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Thermal leakage characteristics of Pt/$SrTiO_3$/Pt structures

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MATERIAL NAMES: strontium titanate ($SrTiO_3$), platinium (Pt), barium strontium titanate (Ba,$SrTiO_3$)

Introduction

Leakage currents of metal-insulator-metal (MIM) structures with ferroelectric and high-permittivity perovskite thin films, such as $(Ba,Sr)TiO_3$ and $SrTiO_3$, have been extensively studied [1-4] because they determine the utility of these materials for dynamic random access memories, tunable microwave circuits and novel non-volatile memories. For structures with large workfunction metals, such as Pt, the leakage current is believed to be dominated by thermionic emission over the reversed biased blocking (Schottky) contact [1-3,5]. The depletion layer widths associated with Schottky barriers determine a wide range of film properties, such as the effective thin film permittivity, which in these materials strongly depends on the electric field [6,7] and has been discussed extensively in the literature [1,6,8,9]. For example, Copel et al. have measured band bending in $(Ba,Sr)TiO_3$ and $SrTiO_3$ with Pt electrodes and estimated the depletion layer width to be greater than 70 nm.

Several studies have recently reported peaks in thermal leakage currents of $(Ba_xSr_{1-x})TiO_3$ films [3,10,11], leading to a positive temperature coefficient of resistance (PTCR) effect within a certain temperature regime. The origin of this effect is still poorly understood. The goal of this brief report is to show that peaks in the thermal leakage can arise from non-steady-state leakage currents, most likely associated with detrapping of charge carriers in the depletion layer. We use a model perovskite MIM system for these studies, consisting of $SrTiO_3$ thin films and Pt electrodes. We have previously shown that these $SrTiO_3$ thin films do not undergo a ferroelectric phase transformation in the temperature range of interest for this study [12].

Experimental

Epitaxial, ~ 100 nm thick (111) Pt served as bottom electrodes and were grown by sputter deposition at ~ 580 °C on c-plane sapphire substrates. Predominantly {110} textured $SrTiO_3$ films with two different thicknesses, ~ 80 nm and 140 nm, respectively, were deposited by radio-frequency magnetron sputtering in an Ar/O_2 gas mixture at a substrate temperature of ~ 700 °C. The deposition parameters and film/electrode microstructures have been reported elsewhere [12-14]. The films had a columnar grain structure with no detectable nucleation layers and grain sizes that were independent of film thickness [14]. A two-step lithography process was used to fabricate MIM structures with an area of 130×150 μm^2. Pt top-contacts were deposited by electron beam evaporation, patterned by lift-off and annealed. Leakage currents were measured using a semiconductor parameter analyzer (HP 4155B, Agilent Technologies, Santa Clara). For measurements between 130 K and 400 K, the analyzer was attached to a probe station with a Joule-Thompson refrigerator (MMR Technologies, Mountain View, California). The bottom electrode was grounded and DC biases were applied to the top electrode. For measurements of the temperature dependent leakage, two sets of experiments were performed for each film thickness. In the first experiment, the films were cooled without an applied bias. After application of a positive bias (bottom electrode injection), the films were heated at a rate of 10 K/min under a constant DC bias and leakage currents were recorded. In the second set of experiments, the same positive bias was applied during cooling and subsequent heating.

Results and Discussion

At low applied voltages (1 – 3 V), leakage measurements as a function of time showed an initial rapid decrease in current (within ~ a few seconds) for both films. This regime was followed first by a more gradual decrease and then an increase in the leakage at longer times (> 100 s). None of the films exhibited steady-state leakage within 1000 s at any voltage (1 – 5 V) and temperature (200 – 300 K).

Figure 1 shows the leakage currents measured during heating from ~ 130 K to 400 K at a rate of 10 K/min under a constant DC bias. For the 140 nm film cooled without applying a bias (Fig. 1a), a distinctive peak (in addition to several minor peaks and changes in slope) was observed in the leakage current at ~ 250 K. In contrast, the large peak at ~ 250 K did not occur if a bias was applied during cooling. This showed that for the 140 nm film that the peaks in the thermal leakage curves could be explained with non-steady-state conditions.

Non-steady-state leakage currents can arise from the strongly temperature-dependent relaxation times of charge carriers emitted from deep trap levels [15-18]. As a bias field is applied to a MIM structures with blocking Schottky contacts electrons are released from traps and the cathodic depletion grows until most of the applied voltage drops across it [15]. Detrapping results in a non-steady-state dielectric relaxation current that decreases as the system approaches steady-state in which the leakage current is controlled by the steady-state current (such as the thermionic emission over the cathodic barrier). Peaks in the thermal leakage arise because the dielectric relaxation time required to reach steady-state after application of a DC bias depends on the trap level depth and strongly on the temperature. If the sample was cooled without an applied voltage it was likely not in steady-state at the lowest temperature, i.e. some traps were not depleted. Peaks in the current versus temperature curve upon heating then occur when traps deplete of their electrons [17]. Conversely, samples cooled under an applied voltage are in (or at least closer to) steady-state at low temperature and no peak is observed as those traps with levels that would give rise to peaks in the measured temperature range were already depleted during cooling.

The large current peak in the thermal leakage due to detrapping of charge shows that the film is not fully depleted, i.e. its thickness is greater that twice the depletion width. This places an upper limit of 70 nm on the depletion length of these films. If films are thinner than twice the depletion length, deep trap levels are expected to be depleted of charge [18]. In this case, no peaks in the non-steady-state thermal leakage due to detrapping should be observed. The absence of the large peak in the thinner (80 nm) film cooled without applied bias (Fig. 1b) may indicate that this trap level is already depleted. In the literature, peaks in the thermal leakage were only observed in thicker films [10,11]. The difference in the thermal leakage of the films with different thickness supports the interpretation that bulk traps (located within the cathodic depletion region), rather than interfacial defect states, are responsible for peaks in the thermal leakage. Further studies are needed to investigate the origin of the shallow peak observed for the thinner film.

In summary, we have shown that anomalous thermal leakage characteristics can be explained with non-steady-state leakage currents. The thermal leakage behavior is consistent with models of detrapping from deep levels and blocking contacts at the electrodes. The results show that care must be taken in the interpretation of anomalous thermal leakage phenomena in thin films of $SrTiO_3$ and related materials, such as $(Ba,Sr)TiO_3$, as samples might not be in steady-state, in particular at low temperatures. We also note that the results are relevant for the interpretation of capacitance measurements as a function of temperature in terms of the film permittivity in these voltage tunable films. The films investigated here likely contain a high density of point defects due to the high-energetic deposition method and high extended defect densities (twin and grain boundaries [13]), all of which could be responsible for traps. More studies are needed to correlate peaks in the thermal leakage with specific defects. Future studies should also address the correlation with the time-dependent leakage observed in these films. Establishing these relationships is complicated, as a result of different parallel processes, including dielectric polarization relaxation [19], and additional traps, each of which may dominate the time-dependent leakage in a different time-domain.

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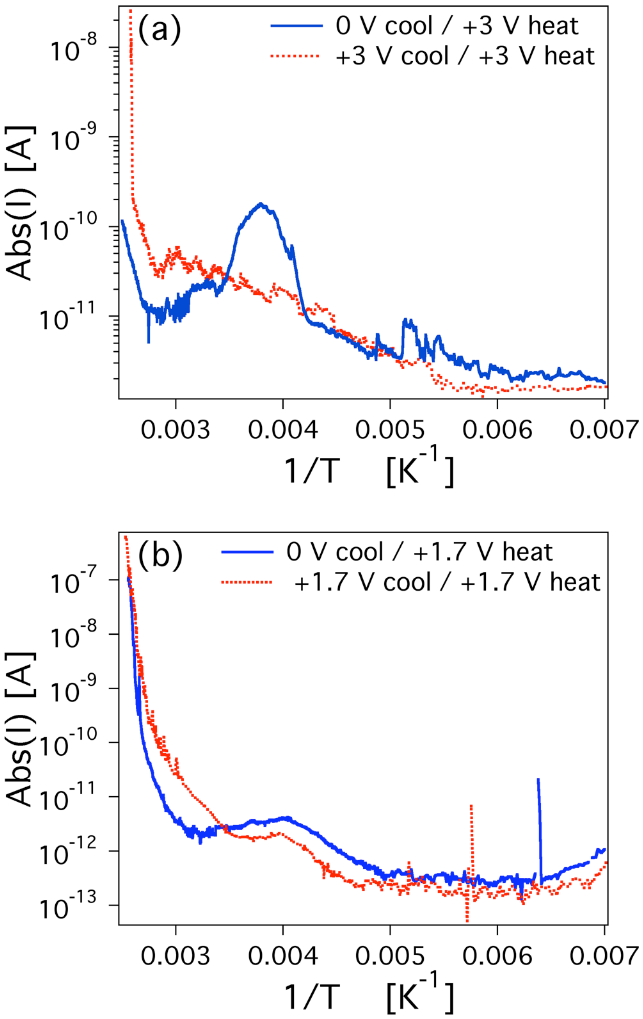
Figure Captions

Figure 1 (color online):

Leakage currents for the 140 nm $SrTiO_3$ film (a) and the 80 nm $SrTiO_3$ (b) measured during heating between ~130 K and 400 K. Solid (blue) curves were measured after no bias was applied during cooling, and dotted (red) lines were measured after cooling under the same bias that was applied during heating.

Figure 1


(a)
0 V cool / +3 V heat
+3 V cool / +3 V heat
Abs(I) [A]
10^{-8}
10^{-9}
10^{-10}
10^{-11}
0.003
0.004
0.005
0.006
0.007
1/T [K^{-1}]
(b)
0 V cool / +1.7 V heat
+1.7 V cool / +1.7 V heat
Abs(I) [A]
10^{-7}
10^{-8}
10^{-9}
10^{-10}
10^{-11}
10^{-12}
10^{-13}
0.003
0.004
0.005
0.006
0.007
1/T [K^{-1}]