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A comprehensive study on the leakage current mechanisms of Pt/SrTiO₃/Pt capacitor

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The leakage current characteristics of SrTiO₃ MIM capacitors, fabricated using atomic layer deposition, are investigated. The characteristics are highly sensitive to the polarity and magnitude of applied voltage bias, punctuated by sharp increases at high field. The characteristics are also asymmetric with bias and the negative to positive current crossover point always occurs at a negative voltage bias. In this work, a model comprising thermionic field emission and tunneling phenomena is proposed to explain the dependence of leakage current upon the device parameters quantitatively. © 2012 American Institute of Physics. [doi:10.1063/1.3673574]

I. INTRODUCTION

Transition metal oxide junctions have attracted considerable attention due to their potential for applications such as dynamic random access memory (RAM),¹ ferroelectric RAM,² and resistive switching RAM.^{3–5} For such applications, an important factor to be addressed is the leakage current that affects both the quality and reliability of the devices. Leakage current through transition metal oxides has been the subject of extensive discussion, and different models have been proposed to explain it.^{6–10} Thermionic field emission equations (i.e., Schottky or Poole-Frenkel) are frequently used to explain temperature dependent characteristics of leakage current, which is mainly attributed to oxygen vacancies.^{4,7,8,11,12} The conduction mechanism of the oxygen vacancies was also found to vary according to the difference in the thermionic field emission types (either Schottky or Poole-Frenkel).

In the case of Poole-Frenkel emission, the oxygen vacancies are regarded as traps inside the bulk.^{7,8} Leakage current originates from the thermionic field emission of electrons between oxygen vacancies (Fig. 1(a)) and can be expressed as

$$J_{PF} = CE \exp \left[-e \left(\phi_{BPF} - \sqrt{\frac{eE}{\pi \epsilon_0 \epsilon_r}} \right) / kT \right], \quad (1)$$

where E is the electric field strength, C is a constant, e is the electron charge, k is Boltzmann's constant, T is the temperature, ϕ_{BPF} is the location of the trap level below the conduction band minimum, and ϵ_r is the dielectric constant.

For Schottky emission, the bulk is regarded as highly conductive where defects such as oxygen vacancies act as dopants.^{4,11–13} The leakage current is then due to thermionic field emission of electrons over the effective Schottky barrier (Fig. 1(b)). The current is then

$$J_{Sch} = A^* T^2 \exp \left[-e \left(\phi_{BSch} - \sqrt{\frac{eE}{4\pi \epsilon_0 \epsilon_r}} \right) / kT \right], \quad (2)$$

where A^* is the Richardson constant and ϕ_{BSch} is the effective Schottky barrier height. An electric field can drive diffusion of charged centers such as oxygen vacancies, changing their concentration profiles at the interface, resulting in a field dependent variation of ϕ_{BSch} , and subsequently, an asymmetry in the leakage current.

Both equations can explain the existence of temperature-dependent leakage characteristics, but the underlying conduction processes are related to the dielectric in fundamentally different ways: Poole-Frenkel is limited by conduction mechanisms in the bulk region without any limit on the carrier injection, whereas Schottky emission is limited by injection at the interface, with bulk conduction being relatively high, mediated by defects such as oxygen vacancies. In order to reduce the leakage current, it is important to determine which of these mechanisms dominates. For instance, if it is Schottky emission, the interface controls the leakage current so that using different electrode materials might improve the leakage current.^{14,15} In contrast, if Poole-Frenkel is the dominant mechanism, then dielectric quality controls the leakage current and bulk treatments such as oxygen annealing might be deployed.¹⁶

However, there are several discrepancies with the existing models in respect of the impact of dielectric film quality or interface on leakage current. The Poole-Frenkel model proposed previously^{7–9,17–19} suggests that bulk controls the leakage current while the Schottky emission model^{18,20–25} indicates interface as a dominant factor.

Moreover, restriction to just the interface mechanism or just the bulk mechanism can lead to inaccurate modeling of leakage current, for example in the calculation of the dielectric constant from the thermionic field emission equations obtained from the leakage current in the positive voltage region.^{8,16,23} From Eqs. (1) and (2), the dielectric constant is defined by the slope of the leakage current curves on a logarithmic scale. Under normal circumstances ϵ_r is greater than

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FIG. 1. (Color online) Scheme thermionic field emission of electrons (a) between the oxygen vacancies in bulk that can be modeled by Poole-Frenkel emission equation and (b) over the effective Schottky barrier height that can be modeled by Schottky emission equation.

unity,²⁶ but dielectric constants obtained from the slope can be unphysically small ($\ll 1$).

In this study, the results of a study into leakage-current mechanisms in SrTiO₃ metal–insulator–metal (MIM) capacitors are presented. A model including both interface and bulk effects upon the leakage current is proposed. We show that this model demonstrates an improved agreement to experimental current density both as a function of voltage and temperature compared to previous published models.^{16,18,20,23,25} The model also indicates the existence of fixed positive charges at the bottom electrode leading to the characteristic asymmetry observed in the current-density–voltage ($J - V$) characteristics.

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. The 10 nm Ti flash-layer provides adhesion between Pt and silicon substrate. On top of the Pt electrode was deposited a 50 nm layer of SrTiO₃ via atomic layer deposition (ALD) at a reactor temperature of 250 °C. After deposition, the sample was annealed at 600 °C for 120 s in N₂. The crystal structure of the annealed film was characterized by x-ray diffraction, which showed strong SrTiO₃ (110) peaks, consistent with the expected perovskite structure. Following the anneal, circular Pt electrodes with a thickness of 100 nm and diameter of 150 μ m were deposited to form MIM capacitors used in electrical characterization.

The leakage current characteristics were measured using an Agilent 4155 C Semiconductor Parameter Analyzer across a temperature range of 300–380 K. Current-atomic force microscopy (C-AFM) was performed using a Park Systems XE-150 employing a CDT-NCHR tip.

III. MODELING THE LEAKAGE CURRENT

Figure 2 shows the $J - V$ characteristics of a SrTiO₃ MIM capacitor. For positive electric fields, there are two distinct regions of behavior: low voltage ($V_b < 0.5$ V) where the leakage current is less than 10⁻⁸ A/cm² and high voltage ($V_b > 1.3$ V) where the leakage current increases with increasing temperature. Each $J - V$ characteristic exhibits an abrupt transition between the two regions at a voltage which increases with temperature ($0.5 < V_b < 1.3$ V). At high neg-

ative voltage bias ($V_b < -1$ V), the measured current shows a temperature-dependence similar to that observed for positive biases. However, the leakage current for negative voltages is lower than the corresponding positive voltages, leading to an asymmetry in leakage-current curves.

The temperature dependence of the leakage current at $V_b > 0.5$ V is suggestive of a thermionic field emission, indicating a current dominated by either Poole-Frenkel emission, Eq. (1), or Schottky emission Eq. (2). The temperature-independence of low leakage current for $V_b < 0.5$ V may imply a current blocked at the metal-insulator interface. Based upon these observations, a likely model might be that the current is dominated by a bulk conduction mechanism where at low voltage, the interface blocks injection of electrons to the bulk. Indeed, at high voltage, the leakage current is modeled very accurately by Eq. (1), where the electrons move through the bulk by hopping between localized states such as might arise from oxygen vacancies inside the insulator (Fig. 1(a)). At low voltage ($V_b < 0.5$ V), the leakage current is dominated by the interface where the injection of electrons into the bulk is blocked. In the range $0.5 < V_b < 1.3$ V, injection most likely occurs by tunneling of electrons from the electrode to the defect states near the interface, which may be modeled by the trap-assisted-tunneling current²⁷

$$J_{TAT} = A \exp \left(- \frac{8\pi\sqrt{2}\epsilon m_s \phi_{BTAT}^{3/2}}{3hE} \right), \quad (3)$$

where A is a constant, m_s is effective mass of the electron, h is Planck's constant, and ϕ_{BTAT} is barrier height. In this model, ϕ_{BTAT} is the difference of ϕ_{BSch} and ϕ_{BPF} , as the electrons tunnel from the electrode to the defect states near the interface.

Based on this model, electrons are injected into deep states near the interface by tunneling and subsequently move through the bulk by thermal emission between traps throughout the dielectric film. At low voltage, the electric field is not high enough for injection and the Poole-Frenkel mechanism is suppressed by the interface. By increasing the voltage, the

FIG. 2. (Color online) $J - E$ characteristics of Pt/SrTiO₃/Pt capacitor for both negative and positive voltage biases over temperature range 300 K to 380 K.

potential across the dielectric is sufficient to result in injection of electrons to the states near the interface, causing an abrupt increase in the leakage current, as shown in Fig. 2. At this point, the thermionic field emission of electrons between the oxygen vacancies limits the leakage current and the dominant mechanism is Poole-Frenkel.

In order to determine values for the physical parameters in the Poole-Frenkel and trap-assisted-tunneling equations, the experimental data is plotted in two ways (Fig. 3). For Poole-Frenkel, plotting $\ln(J/E)$ against $E^{1/2}$ and Fig. 3(a) show linearity for $V_b > 1.3$ V. In the case of trap-assisted-tunneling, plotting $\ln(J)$ against $1/E$ also yields a straight-line fit, but over the complementary range of $V_b < 1.3$ V, as shown in Fig. 3(b). However, the latter plot shows an increase in current density with increasing temperature, inconsistent with Eq. (3), where that trap-assisted-tunneling is temperature independent. Clearly, this experimentally observed temperature dependence shows that at low field, the mechanism cannot be purely tunneling of carriers through an interface barrier.

The temperature dependence can be introduced in Eq. (3) via the constant A , which defines the magnitude of tun-

neling current at high electric field. As the first step, the experimental characteristics were fitted to Eq. (3) for the data obtained at 380 K. From the gradient (Fig. 3(b)), ϕ_{BTAT} is calculated to be 0.18 eV, using $m_s = 0.05m_e$ with m_e being the free electron rest-mass.²⁰ The constant, A , was then obtained for the remaining temperatures using this value of ϕ_{BTAT} . The resulting temperature dependence of A is plotted in Fig. 4, which has an Arrhenius form. The pre-factor, $A(T)$, can, therefore, be expressed as

$$A = A_0 \exp\left(-\frac{e}{kT} \phi_B\right), \quad (4)$$

where from the gradient of the line, the activation energy, ϕ_B , is calculated to be 0.82 eV. Combining Eqs. (3) and (4) provides the current-density for $V_b < 1.3$ V

$$J = A_0 \exp\left(-\frac{e}{kT} \phi_B\right) \exp\left(-\frac{8\pi\sqrt{2em_s}\phi_{BTAT}^{3/2}}{3hE}\right). \quad (5)$$

Equation (5) fits the observations very well. Noting the similarity between the form of the pre-factor and the Poole-Frenkel emission formula suggests the leakage-current can be represented by the product of the two mechanisms, Poole-Frenkel and trap-assisted-tunneling

$$J = J_{PF} \times J_{TAT}. \quad (6)$$

To understand the physical meaning of Eq. (6) further, the parameters of the Poole-Frenkel equation need to be explored in more detail, taking into account the proximity of the electron traps to the interface. Equation (1) consists of a constant, C , and an exponential term. When the deep electron traps lie near the interface, the injection of electrons tunneling through the interface barrier into the nearest trap is highly probable and nearly independent of V_b . Therefore, the leakage current will be limited by the hopping conduction between the deep states, such as those arising from the oxygen vacancies. In this case, C in Eq. (1) represents the current density at low voltage, arising from the hopping of

FIG. 3. (Color online) Extracting the parameters of Poole-Frenkel and trap-assisted-tunneling equations in the temperature range of 300–380 K from (a) $\ln(J/E)$ vs $E^{1/2}$ (Poole-Frenkel plot) and (b) $\ln(J)$ vs $1/E$ (trap-assisted-tunneling plot).

FIG. 4. (Color online) The calculated values of A as a function of $1/kT$.

FIG. 5. The relationship between the electric field and tunneling current, which shows how the injection of electrons to the vacancies is changed by electric field.

electrons that have enough energy to overcome the potential barriers between the defects. By increasing the bias, and hence lowering the effective barrier height for hopping, more electrons can contribute to the leakage current, which thus increases. This bias effect corresponds with the exponential term in Eq. (1).

However, the picture becomes modified when the defects in the SrTiO₃ are located further from the interface. Where this is the case, a significantly higher voltage is required to inject the electrons to the vacancies by tunneling, as the barrier width is effectively much bigger. The injection rate will then begin to become significant at a higher V_b compared to where the traps are in close proximity to the interface, ($0.5 < V_b < 1.3$ V). This can be observed in the experimental characteristics (Fig. 2) where there is an abrupt transition between low and high leakage current. This would correspond to a V_b dependence of C in Eq. (1).

The effect is illustrated in Fig. 5. This shows the calculated injection current from tunneling as a function of applied voltage V_b using Eq. (3), with ϕ_{BTAT} taken to be the value of 0.18 eV extracted from the fit to Fig. 3(b). By increasing the voltage V_b from 0.25 to 1.3 V, the tunneling current increases by more than four orders of magnitude. Above 1.3 V, the current increases much less dramatically and by less than an order of magnitude up to 2 V. As a result, in Eq. (5), the trap-assisted-tunneling term defines the variation of C by voltage and Poole-Frenkel term explains the temperature dependence of the thermionic field emission of electrons. Equation (5) remains valid for $V_b > 1.3$ V where the trap-assisted-tunneling is saturated, and by combining the Poole-Frenkel and trap-assisted-tunneling equations, a more general expression for the leakage current-density is obtained as follows:

$$J = BE \exp \left(- \frac{8\pi\sqrt{2em_s}\phi_{BTAT}^{3/2}}{3hE} \right) \times \exp \left[-e \left(\phi_{BPF} - \sqrt{\frac{eE}{\pi\epsilon_0\epsilon_r}} \right) / kT \right], \quad (7)$$

where B is a constant. In terms of the global agreement with the $J - V$ characteristics, Eq. (7) is a substantial improvement over models proposed previously.^{16,18,20,23,25}

IV. DISCUSSION AND CONCLUSIONS

Figure 6 shows the results of modeling based on Eq. (7) including both the positive and negative voltage ranges. The values of the physical parameters used are listed in Table I. Both these parameters and the $J - V$ traces show a marked polarity asymmetry. Figure 7 shows the $J - V$ data for 300 K, which exhibits a minimum in the leakage current at a negative bias.

It is important that the source of the asymmetry is identified. In the devices used in this study, we propose that the difference between the characteristics with forward and reverse bias is produced by the existence of fixed positive charges at the bottom SrTiO₃/Pt interface. They give rise to an internal electric field at the interface which combines the applied field. At low negative bias, where the internal electric field is greater than the applied electric field, the current is in the direction opposite to that which would be generated by the applied field alone, as observed.

In our proposed model to explain the asymmetry, the fixed positive charges at the bottom electrode repel mobile electron donors, such as positively charged oxygen vacancies, increasing the distance between them and the electrode. Consequently, as outlined above, there will be a wider barrier to injection, and a concomitantly larger bias required to saturate the tunneling injection of carriers into the oxide film. This is reflected in the $J - V$ characteristics where at low voltage, the injection current due to tunneling is very low.

This can also be viewed in terms of the combination of fixed and mobile positively charged defect centers. By increasing the positive voltage bias at the top electrode, positively charged mobile defects are pushed towards the bottom electrode. The positive bias tends to offset the repulsion from the fixed positive charges at the lower electrode, so the positively charged mobile defects are effective in producing a tunneling current. In contrast, when a negative bias is applied, the mobile defects are directed away from the bottom electrode as the internal and external electric fields add to enhance the field driven diffusion. This results in an increase in the distance between the oxygen vacancies and the bottom electrode. Hence, the electron tunneling occurs at higher voltage. However, due to greater distances, the injection of electrons at negative voltage is always lower than for positive voltage. In the model, for the case of the top electrode, there is no fixed positive charge at the interface and as it is exposed to the atmosphere, oxygen vacancies can be created near this interface. Hence, the top electrode does not limit the injection of electrons to the deep traps.

A remaining question exists regarding the nature of the fixed charges at the lower interface. Unfortunately, the experiments performed for this study are not able to reveal their structure or composition. But charge trapping at interface states has been documented previously,^{28,29} with their presence resulting in a measurable conduction-band-bending,

FIG. 6. (Color online) (a) Results of experimental matching based on Eq. (7) at negative and positive voltage. (b) Negative voltage bias at top electrode and low injection of electrons to the bottom electrode due to high distance of oxygen vacancies from bottom electrode. (c) Positive voltage bias at top electrode and high injection of electrons to vacancies due to small distance between oxygen vacancies and bottom electrode.

which in our devices would differ between the top and bottom SrTiO₃/Pt interfaces. With a larger band-bending at the lower electrode, when electrons are injected from the bottom to the mobile traps by tunneling, there is a larger barrier height presented that electrons need to surmount compared with that at the top electrode. The difference of barrier heights is illustrated schematically in Figs. 6(b) and 6(c), where the values of the barrier heights are taken from Table I.

Finally, we have also examined the conduction through the oxide films using C-AFM to resolve the uniformity of the leakage across the film. A typical C-AFM image is shown in Fig. 8(a). There are clearly discrete points through which the leakage current passes most efficiently, which might arise from conductive filaments^{30–32} through the dielectric (Fig. 8(b)). From this analysis, single-point $I - V$ measurement have been carried out by setting the location of AFM tip over a point where the leakage current is particularly high. Fig. 8(c) shows the rectifying behavior of this point, with a comparatively small negative-bias leakage-current compared to the positive-bias case. This is entirely consistent with $J - V$ results that show a lower value of leakage-current at negative-bias. We, therefore, conclude that the main path of leakage current is through more conductive channels distributed across the dielectric film.

In summary, the temperature dependence of the $J - V$ characteristics of Pt/SrTiO₃/Pt structure was investigated

from 300 to 380 K. It was found that the leakage current originates from the injection of electrons to deep traps, possibly oxygen vacancies near the interface, followed by thermionic field emission of electrons between the oxygen vacancies. In order to have an accurate model to explain the leakage current, it is necessary to consider the impact of both the interface and bulk. We, therefore, propose a model that combines both Poole-Frenkel and trap-assisted-tunneling mechanisms in one equation that is able to model the experimental data quantitatively over the whole range of both positive and negative applied bias. It is proposed that the asymmetry in the leakage current with respect to the bias direction is due to the varying distance of mobile electron

FIG. 7. (Color online) Different direction of current where the minimum of leakage current lies at negative voltage.

TABLE I. Calculated parameters used in Eq. (7).

	$\phi_{BPF} \text{ (eV)}$	$\phi_{BTAT} \text{ (eV)}$	ϵ_r
$V_b > 0$	0.90	0.13	7.0
$V_b < 0$	0.55	0.11	6.9

FIG. 8. (Color online) (a) The C-AFM image of SrTiO₃ surface. (b) Schematic path of leakage current through the aligned oxygen vacancies. (c) $I - V$ characteristic of a single-point by sweeping the voltage on top of leaky point.

traps, such as oxygen vacancies, from bottom electrode caused by the presence of a layer of fixed positive charge. This results in a variation in the injection rates of electrons at the two interfaces for a given bias. In addition, C-AFM measurements strongly indicate a highly non-uniform conduction across the dielectric, demonstrating that the leakage current is predominantly through the discrete channels, each of which exhibit asymmetric $I - V$ characteristics consistent with the device $J - V$ results.

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