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Role of interfacial layer on complementary resistive switching in the TiN/HfO_x/TiN resistive memory device

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The role of the bottom interfacial layer (IL) in enabling stable complementary resistive switching (CRS) in the TiN/HfO_x/IL/TiN resistive memory device is revealed. Stable CRS is obtained for the TiN/HfO_x/IL/TiN device, where a bottom IL comprising Hf and Ti sub-oxides resulted from the oxidation of TiN during the initial stages of atomic-layer deposition of HfO_x layer. In the TiN/HfO_x/Pt device, where formation of the bottom IL is suppressed by the inert Pt metal, no CRS is observed. Oxygen-ion exchange between IL and the conductive path in HfO_x layer is proposed to have caused the complementary bipolar switching behavior observed in the TiN/HfO_x/IL/TiN device. © 2014 AIP Publishing LLC. [<http://dx.doi.org/10.1063/1.4903341>]

The resistive random access memory (RRAM) is one of the leading candidates for next-generation nonvolatile memory technology.^{1–4} The simple metal-insulator-metal structure and ease of scalability of an RRAM cell make it an ideal choice for crossbar memory array implementation via back-end-of-line interconnect integration. However, a major issue with the crossbar array is sneak-path current. Parasitic currents flowing through neighboring cells in the low resistance state (LRS) can interfere with the read/write operation performed on a designated cell,⁵ leading to data corruption. A common method by which this problem may be circumvented is to connect a rectifying selector device, such as a diode or transistor, in series with the RRAM cell to block the parasitic current. However, this comes at the expense of increased front-to-back-end integration complexity and reduced memory density.² An alternative approach is found in the concept of complementary resistive switching (CRS).⁵ In the CRS approach, two RRAM cells are connected anti-serially such that when one of the RRAM cells is programmed into the LRS, the other will be programmed into the high resistance state (HRS). In this way, parasitic current associated with a cell in the LRS is suppressed. As the integration of a different rectifying element is not required, this approach is deemed more attractive and has attracted considerable interest lately.^{6–12}

Recently, it was shown by Nardi *et al.*⁶ that CRS could also occur in the TiN/HfO_x/TiN RRAM cell. The result provides the promising outlook that the sneak-path problem may eventually be solved based on just a single RRAM cell. However, the underlying mechanism is not clear. It was reported that CRS could only occur in devices with a uniform Hf concentration in the HfO_x active layer. No CRS was observed in devices with a non-uniform Hf concentration profile. On the other hand, CRS was observed in the Pd/Ta₂O_{5–x}/TaO_y/Pd device, where the functional layer was purposely designed with two tantalum oxide layers of different stoichiometries: an oxygen-rich Ta₂O_{5–x} layer and an

oxygen-deficient TaO_y layer.⁷ CRS was attributed to the exchange of oxygen vacancies between the two layers upon application of external electrical stimuli. In an earlier study, CRS was also observed in the Pt/TiO₂/TiO_{2–x}/Pt device with an asymmetric distribution of oxygen vacancies in the active layer.⁹

In this study, we show that stable CRS can also happen in the asymmetrical TiN/HfO_x/IL/TiN device. IL refers to an interfacial layer comprising Hf and Ti sub-oxides, not grown intentionally but resulted from the oxidation of the bottom TiN electrode during the initial stages of HfO_x formation via atomic layer deposition (ALD). When the bottom electrode is replaced by Pt electrode, the resultant TiN/HfO_x/Pt device does not exhibit CRS. This device only exhibits the usual bipolar resistive switching. High-resolution cross-sectional transmission electron microscopy (HRTEM) shows the absence of a bottom IL in the TiN/HfO_x/Pt device. From these results, it may be concluded that stable CRS in the TiN/HfO_x/IL/TiN device is linked to the presence of the IL. A possible explanation for CRS in TiN/HfO_x/IL/TiN device is proposed.

The test devices were fabricated on SiO₂/p-Si substrates. Physical vapor deposition of either a 150 nm TiN or a 50 nm Pt bottom electrode was first carried out, followed by chemical vapor deposition of SiO₂ to serve as the isolation layer. After active area definition, the wafer was loaded into a Cambridge Nanotech Savannah S200 ALD system for HfO_x growth at a temperature of 250 °C and a pressure of 0.2 Torr. The precursor was tetrakis (dimethylamino) hafnium, and the oxidizing agent was H₂O. After top TiN electrode formation, patterning via reactive ion etching was carried out to form RRAM devices of different sizes. No post ALD and forming gas annealing were carried out. Except for the different bottom electrodes, both the TiN/HfO_x/IL/TiN and TiN/HfO_x/Pt devices underwent identical processing. To facilitate later discussion, the former will be referred as the TiN sample, while the latter as the Pt sample. A schematic cross-section diagram of the test device is shown in Fig. 1(a). The composition of the HfO_x layer was examined

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by x-ray photoelectron spectroscopy and the results are depicted in Fig. 1(b). The ratio of Hf to O is 1:1.74. The binding energy (BE) of Hf 4f_{7/2} and 4f_{5/2} are located at 17.0 eV and 18.6 eV, respectively, (based on the C 1s reference BE of 285.0 eV). The O 1s spectrum comprises two different chemical states. The main component at 530.2 eV is ascribed to hafnium oxide. The smaller component at 531.0 eV may be attributed to non-lattice oxygen atoms.

Results from HRTEM study on the TiN and Pt samples are shown in Figs. 1(c) and 1(d), respectively. The thickness of HfO_x is 8.5 nm. A distinctive feature of the TiN sample is the presence of a region of lighter contrast, between the HfO_x layer and the bottom TiN electrode. This observation implies that an IL (~4 nm thick) is formed between the HfO_x and the bottom TiN electrode. However, no IL formation can be seen in the Pt sample. The clear observation of a bottom IL in the TiN sample may be attributed to the oxidation of the exposed TiN electrode in the initial stages of the ALD process. Studies have shown that TiN can be readily oxidized in an oxygen-rich ambient at relatively low temperatures.¹³ During the initial cycles of our ALD process, where only a few monolayers of HfO_x were formed, the oxidizing species might have penetrated the HfO_x and oxidized the underlying TiN. The growth of a GaO_x IL was similarly observed during the ALD process of Al₂O₃ on a GaN substrate.¹⁴ For non-ALD cases, the formation of a bottom IL may still occur due to the higher thermal budget experienced by the bottom TiN electrode interface. On the other hand, bottom IL formation in the Pt sample is clearly suppressed by the relatively inert Pt metal.

Electrical testing was performed on 50 × 50 μm² devices at room temperature using a Keithley SCS4200 semiconductor parameter analyzer. The voltage-bias was applied to the top electrode and the bottom electrode was always grounded. Forming was necessary to create minute conductive paths within the dielectric stack so that subsequent resistive switching effect could be observed. In order to clearly distinguish the abrupt current jump during forming from the increase of area-dependent background leakage current, the current compliance was set to 5 mA (inset of Fig. 2(a)). It

FIG. 1. (a) Schematic cross-sectional diagram of the TiN/HfO_x/IL/TiN or Pt RRAM device. (b) O 1s XPS spectra of the HfO_x layer (inset: Hf 4f XPS spectra). High resolution cross-sectional transmission electron micrograph of (c) the TiN/HfO_x/IL/TiN and (d) TiN/HfO_x/Pt RRAM device.

FIG. 2. Current-voltage curves for the TiN/HfO_x/IL/TiN RRAM device, depicting the CRS behavior. (a) Conventional bipolar resistive switching characteristics showing a negative reset (step (1)), and positive set (step (2)) behavior after positive forming (the inset). (b) After step (2), a subsequent positive-voltage sweep to a higher value can cause a reset at ~1.2 V (step (3)). (c) Following step (3), the device can only be set under a negative-voltage sweep, at ~-0.95 V (step (4)). The polarities of set/reset have now been reversed, showing clearly a CRS behavior. (d) Increasing the voltage to a more negative value can cause a reset at ~-1.3 V (step (5)). After this, device is changed back to positive set and negative reset (see (a)).

should be mentioned that CRS behavior is also observed for lower current compliance values used on smaller area devices. Comparable current or resistance in LRS is obtained for devices with area ranging from 400 to 40 000 μm² (not shown). This lack of areal dependence of LRS conduction confirms that resistive switching is determined by localized conductive paths created during forming.

Fig. 2 shows the CRS behavior of the TiN sample after the positive-voltage forming operation, as shown in the inset. Immediately after forming, the device is reset using a negative-voltage sweep, followed by a set via a positive-voltage sweep (Fig. 2(a)). The device is switched from LRS (after forming) to HRS at ~-1.3 V (step (1)), and is switched back to the LRS at ~+0.85 V (step (2)), exhibiting the conventional bipolar switching behavior (negative reset/positive set). When the positive-voltage sweep is extended beyond the set, the device can be switched to HRS again at ~+1.2 V (step (3)), as depicted in Fig. 2(b). After this step, the device remains in HRS and cannot be set again by a positive-voltage sweep. Instead, the device can now be set under a negative-voltage sweep, at ~-0.9 V (step (4)), and reset by a positive-voltage sweep (Fig. 2(c)), i.e., the device now operates in a complementary switching mode (negative set/positive reset), opposite to the one (negative reset/positive set) observed earlier. When the negative voltage sweep is extended beyond the set, a reset happens (step (5)) and the switching mode is changed back to negative reset/positive set (Fig. 2(d)). Clearly, the asymmetrical TiN/HfO_x/IL/TiN device can exhibit CRS.

On the other hand, no CRS behavior is observed in the Pt sample, in which the bottom IL is not present (Fig. 1(d)). As can be seen in Fig. 3, only the usual bipolar switching behavior is observed. After the positive forming step, a typical negative reset followed by a positive set can be readily achieved. Extending the positive-voltage sweep beyond the set does not lead to any apparent reset. A subsequent

FIG. 3. (a) Current-voltage curves for the TiN/HfO_x/Pt RRAM device formed under a positive forming. The order of measurements is as indicated by the numbers. In contrast to TiN/HfO_x/IL/TiN RRAM device, only single mode bipolar resistance switching (i.e., negative reset (step (1)) and positive set (step (2))) is evident. Increasing the positive-voltage sweep beyond the first set does not lead to any apparent reset (step (3)). A subsequent negative-voltage sweep yields the usual reset (step (4)). (b) Current-voltage curves of another TiN/HfO_x/Pt RRAM device. Likewise, the device only shows single mode bipolar resistance switching (i.e., negative reset (step (1)) and positive set (step (2))). When the positive-voltage sweep is extended to higher value (step (3)), the device is only set to a lower resistance state, which, in turn, requires a more negative voltage to reset it (step (4)).

negative-voltage sweep yields the usual reset (Fig. 3(a)). Further increasing the positive-voltage sweep sets the device to a lower resistance state, which, in turn, requires a more negative voltage to reset it (Fig. 3(b)). Since the main difference of the Pt sample as compared to the TiN sample lies in the absence of the bottom IL in the former, the corresponding absence of CRS implies that the bottom IL plays a crucial role in enabling the CRS behavior seen in the TiN sample.

The need for a bottom IL to be present in order to exhibit stable CRS for the HfO_x RRAM implies that the underlying mechanism may be similar to that in the Ta₂O_{5-x}/TaO_y,⁷ TiO₂/TiO_{2-x} (Ref. 9) devices. In the latter devices, oxygen ions or vacancies can be alternately transferred between the Ta₂O_{5-x} or TiO₂ layer and the respective TaO_y or TiO_{2-x} layer, depending on the applied voltage polarity, causing either of the two layers to be driven to a high resistance state. A possible explanation for CRS in the TiN sample is schematically illustrated in Fig. 4. Although it may not be as apparent as the bottom IL, a top IL comprising Hf sub-oxide is expected to be formed during device fabrication, due to the oxygen scavenging property of TiN.¹⁵ During positive-voltage forming or set, oxygen ions are drawn from the HfO_x layer towards the top TiN electrode, leading to the formation of a conductive path (comprising oxygen vacancies) in the HfO_x layer (Fig. 4(a)). As a result, the overall oxide resistance decreases, setting the RRAM device to the LRS. Some of the oxygen ions may be captured by vacancies present in the top IL, reducing the vacancy density in the IL region above the conductive path. During a subsequent negative-voltage sweep, these oxygen ions in the top IL are transported back to the conductive path,¹⁶ causing the conductive path near the top IL to be partially reoxidized, and hence, the device is switched to HRS (Fig. 4(b)). The process is reversed during a subsequent positive-voltage set (Fig. 4(a)). The role of the top IL in bipolar resistive switching is corroborated by improved performance achieved via the incorporation of either a thin Ti or Hf cap layer in between the top TiN electrode and the HfO_x layer.¹⁷ It should be noted that the top IL in the TiN sample plays an important role opposite to that of the top Ta₂O_{5-x} layer in the

FIG. 4. Schematic illustration of the proposed mechanism for complementary resistive switching in the TiN/HfO_x/IL/TiN RRAM device. (a) Formation of conductive path in the HfO_x either after positive-voltage forming or set, where oxygen ions from the HfO_x are drawn towards the top electrode, with some captured by the vacancies in the IL. (b) During negative-voltage sweep, oxygen ions captured in the top IL are driven back to the vacancy-filled conductive path. This partially reoxidizes the conductive path next to the top IL and resets the device. Alternating between a positive and negative voltage sweep repeatedly drives the device between low resistance state (Fig. 4(a)) and high resistance state (Fig. 4(b)), respectively. (c) A further increase of the positive voltage after the set (Fig. 4(a)) draws in oxygen ions from the bottom IL, causing the conductive path next to be partially reoxidized and resetting the device (arrow 1). This changes the position of the reoxidized part of the conductive path to the bottom IL. (d) A subsequent negative-voltage sweep drives the oxygen ions in the partially reoxidized part of the conductive path back to the bottom IL, reforming the conductive path and setting the device. A further increase of the negative voltage after the reset in (Fig. 4(d)) drives oxygen ions from the top IL into the underlying conductive path, partially reoxidizing this region of the conductive path and resetting the device (arrow 2). This changes the position of the reoxidized part of the conductive path back to the top IL, as shown in Fig. 4(b).

Ta₂O_{5-x}/TaO_y sample.⁷ In the latter, a negative voltage drives oxygen ions from the relatively oxygen-rich Ta₂O_{5-x} to the oxygen-deficient TaO_y layer, setting the device to LRS during forming, and a positive voltage is needed to reset the device to HRS.

A further extension of the positive-voltage sweep following the set may draw oxygen ions from the bottom IL into the HfO_x layer, and, in turn, cause the conductive path next to the bottom IL to be partially reoxidized, and the device to be reset to HRS (Fig. 4(c)). It is worth noting that the internal configuration of the conductive path is now different from that after negative reset (Fig. 4(b)). The reoxidized part of the conductive path is now situated near the bottom IL, as opposed to case of negative reset, where the reoxidized part is situated near the top IL. It is here where the role of the bottom IL is crucial to realizing stable CRS. Without it, such as in the case of the Pt sample, no supply of oxygen ions to the conductive path can occur. Increasing the positive voltage sweep would lead to more oxygen ions being drawn towards the top IL, further depleting the oxygen content in the conductive path and increasing the current (Fig. 3(b)). Following the positive reset in the TiN sample (Fig. 4(c)), the device now can no longer be set using a positive-voltage sweep, since a moderate positive voltage could not deplete the oxygen ions in the conductive path near the bottom IL. However, a negative-voltage sweep could cause a set

(Fig. 4(d)) since this voltage polarity could drive the oxygen ions back to the bottom IL, making the adjacent conductive path oxygen-deficient again. Due to the absence of the bottom IL in the Pt sample, alternate partial reoxidation and reformation of the conductive path cannot readily happen in this region. Only partial reoxidation/reformation of the conductive path near the top IL can occur, yielding only a single bipolar switching mode.

Sweeping the voltage to more negative values after the negative set (Fig. 4(d)) would drive oxygen ions from the top IL into the conductive path, causing the top part of the conductive path to be partially reoxidized and the device to be reset. When this happens, the internal configuration of the conductive path is changed back to that shown in Fig. 4(b), and the device can now only be set and reset under positive and negative voltages, respectively.

The consistently larger reset voltage, as compared to the set voltage, may suggest that it is more difficult to move oxygen ions from the IL to HfO_x during reset than to move oxygen ions from HfO_x to the IL during set. The difference may be explained as follows. Once the device is reset, most of the applied voltage during a subsequent set measurement will be dropped across the narrow, partially reoxidized part, as this is the most resistive part of the conductive path. Hence, a relatively low voltage is enough to generate the electric field needed to move the oxygen ions from HfO_x to IL. After set, however, the applied voltage during a subsequent reset operation would be more evenly distributed throughout the entire conductive path, and thus, a higher voltage is needed to produce the required electric field to move oxygen ions from IL to HfO_x .

The explanation in Fig. 4 attributes CRS to the alternate exchange of oxygen ions between the HfO_x conductive path and the top and bottom IL. The absence of bottom IL is believed to have caused the CRS behavior to disappear in the Pt sample. To further validate the importance of the bottom IL on CRS, negative-voltage forming was carried out on the Pt sample to determine if the positive reset/negative set switching behavior can happen. Since the Pt would be the anode during forming/set, much poorer switching behavior should result as there is no IL at the Pt electrode interface. This inference is indeed borne out by Fig. 5. Unlike the case of positive-voltage forming, only a very marginal reset is obtained during the positive voltage sweep after negative-voltage forming. Extending the positive voltage sweep to higher values would trigger a set (not shown), and subsequently, the device can be reset by a negative voltage and set by a positive voltage. In this case, it is believed that the partial reoxidation and reformation of the conductive path happen at the top IL interface, similar to the case of positive-voltage forming (Fig. 3). The poor resistive switching behavior after negative forming further supports the proposed crucial role of the bottom IL in determining CRS behavior.

It should be noted that although CRS has been observed in RRAM stacks using Pt as the electrode (e.g., $\text{Pt}/\text{ZrO}_x/\text{HfO}_x/\text{ZrO}_x/\text{Pt}$ (Ref. 18)), the two dissimilar oxide materials (ZrO_x and HfO_x) are believed to have played a role similar to the two layers of the same material but of different oxygen contents (e.g., $\text{Ta}_2\text{O}_{5-x}/\text{TaO}_y$ (Ref. 7) and $\text{TiO}_2/\text{TiO}_{2-x}$

FIG. 5. Evolution of current-voltage characteristics of the TiN/ HfO_x /Pt RRAM device under negative voltage forming for successively higher current compliance (as indicated). After the negative voltage forming (symbol), a negative voltage sweep (red dashed line) reveals a higher current, confirming the transition to a lower resistance state. However, only very marginal reset can be observed under positive voltage sweeps (dashed lines denote a second positive-voltage sweep after the first (solid lines)).

(Ref. 9)), allowing oxygen-ion exchange to occur alternately between them. Hence, CRS could still happen even though the Pt electrode is used, so long as another oxide region exists for oxygen-ion exchange to take place.

In summary, stable CRS is observed in our TiN/ HfO_x /IL/TiN stack. The study shows that the IL between the HfO_x layer and the bottom TiN electrode plays an important role in determining the existence of CRS in the TiN/ HfO_x /IL/TiN RRAM device. It is believed that oxygen-ion exchange between the conductive path and the different ILs causes the position of the partially reoxidized part of the conductive path to change from the top IL to the bottom IL, thus enabling a complementary resistive switching behavior. Replacing the bottom TiN electrode by the inert Pt metal eliminates the bottom IL and also CRS in the TiN/ HfO_x /Pt device. Only the usual positive-set/negative-reset bipolar resistive switching can be observed. The role of the bottom IL is further confirmed by the poor resistive switching behavior of the Pt sample after negative-voltage forming. Interface engineering may thus be explored to achieve high-performance RRAM device with reliable and stable CRS.

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