Non-Destructive Observation of Chemical State in ReRAM by Laser-excited Photoemission Electron Microscopy

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Abstract

We have demonstrated the nondestructive imaging of chemical states of Resistive Random Access Memory (ReRAM) devices using laser-excited photoemission electron microscopy. High resistance states (HRS) and low resistance states (LRS) in oxide layers beneath 10 nm thick top electrodes were observed from top view. We have succeeded in visualization nucleation and the re-oxidization of conductive filaments in the oxide layers. This new technique will accelerate research and development for various types of electronic devices.

1. Introduction

Resistive Random Access Memory (ReRAM) is one of the leading candidates as next generation non-volatile memory. One of the mechanisms of its resistance switching (RS) is considered as oxidation-reduction reaction at its insulating oxide layer. The resistivity change is caused by migration of oxygen ions. This switching mechanism is microscopic intensively researched by destructive techniques [1], and also observation of the samples with a couple of electrodes arranged in-plane [2]. However, these methods need destructive processes for measurements. Recently, in-situ measurement using transmission electron microscope (TEM) have been proposed to observe dynamic process of filament formation [3]. With this technique, it is impossible to observe the state switching in realistic device structures and also to investigate plural devices at the time.

Here, we report on the non-destructive microscopic observation of the conductive filament path in ReRAM by Laser-excited Photoemission Electron Microscopy (Laser-PEEM) [4] as the first step to achieve *operando* measurements. Laser-PEEM is a powerful tool for nanoscale chemical imaging. Since this technique potentially has large probing depth, we have examined the feasibility of the *operando* measurement with Laser-PEEM. In this study, Ta_2O_5/TaO_x structures were used for demonstration, which was reported as highly reliable ReRAM [5] and both the high resistance state (LRS) were microscopically observed.

2. Experimental

We fabricated Pt (10 nm)-Top Electrode (TE)/Ta₂O₅ (7 nm)/TaO_x (30 nm) /Pt (50 nm)-Bottom Electrode (BE) on thermally oxidized Si substrates by RF magnetron sputtering (Fig. 1). TE diameter is 50 µm. After the formation of TE, the sample was annealed at 300°C in air for 1800 sec. We used Laser-PEEM for the real space observation. The photon energy of laser is 4.66 eV, which was considered to be enough probing depth to detect the conductive filament path because of the threshold photoelectron emission [6]. We used the continuous wave laser to avoid a space charge effect and the best spatial resolution is 2.6 nm [4]. Prior to the Laser-PEEM observation, Reset (RS from LRS to HRS) and Set (RS from HRS to LRS) operations were conducted for several devices in order to compare the Laser-PEEM images for the asfabricated state, LRS, and HRS.

3. I-V Characteristic

Shown in Fig. 2 is *I-V* characteristic of the Pt/Ta₂O₅/TaO_x/Pt. In this figure, *I-V* curves for the Forming, Reset, and Set processes are plotted. Forming and Set current are controlled not to exceed 10 mA. The Forming, Set and Reset voltages were -3.8 V, -0.5 V and +1.0 V, respectively. Resistance ratio $R_{\rm HRS} / R_{\rm LRS}$ was about 5 at 0.2 V. In Fig. 2, the current for the 1st Reset operation was larger than that for 2nd Reset operation. This is probably due to the charges accumulated in the crystals when the conductive filaments are created in the Forming operation [7].

4. Laser-PEEM Imaging

The Laser-PEEM images of $Pt/Ta_2O_5/TaO_x/Pt$ from perpendicular to the surface were shown in Fig. 3. Figure 3(a) represents the image of the as-fabricated sample. The photoelectrons were detected from both the area with Pt TE and the exposed Ta_2O_5 layer. Fig. 3(b) shows the device in HRS. We observed a circular and darker area in the Pt TE where the intensity of photoelectrons is lower. This area is considered to be caused by ruptured filament path. All the devices seen in Figs. 3(c)-(e) were observed at the devices in LRS. The electrode in Fig. 3(c) was bright overall, whereas the electrodes in Fig. 3(d) and (e) were partly bright. These bright areas should indicate the existence of conducting filaments, which is created in the Forming processes.

To discuss the results of Laser-PEEM observations, we considered a band diagram of the $Pt/Ta_2O_5/TaO_x$ structures indicated in Fig. 4. As shown by the PEEM images in Fig. 3, the work function of the $Pt/Ta_2O_5/TaO_x$ was lower than laser energy (4.66 eV). However, photo-excitation was not occurred from the Ta_2O_5 layers because the photon energy of the laser was not enough to excite photoelectrons from the valence band electrons of the Ta_2O_5 (Forming and Set operations), the conductive Ta-rich filaments were clearly visible as shown in Figs. 3(c)-(e). As a result, the clear difference between the HRS and the LRS were observed by Laser-PEEM.

5. Conclusions

We have adopted Laser-PEEM technique to visualize the conductive filaments in the $Pt/Ta_2O_5/TaO_x/Pt$ structures. We successfully observed a clear difference between HRS and LRS beneath 10 nm thick top electrodes without any destructive processing. The difference is attributed to the change of the redox state which brings about the creation of conductive filament paths.

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Fig. 1 Cross-section of the fabricated $Pt/Ta_2O_5/TaO_x/Pt$ structure



Fig. 2 I-V Characteristic of Pt/Ta₂O₅/TaO_x/Pt structure



10 um









Fig. 4 Band diagram of Pt/Ta₂O₅/TaO_x structure.