

## Microstructural Investigation of Multi-state Resistive Switching Characteristics in Multi-layered Platinum/Tantalum Oxide using In-situ TEM

Seong-II Kim, Seung-Pyo Hong and Young-Woon Kim

Research Institute of Advanced Materials, Department of Materials Science and Engineering, Seoul National University, Seoul, Korea

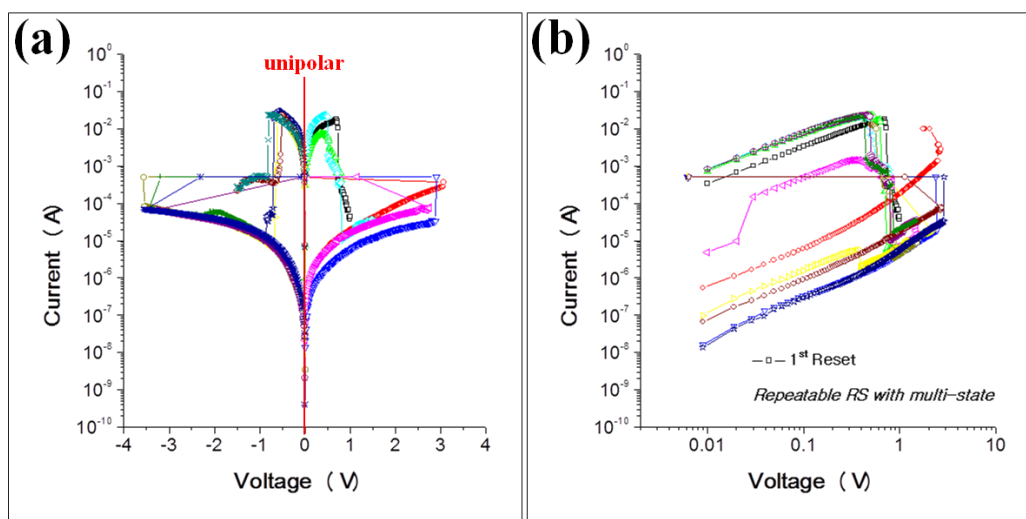
Resistive switching (RS) characteristics of transition metal oxides have been attracted great attention for the next generation non-volatile memory, called resistive random access memory (ReRAM) [1]. Tantalum oxide ( $\text{TaO}_x$ ), one of the most widely studied RS systems, has been reported to show both unipolar and bipolar RS behaviors with multi-state RS characteristics [2-3]. In general, multi-level resistance states is referred when it shows more than 3 distinctive states, for example, low resistance state (LRS), high resistance state (HRS) and states in between them. The bipolar multi-state RS behavior of  $\text{TaO}_x$  has been reported by Hur et al [3]. The unipolar multi-state RS characteristics, on the other hand, are rarely reported. The unipolar multi-state RS characteristics in multi-layered Pt/ $\text{TaO}_x$  will be reported in this presentation. The multi-state RS can provide a simple way to develop multi-bit devices in non-volatile memory storage systems by simple process of forming alternate layers of Pt and  $\text{TaO}_x$ .

A 40-nm-thick  $\text{TaO}_x$  thin film was deposited onto the e-beam-evaporated 200-nm-thick Pt/Ti/SiN/Si substrates by reactive DC magnetron sputtering at room temperature. Subsequently, a 40-nm-thick Pt layer was sputter-deposited onto the former  $\text{TaO}_x$  thin film with same condition above. One more set of  $\text{TaO}_x$  and Pt layers were repeated on the top of the pre-layered Pt/ $\text{TaO}_x$  to make Pt-top-electrode (TE)/ $\text{TaO}_x$ /Pt-middle-electrode (ME)/ $\text{TaO}_x$ /Pt-bottom-electrode (BE) thin film. All depositions were performed in vacuum without exposing to air and retractable patterned metal mask was applied on the substrate during the depositions. By carefully controlling the level of the sweeping voltage, multi-state RS characteristics were obtained from current-voltage (I-V) measurement in the probe station. Microstructural and elemental changes were investigated by using transmission electron microscopy (TEM) equipped with energy dispersive spectroscopy (EDS) and electron energy loss spectroscopy (EELS).

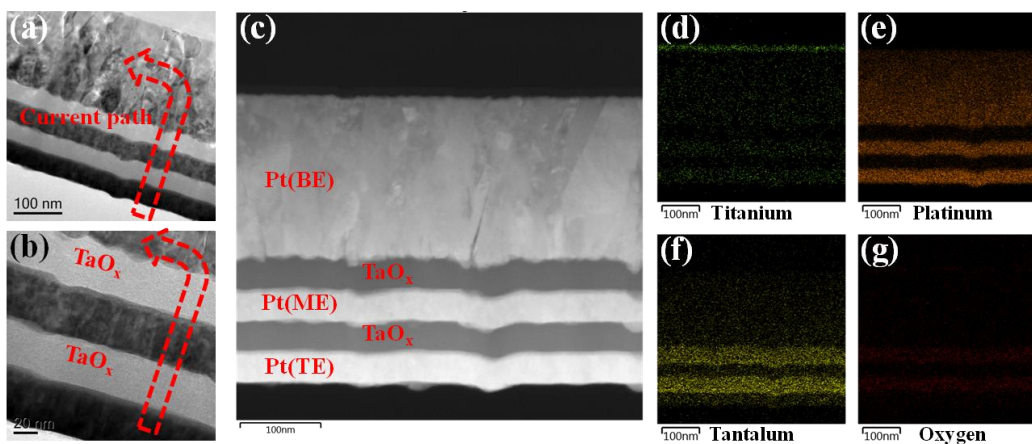
In the typical semi- and double-logarithmic plots of the I-V curve of Pt/ $\text{TaO}_x$ /Pt/ $\text{TaO}_x$ /Pt structure, a pristine state showed LRS in the sample, unlike other ReRAM devices. The electro-forming process, which required initiating the repeatable RS with a compliance current ( $100\mu\text{A}$ ), was not necessary in this performance. Reset and set process occurred with same bias polarity at  $\sim \pm 0.9\text{ V}$  and  $\sim \pm 3.2\text{ V}$ , respectively. With coarse controlling the sweeping voltage, two major resistance states of LRS and HRS were observed, as can be seen in Figure 1, and there showed additional resistance states, which represent resistance ratio of more than 10, were obtained, when the sweeping voltage was manipulated in between voltage. It was believed that RS behavior could be originated from the formation of conducting path through the metal oxide or near the metal/metal oxide interface. Thus, several different conducting paths from each layer or interface in the stacked configuration could explain the multi-state RS. The pristine state of the alternate stacked structure, which was confirmed to be LRS through  $\text{TaO}_x$ , was observed by TEM. According to TEM and EDS investigation in Figure 2, the device was manufactured as planned and amorphous  $\text{TaO}_x$  was formed. Microstructural investigation was made using in-situ probing stage to identify the multi-state RS mechanism. Sequential change of microstructure was recorded and compared, which will be presented in the talk as a movie clip.

## References:

- [1] H. -S. P. Wong *et al*, Proceeding of the IEEE **100** (2012) p. 1951.  
 [2] F. Kurnia *et al*, Phys. Status Solidi RRL **5** (2011) p. 253.  
 [3] J. H. Hur *et al*, Nanotechnology **23** (2012) 225702.  
 [4] This research was supported by the Nano-Material Technology Development Program through the National Research Foundation of Korea funded by the Ministry of Science, ICT & Future Planning (2011-0019984).



**Figure 1.** The semi- and double-logarithmic plots of I-V curve: (a) Unipolar RS behavior of Pt/TaO<sub>x</sub>/Pt/TaO<sub>x</sub>/Pt sample. (b) Multiple resistance states of Pt/TaO<sub>x</sub>/Pt/TaO<sub>x</sub>/Pt sample.



**Figure 2.** (a-b) TEM images of the pristine Pt/TaO<sub>x</sub>/Pt/TaO<sub>x</sub>/Pt sample, current path illustrated during applying the voltage. (c-g) High angle annular dark field (HAADF) image and EDS elemental mapping of (d) titanium, (e) platinum, (f) tantalum, and (g) oxygen.