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Growth and Shrinkage of Conductive Filament in Cu/MoO$_x$ ReRAMs Observed by Means of *In-Situ* TEM

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Abstract

To understand the operation of the ReRAM of Cu and a solid electrolyte, Cu/MoO$_x$ was investigated by means of *in-situ* TEM in sequential switching cycles. Growth and shrinkage of a Cu conductive filament was dynamically observed, but such change was not recognized at the moment of *Set* and *Reset* switching.

1. Introduction

Resistive RAM (ReRAM) shows large resistance change by voltage application and has high potential as a nonvolatile memory [1-4]. The ReRAM of Cu and the solid electrolyte yields such resistance switching. Based on electrochemistry, a conductive filament (CF) is believed to contribute to the switching. However, its mechanism has not been clearly understood yet. By using *in-situ* transmission electron microscopy (TEM) [5, 6], we observed structural changes in Cu/MoO$_x$ during the ReRAM operation.

2. Experimental

Pt/Cu$_{30nm}$/MoO$_{x50nm}$ was on the TiN/Si bottom electrode (BE). It was deposited by conventional or reactive sputtering at RT. The samples were prepared by the ion-shadow method [7] (Figs. 1(a), (b)). The TEM system is shown in Fig. 1(c). A probe was contacted to the Pt/Cu top electrode (TE), and measurements were done by applying voltage to TE. Dynamical images were taken with a CCD camera.

3. Result and Discussion

The TEM sample showed bipolar switching as the conventional ReRAM device (Fig. 2). The resistance became low (or high) with positive (or negative) voltage (*Set* or *Reset*). This indicates that the result described below represents the phenomena in real ReRAMs.

The device initialization is shown in Fig. 3. The resistance decreased without abrupt change. In TEM images, dark contrast near BE advanced to the right. Continuing the I-V cycles, the resistance change became abrupt, and the precipitation was clearly seen (Fig. 4). The round precipitation did not show recognizable change at the *Set* moment, and it grew during subsequent voltage application. No drastic change was seen in the image at the *Reset* moment, and the precipitation became small with further negative voltage. It is thought to act as CF even without connecting TE to BE. After several IV cycles with increasing current compliance, the precipitation (i.e. CF) became large (Fig. 5). It grew towards TE in the *Set* process and connected two electrodes, but no clear image change was seen in *Reset*.

Summarizing these data, the CF grew with positive voltage and shrunk with negative voltage. But the image was not clearly changed at the switching moment. Thus, the ReRAM switching must occur very locally in CF. This result does not deny the *filament model* [1-4]. To erase CF, we needed high negative current (Fig. 6). The CF was erased near BE and went up to TE. Figure 7 shows progress of device destruction in the *Set* process. A precipitation appeared and gushed out of MoO$_x$. At the same time, the Cu TE got lean. Cu dissolved from TE into MoO$_x$ is thought to break the surface. Thus, CF was inside MoO$_x$ and was composed of Cu as recognized in Fig. 8.

By using high compliance current in an almost initial stage, quick CF growth was realized (Fig. 9). At the *Set* moment, a seed of CF appeared and grew towards saliences of TE, where the electric field was concentrated. The CF became fat and bridged between TE and BE within 200 ms.

4. Summary and Conclusion

Evolution of the ReRAM microstructure was dynamically investigated in continuous IV cycles by means of *in-situ* TEM. With positive voltage giving the low resistance state, the CF of Cu grew from BE to TE. With negative voltage giving the high resistance state, the CF shrunk from BE to TE. However, at the switching moment, no large change was recognized in CF. Only the local area of CF contributes to the ReRAM switching. To simulate a realistic ReRAM switching, relatively violent *Set* operation was performed. In this case, CF growth was recognized from BE to the saliences of TE within 200 ms.

Acknowledgements

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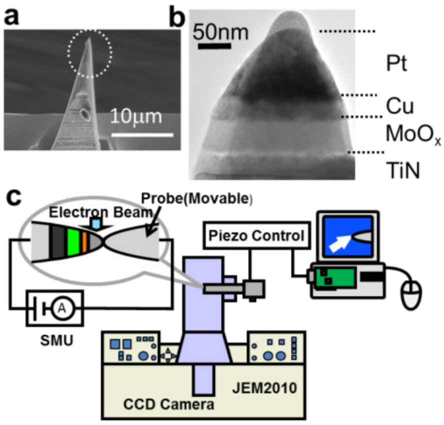

Fig. 1 (a) SEM and (b) TEM images of the ReRAM device investigated. At the top of the needle (circle in a), there is the ReRAM stack. (c) The observation system.

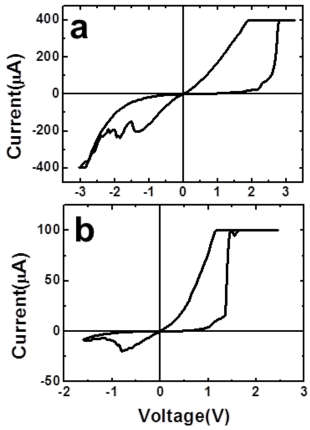

Fig. 2 Pseudo static ReRAM switching of (a) a TEM sample and (b) a conventional device (ϕ16 μm). Both curves showed bipolar switching.

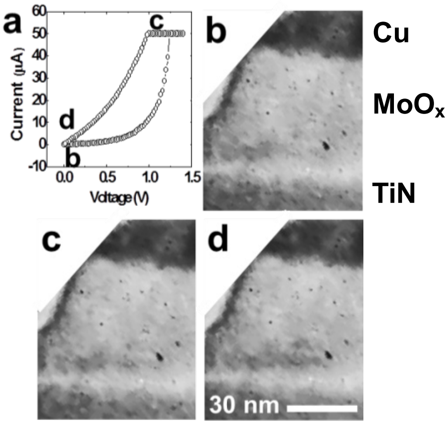

Fig. 3 (a) *Set* curve and (b-d) *in-situ* TEM images almost in the initial state (4th cycle). Evolution of dark contrast is seen near the interface of the bottom electrode.

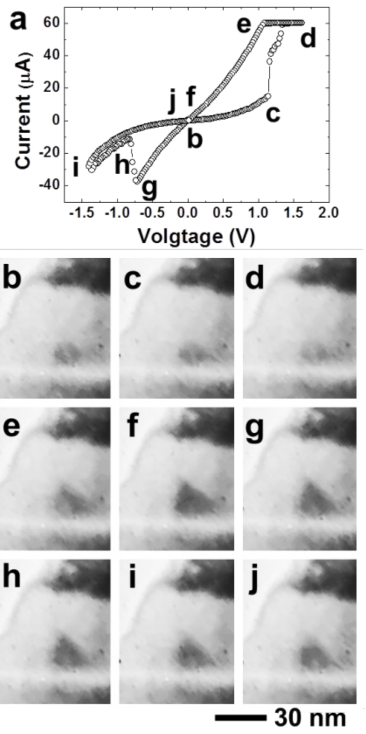

Fig. 4 (a) IV curve and (b-j) TEM images; 2nd cycle after Fig. 3. Growth and shrinkage of the precipitation is seen. No drastic change occurred at the switching moment.

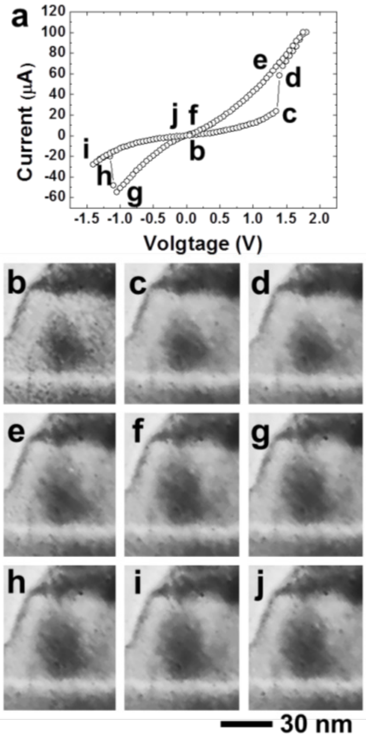

Fig. 5 Data after increasing the compliance current; 6th cycle after Fig. 4. Precipitation became large connecting electrodes. In the *Reset* process, no shrinkage was recognized.

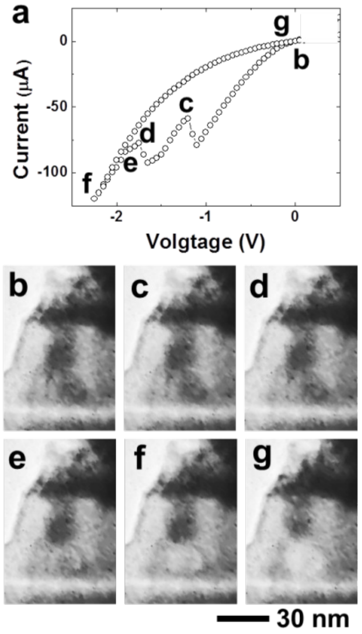

Fig. 6 By increasing the negative bias voltage after *Reset* (c and d), the filament diminished in size; 11th cycle after Fig. 5. The change was seen mainly near the TiN BE. Due to the movement of the filament material, the contrast of the Cu top electrode became slightly darker.

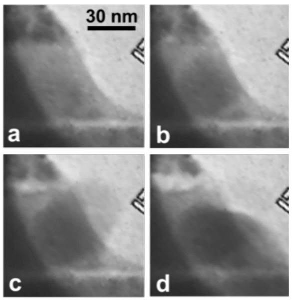

Fig. 7 (a-d) Progress of device destruction with high power injection at *Set*. The filament broke through the surface and grew in the vacuum. The Cu electrode got lean.

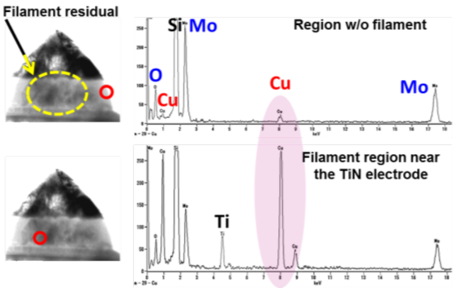

Fig. 8 EDX spectra from regions in and out of the filament. The signals of Si and Ti were from the substrate and the TiN bottom electrode. The filament was composed of Cu.

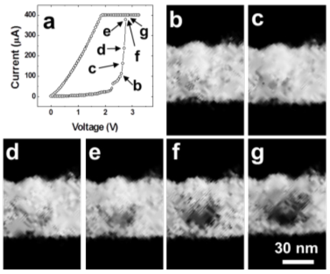

Fig. 9 Quick *Set* operation with high compliance current of 400 μA. Interval between images were 30 ms. The filament grew from BE (TiN) to TE (Cu).