Resistance-switching properties of Bi-doped SrTiO₃ films for non-volatile memory applications with different device structures

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Abstract. SrTiO₃ and Bi-doped SrTiO₃ films were fabricated with different device structures using the sol–gel method for non-volatile memory applications, and their resistance-switching behaviour, endurance and retention characteristics were investigated. SrTiO₃ and Sr_{0.92}Bi_{0.08}TiO₃ films grown on Si or Pt have the same phase structure, morphologies and grain size; however, the grain size of the Sr_{0.92}Bi_{0.08}TiO₃ films grown on Si is slightly larger than those of the SrTiO₃ films grown on Si and the Sr_{0.92}Bi_{0.08}TiO₃ films grown on Pt. The SrTiO₃ or Sr_{0.92}Bi_{0.08}TiO₃ films grown on Si or Pt all exhibit bipolar resistive-switching behaviour and follow the same conductive mechanism; however, the Ag/Sr_{0.92}Bi_{0.08}TiO₃/Si device possesses the highest R_{HRS}/R_{LRS} of 10⁵ and the best endurance and retention characteristics. The doping of Bi is conducive to enhance the R_{HRS}/R_{LRS} of the SrTiO₃ films; meanwhile, the Si substrates help improve the endurance and retention characteristics of the Sr_{0.92}Bi_{0.08}TiO₃ films.

Keywords. SrTiO₃; Bi doping; resistance-switching properties; device structure; sol–gel.

1. Introduction

To meet the constantly increasing data storage requirements, researchers have long been searching for faster and smaller non-volatile memory devices. Some new non-volatile memory devices, such as phase-change random access memory (PCRAM), magnetic random access memory (MRAM), ferroelectric random access memory (FRAM) and resistive random access memory (RRAM), have been widely investigated. Among them, RRAM, based on electrically induced ‘resistive-switching’ effects, has generated considerable interest among researchers due to its simpler and smaller cell structure compared with MRAM and FRAM, as well as lower driving voltage compared with PCRAM [1–3]. Thus far, various materials presenting resistive-switching characteristics, such as binary transition metal oxides [4–7], perovskite oxides [8,9], manganites [10,11] and organic compounds [12], have been reported. Among them, SrTiO₃ has been investigated intensively because of its good switching properties and interesting bipolar/unipolar resistive-switching behaviour [13–15]. Many previous studies suggested that rare-earth element doping is an effective method to enhance the properties of materials [16–18]. SrTiO₃ belongs to the perovskite family with the general formula ABO₃, and its conductivity can be improved by doping a rare-earth element into the A sublattice or a transition metal into the B sublattice [17,18]. Doping of some elements into SrTiO₃ films for resistive-switching device application, such as Nb [19–23], Fe [24–26], V [27] and Cr [15], has been widely

researched. Because the radius of Bi³⁺ is close to that of Sr²⁺ [28], if Bi³⁺ ion doping is used to replace Sr²⁺ in SrTiO₃, electrons can be released to form an ionized and non-movable positive charge centre, so as to complete donor doping. Compared to La³⁺ doping, adding Bi³⁺ into SrTiO₃ can improve the carrier concentration [29], but research on Bi-doped SrTiO₃ films has still been very scarce. Meanwhile, resistive-switching behaviours in different thin films, such as NiO [30,31], CuO [32], ZnO [33], ZnMn₂O₄ [34] and SrTiO₃ [20], have been explored in previous reports by altering the device structure. However, the device structure-dependent resistive-switching behaviour and properties of Bi-doped SrTiO₃ thin films have yet to be reported; therefore, investigation of the resistive-switching behaviour and properties of Bi-doped SrTiO₃ films with different device structures requires attention.

In this study, SrTiO₃ and its Bi-doped films used for RRAM application were fabricated on different bottom electrodes *via* the sol–gel method, and the contrastive study of resistive-switching behaviour and properties of pure SrTiO₃ and Bi-doped SrTiO₃ thin films with different device structures is reported. The dependence of these structures on current–voltage (I – V), endurance and retention characteristics of Bi-doped SrTiO₃ films are explored in detail.

2. Experimental

By using the sol–gel method, SrTiO₃ and Sr_{0.92}Bi_{0.08}TiO₃ films were deposited on Si and Pt/Ti/SiO₂/Si (hereafter

abbreviated as Pt) substrates with sizes of $10\text{ mm} \times 10\text{ mm}$, respectively. The SrTiO_3 and $\text{Sr}_{0.92}\text{Bi}_{0.08}\text{TiO}_3$ solutions were prepared using $\text{C}_{16}\text{H}_{36}\text{O}_4\text{Ti}$, $\text{C}_4\text{H}_6\text{Sr} \cdot 1/2\text{H}_2\text{O}$ and $\text{Bi}(\text{NO}_3)_3 \cdot 5\text{H}_2\text{O}$ as source materials. Acetic acid and $\text{CH}_3\text{OCH}_2\text{COCH}_3$ were used as solvents, acetylacetone as a chelating agent and diethanolamine as a stabilizer. The SrTiO_3 and $\text{Sr}_{0.92}\text{Bi}_{0.08}\text{TiO}_3$ films were prepared on Si or Pt substrates by spin coating the solution at 4000 rpm for 30 s, followed by preheating at 120°C for 5 min and then at 400°C for 10 min to remove volatile materials before spinning the next layer. The above processes were repeated five times. Finally, the SrTiO_3 or $\text{Sr}_{0.92}\text{Bi}_{0.08}\text{TiO}_3$ films were annealed at 800°C for 10 min in ambient air. The top Ag electrodes with a diameter of 200 nm were deposited by the vacuum evaporation method with a metal shadow mask to form a device structure of $\text{Ag}/\text{SrTiO}_3/\text{Si}$, $\text{Ag}/\text{Sr}_{0.92}\text{Bi}_{0.08}\text{TiO}_3/\text{Si}$ or $\text{Ag}/\text{Sr}_{0.92}\text{Bi}_{0.08}\text{TiO}_3/\text{Pt}$.

The crystalline structures of the SrTiO_3 and $\text{Sr}_{0.92}\text{Bi}_{0.08}\text{TiO}_3$ films were characterized by X-ray diffraction (XRD; AXS D8-ADVANCE, Bruker). The surface morphologies of these films were observed by using a scanning electron microscope (SEM; Hitachi S4800). The measurement of I – V characteristics of the films with different device structures was performed using a Keithley 2400 Source Meter at room temperature.

3. Results and discussion

Figure 1 shows the typical XRD patterns of the SrTiO_3 and $\text{Sr}_{0.92}\text{Bi}_{0.08}\text{TiO}_3$ films grown on Si and Pt substrates. From figure 1a, it can be seen that the SrTiO_3 and $\text{Sr}_{0.92}\text{Bi}_{0.08}\text{TiO}_3$ films grown on Si and Pt substrates all possess a polycrystalline structure and high degree of crystallinity. The (110) and (200) peaks of the films are clearly observed, and no peaks from other phases are detected. It is worth noting that the characteristic diffraction peaks of both the $\text{Sr}_{0.92}\text{Bi}_{0.08}\text{TiO}_3$ films grown on Si and Pt substrates shift slightly to the higher angles compared with those of the SrTiO_3 films, as shown in figure 1b, which indicates that the SrTiO_3 lattice is reduced due to the fact that the Bi^{3+} ions (ionic radius: 1.03 pm [28]) substituted the Sr^{2+} ions (ionic radius: 1.18 pm [28]), but not the Ti^{4+} ions (ionic radius: 0.605 pm [28]). In addition, the diffraction peaks of (110) and (200) in the $\text{Sr}_{0.92}\text{Bi}_{0.08}\text{TiO}_3$ films grown on Si are higher and more sharper than those in the $\text{Sr}_{0.92}\text{Bi}_{0.08}\text{TiO}_3$ films grown on Pt and the SrTiO_3 films grown on Si, indicating the nuance in the growth-oriented direction and grain size for the films grown on different substrates but with the same phase structure.

SEM surface and cross-section images of the SrTiO_3 and $\text{Sr}_{0.92}\text{Bi}_{0.08}\text{TiO}_3$ films grown on Si and Pt are shown in figure 2. The surfaces of these films are all smooth, showing no flaws and cracks. Crystal grain is uniform with a spherical shape, arranged closely with an average size of 30 nm for the $\text{Sr}_{0.92}\text{Bi}_{0.08}\text{TiO}_3$ films grown on Si substrates and ~ 25 and

Figure 1. (a) XRD patterns of SrTiO_3 and $\text{Sr}_{0.92}\text{Bi}_{0.08}\text{TiO}_3$ films grown on Si and Pt and (b) (110) peak expansion.

22 nm for the SrTiO_3 films grown on Si and Pt, respectively. The grain size of the $\text{Sr}_{0.92}\text{Bi}_{0.08}\text{TiO}_3$ films grown on Si is slightly larger than that of the SrTiO_3 films grown on Si and the $\text{Sr}_{0.92}\text{Bi}_{0.08}\text{TiO}_3$ films grown on Pt; these findings are in agreement with the XRD results. The similarity in grain morphologies is due to the same phase structure, and the difference in grain size of these films may be related to the substrates. In addition, the cross-sectional SEM image shown in figure 2d reveals a sharp interface structure with a thickness of ~ 180 nm for the SrTiO_3 films grown on Si.

To evaluate the resistance switching memory effects of the SrTiO_3 and $\text{Sr}_{0.92}\text{Bi}_{0.08}\text{TiO}_3$ films grown on different substrates, the I – V characteristics of the three-structure device cells are studied *via* dc voltage sweep measurements, and the typical results are illustrated in figure 3. During the I – V measurements, the bias voltage $0\text{ V} \rightarrow 6\text{ V} \rightarrow 0\text{ V} \rightarrow -6\text{ V} \rightarrow 0\text{ V}$

Figure 2. SEM surface images: (a) SrTiO_3 on Si; (b) $\text{Sr}_{0.92}\text{Bi}_{0.08}\text{TiO}_3$ on Si and (c) $\text{Sr}_{0.92}\text{Bi}_{0.08}\text{TiO}_3$ on Pt. SEM cross-section image of (d) $\text{Sr}_{0.92}\text{Bi}_{0.08}\text{TiO}_3$ on Si.

is applied to the top electrode of the device cell while the bottom electrode was grounded. Figure 3 shows that two distinct resistance states are observed in all samples, exhibiting bipolar resistive-switching behaviour and no electroforming process. In the initial state, the current of the films is extremely low, indicating that it is in a high-resistance state (HRS). By steadily increasing the positive bias voltage, the current suddenly increases, indicating that it has changed from the HRS to a low-resistance state (LRS) at the set voltage (V_{set}) and that the ‘set’ progress is accomplished. After reducing the voltage to 0 V, a reverse bias voltage is applied to the films. When the reset voltage (V_{reset}) is achieved, the current abruptly decreases, and the films change from the LRS to HRS. For all examples, the V_{set} is about 3–4 V, which is similar to those of the Nb-doped SrTiO_3 (100 nm thickness) thin films [19] and Cr-doped SrTiO_3 (100 nm thickness) thin films [27] on Si, but it is higher than that of the Fe-doped SrTiO_3 (500 nm thickness) thin films [25] on the Nb-doped SrTiO_3 substrate and much lower than that of the V-doped SrTiO_3 (150 nm thickness) thin films on Si or Pt [15]. Although the resistive-switching behaviour in the three-structure devices is similar, the resistance ratio ($R_{\text{HRS}}/R_{\text{LRS}}$) of HRS to LRS of the $\text{Sr}_{0.92}\text{Bi}_{0.08}\text{TiO}_3$ films grown on Si is over 10^5 , higher than those of the SrTiO_3 films grown on Si (10^3) and the

$\text{Sr}_{0.92}\text{Bi}_{0.08}\text{TiO}_3$ films grown on Pt (10^4), which is higher than those of Nb-doped SrTiO_3 [19] and V-doped SrTiO_3 [15] thin films on Si. In addition, for SrTiO_3 and $\text{Sr}_{0.92}\text{Bi}_{0.08}\text{TiO}_3$ films grown on Si, when the bias voltage sweeps from -6 to 0 V, the current notably reaches a minimum value at a certain voltage, which indicates the hysteresis of the capacitance of the films [35].

To investigate the conduction mechanisms of Bi-doped SrTiO_3 films grown on different substrates, the positive $I-V$ curves of the $\text{Sr}_{0.92}\text{Bi}_{0.08}\text{TiO}_3$ films grown on Pt in the LRS and HRS are replotted as $\ln|I|-\ln|V|$, as shown in figure 4. The slope of the $\ln|I|-\ln|V|$ curve is ~ 1 in the LRS, which indicates that the conduction behaviour undergoes ohmic transport and can be explained by the conducting filament mechanism [36]. However, the conduction behaviours of the $\text{Sr}_{0.92}\text{Bi}_{0.08}\text{TiO}_3$ films in the HRS are complicated. In the lower electric-field region of the HRS, the $\ln|I|-\ln|V|$ curve is linear with a slope of ~ 1 and the conduction behaviour is governed by Ohm’s law; however, the current increases rapidly with a slope of ~ 2 in the higher electric-field region of the HRS, indicating that the conduction behaviour of the $\text{Sr}_{0.92}\text{Bi}_{0.08}\text{TiO}_3$ films can be explained by the space-charge-limited current conduction mechanism. Given that the density of thermally generated free carriers in the films is predominant

Figure 3. Typical I - V curves of SrTiO_3 and $\text{Sr}_{0.92}\text{Bi}_{0.08}\text{TiO}_3$ films grown on Si and Pt.

over the injected charge carriers with the increasing applied voltage, the carriers hop into the traps and the excess electrons are trapped, leading to space charge [37]. A similar phenomenon on the $\ln|I|$ - $\ln|V|$ curves of the SrTiO_3 and $\text{Sr}_{0.92}\text{Bi}_{0.08}\text{TiO}_3$ films grown on Si substrates is also obtained.

Figure 4. Typical $\ln I$ - $\ln V$ plots of $\text{Sr}_{0.92}\text{Bi}_{0.08}\text{TiO}_3$ films grown on Pt.

Good endurance characteristics are vitally important for RRAM devices to ensure a reproducible switching between on and off states. To evaluate the endurance characteristics of SrTiO_3 and $\text{Sr}_{0.92}\text{Bi}_{0.08}\text{TiO}_3$ grown on different substrates, a switching test for 2×10^3 cycles is performed under a reading voltage of 2 V; the results are shown in figure 5. The $\text{Ag}/\text{Sr}_{0.92}\text{Bi}_{0.08}\text{TiO}_3/\text{Si}$ structure device does not show any resistance degradation at 2000 repeated switching cycles with an $R_{\text{HRS}}/R_{\text{LRS}}$ of 10^5 , the endurance performance of $\text{Ag}/\text{Sr}_{0.92}\text{Bi}_{0.08}\text{TiO}_3/\text{Si}$ is equivalent to that of the Nb-doped SrTiO_3 thin films on Si using TiN as a buffer layer [19], but less than that of $\text{Al}/\text{Cr}-\text{SrTiO}_3/\text{p-Si}$ structure devices [27]. The resistance ratio of the HRS to LRS of the $\text{Sr}_{0.92}\text{Bi}_{0.08}\text{TiO}_3$ films grown on Pt is higher than that of the SrTiO_3 films grown on Si by one order of magnitude; however, the stable repeated switching cycles are only ~ 1200 , which is less than that of 1600 cycles for the latter. The $\text{Ag}/\text{Sr}_{0.92}\text{Bi}_{0.08}\text{TiO}_3/\text{Pt}$ structure device undergoes obvious degradation of resistance in the HRS after 1200 test cycles. Then, the low resistance increases during the next 800 test cycles. Finally, high and low resistances have no significant difference when the switching test is over 2000 cycles. The endurance performance of the $\text{Ag}/\text{SrTiO}_3/\text{Si}$ structure device possesses a similar change trend; however, the number of stable repeated switching cycles is more than 1600. These results indicate that the SrTiO_3 and $\text{Sr}_{0.92}\text{Bi}_{0.08}\text{TiO}_3$ films grown on Si have better endurance characteristics than the $\text{Sr}_{0.92}\text{Bi}_{0.08}\text{TiO}_3$ films grown on Pt.

Retention characteristics imply the ability to maintain a state of high and low resistances without electrical power. The retention performances of the three-structure device at room temperature are displayed in figure 6, where the resistance values of two states are read out at 1 V. After the device is switched from the on to the off state, no electrical power is applied to maintain the resistance within a given

Figure 5. Endurance characteristics of SrTiO_3 and $\text{Sr}_{0.92}\text{Bi}_{0.08}\text{TiO}_3$ films grown on Si and Pt.

state. Although a slight fluctuation of resistance occurs in the LRS and HRS, a stable $R_{\text{HRS}}/R_{\text{LRS}}$ of 10^5 is maintained for longer than 10^6 s for the $\text{Ag}/\text{Sr}_{0.92}\text{Bi}_{0.08}\text{TiO}_3/\text{Si}$ structure device (figure 6b), which is equivalent to that of the Nb-doped SrTiO_3 thin films on Si using TiN as a buffer layer [19], but longer than that of the $\text{Al}/\text{Cr}-\text{SrTiO}_3/\text{p-Si}$ structure devices [27]. However, when the time persists for

Figure 6. Retention characteristics of SrTiO_3 and $\text{Sr}_{0.92}\text{Bi}_{0.08}\text{TiO}_3$ films grown on Si and Pt.

10^2 s for the $\text{Ag}/\text{Sr}_{0.92}\text{Bi}_{0.08}\text{TiO}_3/\text{Pt}$ device and 10^4 s for the $\text{Ag}/\text{SrTiO}_3/\text{Si}$ device, the R_{HRS} in both devices begin to decrease, indicating that the information stored in these two structure devices is unlikely to persist for a longer time based on the present data trend. According to the retention performance of these devices, $\text{Ag}/\text{Sr}_{0.92}\text{Bi}_{0.08}\text{TiO}_3/\text{Si}$ as a memory

Table 1. Resistive-switching property parameters of a batch of samples for the Ag/Sr_{0.92}Bi_{0.08}TiO₃/Si device.

Example	V _{set} , V	R _{HRS} /R _{LRS}	Cycles without degradation, #	Retention time, s
1	3.55	1.2 × 10 ⁵	> 2 × 10 ³	> 10 ⁶
2	3.41	0.9 × 10 ⁵	> 2 × 10 ³	> 10 ⁶
3	3.47	1.1 × 10 ⁵	> 2 × 10 ³	> 10 ⁶
4	3.52	1.0 × 10 ⁵	> 2 × 10 ³	> 10 ⁶
5	3.59	0.8 × 10 ⁵	> 2 × 10 ³	> 10 ⁶
6	3.43	1.2 × 10 ⁵	> 2 × 10 ³	> 10 ⁶
7	3.61	1.3 × 10 ⁵	1.5 × 10 ³	> 10 ⁶
8	3.39	1.1 × 10 ⁵	> 2 × 10 ³	> 10 ⁶
9	3.45	1.0 × 10 ⁵	> 2 × 10 ³	> 10 ⁶
10	3.51	0.9 × 10 ⁵	> 2 × 10 ³	> 10 ⁶

device cell has better retention characteristics for RRAM than Ag/Sr_{0.92}Bi_{0.08}TiO₃/Pt.

Table 1 shows the main resistive-switching property parameters of a batch of samples for the Ag/Sr_{0.92}Bi_{0.08}TiO₃/Si device. From table 1, it can be seen that the resistive-switching properties of the Ag/Sr_{0.92}Bi_{0.08}TiO₃/Si device is stable and repeatable, which exhibits the application potentiality in non-volatile memory devices.

4. Conclusions

SrTiO₃ films grown on Si and Sr_{0.92}Bi_{0.08}TiO₃ films grown on Si or Pt *via* the sol–gel method have the same phase structure and morphologies. However, the grain size of the Sr_{0.92}Bi_{0.08}TiO₃ films grown on Si is slightly larger than those of the SrTiO₃ films grown on Si and the Sr_{0.92}Bi_{0.08}TiO₃ films grown on Pt. The SrTiO₃ or Sr_{0.92}Bi_{0.08}TiO₃ films grown on different substrates all exhibit bipolar resistive-switching behaviour and follow the same conductive mechanism. Nevertheless, the Ag/Sr_{0.92}Bi_{0.08}TiO₃/Si device possesses the highest R_{HRS}/R_{LRS} of 10⁵ and the best endurance and retention characteristics. The R_{HRS}/R_{LRS} of both the Ag/Sr_{0.92}Bi_{0.08}TiO₃/Si and Ag/Sr_{0.92}Bi_{0.08}TiO₃/Pt devices are higher than that of the Ag/SrTiO₃/Si device. Meanwhile, the number of stable repeated switching cycles and the retention time of the Ag/Sr_{0.92}Bi_{0.08}TiO₃/Pt device are poorer than that of the Ag/Sr_{0.92}Bi_{0.08}TiO₃/Si or Ag/SrTiO₃/Si device. This result indicates that the doping of Bi is conducive to enhance the R_{HRS}/R_{LRS} of the SrTiO₃ films and that the Si substrates help improve the endurance and retention characteristics of the Sr_{0.92}Bi_{0.08}TiO₃ films.

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