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In-situ TEM Observation on Cu/MoOx Resistive Switching RAM

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ABSTRACT

In-situ transmission electron microscopy (TEM) observation was applied to the resistive RAM (ReRAM) in order to investigate detailed resistive switching mechanism. The ReRAM material studied in this work is MoOx sandwiched between Cu and TiN electrodes. By applying a positive voltage to the Cu electrode, resistance switching from high- resistance state (HRS) to low-resistance state (LRS) occurred, and formation of a precipitation near the bottom electrode was observed. On the other hand, negative voltage induced switching from LRS to HRS, and the precipitation was diminished. A narrow current path through this precipitation seems to contribute the ReRAM switching.

1. INTRODUCTION

Since having high operating speed, simple capacitor structure, and also non-volatility, ReRAM has high potentials as the next generation non-volatile memory. (Sawa 2008) Usually, ReRAM devices are composed of the metal/isolator/metal (MIM) structure, the resistance changes from a high resistance state (HRS) to a low resistance state (LRS) and vice versa by applying a certain voltage to the electrodes. However the mechanism of the resistance switching is not well known yet. In order to clarify this mechanism, we used the in-situ TEM method. It is a powerful method with which nano-structural changes during electrical measurements can be investigated. (Kwon 2010, Fujii 2011, Lui 2012) In this work, we used MoOx as the resistive switching layer and Cu as the top electrode. (Lee 2007) In this type of ReRAM, formation and rupture of a conductive filament inside the MoOx layer by Cu ion migration is expected during a voltage is applied.

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2. EXPERIMENTAL DETAILS

In order to perform TEM observations, the ReRAM samples were prepared by using the ion-shadow method. (Arita 1999, Kudo 2013) It is an ion milling process with using mask particles as schematically shown in Figure 1. In this work, carbon particles with a diameter of 10-30μm were used as the mask material, which has a low milling rate.

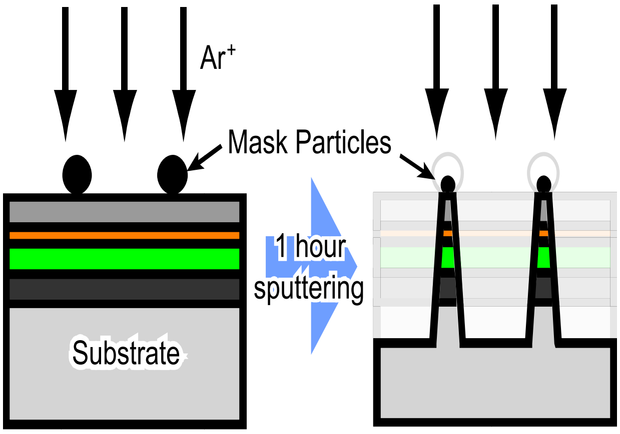

Fig. 1 Schematic image of the ion-shadow method

Placing these particles over the ReRAM film, Ar^+ milling was performed for 1 hour (4kV, 1mA). The ion beam was irradiated perpendicularly to the substrate surface. During the milling of the film and the substrate, the mask particles gradually become smaller, and sharp needles with the ReRAMs are formed. An example is shown in Figure 2.

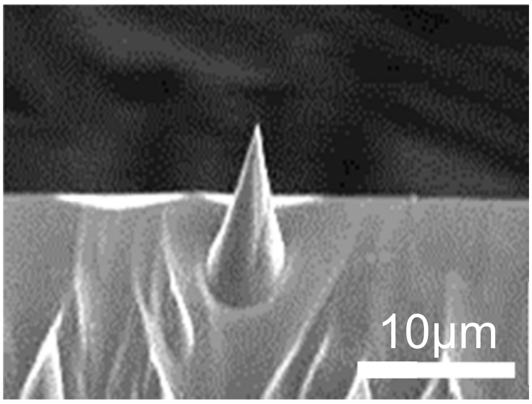

Fig. 2 SEM image of a needle formed upon
a Si substrate by ion-shadow method

Miniaturized ReRAM devices of a few tens to a few hundreds of nanometers (nm) in size were obtained on top of this needle.

For the in-situ TEM observation, the TEM system schematically shown in Figure 3

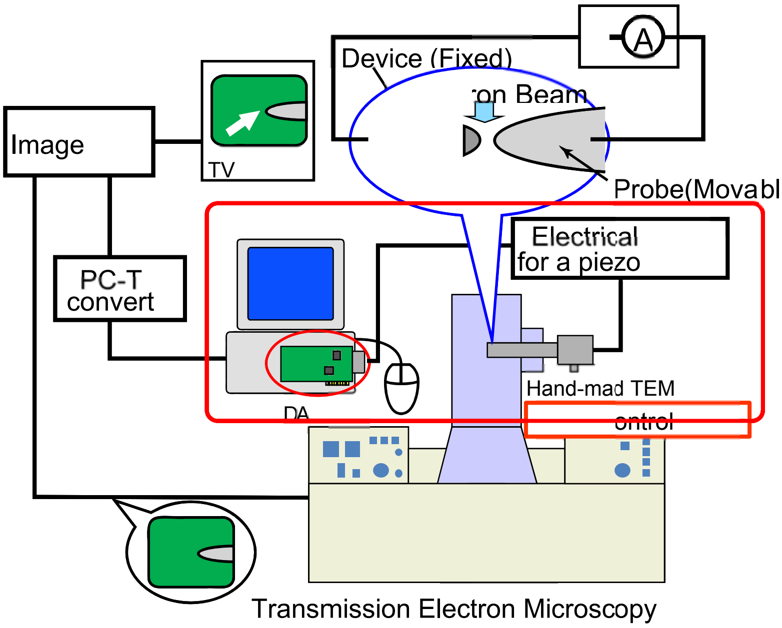

Fig. 3 Schematic image of the in-situ TEM experimental system

was used. The hand-made TEM specimen holder shown in Figure 4 has a piezo actuator which can control a probe electrode at a precision of a few nm or less. By contacting the probe electrode to the ReRAM top electrode (TE), electrical measurements were performed. The current between the TE and the bottom electrode (BE) were measured by using a Yokogawa GS820 source measure unit (SMU). (Figure 5) To reduce the resistance of the substrate, heavily doped silicon wafers was used. Samples used in this work was Pt/Cu/MoOx/TiN deposited by RF sputtering. TiN and Cu were the BE and the TE, respectively, whereas Pt acts as a protective layer to prevent excessive oxidation of the Cu TE. The MoOx insulating layer was prepared by reactive sputtering (Ar-O_2).

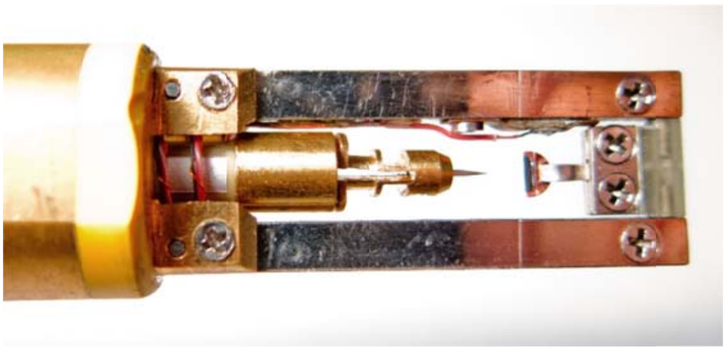

Fig. 4 TEM specimen holder with a piezo actuator

3. RESULTS AND DISCUSSION

Figure 6 shows the TEM image of a Pt/Cu/MoOx/TiN miniaturized ReRAM device in initial state. Before applying the ion-shadow method, each layer had a thickness of 100nm, 30nm, 60nm, 10nm, respectively. However, the thickness of the Pt layer is less than 100nm in this TEM image because its surface was sputtered out during the milling process.

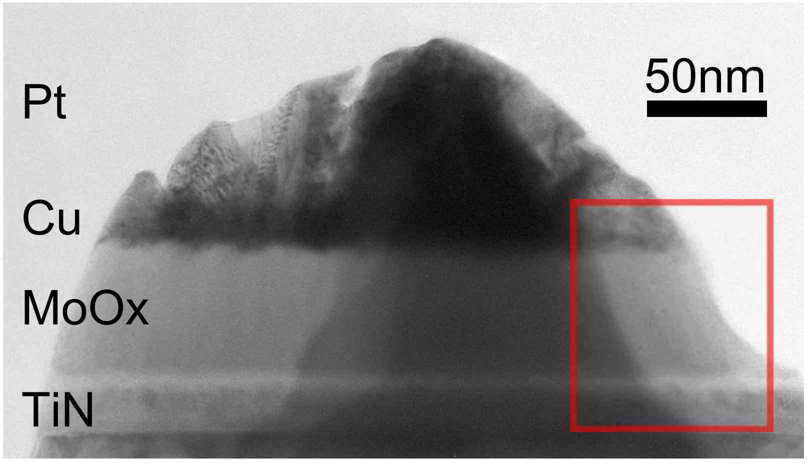

Fig. 6 TEM image of a Pt/Cu/MoOx/TiN miniaturized ReRAM device in initial state. Red boxed area shows the magnified position in Figure 8.

As a similar method to create TEM samples, the focus ion beam (FIB) method is known, but because of the high ion accelerating voltage, surface damage of a few nanometers is usually present. Moreover, using the FIB method on highly insulated in-situ TEM samples is difficult because the Ga^+ ion used has conductivity which creates short paths over the layer. Using the ion-shadow method which uses a much lower acceleration voltage, surface damage layers were rarely observed in ReRAMs, and high initial resistance of the MoOx isolating layer was confirmed.

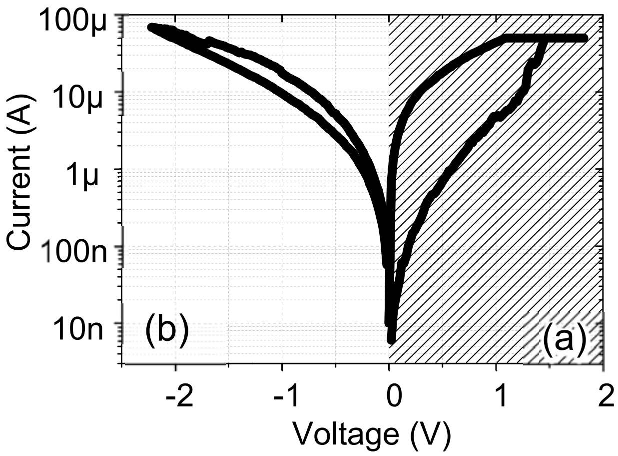

Fig. 7 I-V characteristics obtained during ReRAM operation.

Sweeping the voltage applied to the Cu TE in a positive direction, a sharp resistive switching from HRS to LRS occurred at +1.3 Volts. (Figure 7(a)) To prevent destruction of the device, the current compliance of the SMU was set to 50 µA which can be seen in the current of LRS. During this resistive switching (set process), a precipitation in the MoOx region appeared near the TiN BE. (Figure 8(a)) This precipitation remained even after the applied voltage returned to zero while the resistance also remained in LRS, which shows that this switching has non-volatility.

Subsequently, a voltage sweep in the negative direction was applied and a resistive switching from LRS to HRS (reset process) was observed. (Figure 7(b)) Simultaneously from the TEM image, diminishing of the precipitation was observed. (Figure 8(b)) Here, the precipitation did not completely disappear, and the slight contrast of the precipitation was still observed. In addition, the HRS resistance had not returned to the initial HRS value. This agrees with the fact that the precipitation partly remained after the reset process. Afterwards a few resistive switching cycles was applied to this device, but complete erase of the precipitation was not realized.

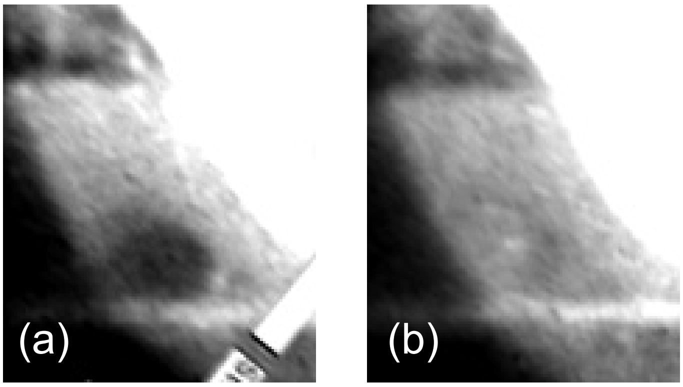

Fig. 8 TEM image after resistive switching of (a) HRS to LRS and (b) LRS to HRS.

From these results, this precipitation seems to be working as a conducing path which grows and diminishes by reversing the voltage polarity applied to the Cu TE. Once a fat conducting path is formed, wiping it out completely to the initial state was difficult.

4. CONCLUSIONS

In-situ TEM observation was applied on a miniaturized Cu/MoOx ReRAM device formed by the ion-shadow method. At resistance switching from high-resistance state (HRS) to low-resistance state (LRS), formation of a precipitation was observed. Subsequently, a voltage of reversed polarity was applied and resistance switching from LRS to HRS occurred while the precipitation diminished. From these results, this precipitation seems to work as a conducting filament in Cu/MoOx ReRAMs.

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