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Space electric field concentrated effect for Zr:SiO₂ RRAM devices using porous SiO₂ buffer layer

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Abstract

To improve the operation current lowering of the Zr:SiO₂ RRAM devices, a space electric field concentrated effect established by the porous SiO₂ buffer layer was investigated and found in this study. The resistive switching properties of the low-resistance state (LRS) and high-resistance state (HRS) in resistive random access memory (RRAM) devices for the single-layer Zr:SiO₂ and bilayer Zr:SiO₂/porous SiO₂ thin films were analyzed and discussed. In addition, the original space charge limited current (SCLC) conduction mechanism in LRS and HRS of the RRAM devices using bilayer Zr:SiO₂/porous SiO₂ thin films was found. Finally, a space electric field concentrated effect in the bilayer Zr:SiO₂/porous SiO₂ RRAM devices was also explained and verified by the COMSOL Multiphysics simulation model.

Keywords: RRAM; Porous SiO₂; Space charge limited current; Zr

Background

Recently, various non-volatile random access memory (NvRAM) such as magnetic random access memory (MRAM), ferroelectric random access memory (FeRAM), phase change memory (PCM), and resistive random access memory (RRAM) were widely investigated and discussed for applications in portable electronic products which consisted of low power consumption IC [1], non-volatile memory [2-6], and TFT LCD display [7-10]. To overcome the technical and physical limitation issues of conventional charge storage-based memories [11-18], the resistive random access memory (RRAM) device which consisted of the oxide-based layer sandwiched by two electrodes was a great potential candidate for the next-generation non-volatile memory because of its superior properties such as low cost, simple structure, fast operation speed, low operation power, and non-destructive readout properties [19-42].

In our previous report, the resistive switching stability and reliability of RRAM device can be improved using a

high/low permittivity bilayer structure [43]. Because the permittivity of porous SiO₂ film is lower than that of SiO₂ film, the zirconium metal doped into SiO₂ (Zr:SiO₂) thin film fabricated by co-sputtering technology and the porous SiO₂ buffer layer prepared by inductively coupled plasma (ICP) treatment were executed to form Zr:SiO₂/porous SiO₂ RRAM devices in this study. In addition, the resistive switching behaviors of the Zr:SiO₂ RRAM devices using the bilayer structure were improved and investigated by a space electric field concentrated effect.

Methods

To generate a space electric field concentrated effect in RRAM devices, the porous SiO₂ buffer layer in the bilayer Zr:SiO₂/porous SiO₂ structure was proposed. The patterned TiN/Ti/SiO₂/Si substrate was obtained by standard deposition and etching process; after which, 1 μm × 1 μm via holes were formed. After that, the C:SiO₂ film was prepared by co-depositing with the pure SiO₂ and carbon targets, and the porous SiO₂ thin film (about 6 nm) was formed by ICP O₂ plasma technology. Then, the Zr:SiO₂ thin film (about 20 nm) was deposited on the porous SiO₂ thin film by co-sputtering with the pure SiO₂ and zirconium targets. The sputtering power

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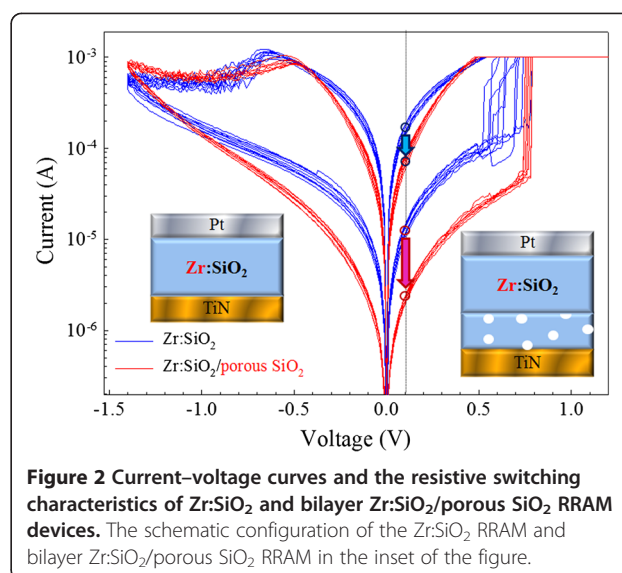
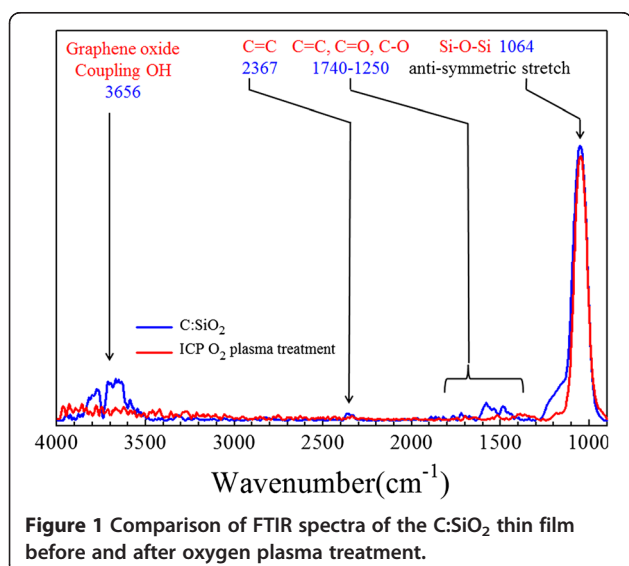
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was fixed with rf power 200 W and direct current (DC) power 10 W for silicon dioxide and zirconium targets, respectively. A Pt electrode of 200-nm thickness was deposited on all samples by DC magnetron sputtering. Finally, all electrical devices were fabricated through lithography and lift-off techniques. Besides, the Fourier transform infrared spectroscopy (FTIR) was used to analyze the chemical composition and bonding of the Zr:SiO₂ thin films, and the entire electrical measurements of devices with the Pt electrode were performed using Agilent B1500 semiconductor parameter analyzer (Santa Clara, CA, USA).

Results and discussion

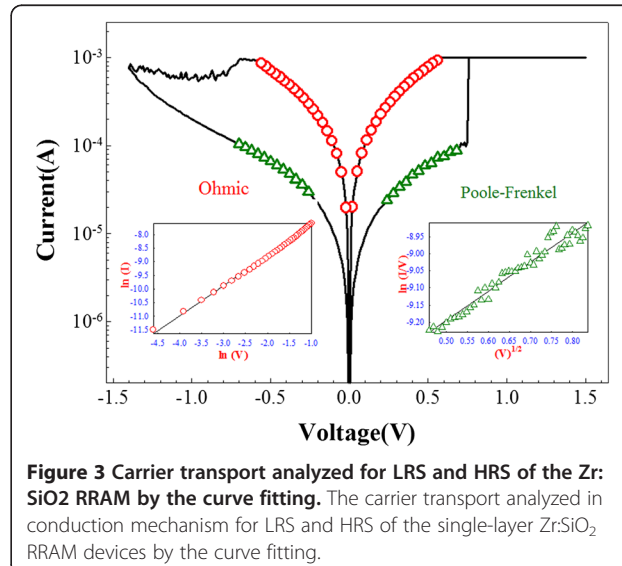
To verify the porous SiO₂ layer generated and formed, the FTIR spectra of the non-treated and treated C:SiO₂ thin film prepared by the oxygen plasma treatment was compared and showed in Figure 1. It was clearly observed that the absorption of anti-symmetric stretch mode of Si-O-Si bonding was at 1,064 cm⁻¹ in the non-treated and treated C:SiO₂ thin film by oxygen plasma treatment. In addition, the C = C bonding at 2,367 cm⁻¹, C:SiO₂ coupling OH bonding at 3,656 cm⁻¹, C-O bonding, and C-C bonding from 1,250 to 1,740 cm⁻¹ were found. This result implicated that the porous SiO₂ thin film was formed by the chemical reaction between carbon and oxygen plasma treatment.

The forming process for the compliance current of 1 μA was required to activate all of the single-layer Zr:SiO₂ and bilayer Zr:SiO₂/porous SiO₂ thin film RRAM devices. For Zr:SiO₂ RRAM devices, the sweeping voltage was applied on TiN electrode with the grounded Pt electrode. Figure 2 shows the resistive switching characteristics of the single-layer Zr:SiO₂ and the bilayer Zr:SiO₂/porous SiO₂ RRAM devices, respectively. The



single-layer Zr:SiO₂ and the bilayer Zr:SiO₂/porous SiO₂ RRAM device structure were also shown in the inset of Figure 2. At the reading voltage of 0.1 V, the operation current of the LRS and HRS in Zr:SiO₂ RRAM devices using the porous SiO₂ buffer layer was smaller than that of others. A space electric field concentrated effect was testified to cause the operation current lowering of the RRAM devices using the porous SiO₂ buffer layer.

In order to further discuss the resistive switching mechanism in single-layer Zr:SiO₂ and bilayer Zr:SiO₂/porous SiO₂ RRAM devices, the conduction mechanism of current-voltage (*I-V*) curves in LRS and HRS were analyzed to discuss the carrier transport in the switching layer in Figures 3 and 4. The carrier transport of the LRS in Zr:SiO₂ RRAM devices dominated by ohmic



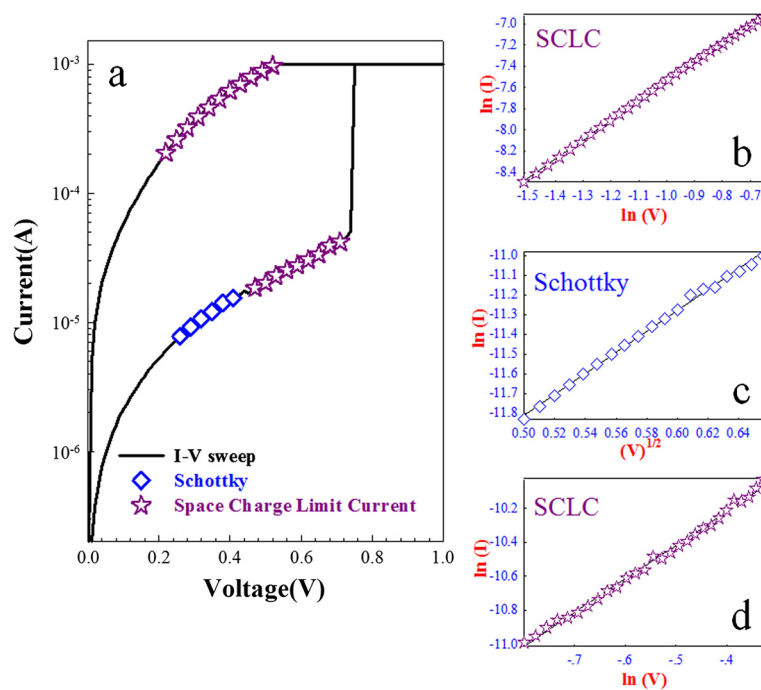


Figure 4 Carrier transport and I-V plots. (a) The carrier transport analyzed in conduction mechanism for LRS and HRS of the single bilayer Zr:SiO₂/porous SiO₂ RRAM devices by the curve fitting. (b) $\ln(I)$, (c) $\ln(I/V^{1/2})$, and (d) $\ln(I/V)$ plots.

conduction mechanism is shown in the left inset of Figure 3. The result revealed that the conductive filament formed by the defect is induced by the zirconium atoms as the current flows through the Zr:SiO₂ film. As shown in the right inset of Figure 3, the carrier transport

in HRS of Zr:SiO₂ RRAM was dominated by Pool-Frenkel emission, which resulted from the thermal emission of trapped electrons in the Zr:SiO₂ film. However, for the bilayer Zr:SiO₂/porous SiO₂ structure, the current mechanism of the LRS in Zr:SiO₂ RRAM devices was dominated

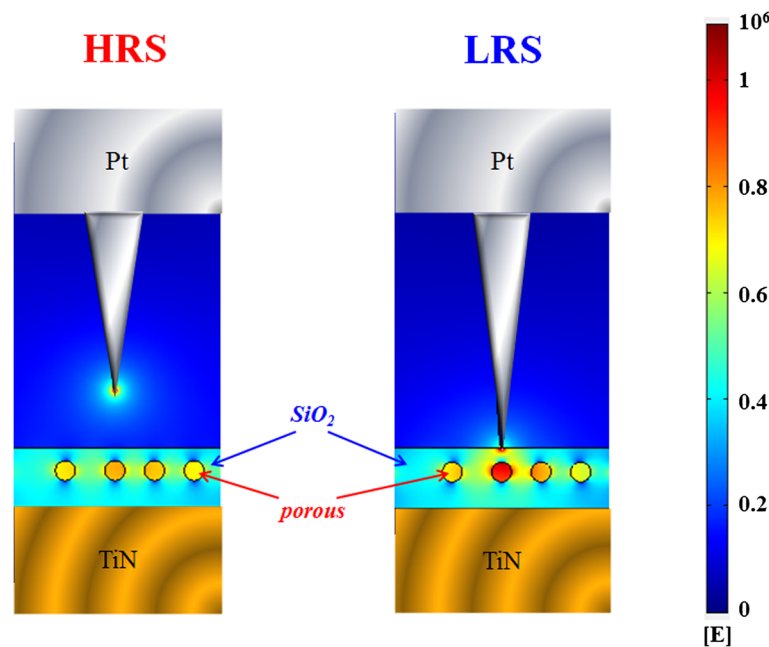


Figure 5 Electric field simulation in LRS and HRS for Pt/Zr:SiO₂/porous SiO₂/TiN RRAM devices.

by the space charge limited current (SCLC) conduction (Figure 4b). Additionally, the current conduction mechanism of the HRS in Zr:SiO₂/porous SiO₂ RRAM devices was transferred from Schottky emission to SCLC conduction in Figure 4c,d. These results indicated that the filament is connected to the pore of porous SiO₂ film after the forming process and the SCLC conduction mechanism is caused by an electric field concentrated effect.

To clarify and discuss the SCLC conduction mechanism in bilayer Zr:SiO₂/porous SiO₂ RRAM devices, the COMSOL Multiphysics simulation model was employed to analyze the distribution of electric field concentrated effect. Figure 5 shows the distribution of the electric field in the bilayer Zr:SiO₂/porous SiO₂ RRAM devices for LRS and HRS. A high density of electric field exists in and around the area of the pore in porous SiO₂ film, which confirms the electric field concentrating capability of nanopores. Thus, during the set process, the metal conduction filament has an inclination to form towards the direction of the pore, and the conduction of the electron was dominated by the SCLC conduction in the porous SiO₂ film.

Conclusion

In conclusion, a space electric field concentrated effect was demonstrated to cause the operation current lowering for the Zr:SiO₂ RRAM devices. In addition, the single-layer Zr:SiO₂ and bilayer Zr:SiO₂/porous SiO₂ were prepared to investigate the resistive switching characteristics of RRAM devices. Compared with the conduction mechanism of the bilayer Zr:SiO₂/porous SiO₂ RRAM with single-layer Zr:SiO₂ RRAM, the conduction mechanism of the LRS was transferred from ohmic to SCLC conduction mechanism. Besides, the conduction mechanism of the HRS was transferred from Pool-Frenkel emission to Schottky emission at low field and dominated by SCLC at high field. Through a space electric field concentrated effect, the SCLC conduction of the Zr:SiO₂ RRAM devices using the porous SiO₂ buffer layer was explained and discussed by the COMSOL Multiphysics simulation model.

Competing interests

The authors declare that they have no competing interests.

Authors' contributions

K-CC designed and set up the experimental procedure. J-WH and T-CC planned the experiments and agreed with the paper's publication. T-MT, K-HC, T-FY, J-HC, D-SG, and J-CL revised the manuscript critically and made some changes. RZ fabricated the devices with the assistance of S-YH. Y-CP conducted the electrical measurement of the devices. H-CH and Y-ES performed the FTIR spectra measurement. SMS and DHB assisted in the data analysis. All authors read and approved the final manuscript.

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