Stability-Enhanced Resistive Random-Access Memory via Stacked \( \text{In}_x\text{Ga}_{1-x}\text{O} \) by the RF Sputtering Method

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**ABSTRACT:** The stability of a resistive random-access memory (RRAM) device over long-term use has been widely acknowledged as a pertinent concern. For investigating the stability of RRAM devices, a stacked \( \text{In}_x\text{Ga}_{1-x}\text{O} \) structure is designed as its switching layer in this study. Each stacked structure in the switching layer, formed via sputtering, consists of varying contents of gallium, which is a suppressor of oxygen vacancies; thus, the oxygen vacancies are well controlled in each layer. When a stacked structure with layers of different contents is formed, the original gradients of concentration of oxygen vacancies and mobility influence the set and reset processes. With the stacked structure, an average set voltage of 0.76 V, an average reset voltage of \(-0.66\) V, a coefficient of variation of set voltage of 0.34, and a coefficient of variation of reset voltage of 0.18 are obtained. Additionally, under DC sweeps, the stacked RRAM demonstrates a high operating life of more than 4000 cycles. In conclusion, the performance and stability of the RRAM are enhanced herein by adjusting the concentration of oxygen vacancies via different compositions of elements.

**INTRODUCTION**

Recently, resistive random-access memory (RRAM) devices have garnered significant interest owing to their advantages, such as a high operation speed, a high integration density, and a simple metal–insulator–metal or metal–semiconductor–metal structure, compared to other emerging memory devices. Various materials, such as HfO\(_2\), TiO\(_2\), ZrO\(_2\), and NiO, have reportedly been used as switching layers. However, the lifetime of a RRAM device has been a critical issue. Oxygen vacancies (VO) play a crucial role in oxide-based resistive switching RAM. Conductive filaments of RRAM devices can be formed by the redistribution of oxygen vacancies in response to the applied electric field. However, the arrangement of oxygen vacancies in an oxide-based semiconductor is highly variable and hard to control and is related to its chemical composition and physical properties. As a result, the performance and stability of the resistive switching behavior is influenced by the oxygen vacancies.

Gallium oxide (Ga\(_2\)O\(_3\)) has been widely used in the academia owing to its advantages such as an ultrawide bandgap of 4.8 eV, high thermal stability, and capability for mass production. Research on InGaO (IGO) as a switching layer in RRAM devices is still in its infancy, and our previous work has indicated that IGO is a good candidate for use in RRAM devices that exhibits the best performance with an Al top electrode (TE). In this work, IGO is specifically investigated for use as the switching layer of the RRAM device. Indium ions (In\(^{3+}\)) contribute to distinct mobility owing to their particular electronic configuration of \((n-1)d^{10}ns^{0}\), where \(n\) is the principal quantum number. However, oxygen vacancies in indium oxide (In\(_2\)O\(_3\)) are hard to control. Gallium is well known as a suppressor of oxygen vacancies. The oxygen vacancies can then be controlled via introducing the gallium element in the switching layer. The bond-dissociation energy of Ga–O (374 kJ/mol) is larger than that of In–O (346 kJ/mol). Moreover, the atomic size of Ga is similar to that of In, which causes only little effect upon substitution.

As everyone knows, RRAM devices were operated between a high-resistance state (HRS) and a low-resistance state (LRS). The switching behavior from the HRS to the LRS is caused by a set process, which arranges the oxygen vacancies and forms a conductive filament. In contrast, the reset process destroys the filament arrayed by oxygen vacancies and changes the RRAM device from the LRS back to the HRS. The applied voltages...
when the RRAM device switches from each state can then be obtained as the set voltage and reset voltage, respectively.

Because oxygen vacancies play a crucial role in RRAM devices as mentioned earlier, this work focuses on how the arrangement of oxygen vacancies influences the performance, including the set voltage, reset voltage, HRS, LRS, and stability of different structures of the switching layer. Though there are plenty of research conducted on RRAM devices using different materials, most of them are on devices with a single layer. Stacked IGO RRAM with gradually introduced elements of In and Ga is expected to have a better arrangement of oxygen vacancies. Based on the above concerns, a RRAM device with IGO stacked layers was fabricated. Moreover, the current−voltage (I−V) characteristics and stability of the IGO RRAM are investigated.

RESULTS AND DISCUSSION

Figure 1a shows the IGO cross-sectional image of the stacked IGO RRAM obtained using a transmission electron microscope. The total thickness of the IGO stack layers was approximately 50.1 nm, with no significant interface observed between each layer. It is observed that the In element decreases and the Ga element increases from the TE to the BE, as shown in Figure 1b. The transmission electron microscopy (TEM) results indicate that the thickness and elements of the fabricated IGO RRAM are well controlled. Through Gaussian fitting, the O 1s peak can be deconvoluted into two peaks at 530.1 and 531.9 eV from the X-ray photoelectron spectroscopy (XPS) results. The peak at 530.1 eV is assigned to the well-bonded oxygen with the metal cation (M−O) and the other is assigned to the oxygen vacancies (VO). Through calculation of the ratio of the area below the curve represented for VO to the total area below the O 1s curve, the amount of oxygen vacancies of each IGO layer can be extracted. As shown in Figure 2a−e, the amounts of oxygen vacancies of the five layers increase with values of 15.1, 20.4, 39.1, 52.3, and 55.6% when the ratio of In2O3 increases from 0 to 100%. The XPS results demonstrate that the Ga element well suppresses the oxygen vacancies in the IGO thin film. Further impact will be discussed later.

Figure 3a−d illustrates the I−V characteristics, endurance test, and retention test of the stacked IGO RRAM. The forming voltage of the stacked IGO RRAM is 1.5 V. In addition, the endurance and retention tests indicate that the IGO RRAM can operate under a DC sweep of more than 4000 cycles, and its LRS is still separate from the HRS after 10,000 s, which ensures durability and reliability. Some basic parameters are calculated from the 4090 cycles, where the average set voltage is 0.76 V, the average reset voltage is −0.66 V, the average HRS is approximately 3236 Ω, and the average LRS is approximately 14 Ω, respectively.

For comparing and analyzing the stability enhancement by the stacked InxGa1−xO layer, RRAM devices with five single layers and different compositions of IGO were fabricated using five single targets, namely, pure In2O3, IGO (In2O3/Ga2O3 = 9:1/4:6/1:9 in at. %), and pure Ga2O3. The experimental details are the same as those of our previous work with TE of Al.28 Table 1 summarizes several basic parameters of the RRAM device with different IGO switching layers. A significant difference in the forming voltages between stacked IGO and the others can be observed. While the forming voltages of the single-layer IGO RRAM devices are all greater than 4 V, that of the stacked IGO RRAM is only 1.5 V. A small forming voltage is desirable in order to protect the RRAM device from hard breakdown. However, the forming process, as the first set process, usually needs a larger electric field to arrange the oxygen vacancies (LRS) from random distribution (original state). It is observed that the concentrations of the oxygen vacancies in the stacked IGO layer vary between each layer; these vacancies are suppressed by the gallium element as mentioned above. As a result, there exists a gradient in the concentration of oxygen vacancies at the original state of the stacked IGO RRAM. This implies that the stacked IGO RRAM only needs a little electric field to arrange the oxygen vacancies and form the conductive filament from the original state. Furthermore, the average set voltage of the stacked IGO RRAM is only 0.76 V, while that of all the other single-layer IGO RRAMs is greater than 1.7 V. The smaller average set voltage can also be attributed to the stacked structure of the InxGa1−xO layer. As mentioned, bond-dissociation energy of Ga−O (374 kJ/mol) is larger than that of In−O (346 kJ/mol). In other words, the activity of gallium toward oxygen is larger than that of indium. Oxygen ions easily bond with gallium, and lesser oxygen vacancies are left behind. On the contrary, there may be more oxygen vacancies and lesser oxygen ions in the indium oxide layer. Moreover, the mobility of the oxygen ions in indium is larger than that of gallium, which may accelerate
the oxygen ions to migrate from the stacked IGO layer with less indium to the stacked IGO layer with more indium. Considering the set process, a significant number of oxygen ions are attracted from the Ga$_2$O$_3$ layer, accelerating to accumulate at the In$_2$O$_3$ layer owing to the Coulombic force. A conductive filament is formed when a positive bias is applied on the TE as shown in Figure 4a. With the aid of the original gradient of the oxygen ions and mobility between different layers, a smaller average set voltage of the stacked IGO RRAM can be explained. On the other hand, there is no significant difference in the average reset voltage between the stacked RRAM and the others. In previous work, we have discussed different mechanisms of the reset process with different TEs. The redox reaction is due to oxidation and reduction of oxygen vacancies in the switching layer to form or destroy the filament. The Joule heat effect only needs a small electric field to burn the filament owing to the current crowding effect. If the reset process is dominated by the redox reaction, the maximum set current would be almost the same as the maximum reset current because the reaction is reversible. On the contrary, if

Figure 2. XPS O 1s spectra of (a) Ga$_2$O$_3$, (b) I$_{0.1}$Ga$_{0.9}$O, (c) I$_{0.4}$Ga$_{0.6}$O, (d) I$_{0.9}$Ga$_{0.1}$O, and (e) In$_2$O$_3$ thin films.
the reset process is dominated by the Joule heat effect, the maximum reset current would be much larger than the maximum set current, and this would crucially affect the stability. From Figure 3a, it is observed that the maximum currents in the set and reset processes are almost the same, which signifies that the reset process of the stacked IGO RRAM is dominated by the redox reaction instead of the Joule heat effect. Figure 4b indicates the redox reaction schematic of the reset process. Because a negative bias is applied on the TE, the accumulated oxygen ions are excluded rapidly by the Coulombic force and recombine with the oxygen vacancies. The conductive filament is finally destroyed under a small reset voltage.

For verifying the stability of the RRAM devices, the coefficient of variation (CV) values of set voltage and reset voltage of each of the IGO RRAM devices are summarized in Table 1. The CV of set voltage is 0.34 for the stacked IGO RRAM, which is smaller than those of the other single-layer IGO RRAM devices. Moreover, the CV of reset voltage is 0.18 for the stacked IGO RRAM. As mentioned before, the variation of the set process is improved by the gradient of the concentration of oxygen vacancies and mobility. Besides, owing to a larger activity of gallium toward oxygen, oxygen ions bond with gallium tighter in the gallium oxide layer. Namely, the bottom filament paths (InG\textsubscript{x}O\textsubscript{1-x} switching layer with x < 0.5) for every set process may be relatively regular and can be followed by the upper filament (InG\textsubscript{x}O\textsubscript{1-x} switching layer with x > 0.5), which will not happen in a single-layer structure. As a result, a stabler set process can be achieved. Although the average reset voltage of the stacked IGO RRAM is similar to that of other single-layer IGO RRAM devices, the variation of the reset voltage shows a significant improvement in the stacked IGO RRAM. This result is owing to the mechanism of the reset process. In comparison with the Joule heat effect of the single-layer structure, the stacked structure brings forth the dominance of the redox reaction during the reset process. When the conductive filament is destroyed owing to the Joule heat effect, the arrangement of the oxygen vacancies is majorly interrupted. This process is irreversible, implying that, the path of the filament would be different subsequently. In contrast, the reset process of the stacked IGO RRAM is dominated by the redox reaction, which is a reversible reaction, leading to a similar absolute value of the set voltage and reset voltage. In this case, the path of the conductive filament would be more stable. As a result, the variations of set voltage and reset voltage of the single-layer

| switchinglayer forming voltage (V) | set voltage (V) | C.V. of V\textsubscript{set} | reset voltage (V) | C.V. of V\textsubscript{reset} | cycle | on/off ratio | mechanism |
|------------------|---------------|-----------------|-----------------|-----------------|------|------------|-----------|
| single In\textsubscript{2}O\textsubscript{3} | 13.2 | 4.5 | 0.756 | -0.7 | 0.456 | 608 | >10\textsuperscript{4} | Joule heat effect |
| single In\textsubscript{0.9}Ga\textsubscript{0.1}O | 13.1 | 6.1 | 0.746 | -0.7 | 0.366 | 247 | >10\textsuperscript{5} | Joule heat effect |
| single In\textsubscript{0.4}Ga\textsubscript{0.6}O | 11.5 | 3.8 | 0.945 | -0.5 | 0.553 | 129 | >10\textsuperscript{6} | Joule heat effect |
| single Ga\textsubscript{2}O\textsubscript{3} | 5.2 | 1.8 | 0.64 | -0.5 | 0.334 | 169 | >10\textsuperscript{1} | Joule heat effect |
| single I\textsubscript{0.1}G\textsubscript{0.9}O | 4 | 2.2 | 0.45 | -0.9 | 0.724 | 336 | >10\textsuperscript{3} | Joule heat effect |
| stacked I\textsubscript{1}G\textsubscript{1-x}O | 1.5 | 0.8 | 0.335 | -0.7 | 0.180 | 4090 | >10\textsuperscript{2} | redox |
IGO RRAM devices are larger than those of the stacked IGO RRAM. Another result worth noting is that the stacked IGO RRAM can operate at a DC sweep of more than 4000 cycles, which is much greater than that observed for other single-layer IGO RRAM devices. The remarkable operating times can also be attributed to the redox reaction and the original gradient of the stacked structure. Because the Joule heat effect is an irreversible action as mentioned, the reset process may cause repeated damage to the switching layer rendering it unable to work anymore. As a result, the operating cycles of the single-layer IGO RRAM devices are much lesser than the stacked IGO RRAM.

**CONCLUSIONS**

In this work, we fabricated a stacked structure IGO RRAM with several IGO targets using a radio frequency (RF) sputtering system. The stacked IGO RRAM can operate more than 4000 times with a small average set voltage of 0.76 V and an average reset voltage of −0.66 V. Compared to other single-layer IGO RRAM devices, the coefficients of variation of set voltage and reset voltage of the stacked RRAM are smaller. The improved result is due to the original gradient of the concentration of oxygen vacancies and mobility of the stacked layers, which stabilized the set and reset processes. Furthermore, the reset process of the stacked IGO RRAM is dominated by the reversible redox reaction. The stability of the IGO RRAM is significantly improved by controlling the oxygen vacancies and composition of elements.

**EXPERIMENTAL SECTION**

Figure 5 shows the schematic of the IGO RRAM, for which a 10 nm-thick Ti adhesion layer and a 50 nm-thick Pt bottom electrode were deposited using an e-beam evaporator on a 2 cm × 2 cm quartz substrate. The substrate was cleaned via ultrasonication, using acetone, isopropyl alcohol, and deionized water for 5 min each. For fabricating the five switching layers with different compositions of In and Ga elements, a pure In$_2$O$_3$ target, a pure Ga$_2$O$_3$ single target, and a three single IGO targets with different In$_2$O$_3$ and Ga$_2$O$_3$ ratios (In$_2$O$_3$/Ga$_2$O$_3$ = 9:1/4:6/1:9 in at. %) were used. When it comes to the switching layer, a RF sputtering system with a frequency of 13.56 MHz was used. The Ga$_2$O$_3$ layer was first deposited via RF sputtering on the substrate, followed by the IGO layers in the decreasing order of the Ga content, with the In$_2$O$_3$ layer being deposited last. The thickness of each layer was fixed at 10 nm, implying that the total thickness of the IGO switching layer was 50 nm. For the sputtering process, the sputtering power was set at 80 W, and the gas flow ratio of O$_2$/Ar was 10/90 sccm. Finally, a 100 nm-thick Al TE was deposited using an e-beam evaporator. On the other hand, the switching layer and TE were defined by metal masks, which measured 1500 μm × 1500 μm for the switching layer and 50 μm × 50 μm for the TE.

The current–voltage (I–V) characteristics were measured at room temperature using a B1500A semiconductor parameter analyzer (Agilent, Santa Clara, CA, USA). TEM (JEM-2100F electron microscope, JEOL, Tokyo, Japan) and XPS (PHI 5000 VersaProbe, ULVAC-PHI, Chigasaki, Japan) analyses were conducted to verify the composition of the elements and oxygen vacancies.

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