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Anomalous resistive switching phenomenon

Shahin A. Mojarad,¹ J. P. Goss,¹ Kelvin S. K. Kwa,¹ Peter K. Petrov,² Bin Zou,² Neil Alford,² and Anthony O'Neill¹

¹*School of Electrical and Electronic Engineering, Newcastle University, Newcastle upon Tyne NE1 7RU, United Kingdom*

²*Department of Materials, Imperial College London, London SW7 2AZ, United Kingdom*

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Resistive switching was observed in Pt/SrTiO₃/Pt capacitor devices. The switching depends on both the amplitude and polarity of the applied voltage and cannot be described as either bipolar or unipolar resistive switching. We term this behavior antipolar due to the opposite polarity of the set voltage relative to the previous reset voltage. A model based on electron injection by tunneling at interfaces and a Poole-Frenkel mechanism through the bulk is proposed. This model is quantified by use of a simple mathematical equation to simulate the experimental results. © 2012 American Institute of Physics. [<http://dx.doi.org/10.1063/1.4770489>]

I. INTRODUCTION

In the pursuit of emerging non-volatile memory devices, metal oxides have shown significant promise.^{1–3} Ideal memory characteristics should display low power-consumption, fast programming, non-destructive readout, high-density integration, and low fabrication-cost. Resistive random access memory (RRAM) devices have attracted considerable attention due to exhibiting most of these features.^{4–6} A RRAM cell is constructed using metal/insulator/metal capacitors, where the insulator is typically constituted from metal oxide dielectrics. The resistivity of metal oxides can be electrically switched between a low resistance state (LRS) and high resistance state (HRS), where the current-voltage ($I-V$) characteristics exhibit a pronounced hysteresis.

Several models have been proposed to explain the underpinning mechanisms of hysteresis in $I-V$ characteristics in metal oxides. The models cover a wide range of chemical and physical mechanisms,^{7–11} but it has proven difficult to find a simple unified model. However, the hysteresis in $I-V$ characteristics can be broadly grouped into two main categories: bipolar and unipolar resistive switching.

In bipolar resistive switching (BRS), the switching depends upon the polarity of the applied voltage (V). A cell can be set to the LRS by $V > V_{\text{set}}$ or reset to the HRS by $V < -V_{\text{reset}}$ (Fig. 1(a)). In contrast, during unipolar resistive switching (URS), the switching depends only on the amplitude of the applied voltage, and not the polarity. Fig. 1(b) shows a schematic for URS, where the oxide resistance can be reset to the HRS by applying $V_{\text{reset}} < |V| < V_{\text{set}}$. The LRS is then achieved via application of $|V| > V_{\text{set}}$.

Binary oxides including NiO,^{12,13} ZnO,^{14–16} HfO₂,^{17–19} TiO₂,²⁰ and ZrO₂²¹ have been reported to show URS and/or BRS. BRS behavior has been also observed in perovskite structure metal oxides such as BiFeO₃,²² SrZrO₃,²³ SrTiO₃,⁷ PrCaMnO₃,²⁴ as well as in perovskite heterostructures.²⁵

In this paper, we report resistive switching in Pt/SrTiO₃/Pt capacitors with switching characteristics that cannot readily be categorized as either BRS or URS. The $I-V$ characteristics show that the resistive switching depends on both the

polarity and amplitude of the applied voltage (Fig. 1(c)). The SrTiO₃ resistance can be reset to the HRS by application of a high voltage with either polarity. A voltage of the opposite polarity to that used to generate the HRS sets the LRS. Thus, if a high positive voltage has reset the device, the polarity of the set voltage must be negative. Similarly, if the reset voltage was negative, the polarity of set voltage must be positive. We call this behavior antipolar resistive switching (ARS) due to the opposite polarity of the set voltage with respect to previous reset voltage.

The differences between the operation of BRS, URS, and ARS devices are summarized in Table I. The contrast between the set and reset voltage magnitudes and polarities shows that the ARS behavior of the SrTiO₃ devices characterized in this study do not follow either the BRS or URS behavior of oxides. In this paper, we propose a model based on electron injection from the electrode to trap states near the interface by tunneling, followed by hopping conduction between traps via thermionic emission, supported by a simple mathematical framework that quantitatively supports the conduction model.

II. EXPERIMENT

A 100 nm thick Pt film was deposited on a Ti/SiO₂/Si substrate to act as the bottom electrode in the capacitor. A 25 nm thick SrTiO₃ thin film was deposited on the Pt by pulsed laser deposition at a substrate temperature of 700 °C. The Pt/Ti/SiO₂/Si substrate was secured by silver paste onto the stainless-steel resistive heater with a target to substrate distance of 50 mm. The SrTiO₃ film was grown by laser ablation (Neocera pulse laser deposition system with a Lambda Physik KrF laser, $\lambda = 248$ nm) using a 20 mm diameter stoichiometric SrTiO₃ target in an oxygen pressure of 300 mTorr. The substrate temperature during deposition was controlled using a thermocouple embedded in the heater. The energy density of the laser spot ($1.5 \times 8 \text{ mm}^2$) was 1.2 J/cm^2 , while the laser pulse repetition rate was 8 Hz. Once the ablation was over, the sample was cooled down at a rate of 10 °C/min in an oxygen rich environment

FIG. 1. Schematic representations of the ideal $I-V$ and $R-V$ hysteresis behavior of (a) bipolar resistive switching, (b) unipolar resistive switching, and (c) antipolar resistive switching.

(700 Torr). The sample thickness was measured using a Dek-tak 150 contact profiler. The Pt top electrodes (100 nm) were deposited by electron beam evaporation, and patterned by a lift-off process for electrical characterization.

The electrical characteristics were obtained using a HP4155C semiconductor parameter analyzer. The voltage biases were applied on top electrodes while the bottom electrode was grounded.

III. ELECTRICAL RESULTS

Figure 2(a) represents 100 switching loops, illustrating the repeatability of the resistive switching behavior. The cycling between the low and high resistance states can be characterized by switching in the following fashion.

By increasing the positive bias from zero, the SrTiO_3 resistive state changes from HRS to LRS with $V > 0.2$ V, as shown in Fig. 2(b). As the bias is further increased, the SrTiO_3 switches to the HRS at ≈ 1 V. Subsequently, during reduction of the bias to zero the device remains in the HRS.

From zero bias, the sweeping of the voltage in the opposite direction results in switching to the LRS at $V < -0.2$ V. In accord with the magnitude of the reset voltage in positive

bias, the device is switched to the HRS around -1 V, and the device remains in the HRS until a set voltage of >0.2 V is applied.

FIG. 2. (a) $J-V$ characteristics of SrTiO_3 for 100 consecutive sweeping loops between -3 and $+3$ V. (b) The $I-V$ characteristics in the antipolar resistive switching behavior of the Pt/ SrTiO_3 /Pt capacitive devices.

TABLE I. Characteristics of bipolar, unipolar, and antipolar resistive switching in terms of the conditions required to set the devices to the low resistance state and reset to the high resistance state.

	Reset voltage	Set voltage	$V_{\text{set}} \Leftarrow V_{\text{reset}}$
Bipolar	$V < -V_{\text{reset}}$	$V > +V_{\text{set}}$	$V_{\text{set}} \approx V_{\text{reset}}$
Unipolar	$V_{\text{reset}} < V < V_{\text{set}}$	$ V > V_{\text{set}}$	$V_{\text{set}} > V_{\text{reset}}$
		If $V > V_{\text{reset}}$:	
		$-V_{\text{reset}} < V < -V_{\text{set}}$	
		If $V < -V_{\text{reset}}$:	
Antipolar	$ V > V_{\text{reset}}$	$V_{\text{set}} < V < V_{\text{reset}}$	$V_{\text{set}} < V_{\text{reset}}$

In order to further rule out BRS and URS, the device has been cycled 25 times in three different bias ranges (Fig. 3(a)). The voltage cycling in the HRS only with a positive polarity between $0 < V < 2.5$ V (red curves) shows no hysteresis. Since the resistive state of the structure does not change with the magnitude of the bias, the resistive switching behavior is not unipolar. Similar behavior is observed sweeping the voltage with negative polarity (green curves). The SrTiO_3 shows the same resistance for either polarity of voltage which is in contrast with BRS where the resistive state of the structure is switched by changing the polarity of the voltage, and as a result BRS is also ruled out. By cycling the voltage over the range $-1 < V < 1$ V (blue curves), the SrTiO_3 is in the LRS because the device is never exposed to the reset threshold voltage, and although there is some hysteresis, it is much less significant than that seen in the large voltage range (Fig. 2).

After each set of experiments depicted in Fig. 3(a), the resistive state of SrTiO_3 has been checked by cycling the voltage in the range of $-0.2 < V < 0.2$ V 25 times, with the resulting characteristics shown in Fig. 3(b). Sweeping the voltage between zero and high positive or negative voltage ($|V| > 1$ V) resets the SrTiO_3 into the HRS. By sweeping the voltage between -1 and 1 V, the SrTiO_3 is maintained in the LRS.

IV. MODELING THE ANTIPOLAR RESISTIVE SWITCHING

Conduction through metal oxides can be modeled using the Poole-Frenkel equation.²⁶ The electrical current originates from electron transport via the conduction band by trapping and detrapping of carriers at localized states, expressed by

$$J_{PF} = C\bar{E} \exp \left[-e \left(\phi_T - \sqrt{\frac{e\bar{E}}{\pi\epsilon}} \right) / kT \right], \quad (1)$$

where C is a constant, $\bar{E}$ is the electric field strength, e is the electron charge, k is Boltzmann's constant, T is the absolute temperature, ϕ_T is the location of the trap below the conduction band minimum, and ϵ is the permittivity of the material. Application of Eq. (1) assumes that the interface is ideal, so that there is efficient injection of electrons from the electrode into the dielectric film.

In several metal oxides, it has been observed that at low electric field there is a small current that increases abruptly

to high values by increasing the voltage.²⁷ The abrupt transition between low and high leakage-current may indicate that the injection of electrons into the bulk is suppressed at low voltages. Hence, the impact of the interface on injection of electrons must be combined with the Poole-Frenkel equation to account for this phenomenon. This had previously been modeled in a theory of trap-assisted charge-injection.²⁸ Recently, another model has been proposed based upon a tunneling-assisted Poole-Frenkel equation.²⁹ Both studies suggest that the conduction mechanisms in metal oxides originate from the injection of electrons into the bulk by tunneling to trap states near the interface, followed by thermal activation of electrons between the traps inside the dielectric film. The equation for conduction may be summarized by

$$P = P_{\text{injection}} \times P_{\text{excitation}}, \quad (2)$$

where P is the probability of an electron will pass through the oxide, which is equal to the product of electron injection probability at the interface ($P_{\text{injection}}$) and the probability that an electron will pass through the bulk by thermal excitation via trap bound-states and the conduction band of the insulator ($P_{\text{excitation}}$). At low voltage, P is limited by $P_{\text{injection}}$ due to low electron injection at the interface. At high voltages, $P_{\text{excitation}}$ limits the total current where there is sufficient tunneling injection of electrons into the dielectric. $P_{\text{excitation}}$ is given by the Poole-Frenkel equation (Eq. (1)), where the exponential term shows the probability of electron thermal activation from the trap states into the conduction band. The thermal excitation of electrons can be increased by lowering the potential barriers between traps, where the term $\sqrt{e\bar{E}/\pi\epsilon}$ in Eq. (1) describes the effect of an electric field upon the barriers. $P_{\text{injection}}$ has been modeled by different tunneling equations,^{28,29} where the location of traps relative to the interface have been assumed to be fixed. The electric field increases electron injection by lowering and/or narrowing the barrier between the traps and the interface.

By using Eq. (2), the current characteristics in metal oxides can be accurately modeled.³⁰ However, in order to model the ARS behavior of SrTiO_3 , an additional mechanism must be added into Eq. (2). First, in a metal/insulator/metal structure, the leakage current occurs by electron injection from an electrode to the bulk and from the bulk to the opposite electrode (Fig. 4). As a result, both interfaces must be included independently, and Eq. (2) is modified to

$$P = P_{BE} \times P_{\text{excitation}} \times P_{TE}, \quad (3)$$

where P_{BE} and P_{TE} are the probabilities of electron injection at the bottom and top electrodes, respectively.

FIG. 3. (a) Current-density-voltage characteristics for 25 cycles under different conditions: positive bias (red curves), negative bias (green curves), and $-1 < V < 1$ V (blue curves). (b) Confirmation of the resistive state in each case from (a) via variation of the bias in the $-0.2 < V < 0.2$ V range.

FIG. 4. The electron injection from an electrode to the bulk and from the bulk to the opposite electrode at different voltage polarities.

Second, in resistive switching metal oxides, it is currently understood that there is migration of traps during the application of a bias, changing the spatial distribution of the electron traps. In SrTiO₃ the traps probably arise from oxygen vacancies, which act as deep double donors.^{31,32} By changing the polarity of the voltage, the oxygen vacancies migrate nearer to the top or bottom electrode (Fig. 4). Consequently, the distance between oxygen vacancies and the electrodes can be changed electrically. If this is the case, the electric field alters the tunneling barrier width at the interfaces, by affecting the distance between traps and the electrodes. Thus, the tunneling equation needs modification, since they are based upon fixed location of traps.^{28,29}

To incorporate the mechanisms involving the charge carriers and mobile deep traps, we have constructed a simple, one-dimensional model, depicted in Fig. 5(a), which can be divided into two parts. The first part relates to the carrier injection at both interfaces, and the second part relates to the movement of oxygen vacancies driven by the electric field inside the dielectric film.

For the tunneling injection, the simplified approach is to assume a square potential of fixed height, V_0 , and width defined by the distance from the interface to the closest deep trap, denoted d_{BE} and d_{TE} corresponding to the bottom and top electrodes, respectively. The resulting tunneling probability is given by

$$P_T(d) = 16 \left(\frac{E}{V_0} \right) \left(1 - \frac{E}{V_0} \right) \exp(-2Kd), \quad (4)$$

where E is the electron energy, V_0 is the potential barrier height, d is barrier width, and $K = \sqrt{2m(V_0 - E)/\hbar^2}$, where m is the electron effective mass. As a result of the presence of the two, in general, non-equivalent junctions, the total $P_{injection}$ is

$$P_{injection} = P_T(d_{BE}) \times P_T(d_{TE}). \quad (5)$$

The barrier widths, d_{BE} and d_{TE} , are modeled by allowing a fixed number of point charges to respond to the applied electric field. Based upon the probable oxidation states of oxygen vacancies, three $+2e$ point charges were allowed to move between the electrodes without any barrier (Fig. 5(a)).³² In addition, to confine the movement of charges between electrodes, two fixed small charges of $+2e/100$ were placed at the interfaces. The total distance between the electrodes was taken to be 25 nm, equal to that of the experimental sample thickness.

Based upon this model, values of d_{BE} and d_{TE} were obtained, along with the nominal distances between the point charges, d_1 and d_2 , by varying the voltages and minimizing the electrostatic forces between the charges (Fig. 5(b)). As would be expected, a positive voltage applied to the top electrode results in the oxygen vacancies migrating towards the bottom electrode so that d_{BE} decreases and d_{TE} increases. For a negative voltage polarity, the oxygen vacancies are attracted to the top electrode, and d_{BE} increases while d_{TE} decreases. Using the geometries obtained by this model, the current density can be expressed as

$$J = P_T(d_{BE}) \times P_T(d_{TE}) \times J_{PF}. \quad (6)$$

Fig. 5(c) depicts the variation of injection current where the applied field results in changing d_{BE} and d_{TE} . The maximum value for $P_{injection}$ occurs at zero bias where d_{BE} and d_{TE} are equal. By increasing amplitude of the voltage, J_{PF} increases while $P_{injection}$ decreases resulting in maxima current at ± 1 V.

In the above analysis, the point charges are free to move in response to the electric field, but in real materials, in addition to the long-range electrostatic force between oxygen vacancies, there are also diffusion barriers. Over short time-scales the existence of diffusion barriers may inhibit the redistribution of the defects, and equilibration may not have sufficient time take place. As a result, the oxygen vacancies can be driven near to one electrode by increasing the bias, but they do not necessarily redistribute uniformly through the film upon removal of the bias.

To include the kinetics of migration in a fashion consistent with our observations (Fig. 2(b)), we include two threshold values of bias. The oxygen vacancy migration towards the opposite electrode occurs when a voltage with opposite polarity is applied with an amplitude of 0.2 V, which is equal to the “set” voltage. In addition, increasing the voltage moves the oxygen vacancies towards the bottom or top electrodes up to ± 2.2 V (Fig. 6). Above this voltage, the oxygen vacancies do not move nearer to the interface and $P_{injection}$ does not change. Fig. 7(a) shows the simulation results by

FIG. 5. (a) A simple model with square barriers between the oxygen vacancies and interfaces. (b) Schematic of the location of the deep traps within the oxide film under different voltage biases. (c) The variation of injection current due to the movement of oxygen vacancies.

FIG. 6. Specification of the injection probability, $P_{injection}$, as a function of the bias magnitude and polarity.

using the values of Fig. 6 for $P_{injection}$ assuming $\phi_T = 0.6$ eV and $V_0 = 1$ eV.³⁰

Fig. 7(b) illustrates the movement of oxygen vacancies through the bulk at different voltage biases. Application of a high voltage drives the oxygen vacancies away from one electrode, resulting in the HRS as a consequence of the low electron injection at this electrode. The LRS can be achieved by redistributing the oxygen vacancies through the bulk by reversing the low voltage polarity.

V. SCHOTTKY AND POOLE-FRENKEL SWITCHING BEHAVIOR

The antipolar behavior presented in this paper has been modeled based upon a Poole-Frenkel conduction mechanism. However, we note that resistive switching might be due to a Schottky mechanism. Yang *et al.* recently showed that the oxygen vacancies in BiFeO₃ can accumulate near one electrode by application of different voltage polarities.³³

Sweeping the voltage produced a hysteretic I - V characteristic caused by changes in the distribution of oxygen vacancies through the film.

In order to discriminate between Schottky and Poole-Frenkel mechanisms, we present illustrations of both hysteresis switching behaviors in Fig. 8. For Schottky devices (Fig. 8(a)), when a high negative voltage is applied to one electrode, the oxygen vacancies respond electrostatically and move towards this electrode, increasing the n-type carriers near the interface.³³ This would result in an increase in electrons tunneling into the conduction band due to narrowing of the potential barrier at the Schottky contact. Driving the oxygen vacancies near one electrode also reduces the n-type carriers at the opposite electrode, which in turn yields a wider Schottky barrier at this electrode. Then, the negatively biased electrode is a more Ohmic-like contact while the grounded electrode is a Schottky contact (Fig. 8(a)). Therefore, upon application of a small, positive voltage, there would be low electron injection from the grounded electrode to the film due to the Schottky contact, resulting in a low leakage current. By increasing the voltage in the positive direction, the oxygen vacancies move towards the grounded electrode and transform the contact from Schottky to Ohmic-like which increases the leakage current up to the point that the injection at the biased electrode becomes the limiting factor. Therefore, in Schottky devices, the oxygen vacancies act as the mobile donors,³³ where the leakage current through the film is dominated by movement of electrons through the conduction band and not via hopping between defect states.

In contrast, in the case of Poole-Frenkel conduction (Fig. 8(b)), the oxygen vacancies act as the traps where the leakage current occurs by electrons tunneling from one electrode to the trap states near the interface, followed by trap and detrapping between the trap states through the bulk by thermionic emission, and tunneling from the trap states to the opposite electrode.

As a result, in Schottky devices a high voltage bias increases the leakage current by increasing the injection of electrons from electrode to the bulk, but it reduces the leakage current in Poole-Frenkel devices by reducing the injection of electrons from the bulk to the electrode. Hence, although Schottky devices have similar hysteresis switching curves to Poole-Frenkel devices, the sense of operation with bias is reversed, allowing the mechanisms to be distinguished.

FIG. 7. (a) The simulation results based on the calculated values of $P_{injection}$. (b) Scheme movement of oxygen vacancies through the SrTiO₃ at different voltage bias by including the threshold values.

FIG. 8. The resistive switching behavior of metal oxides when oxygen vacancies act as the mobile (a) donors and (b) traps.

VI. THREE-STATE RRAM

The peculiar ARS behavior might give rise to some novel applications in electronic devices. From the RRAM application view point, ARS exhibits three stable states. In conventional BRS and URS RRAM, a bit can be stored by applying different voltages to the MIM structure which result in reversible switching between LRS (representing digital “1”) and HRS (representing digital “0”). In ARS, redistribution of oxygen vacancies through the bulk leads to the LRS, which represents digital “1” (Fig. 9(a)). However, the HRS includes two distinct states that originate from the polarity of the “reset” voltage. A positive “reset” voltage drives the oxygen vacancies close to the bottom electrode (Fig. 9(b)) while a negative “reset” voltage drives oxygen vacancies near the top electrode (Fig. 9(c)). These two different high resistant states can be revealed by application of a “set” voltage. For instance, if the device has been reset to its HRS by a positive voltage, then application of a positive “set” voltage does not switch the device to LRS. And if the device has been reset to its HRS by a negative voltage, then application of positive “set” voltage switches the device to LRS. In other words, the device memorizes the polarity of “reset” voltage and ARS can be used as a device with three states of a LRS, a positive HRS (when $V > V_{\text{reset}}$ has been used to “reset” the device) and a negative HRS (when $V < -V_{\text{reset}}$ has been used to “reset” the device).

FIG. 9. Three stable states of RRAM: (a) low resistance state, (b) positive high resistance state, and (c) negative high resistance state.

(b) Poole-Frenkel

VII. CONCLUSIONS

The conduction mechanism of antipolar resistive switching has been elucidated by developing a simple mathematical model, where the leakage current is sensitive to the location of oxygen vacancies relative to the electrodes. The application of a high voltage bias can drive oxygen vacancies near one electrode and switch the oxide into a high resistance state. The low resistance state can be achieved by redistributing the oxygen vacancies throughout the film by a low voltage with opposite polarity relative to the previous “reset” voltage.

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