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Leakage current asymmetry and resistive switching behavior of SrTiO₃

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Raied A. S. Al-Hamadany, Daniel J. R. Appleby, Nikhil K. Ponon, and Anthony O'Neill
*School of Electrical and Electronic Engineering, Newcastle University, Newcastle upon Tyne NE1 7RU,
United Kingdom*

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The resistive switching characteristics of SrTiO₃ metal-insulator-metal capacitors are investigated. The current-density versus voltage (J - V) characteristics show asymmetry at all temperatures examined, with resistive switching behavior observed at elevated temperatures. The asymmetry is explained by the relative lack of electron traps at one electrode, which is determined from the symmetric J - V curve obtained at room temperature due to the redistribution of the dominant electrical defects in the film. We present evidence for the model of resistive switching originating from defect diffusion (possibly oxygen vacancies) at high temperatures. © 2012 American Institute of Physics. [<http://dx.doi.org/10.1063/1.4764544>]

Resistive switching phenomena in metal oxide metal-insulator-metal (MIM) capacitors have attracted significant attention for nonvolatile memory devices,^{1,2} with the underpinning mechanisms thought to depend upon the material.³ However, there are two main models in the literature. The first concerns variation of the Schottky barrier height due to a change in the oxygen-vacancy (OV) concentration at the metal/oxide interface that can be controlled electrically,^{4,5} while the second involves electrically conducting filaments formed by diffusion of oxygen in or out of the metal oxide.^{6–8}

Although these models can explain many features of resistive switching in metal oxides, the relationship between the observed leakage current asymmetry and resistive switching of oxides has not been clearly elucidated. This is partly caused by the asymmetry in leakage current being confused with the resistive switching behavior of oxides.^{9,10} In this letter, J - V characteristics of SrTiO₃ are presented that show both the asymmetry of J - V characteristics and resistive switching at high temperatures. In order to explain the behavior, we extend a previous model¹¹ by considering the role of OVs at the bottom electrode (BE) in more detail.

50 nm SrTiO₃ films were deposited on Pt/TiO₂/SiO₂/Si substrate by atomic layer deposition, followed by rapid thermal annealing at 600 °C in N₂ for 120 s. Circular Pt top electrode (TE) of 100 nm thickness and 150 μm diameter were deposited by electron beam evaporation through a shadow mask. The leakage current-density-voltage (J - V) characteristics were investigated using a HP4155C semiconductor parameter analyzer.

Figure 1 shows J - V characteristics of the leakage current at temperatures between 280 K and 400 K. The voltage is swept at a constant rate from zero bias to 4 V, then to -4 V before returning to zero bias. An asymmetry in the leakage current was observed at all temperatures examined. In addition, the leakage current curves exhibit a pronounced hysteresis between the forward and reverse sweep under both positive and negative directions of the bias variation that increases by temperature.

Previously¹¹ we proposed that the leakage current is through the aligned OVs from TE to BE. The leakage current occurs by electron injection from one electrode to OVs near

the interface by tunneling, followed by hopping conduction between OVs by thermionic emission before tunneling from OVs to the opposite electrode (inset in Fig. 1). Due to the existence of positive charge trapped at the BE interface, the positively charged OVs are electrostatically repelled, and in equilibrium located far from BE. In this case, the tunneling current limits the injection of electrons to the bulk, being highly sensitive to the distance of OVs from the BE. In the model, due to the difference in preparation of the two interfaces, there is no fixed positive charge at the TE, leading to the potential for asymmetric electrical characteristics with respect to bias direction. The absence of the positive fixed charge at the TE allows OVs to lie close to the interface, so that injection does not limit the leakage current here.

In order to describe the processes underpinning resistive switching, we use the displacement and migration of OVs at the BE. Displacement is defined as the reversible movement of OVs under electrical bias, where upon removal of the applied voltage the OVs return to their initial locations. Migration occurs when the field-driven movement of the vacancies is not reversible upon removal of the bias.

At low temperature (<320 K), the OVs cannot surmount the diffusion barriers. Application of a positive voltage bias displaces OVs towards the BE that enhances electron injection and increases the leakage current. A negative voltage displaces OVs away from the BE that suppresses electron injection (Fig. 2(a)). The displacement of OVs at low temperatures leads to small leakage current asymmetry that is in order of 1.4 at 280 K (Fig. 1). By increasing the temperature, there is more thermal energy, increasing the rate of migration. When the voltage increases in the positive direction, the OVs migrate towards the BE leading to a higher electron injection relative to low temperatures (Fig. 2(b)).

At 400 K, by decreasing the voltage from 4 V to 0.5 V and removing the external electric force on OVs, the positive charge trapped at the BE interface repels the OVs, leading to a reduction in current by four orders of magnitude (Fig. 1). However, while approaching zero bias, there remains a remnant of the OVs population close to the BE. When the voltage is swept to negative bias, this remnant is removed, so that upon return to low voltage from the negative bias the

FIG. 1. J - V characteristics for the SrTiO_3 MIM capacitors from 280 to 400 K. Insets are illustrations of conduction mechanism under different voltage polarities.

leakage current is significantly lower than when the remnant is present. This effect is indicated by the vertical arrow at 0.2 V in Fig. 1 where there is a hysteresis of around two decades of current. As a result, when the applied voltage is swept from negative to positive, the leakage current does not follow the same trend as it does when the voltage is swept from positive to negative, demonstrating hysteresis.

In order to examine this model, an experiment was carried out increasing the temperature from 300 K to 400 K and then back to 300 K with the DC voltage fixed at 4 V. The leakage current-density as a function of the changing temperature is shown in Fig. 3(a). Initially it increases in the range from 300 K to 400 K. Upon the subsequent reduction of the temperature under bias, the leakage current-density decreased, but did not return to its initial value: there is nearly two orders of magnitude difference in the initial and final current densities at 300 K.

Figure 3(b) shows the initial and final leakage-current curves at 300 K, as well as the J - V curve at 400 K. The variation is interpreted as follows. Increasing the temperature under the fixed dc voltage of 4 V causes OV's to migrate nearer to the BE interface. In the phase where the temperature is reduced to 300 K, the OV's maintain their location closer to the BE interface due to the applied bias (Fig. 3(a), inset). As a result, the final injection current is greater than at the beginning. By comparing the final 300 K and 400 K J - V characteristics, it can be seen that at higher voltages the

FIG. 3. (a) The leakage-current density at +4 V during the temperature variation from 300 K $\rightarrow$ 400 K $\rightarrow$ 300 K. Inset are OV's locations in relation to the BE before and after annealing cycle. (b) The leakage current at 300 K before and after the annealing cycle, and at the peak temperature of 400 K. Insets are illustrations of the model involving OV's in relation to the BE.

thermionic emission starts to dominate, causing divergence of the two J - V curves (Fig. 3(b)).

Sweeping the voltage to negative values displaces the OV's away from the BE interface. However, at 300 K, subsequent to the thermal excursion to 400 K, the OV's do not have sufficient energy to surmount the diffusion barriers. Therefore, migration does not take place and the electron injection is similar to that seen in the positive voltage sweep.

FIG. 2. A model for the displacement and migration of oxygen vacancies near the bottom electrode interface under an applied voltage at (a) low temperature and (b) high temperature relative to the onset of migration of OV's.

FIG. 4. Effect of temperature on the leakage-current density after annealing the MIM device at 400 K under a positive dc bias of 4 V. Inset is the leakage current variation by increasing temperature at -4 V.

Fig. 3(b) shows that the leakage current at 300 K after the thermal cycle is almost symmetric, with only small differences between the positive and negative bias sides, probably due to OV's displacement. We note that under the negative voltage subsequent to the thermal cycle, the leakage current at 300 K is greater than at 400 K. We interpret this as arising from the migration of vacancies at 400 K away from the BE, which reduces injection, but at 300 K after a thermal cycle under positive bias the vacancies are fixed at the BE interface, enhancing the injection.

The J - V characteristics of the sample that had received a thermal cycle under a fixed $+4$ V bias were investigated at temperatures between 300 K and 380 K. Figure 4 shows the impact upon the leakage current-density characteristics. The leakage-current under positive bias increases with temperature in a fashion similar to the normal operation shown in Fig. 1. However, the leakage-current decreases in the $-4 < V < 0.5$ V range, an effect also observed by Shang *et al.*⁹ This effect is explained by the same mechanism as proposed for the temperature dependence shown in Fig. 3. By increasing the temperature, the OV's can migrate away from the BE under negative bias, allowing them to move to their equilibrium locations. This reduces the injection of carriers, and therefore the leakage current.

In order to estimate the activation energy of OV's, the experiment (Fig. 3(a)) had been repeated. After driving the OV's near to the BE, the temperature was increased under a fixed -4 V bias. The inset in Fig. 4 shows the variation in leakage current with temperature in this case. By increasing the temperature, the leakage current increases by enhancing the thermionic emission of electrons between the OV's. However, after 340 K, OV's begin to migrate away from the BE, leading to a reduction in the injection-current, which in turn reduces the leakage current. The onset of migration at around 340 K is followed by an abrupt decrease in the leakage cur-

rent after 385 K, which is due to a rapid migration of the majority of the population of OV's. After the migration of OV's, the injection-current reduces to a constant value. Therefore, by increasing the temperature, the thermionic emission dominates current, due to a fixed value of injection-current at the BE, which increases the leakage current. A rough estimate of the activation energy can be made by Urbach's approximation¹² $E_T = 23kT$ where k is the Boltzmann constant and T is the temperature. Using this equation, the activation energy lies between 0.67 eV ($T = 340$ K) and 0.76 eV ($T = 385$ K). We note that the calculated OV's migration energy is 0.757 eV,¹³ consistent with our observation of rapid migration at 385 K.

In summary, resistive switching in SrTiO₃ MIM capacitor structures was investigated. By constructing a simple model based upon the movement of electrically active point defects, it is possible to account for all the key features observed. A critical condition is that the leakage current is sensitive to the distance of OV's from the BE due to the positive charge trapped at the interface.¹¹ The asymmetry of leakage current originates from the displacement of OV's relative to the BE, depending upon the temperature and applied bias. At high temperatures, OV migration towards the BE leads to the resistive switching behavior of leakage current, with OV's driven to the BE resulting in a symmetric J - V curve at room temperature.

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