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Failure and Recovery of Double-Layer CBRAM Studied by *In-Situ* TEM

Masashi Arita[1,*], Ryusuke Ishikawa[1], Katsunori Arima[1], Atsushi Tsurumaki-Fukuchi[1], Yasuo Takahashi[1], Masaki Kudo[2], and Syo Matsumura[2]

1 Graduate School of Information Science and Technology, Hokkaido University, Sapporo 060-0814, Japan
2 The Ultramicroscopy Research Center, Kyushu University, Fukuoka 819-0395, Japan
* Phone: +81-11-706-6457 E-mail: arita@nano.ist.hokudai.ac.jp

Abstract

***Reset* failure and recovery of double-layer CBRAM of Cu/MoO_x/Al_2O_3 was investigated using *in-situ* TEM. With *Set/Reset* (i.e. write/erase) operation, Cu was accumulated at the MoO_x/Al_2O_3 interface, and Cu drift into Al_2O_3 was limited. This limitation is effective to recover the device from *Reset* failure, even when much Cu drifts into MoO_x. It was experimentally confirmed that this is one of the advantages of the double layer structure to achieve stable operations.**

1. Introduction

ReRAM (resistive RAM) device of a solid electrolyte and Cu called CBRAM (conductive bridging RAM) yields ReRAM switching, where the *Set*/*Reset* operation giving the high/low resistance state (HRS/LRS) is caused by formation/rupture of Cu conductive filaments (CFs) [1-3]. For stable use of CBRAM, devices composed of double insulator layers have been widely investigated [4]. Regarding to this type of CBRAM, we performed *in-situ* transmission electron microscopy (TEM) on the MoO_x/Al_2O_3 and discussed the Cu movement and degradation [5]. In this report, recovery from the *Reset* failure state will be under attention, and the role of the Al_2O_3 layer will be discussed.

2. Experimental

The Cu/MoO_x/Al_2O_3/TiN ReRAM film was sputter deposited on SiO_2/Si at room temperature (Fig. 1a). The sample film was processed into needle-shaped devices of which the bottom electrode (BE) is common (Fig. 1b). An example of the *in-situ* TEM sample is shown in Fig. 1c. Clear layer stacking was confirmed. Fig. 2 is the experimental system. A probe was contacted to the Pt cap layer deposited by FIB, and measurements were done by applying voltage to the top electrode (TE).

3. Result and Discussion

For initialization of the device, voltage sweeps (16 cycles) were performed with increasing maximum voltage from 1 to 2.85 V, and the resistance gradually decreased (ca. 50 to 1 MΩ at 0.5V) while there were no clear ReRAM switching. Afterwards, the operation started to appear. Some examples of the *I-V* curves are shown in Fig. 3a. While the *Set* voltage scattered, it was less than the value of the 1st cycle as widely reported. The cyclic endurance graph is presented in Fig. 3b. The on/off ratio was about 10, and it reduced gradually with cycles. Suddenly at the 10th cycle, *Reset* giving the HRS was not realized (10th to 13th cycles). After this *Reset* failure, three *Reset* cycles without applying positive voltage were performed. The on/off ratio recovered to be about 10 in the 14th cycle.

TEM photographs picked up from the recorded video are compared with the *I-V* curves in Figs. 5-7. The 1st cycle is shown in Fig. 5. By the *Set* operation, a dark contrast appeared near the right-bottom edge of the MoO_x layer (broken circle). Cu came from the TE, and accumulated at the MoO_x/Al_2O_3 interface. No clear contrast change in MoO_x was seen in *Reset*. A tiny CF contributing to operation is expected in the Al_2O_3 layer, as schematically shown in Fig. 8a. Until the 9th cycle, relatively stable operation was identified, and there were no drastic change in the TEM images. In the 10th cycle, current reduction was seen at about +2 V. The CF giving stable switching in the 1st to 9th cycles is thought to be broken. Afterwards, a clear *Set* happened at 2.6 V, and *Reset* failure occurred (red curve in Fig. 6a). In the corresponding TEM image, the lower edge of the Cu TE became bright, and the overall region of the MoO_x layer was dark after *Set* (Figs. 6b and 8b). On the other hand, the contrast of the Al_2O_3 layer did not show a drastic change except the central part marked with the triangle. At this position, the slightly dark region (CF) expanded after *Set*, which must be the origin of the *Reset* failure. Afterwards, three *Reset* operations (black curves in Fig. 6a) were performed and the HRS was recovered. The CF in the TEM image became slightly narrower while the Cu in the MoO_x layer did not show clear change (schematically drawn in Fig. 8c). Much Cu diffused into MoO_x remained in subsequent operations, but stable *Set*/*Reset* switching continued (Fig. 7).

4. Summary and Conclusion

Cyclic switching of Cu/MoO_x/Al_2O_3/TiN double layer CBRAM was investigated using *in-situ* TEM. In the *Set* process, Cu drifted from the TE to the MoO_x/Al_2O_3 interface and formed a deposit. A tiny CF contributing memory operation is expected in Al_2O_3. When accidental high power was injected, much amount of Cu was introduced in MoO_x, but it was stopped at the interface. Because of limitation of Cu drift into Al_2O_3, device recovery could be done. This must be caused by difference of degree of Cu solubility and mobility between MoO_x and Al_2O_3. It was experimentally confirmed using *in-situ* TEM that the insertion of the second switching layer like Al_2O_3 prevents needless Cu drift and achieves stable ReRAM operation.

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References

[1] R. Waser *et al.*, *Nature Mater.* **6** (2007) 833.
[2] A. Sawa, *Mater. Today* **11** (2008) 28.
[3] H. Akinaga *et al.*, *Proc. IEEE* **98** (2010) 2237.
[4] D. Kumar *et al.*, *Appl. Phys. Lett.* **110** (2017) 203102.
[5] M. Arita *et al.*, *SSDM.* (2017) 39.

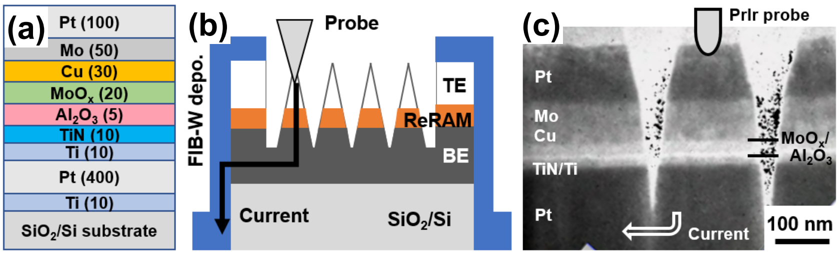

Fig. 1 (a) CBRAM composed of the double switching layers; MoO_x and Al_2O_3. (b, c) FIB processed TEM sample. The cut to divide devices is stopped at the middle of the BE, and the current flows via the FIB deposited thick W layer at the side of the TEM sample. Because of this structure, the parasitic resistance at the BE/substrate interface is removed from the current path.

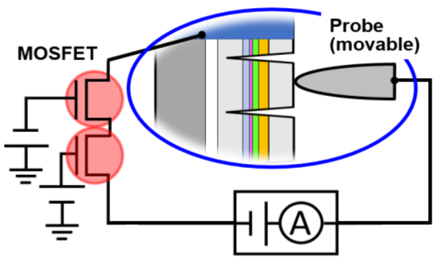

Fig. 2 *In-situ* TEM experimental system. For prevention of strong current overshoot at electroforming and *Set*, MOSFETs are serially connected near the device.

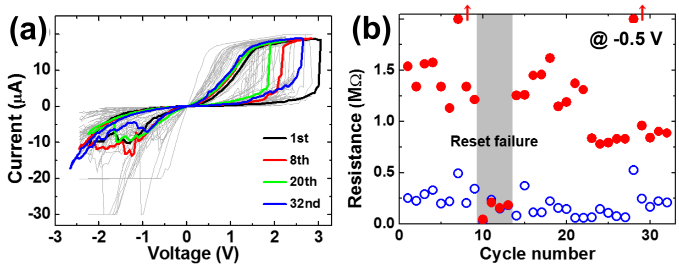

Fig. 3 (a) *I-V* switching curves obtained during *in-situ* TEM experiments. (b) Corresponding cyclic endurance plots where cycles with *Set* failure are omitted. The red filled circles and blue open circles denote HRS and LRS, respectively. Data points with arrows are out of the scale.

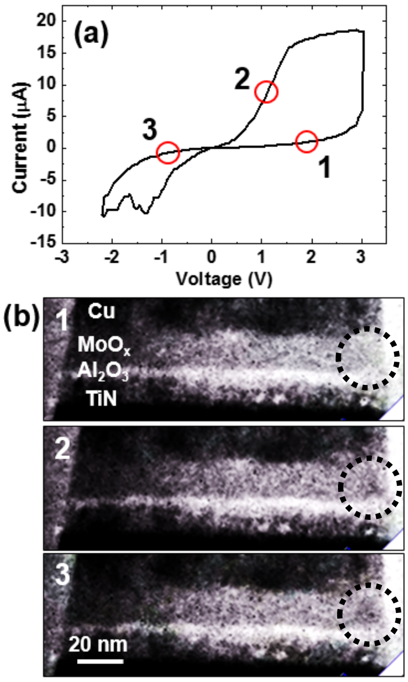

Fig. 5 (a) *I-V* curve of the 1st *Set*/*Reset* cycle. (b) Corresponding TEM images. Contrast change of the CF is identified at the right end of the device (broken circle).

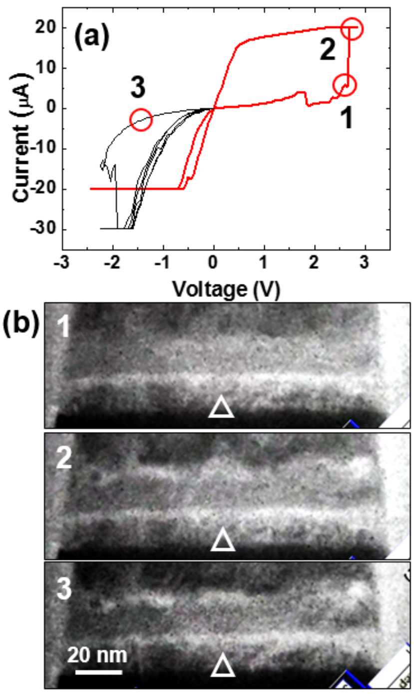

Fig. 6 (a) *I-V* curve of the 10th *Set*/*Reset* cycle and subsequent *Reset* cycles. (b) Corresponding TEM images. Expected filament is marked with the triangle.

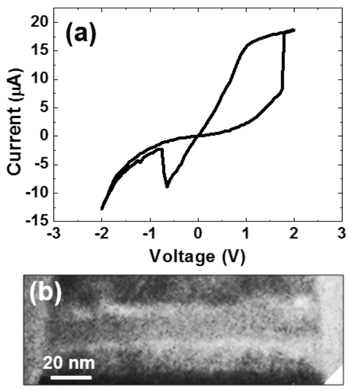

Fig. 7 *I-V* curve and TEM image of the 26th *Set*/*Reset* cycle.

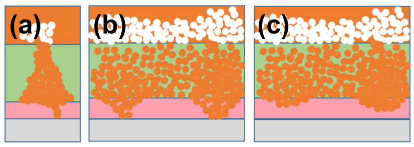

Fig. 8 Schematics of the CF, where Cu (orange), MoO_x (green), Al_2O_3 (pink) and TiN (grey) are stacked. (a) Typical CF with Cu accumulation at the interface. (b, c) Microstructure at *Set* and *Reset* when much Cu drifts in MoO_x. While Cu spreads widely in MoO_x, Cu drift into Al_2O_3 is limited and recovery is possible.