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Operando observation of resistive switching in ReRAM by laser-excited photoemission electron microscope

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Abstract

We have developed Laser-excited PhotoEmission Electron Microscope (Laser-PEEM) for non-destructive *operando* observation of electronic devices. By utilizing Lase-PEEM, the non-volatile resistance change in resistive random access memory (ReRAM) was clearly visualized.

1. Introduction

Resistive random access memory (ReRAM) is a device recording data as the resistance that has the simple structure consisting of an oxide layer with two electrodes at the both ends. It is considered that the resistance change is caused by formation and annihilation of the conductive path generated by oxygen vacancies. The formation occurs by the redox reaction generated when a voltage is applied between the electrodes. However, it is known that there is a bottleneck for ReRAM developments to improve its stability and performance because it is difficult to observe oxygen vacancy paths directly.

From the point of view of ReRAM developments, non-destructive imaging methods are indispensable to compare changes in chemical states before and after operations. Previously Kumar *et al*. adopted X-ray microscopy technique to ReRAM device [1]. However, they had to repeat operations to enhance the contrast by degrading the sample. TEM technique was successfully used to observe conductive path of metal ions [2, 3]. However, this method cannot be applied for actual device structures.

Laser-PEEM can visualize chemical states attributed from density of states near the Fermi level as contrast [4]. This method enables us to detect changes in density of oxygen vacancies very sensitively as well as metal ions, consequently shows distinct difference between metallic and insulating areas. Owing to the UV laser excitation, the Laser-PEEM has the appropriate probing depth, which allows us to investigate buried nanostructures in ReRAM devices. Recently we visualized that the change of chemical state in Ta_2O_5-ReRAM [5]. Without any destructive process, the Laser PEEM revealed the change occurring with accompanying the resistance change. However, the operand observation of the resistance transition has not yet fully demonstrated because of the huge drift and distortion of the PEEM image due to the voltage application. In this contribution, we demonstrate the stable Lase-PEEM observation of ReRAM operations. Newly developed system to apply a pulse-voltage to the ReRAM device suppressed the drift and the distortion of the PEEM images.

2. Experimental

The ReRAM stacking structure, Pt-top electrode (10 nm) / Ta_2O_5 (5 nm) / TiN-bottom electrode (20 nm), was deposited on a Si substrate by the method of RF sputtering in crossbar shape. This sample was loaded into the PEEM chamber with the electrical connections to a source measure unit (SMU). The SMU was utilized to apply pulse voltages between the Pt electrode and the TiN-bottom electrode grounded. Laser-PEEM measurements were performed from the sample normal as shown in Fig. 1. The image distortion was avoided by shifting the timing of applied pulse voltage and image acquisition. We have calculated the difference in photoelectron intensity between each image and the initial image. This allows us to elucidate information on their contrast changes for each step.

3. *I-V* characteristic

Fig. 2 shows a typical example of the current-voltage (*I-V*) curve for the RESET (resistance increase) process generated by the negative voltage. This *I-V* curve was obtained with 1 ms pulses applied ranging from 0 V to 7 V. It shows the resistance change from Point [A]: 75 kΩ to Point [G]: 184 kΩ during the RESET process. This behavior was observed regardless of whether pulsed-voltage or DC voltage.

4. Laser-PEEM imaging

Figs. 3 (a) and (b) show PEEM images acquired in Point [A] and [G] of Fig. 2, respectively. Fig. 3(c) is the difference in photoemission intensity between the two images before and after the RESET process, respectively. We have found the clear decrease in photoelectron intensity (blue color spots) in two areas at the corners of device. As shown in this result, *operando* measurements can visualize such a very small change. Since the PEEM images were acquired between the pulse voltages, the image was clear and stable without the drift and distortion. Accordingly, we obtained the fine changes in acquired images for each voltage step.

Fig. 4 (a) shows the PEEM image corresponding to Point

[A], and Figs. 4(b) - (g) are differential images comparing Point [B] to [G] with Point [A], respectively. These results clearly show that the images of the entire process of resistance change have been successfully acquired without any image distortion and drift. As shown in Figs. 4 (b), (c), and (d), the transitions to high resistive state occurs during Points [B], [C], and [D]. These images clearly indicate the decrease in intensity at the right-bottom corner of the device (in 3 o'clock direction in the field of view). It should be noted that the faint but clear blue area is evidently observed in Point [C]. The change is considered to be reflected to the sudden current decrease just after Point [B], shown in Fig. 2. Even after the reset process, the other blue area appeared at the lower left part of the device (in 7 o'clock direction in the field of view). The decrease in the photoelectron intensity is also considered to be related to the sudden current decrease at Point [E]. These behaviors indicate the multiple reset processes in the device. The further investigation, such as the pulse width dependence, is certainly required. The relatively long and complex electric connection to the device should be carefully taken into account in the analysis.

The change in the PEEM image corresponds one-to-one with the change in the *I-V* curve. The operando technique we have developed is expected to be a strong tool for characterizations to understand the relationship between the electric behavior and microscopic changes in chemical states. This technique can also be applied to a variety of devices such as NAND-flash, PCM, and MRAM, because Laser-PEEM can get various information not only on chemical states but also magnetism inside the device.

5. Conclusions

We have developed the Laser-PEEM microscope with the pulse-voltage application system for *operando* observation. The microscope enabled us to visualize the change of the chemical states in ReRAM followed by the electrical change in real time. This non-destructive observation method makes it possible to observe the fine changes in the device, which could not be observed by the conventional microscopies and will give us the understandings of the operation mechanism as well as the improvement of device operations.

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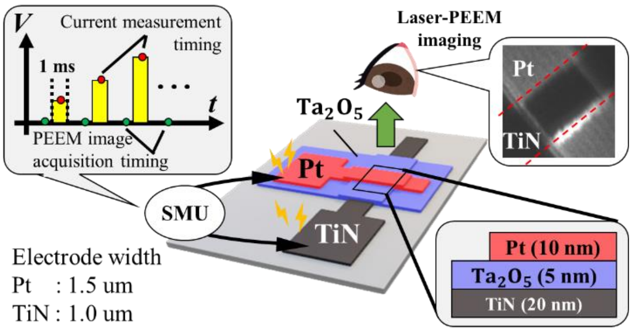

Fig. 1 Sample structure and observation geometry. An image is acquired without effect of voltage because timing of voltage application and image acquisition is different.

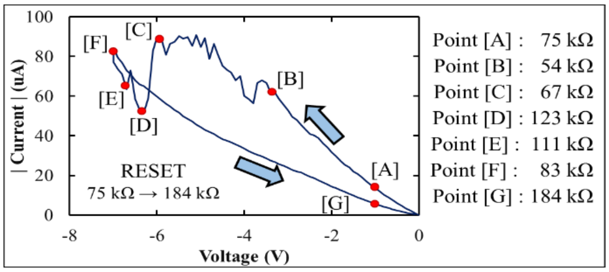

Fig. 2 *I-V* curve for the RESET process in the Pt/Ta_2O_5/TiN device.

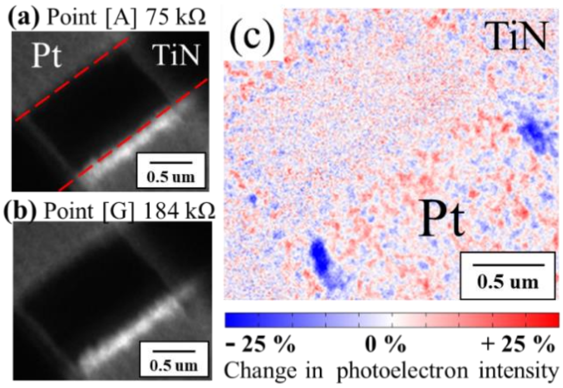

Fig. 3 (a) and (b) Laser-PEEM images of Point [A] and [G] (c) Differential image of photoelectron intensity between Point [A] and [G]. Blue and red show the decrease and increase in photoelectron intensity, respectively.

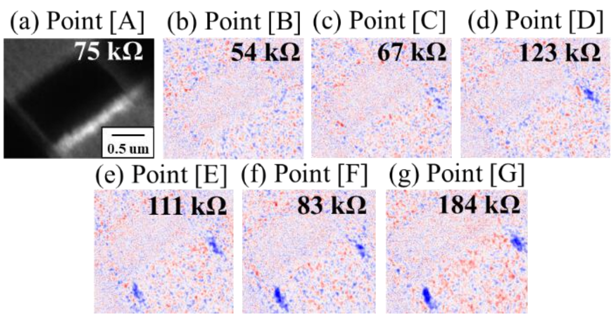

Fig. 4 (a) Laser-PEEM images of Point [A], (b)~(g) Differential images between Point [A] and each Point [B] ~ [G].