

# Electronic states in the neutral-beam-formed Ta<sub>2</sub>O<sub>5</sub> film measured by thermally stimulated current method

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## Abstract

In this paper, we form a nanometer-thick Ta<sub>2</sub>O<sub>5</sub> film by room temperature neutral beam oxidation and show a thermally stimulated current (TSC) spectrum obtained from an Au/Ta<sub>2</sub>O<sub>5</sub>/Au structure. After irradiation of a Ta metal film with a neutral oxygen beam, a nm-thick Ta<sub>2</sub>O<sub>5</sub> film was formed. By applying a TSC measurement, the existence of tightly distributed shallow trap levels was confirmed. In addition, fabricated Cu/Ta<sub>2</sub>O<sub>5</sub>/Pt resistive memory (ReRAM) structure showed a high OFF resistance value due to low trap density. These results indicate the effectiveness of neutral beam oxidation for the development of metal oxide-based ReRAM.

## 1. Introduction

Electronic nanodevices such as memory and transistor are key device for future information technology. Among of memories, resistive switching memory (ReRAM) [1] has been proposed as a candidate for future generation non-volatile memory because of unique functions, such as memristive operation [2] and synaptic behavior [3]. Typically, a metal oxide film such as tantalum oxide (Ta<sub>2</sub>O<sub>5</sub>) is used as an ionic transport material in ReRAM structure, and metal oxide-based electrochemical metallization memory as well as valence change memory requires a high quality oxide film with nm thickness to improve device performance.

To form a nm-thick metal oxide film, we have proposed novel metal oxidation process named "Neutral Beam Oxidation" [4]. This technique has advantages for thin film oxidation such as elimination of UV photons, prevention of charge build-up of sample and low-temperature process. By applying this oxidation method, we have demonstrated a resistive switching in a Ta<sub>2</sub>O<sub>5</sub> film [5] and a metal-oxide-semiconductor (MOS) gate operation of the aluminum oxide/germanium oxide structure [6–7].

Metal oxide formed by sputtering method, which is widely used in the ReRAM fabrication, is well known. However, film property of neutral-beam-formed metal oxide is unknown. In this paper, we report the characteristics of Ta<sub>2</sub>O<sub>5</sub> film formed by neutral beam oxidation, analyzed by

the thermally stimulated current (TSC) method [8–9].

## 2. Experimental

Fig. 1 shows a generation system of the neutral oxygen beam. This system consists of an oxygen plasma chamber and a sample chamber for oxidation separated by a silicon (Si) aperture electrode with numerous holes and a high aspect ratio (1 mm in diameter and 10 mm in thickness). In this system, oxygen ions in the plasma can be neutralized by collision with the sidewalls of the Si aperture while passing through the holes, and only neutral oxygen particles arrive at the sample. As a result, the sample can be oxidized by irradiation with a neutral oxygen beam.

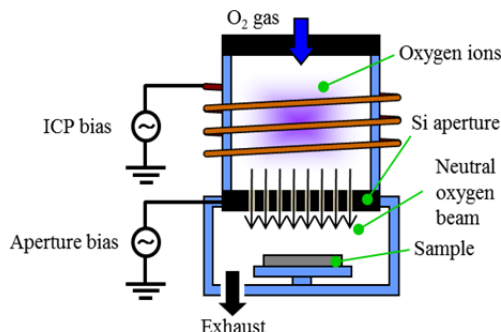


Fig. 1. Neutral beam system.

Fig. 2 shows a sample structure for TSC method. Gold (Au)/Ta<sub>2</sub>O<sub>5</sub>/Au is a current pathway, and this structure was fabricated on a SiO<sub>2</sub>/Si substrate. After deposition of a Ta metal film by conventional sputtering deposition, neutral beam oxidation of Ta was carried out at room temperature. The thickness of Ta<sub>2</sub>O<sub>5</sub> was 2.5 nm. The electronic state of the above-mentioned sample was measured using a TSC measurement system. The temperature range, the temperature ramping rate, and the bias voltage were 100–600 K, 0.15 K/s and 0V, respectively.

ReRAM device of copper (Cu)/Ta<sub>2</sub>O<sub>5</sub>/platinum (Pt) structure was fabricated in this paper. A Pt inert bottom electrode with a thickness of 30 nm was formed on a SiO<sub>2</sub>/Si substrate by sputtering, followed by formation of the Ta<sub>2</sub>O<sub>5</sub>

film. Finally, a 30-nm-thick active Cu electrode was formed by sputter deposition.

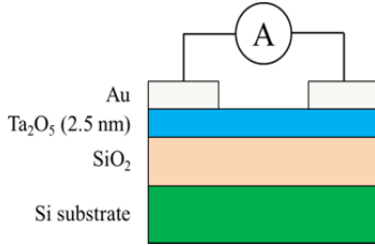


Fig. 2. Sample structure for TSC measurement.

### 3. Results and Discussion

Fig. 3 shows the temperature dependence of TSC in a Ta<sub>2</sub>O<sub>5</sub> film on SiO<sub>2</sub>/Si substrate. In case of Ta<sub>2</sub>O<sub>5</sub> film, three distinguishing peaks (160, 210 and 530 K) were confirmed. The horizontal axis of this figure corresponds to the activation energy of the trapped carrier, calculated using the following equation [10–11]:

$$E = k_B T_m \ln(T_m^4 / \beta)$$

where  $k_B$  is the Boltzmann constant,  $T_m$  is the TSC peak temperature, and  $\beta$  is a temperature ramping rate of 0.15 K/s. Therefore, activation energy of 0.31, 0.42, and 1.23 eV below the conduction band could be obtained from the Ta<sub>2</sub>O<sub>5</sub> sample, and it was confirmed that the shallow trap levels of 0.31 and 0.42 eV exists in the Ta<sub>2</sub>O<sub>5</sub> film formed by neutral beam oxidation at room temperature.

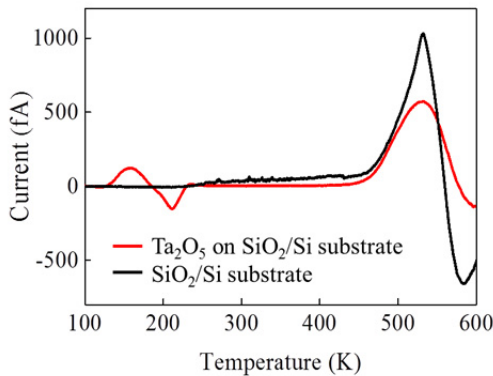


Fig. 3. TSC spectrum of Ta<sub>2</sub>O<sub>5</sub> film.

For reference, a SiO<sub>2</sub>/Si with Au electrodes sample was also prepared. About this sample, the TSC spectrum includes a current flow only through a SiO<sub>2</sub> film. It was confirmed that the temperature dependence of TSC in the SiO<sub>2</sub> film is gentle over a wide temperature range (220–450 K) and 530 K (1.23 eV). As the energy of 1.23 eV is common, the corresponding peak of Ta<sub>2</sub>O<sub>5</sub> sample may be caused by a current through the SiO<sub>2</sub> film. Therefore, it is expected that dominant electronic states in the fabricated Ta<sub>2</sub>O<sub>5</sub> film is shallow trap levels. In addition, the amount of TSC at low temperature was less than 120 fA, meaning that low trap density. This causes high OFF resistance in the ReRAM

switching operation. Fig. 4 indicates a typical switching curve of a fabricated Ta<sub>2</sub>O<sub>5</sub>-based ReRAM structure measured at room temperature. As shown in this result, an OFF resistance of GΩ order was obtained although the thickness of Ta<sub>2</sub>O<sub>5</sub> film is only 2.5 nm.

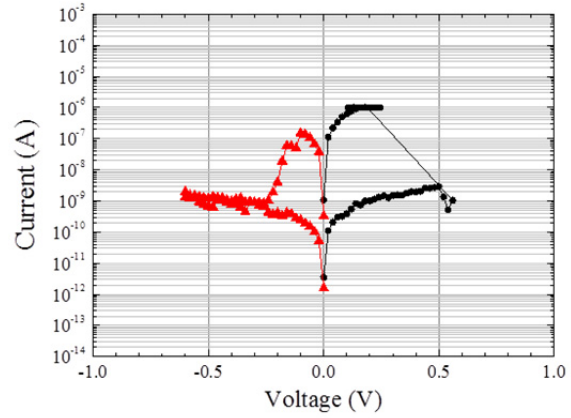


Fig. 4. Switching characteristic of Cu/Ta<sub>2</sub>O<sub>5</sub>/Pt structure.

### 4. Conclusions

We performed a TSC method for the investigation of trap levels in the Ta<sub>2</sub>O<sub>5</sub> film formed by neutral beam oxidation of Ta metal. Electronic states 0.31, 0.42, and 1.23 eV were observed in the 2.5-nm-thick Ta<sub>2</sub>O<sub>5</sub> film, and it was confirmed that the shallow trap levels with low trap density is mainly dominate. This result we obtained shows the great potential of neutral beam oxidation for forming a high-quality metal oxide-based ReRAM with higher OFF resistance.

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### References

- [1] H. Akinaga and H. Shima, *Proceedings of the IEEE* **98** (2010) 2237.
- [2] H. Shima *et al.*, *Appl. Phys. Lett.* **94** (2009) 082905.
- [3] T. Ohno *et al.*, *Nature Mater.* **10** (2011) 591.
- [4] S. Samukawa, *ECS J. Solid State Sci. Technol.* **4** (2015) N5089.
- [5] T. Ohno and S. Samukawa, *Appl. Phys. Lett.* **106** (2015) 173110.
- [6] T. Ohno *et al.*, *Appl. Phys. Lett.* **107** (2015) 133107.
- [7] T. Ohno *et al.*, *Extended Abstracts of the 2015 International Conference on Solid State Devices and Materials* (2015) 1158.
- [8] H. Shima and H. Akinaga, *MRS 2010 Fall Meeting Symposium K* (K8.20).
- [9] Nigo *et al.*, *J. Appl. Phys.* **112** (2012) 033711.
- [10] M. G. Buehler, *Solid-State Electron.* **15** (1972) 69.
- [11] Z. Fang *et al.*, *Mater. Sci. Eng. B* **5** (1990) 397.