

Dependence of reactive metal layer on resistive switching in a bi-layer structure Ta/HfO_x filament type resistive random access memory

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The dependence of reactive metal layer on resistive switching characteristics is investigated in a bi-layer structural Ta/HfO_x filament type resistive random access memory (ReRAM). By increasing the oxygen absorption rate of the reactive metal layer, formation of an induced resistive switching region that led to significant changes in the resistive switching characteristics of the ReRAM was observed. Electrical and physical analyses showed that the induced TaO_x-resistive switching region can result in self-compliance behavior, uniform resistive switching, and a gradual set process, which can be utilized for low power and analog operations. © 2014 AIP Publishing LLC.

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Resistive random access memory (ReRAM) is considered to be one of the most promising emerging non-volatile memory technologies, a group which also includes phasechange random access memory, magnetoresistive random access memory, and ferroelectric random access memory. The proposed emerging non-volatile memories, which allow information storage without employing electron charging behavior, can overcome the limitations of current electron storage based memories. Among these proposed emerging non-volatile memory technologies, recently, ReRAM has been intensively studied because of its advantages, which include a simple structure, low-power operation, and its feasibility for 3D structural high density applications.¹⁻³ However, switching variability is still one of the main obstacles to its commercialization.⁴ Therefore, a bi-layer structure comprising a reactive metal layer and an active layer was proposed as a simple and effective method for improving the switching variability.^{5,6} During deposition of the reactive metal layer, oxygen absorption can occur from the active layer to the reactive metal layer. On the basis of the chemical reactivity of the reactive metal layer, the oxygen absorption that generates the oxygen vacancy in the active layer can be sensitively controlled by modifying the reactive metal layer. Thus, the resistive switching characteristics of a bi-layer structure ReRAM can be accurately controlled by optimizing the reactive metal layer. In this study, to demonstrate the influence of the reactive metal layer, we investigated the dependence of the reactive metal layer on the resistive switching characteristics (the reactive metal dependence) of ReRAM.

 HfO_x based bi-layer ReRAM was fabricated to examine its reactive metal dependence, as shown in Fig. 1. To fabricate the HfO_x based ReRAM, into a 250 nm diameter hole patterned substrate, an 11 nm thick-Ta layer was sputtered as the reactive metal layer onto a 4 nm thick HfO_2 layer which was deposited by an atomic layer deposition system. Next, a Pt layer was deposited onto the Ta layer as a capping layer. Fig. 1(a) shows the cross-sectional transmission electron microscopy image of fabricated sample. The reactive metal dependence was investigated for various thicknesses of the Ta layer (11, 19, and 27 nm). This HfO_x based ReRAM exhibited typical filament switching, as shown in Fig. 1(b). Under the first positive sweep, the forming process which is the first formation of the conducting path (conducting filament: CF) occurred. Subsequently, the reset process that is the resistance change from a low resistance state (LRS) to a high resistance state (HRS) was conducted under a negative bias. After the first cycle (forming process and reset process), the device exhibited consistent resistive switchings. The electrical characterizations and physical analyses were conducted by a semiconductor parameter analyzer (Agilent B1500) and X-ray photoelectron spectroscopy (XPS) depth profiling, respectively.

Fig. 2(a) shows the initial currents of the various samples, including 11 nm thick-Ta/HfO_x (11 nm sample), 19 nm thick-Ta/HfO_x (19 nm sample), and 27 nm thick-Ta/HfO_x (27 nm sample). As the thickness of Ta layer increased, the initial current also increased, which means that a thicker reactive metal layer can absorb more oxygen from the active layer (HfO_x). In other words, during depositing Ta layer, the oxygen absorption generating oxygen vacancies in the HfO_x layer can be occurred from HfO_x layer to Ta layer. Considering that the induced oxygen vacancies can assist leakage current (initial current) of the HfO_x, the higher leakage current can be considered as the higher number of oxygen vacancies in the HfO_x layer. Fig. 2(b) shows a statistical analysis of the initial resistance derived from more than 15 samples of each thickness. This statistical analysis strongly supports the experimental result showing that a thicker Ta layer results in a higher oxygen absorption.

Among the thicknesses examined (11, 19, and 27 nm), the 27 nm sample exhibited significantly different switching characteristics, as shown in Fig. 3. The first resistive change

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FIG. 1. (a) Cross-sectional transmission electron microscopy image of the fabricated 11 nm thick-Ta/4 nm thick-HfO_x bi-layer structure ReRAM. (b) Current versus voltage (I-V) characteristics of an 11 nm thick-Ta/4 nm thick-HfO_x bi-layer structure ReRAM. The device exhibited typical filament switching (indicated with the numbered blue arrows), including (#1) a forming process, (#2) a set process, and (#3) a reset process. The inset is a simplified illustration of the fabricated sample.

of the 27 nm sample was a reset process under a positive bias (#1), as shown in Fig. 3(a). Subsequently, the device exhibited an additional reset process under a negative bias (#2). After the second resistive change, the device exhibited consistent resistive switching, including a positive set process (#3) and a negative reset process. Fig. 3(b) shows a comparison of the current versus voltage (I-V) characteristics of the 11 nm sample and the 27 nm sample. The 27 nm sample exhibited several different resistive switching characteristics such as a gradual set process, uniform resistive switching, self-compliance behavior, and a smoother LRS.

On the basis of the results shown in Fig. 4, it can be seen that these changes result from the higher oxygen absorption of the Ta layer. The complementary resistive switching (CRS) shown in Fig. 4(a) strongly indicates the existence of two resistive switching regions.^{7,8} When two filament type ReRAMs are serially connected in opposite directions, the connected device (two ReRAMs) exhibits CRS behavior because a set process in one ReRAM occurs, while a reset process in the other ReRAM occurs in the same bias region. In other words, CRS behavior can be considered as strong evidence for the existence of two resistive switching regions (two oxide layers). In addition, the results shown in



FIG. 2. (a) Comparison of the initial currents of the 11, 19, and 27 nm samples. As the thickness of the Ta layer increased, the initial current increased. It indicates that the relative amount of oxygen vacancy also increased. (b) Statistical analysis of the initial resistance. The 27 nm thick sample exhibited the smallest initial resistance.

Figs. 4(b) and 4(c) firmly support the existence of two oxide layers. Figs. 4(b) and 4(c) show XPS depth profiling of the as-deposited 27 nm sample for (b) the whole sample region and (c) the interface region between the Ta and HfO_x layers. The result presented in Fig. 4(c) clearly shows the oxygen absorption of the Ta layer, which can form an additional oxide layer (induced TaO_x layer that can play a role of an internal resistor or 2nd switching layer). Thus, the induced TaO_x layer can affect the resistive switching characteristics of the 27 nm sample.

On the basis of these results, simple switching mechanisms of the 27 nm sample are illustrated in Fig. 5. The 27 nm thick Ta layer can absorb a high amount of oxygen from the HfO_x layer during deposition of the Ta layer. Therefore, a high amount of oxygen vacancies are generated in the HfO_x layer, as shown in Fig. 5(a). The initially ohmic characteristic of the 27 nm sample can be explained by the initially generated substantial amount of oxygen vacancy. Under the first positive bias, the mobile oxygen of the HfO_x layer can move to the induced TaO_x layer, then fills oxygen vacancies of the TaO_x layer (the first reset process under the 1st positive bias), as shown in Fig. 5(b). As a result, the TaO_x layer can become an internal resistor that leads to the gradual set process, uniform resistive switching, self-compliance behavior, and a smoother LRS of the 27 nm sample. However, under the first negative bias, the mobile oxygen of the TaO_x layer move to the HfO_x layer, then cause the set process of the TaO_x layer



FIG. 3. (a) The first and second sweep of the 27 nm thick sample. As indicated by the blue arrows, the sample exhibited (#1) the first reset process when a positive bias was first applied. Then, under a negative bias, the sample exhibited (#2) a second reset process. After the second reset process, the sample exhibited (#3) consistent resistive switching including a set process under a positive bias and a reset process under a negative bias. (b) A comparison of the I-V characteristics of the 11 nm sample and the 27 nm sample. The 27 nm sample exhibited several different resistive switching characteristics such as a gradual set process, uniform resistive switching, self-compliance behavior, and a smoother LRS.



FIG. 4. (a) I-V characteristics of the 27 nm-thick sample. Under the first applied negative bias, the sample exhibited CRS behavior, which is strong evidence for the existence of two resistive switching layers. (b) XPS depth profiling of the as-deposited 27 nm thick sample for (a) the whole region and (b) the interface between the Ta layer and the HfO_x layer. At the interface, formation of TaO_x which can act as an additional resistive switching layer was clearly observed.



FIG. 5. Simplified illustration of resistive switching in the 27 nm thick sample (a) at its initial state. A large number of oxygen vacancies can be generated by the high oxygen absorption of the thick Ta layer. (b) Under the first positive bias, mobile oxygen can move to the TaO_x layer, then fill the oxygen vacancies, which can make the TaO_x layer act as an internal resistor. Thus, under the first positive bias, the sample exhibited the first reset process. (c) Under the first negative bias, the TaO_x layer can act as a resistive switching layer.

in the negative bias region. At the same time, the reset process of the HfO_x layer occurs due to injection of the oxygens from the TaO_x layer. Consequently, the sample exhibits CRS behavior when the first negative bias is applied.

In summary, bi-layer structural HfO_x based filament type ReRAMs were investigated with various thicknesses of the reactive metal (Ta) layer in order to understand the dependence of the Ta layer on the resistive switching characteristics. The oxygen absorption rate of the Ta layer increased as the thickness of the Ta layer increased. Consequently, the TaO_x layer was formed by the oxygen absorbed from the HfO_x layer. The induced TaO_x layer led to a gradual set process, uniform resistive switching, self-compliance behavior, and a smoother LRS. These induced switching characteristics can facilitate low power switching and analog operations. Thus, not only low power operation that can be carried out by utilizing a TaO_x layer as an internal resistor but also analog operations that can be adapted to neuromorphic applications are possible.^{2,9}

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