a factor of $>10^3$ at the median value [Fig. 3(a)]. This is in contrast to the comparable HRS1 and HRS2 of the TiN device [Fig. 3(b)]. Clearly, the lower HRS2 resistance of the ITO device limits the memory window. But it is pertinent to clarify that this lower HRS2 resistance (or higher current) of the ITO device would not pose any problem for array level CRS operation. Electrically, HRS1 and HRS2 are not distinguished by the difference in their current or resistance values but by their different responses to a read voltage bias. A positive voltage larger than V_{set}^+ would trigger a HRS1 → LRS transition [Fig. 2(a)], since the ruptured filament next to the ITO interface is reconnected to the ITO (via oxygen migration from the ruptured filament to the ITO). However, it has no effect if the device is in the HRS2 state, since the voltage polarity does not favor oxygen migration from the ruptured filament back to the adjacent TiN electrode. Similarly, a read voltage more negative than V_{reset}^- would trigger a HRS2 → LRS transition but has no effect if the device is in the HRS1 state.

Second, the V_{set} and V_{reset} of the ITO device show a larger difference of 1.6 V (median) in the positive-voltage

FIG. 2. Typical complementary resistive switching behavior of the (a) ITO/HfO_x/TiN and (b) TiN/HfO_x/TiN RRAM devices. “TE” denotes the top (ITO) electrode.

FIG. 3. Distributions of the low-resistance state and high-resistance state (HRS1 and HRS2) resistances in the positive- and negative-voltage regimes for the (a) ITO/HfO_x/TiN and (b) TiN/HfO_x/TiN RRAM devices.

FIG. 4. Distribution of the set and reset voltages in the positive- and negative-voltage regimes for the (a) ITO/HfO_x/TiN and (b) TiN/HfO_x/TiN RRAM devices. As indicated are the respective read windows and margins in both voltage regimes.

regime as compared to 0.7 V in the negative-voltage regime [Fig. 4(a)], whereas they differ comparably (0.5 V) in both voltage regimes for the TiN device [Fig. 4(b)]. After accounting for the spread in voltages, the difference between the upper end of V_{set}^+ and the lower end of V_{reset}^+ distributions (defined as the read margin) in the positive-voltage regime is 1.1 V for the ITO device, substantially larger than 0.13 V for the TiN device. On the other hand, the read margin in the negative-voltage regime is comparable for both devices (0.2 V versus 0.15 V). Table I summarizes, for both devices, the median V_{set} and V_{reset} values for the positive- and negative-voltage regimes, together with the filament change that occurred for each voltage, and a notation denoting the direction of oxygen transfer between the filament and the electrode. Take the V_{set}^+ of the ITO device for instance, which corresponds to the reconnection of the ruptured filament next to ITO [Figs. 2(a) and S2]. This change arises from the migration of oxygen from the ruptured filament (HfO_x) to the ITO, and the process is denoted as $\text{O}_{\text{HfO}_x} \rightarrow \text{ITO}$. At V_{reset}^+ , the filament next to TiN is ruptured and the oxygen transfer process is denoted as $\text{O}_{\text{TiN}} \rightarrow V_{\text{O}}$ (V_{O} denotes the vacancy sites of the filament). It is clear from Table I that the enlarged positive read window of the ITO device is due to the reduction in V_{set}^+ and the increase of V_{reset}^+ , relative to those of the TiN device. The analysis implies that (1) it is easier for oxygen to migrate from the ruptured filament to ITO ($V_{\text{set}}^+ = 0.2$ V) than to TiN ($V_{\text{set}}^+ = 0.6$ V); (2) it is more difficult for oxygen to migrate from the bottom TiN to adjacent vacancy sites of the filament in the ITO device ($V_{\text{reset}}^+ = 1.8$ V) than in the TiN device ($V_{\text{reset}}^+ = 1.1$ V).

Since both devices have the same bottom TiN electrode, the above differences may be ascribed to the different top electrodes. To understand point (1), X-ray photoelectron

spectroscopy (XPS) measurement was made on separately prepared test samples with a 7-nm HfO_x blanket-deposited over either a bottom TiN or ITO electrode (Fig. S3, [supplementary material](#)) to check the effect of the electrode material on the HfO_x stoichiometry. Details on the XPS test samples, measurement, and data analysis are provided in the [supplementary material](#). The Hf 4f and O 1s core-level spectra, and their corresponding constituent component spectra, are shown in Fig. S4. Analysis revealed a Hf:O ratio of 1:1.4 for the HfO_x layer in the HfO_x/ITO stack, as compared to a ratio of 1:1.7 in the HfO_x/TiN stack. The lower oxygen ratio for the HfO_x/ITO stack indicates that oxygen in the HfO_x may have been “scavenged” by the bottom ITO layer. Studies^{36,37} have found that an ITO film sputter-deposited in an argon-only (i.e., oxygen-absent) ambient has a high concentration of oxygen vacancies, which give the film an n-type conductivity. An oxygen deficient ITO layer in turn promotes the migration of oxygen from the HfO_x layer,³⁸ thus making the latter also more oxygen deficient. The XPS analysis shows that the ITO electrode has a stronger affinity for oxygen than the TiN electrode. This may explain the smaller V_{set}^+ of the ITO device.

Next, we turn our attention to point (2). It should be noted that the V_{reset}^+ of the TiN device (1.1 V) is smaller than that of the ITO device (1.8 V), despite in both cases, the reset involves the rupture of filament due to oxygen migration from the bottom TiN electrode. The large difference between HRS1 and HRS2 of the ITO device provides a clue on the effect of the ITO electrode on electrical conduction through the stack. To shed further light, a study on the current conduction mechanism was made. Among the various transport models (Poole-Frenkel, Schottky, space-charge limited, and quantum point contact) proposed for RRAM current conduction in HRS, the Schottky model is found to give the best fit to the experimental data. According to the Schottky conduction model, the current density J may be expressed as^{39,40}

$$J = A^* T^2 \exp \left[\frac{-q(\phi_B - \sqrt{qE/4\pi\epsilon})}{kT} \right] = C_1 \exp \left(-\frac{q\phi_B}{kT} + C_2 \sqrt{V} \right), \quad (1)$$

where $C_1 (= A^* T^2)$ and $C_2 \left(= \frac{1}{kT} \sqrt{\frac{q}{4\pi\epsilon}} \right)$ are constants for a given interface structure and temperature T ($A^* = 120 \text{ A cm}^{-2} \text{ K}^{-2}$ is the Richardson constant;⁴¹ k is the Boltzmann constant; q is the electronic charge; d is the effective barrier thickness; and ϵ is the permittivity) and $q\phi_B$ is the Schottky barrier (in eV). As is apparent in Fig. 5, a very good least-square linear regression fit ($R^2 > 0.98$) to the experimental data can be obtained on a $\ln I$ versus $V^{1/2}$ plot, implying that conduction in HRS1 and HRS2 is limited by Schottky emission. The extracted Schottky barrier is 0.49 eV and 0.17 eV for the ITO/HfO_x and HfO_x/TiN interface, respectively. The substantially lower current (or higher resistance) for HRS1 may thus be explained by the larger Schottky barrier at the ITO/HfO_x interface. Studies on the tunneling current characteristics of ITO/HfO₂/Si and TiN/HfO₂/Si structures have also found a larger ITO/HfO₂ Schottky barrier.^{42,43} The

TABLE I. Summary of CRS voltages, location of filament change, and direction of oxygen transfer between filament and electrode.

Device	Median switching voltage	Filament change	Oxygen transfer
ITO/HfO _x /TiN	$V_{\text{set}}^+ = 0.2$ V	Reformed @ITO	$\text{O}_{\text{HfO}_x} \rightarrow \text{ITO}$
	$V_{\text{reset}}^+ = 1.8$ V	Ruptured @TiN	$\text{O}_{\text{TiN}} \rightarrow V_{\text{O}}$
	$V_{\text{set}}^- = -0.3$ V	Reformed @TiN	$\text{O}_{\text{HfO}_x} \rightarrow \text{TiN}$
	$V_{\text{reset}}^- = -1.1$ V	Ruptured @ITO	$\text{O}_{\text{ITO}} \rightarrow V_{\text{O}}$
TiN/HfO _x /TiN	$V_{\text{set}}^+ = 0.6$ V	Reformed @TiN _{top}	$\text{O}_{\text{HfO}_x} \rightarrow \text{TiN}_{\text{top}}$
	$V_{\text{reset}}^+ = 1.1$ V	Ruptured @TiN _{bot}	$\text{O}_{\text{TiNbot}} \rightarrow V_{\text{O}}$
	$V_{\text{set}}^- = -0.6$ V	Reformed @TiN _{bot}	$\text{O}_{\text{HfO}_x} \rightarrow \text{TiN}_{\text{bot}}$
	$V_{\text{reset}}^- = -1.1$ V	Ruptured @TiN _{top}	$\text{O}_{\text{TiNtop}} \rightarrow V_{\text{O}}$

FIG. 5. $\ln I$ versus $V^{1/2}$ plot showing a good fit of the Schottky conduction model to the current-voltage characteristic of the (a) HRS1 and (b) HRS2 states of the ITO/HfO_x/TiN device. The extracted Schottky barrier $q\phi_B$ for each case is as indicated. (c) Schematic energy band diagram of the ITO/filament/TiN configuration, showing the presence of a Volta potential $\Delta\psi$ arising from the different work functions of the ITO and TiN electrodes. The band structure of the filament is omitted due to uncertainties on its chemical composition and shape. Variation in $\Delta\psi$ across the structure is represented by a simplified linear variation of the vacuum level E_{vac} . $E_{F,ITO}$ and $E_{F,TiN}$ denote the Fermi level of the ITO and TiN electrode, respectively.

smaller HfO_x/TiN Schottky barrier gives rise to a lower HRS2 resistance which limits the memory window. While it may be possible to increase the HfO_x/TiN Schottky barrier (e.g., by inserting an ultrathin wide bandgap oxide such as Al₂O₃ between the HfO_x and TiN), this could potentially impact the effective TiN work function and hence the read margin, as elaborated below.

The larger Schottky barrier at the ITO/HfO_x interface implies that the ITO electrode has a larger work function than the TiN electrode. This inference is in general agreement with the work function of ITO and TiN reported elsewhere; e.g., 5.3 eV for ITO⁴⁴ and 4.5 eV for TiN.⁴⁵ Due to the work function difference, a Volta potential, $\Delta\psi$, exists between the ITO and TiN electrodes in both the high- and low-resistance states [Fig. 5(c)]. Because of the larger ITO work function, the polarity of $\Delta\psi$ is such that it opposes (reinforces) a positive (negative) voltage applied to the ITO with respect to the TiN. The opposition between the $\Delta\psi$ and positive applied voltage means that a more positive voltage is needed to cause a reset, which explains the increase in V_{reset}^+ of the ITO device. An increase in V_{set}^+ should also be expected but the stronger oxygen affinity of ITO may have offset the increase, resulting in an overall decrease relative to the TiN device. A comparison of V_{set}^- and V_{reset}^- of the two devices (Table I) also consistently reflects the possible role of $\Delta\psi$ in the resistive switching voltages of the ITO device in the negative-voltage regime. The V_{set}^- of the ITO device is -0.3 V, less negative than that of the TiN device (-0.6 V), despite in both cases, the set involves reconnection of the ruptured filament at the TiN interface, by oxygen migration from the ruptured filament to TiN. But this difference in V_{set}^- of the two devices is smaller than the difference in V_{reset}^+ . The reason for this discrepancy is not clear and is a subject of further investigation. As for V_{reset}^- , a more negative value should

be expected for the ITO device, in view of the stronger oxygen affinity of ITO and hence a greater difficulty for oxygen to migrate from ITO back to the filament. However, the V_{reset}^- of both devices is similar (1.1 V), implying that the Volta potential in the ITO device may have helped reduce the reset voltage (as it reinforces the negative applied voltage). On the other hand, no Volta potential should exist in the symmetrical TiN/HfO_x/TiN stack and thus the TiN device shows identical set and reset voltages in both voltage regimes (Table I). Our explanation on the effect of the electrode work-function difference on the switching operation of the ITO device is supported by the study of Hadi *et al.*,⁴⁶ who reported a set-voltage increase for the W/CeO_x/SiO₂/n⁺Si stack relative to the W/CeO_x/SiO₂/p⁺Si stack, arising from a decrease in the work function of the n⁺Si electrode relative to the top W electrode.

In summary, highly asymmetrical CRS characteristics are observed in the ITO/HfO_x/TiN RRAM device. These behaviors are manifested in the significant difference (by a factor of $>10^3$) in the two high-resistance states (arising from the different interface barriers) and the much wider positive read margin of 1.1 V as compared to the negative read margin of 0.2 V. The former elucidates the important role of the interface barrier in the memory window, while the latter represents an important improvement over the much narrower read margin of 0.1 V obtained for the symmetric TiN/HfO_x/TiN as well as other CRS devices reported to-date, and it may be ascribed to a combination of two effects: (1) a reduced set voltage due to the greater oxygen affinity of ITO and (2) an increased reset voltage due to an opposing Volta potential arising from the larger work function of ITO, as evidenced by the larger ITO/HfO_x interface barrier. From the insights achieved, any attempt to modulate the HfO_x/TiN interface barrier for increasing the memory window should be made in the light of a possible impact on the read margin.

See [supplementary material](#) for a figure showing the repeated CRS switching cycles (Fig. S1) and a schematic illustration of the proposed mechanism for CRS (Fig. S2) in the ITO/HfO_x/TiN RRAM device. Details on XPS measurement (Fig. S3) and data analysis (Fig. S4) are also provided.

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