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In-situ Observation of Cu Residuals in Resistance Switching Failure of MoO_x/Al_2O_3 CBRAM

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Abstract

***In-situ* TEM was performed on a double-layered CBRAM. A Cu conductive filament appeared in the switching layer at *Set*. With *Set/Reset* repetition, Cu was accumulated in the switching layer, the resistance decreased, and the on/off switching ratio became small. This is an indication of the endurance failure.**

1. Introduction

Resistive RAM (ReRAM) has a high potential as a next-generation nonvolatile memory and an artificial neural network device [1-3]. The device of a solid electrolyte and Cu called the conductive bridging RAM (CBRAM) yields ReRAM switching, where the *Set*/*Reset* operation giving the high/low resistance state (HRS/LRS) is caused by appearance/disappearance of Cu filaments. To understand this process, *in-situ* transmission electron microscopy (TEM) has been applied [4]. However, the device degradation is still ambiguous. In this work, *in-situ* TEM was performed to understand microstructural evolution in the MoO_x/Al_2O_3 double-layer CBRAM during switching cycles.

2. Experimental

The $Pt/Mo/Cu/MoO_x/Al_2O_3$ ReRAM film was sputter deposited on TiN/Si at room temperature. Afterwards, the focused ion beam method (FIB) was used to fabricate *in-situ* TEM samples. Many needle-shaped CBRAM samples were formed (Fig. 1a), and a clear layer stacking was confirmed (Fig. 1b). Fig. 2 is the experimental system. A probe was contacted to the W cap layer deposited on Pt/Cu, and measurements were done by applying voltage to W.

3. Result and Discussion

A current-voltage (*I-V*) switching curve for *Set* is shown in Fig. 3a. Increasing the voltage, the current in HRS was enhanced gradually and steeply increased at 3.7 V to realize LRS. With reducing voltage, the *I-V* curve was hysteretic. The corresponding TEM image is shown in Fig. 3b where a filament is seen in the square region. The filament appeared in the Al_2O_3 layer was about 20 nm in width (Fig. 3c). The EDX (energy dispersive X-ray spectroscopy) mapping images showed that this filament was of Cu (Fig. 3d-e)

Using another sample, 50 switching cycles were performed in TEM. Three *Set/Reset* curves are compared in Fig. 4a. Though the switching window was narrow, clear hysteretic curves were identified. This narrowness of the window was due to the small HRS resistance caused by the redeposits of electrode materials during FIB. The degree of the hysteretic property became low in the 13th and 24th cycles. This must relate to the endurance failure. In Fig. 4b, the resistance values are summarized. The resistance gradually decreased and the switching window became narrow.

In Fig.5, TEM images of the (a) initial state, (b) after the 3rd *Set* and (c-i) after subsequent *Set/Reset* are compared. At the 3rd *Set*, filament appeared in the Al_2O_3 layer (a triangle in Fig. 5b). Continuing the switching cycles, the center of the Cu layer turns brighter, while the Al_2O_3 became darker (Fig. 5c-h). Cu moved into the MoO_x/Al_2O_3 layer at *Set* was not back to the original region even after *Reset*. As the result, resistance decreased, and endurance failure should occur. To remove Cu from the switching layer, high negative current was required (200 μA in Fig. 5i). The Al_2O_3 layer became brighter indicating removal of Cu. However, this operation induced the "*negative Set*" giving lower resistance and destruction as occasionally occurred in CBRAMs, though details are still unknown.

4. Summary and Conclusion

Cyclic switching of $Cu/MoO_x/Al_2O_3/TiN$ was investigated using *in-situ* TEM. In the *Set* process, Cu filament appeared in the switching layer. It did not disappear in *Reset*, and Cu was accumulated with switching repetition. As the result, the resistance gradually decreased showing an indication of the endurance failure. While high power *Reset* was effective to remove Cu from Al_2O_3, it may destroy the structure. Power balance of *Set* and *Reset* in each cycle must be important for stable switching continuity.

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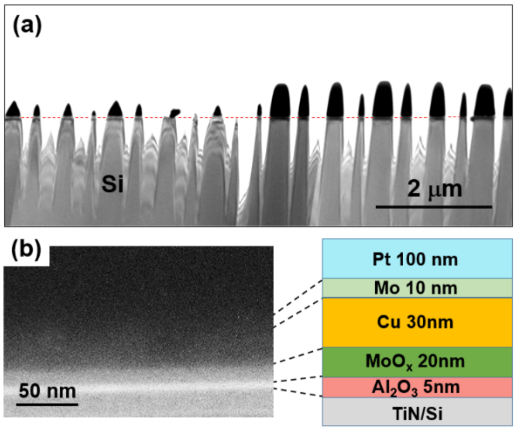

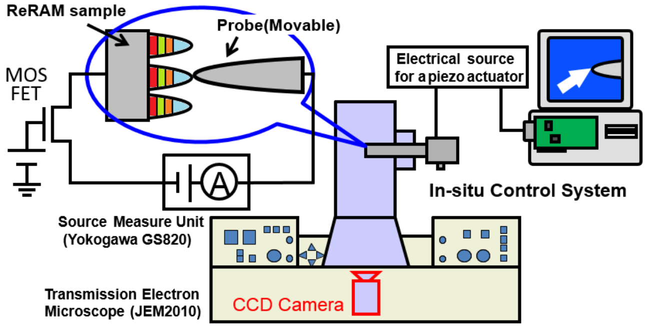

Fig. 2 *In-situ* TEM experimental system.

Fig. 1 (Left) (a) TEM photograph of the *in-situ* TEM sample processed using FIB. In each needle, there is the ReRAM layer at the level of the red broken line. The dark region at the top of each needle corresponds to the W cap layer used for FIB. (b) Enlarged TEM photograph of the area for switching, and layer schematic.

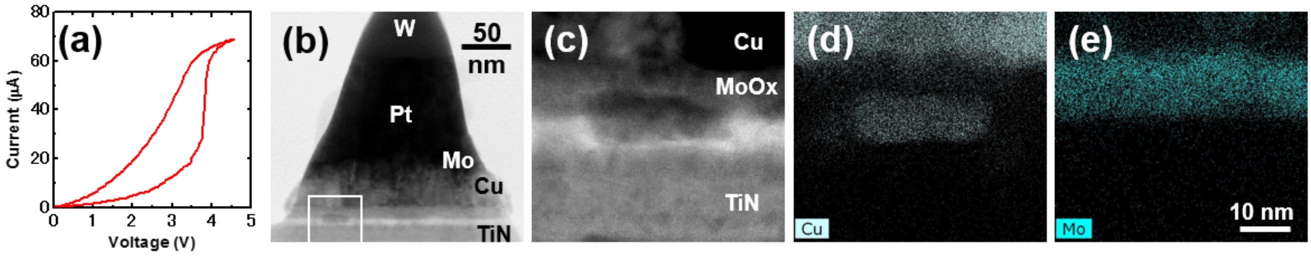

Fig. 3 (a) An example of the *I-V* switching curve in the *Set* process measured in TEM. (b) A TEM image after the *Set* and (c) an enlarged STEM image of the square region in (b). Corresponding EDX mapping images of (d) Cu and (e) Mo taken from a region with a square in (b). Formation of a Cu conductive filament was identified after the *Set* process.

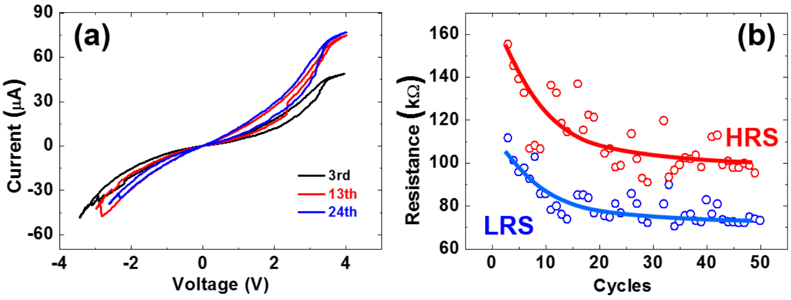

Fig. 4 (a) *I-V* switching curves of the 3rd, 13th and 24th cycles measured during TEM observation. (b) Resistance evaluated at -1 V as a function of the switching cycle. It decreased with the cycle.

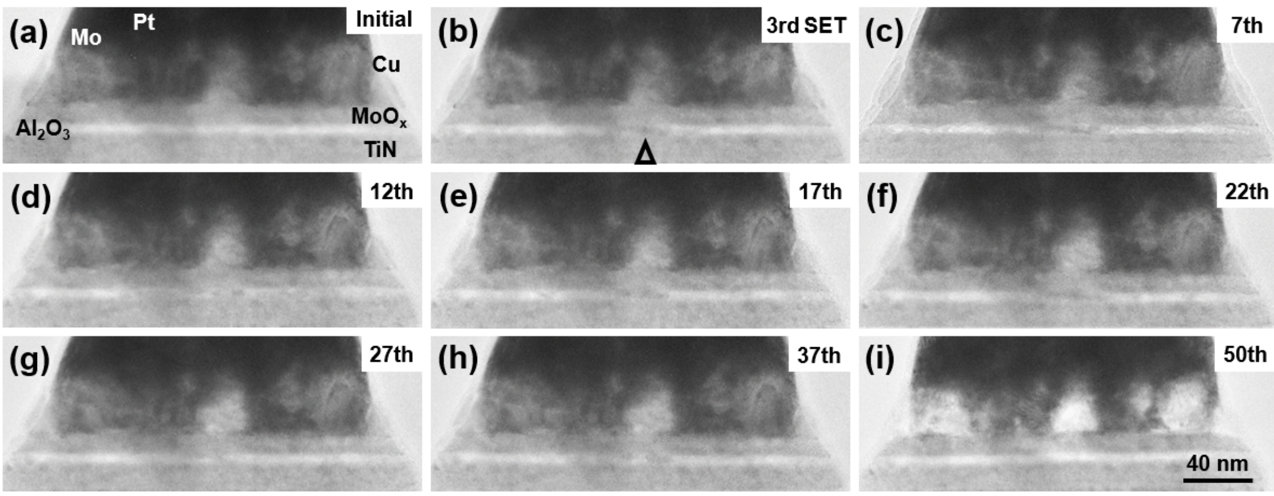

Fig. 5 TEM images (a) in the initial state, (b) after the 3rd *Set* and after *Reset* of the (c) 7th, (d) 12th, (e) 17th, (f) 22nd, (g) 27th, (h) 37th and (i) 50th cycles. At the arrow head in (b), a dark contrast corresponding to the Cu filament appeared in the Al_2O_3 layer.