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Interfacial defects in resistive switching devices probed by thermal analysis

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Resistive switching mechanism is investigated by thermal analysis of metal electrodes in the planar Al/$Pr_{0.7}Ca_{0.3}MnO_3$(PCMO)/Ni resistive switching device geometry. Two microthermocouples are used to monitor the electrode temperatures under different electrical bias conditions. Comparison of temperature differences between Al and Ni electrodes at high and low resistance states suggests that local heat source exists under the Al electrode at high resistance state. It agrees well with the recent finding in which AlO_x presents at the Al/PCMO interface and it can be the origin of the resistance switching mechanism [Li *et al.*, J. Phys. D **42**, 045411 (2009)]. Thermal measurements demonstrate excellent capability on characterizing resistance switching devices. © *2009 American Institute of Physics*. [DOI: 10.1063/1.3157207]

I. INTRODUCTION

The resistive switching (RS) effect has gained great attention recently due to its potential application for nonvolatile memory devices.[1,2] Upon the application of a voltage pulse, the device resistance can be reversibly modulated by orders of magnitude. RS has been observed in various oxides and measurement geometries.[3–5] Due to the short writing times (<10 ns), high storage densities (multistates possible) and low operating voltages (<2 V), resistance random access memories based on RS are being investigated as potential candidates for the next generation memories technology.[6–8]

Although RS memory devices have been successfully demonstrated by different research groups, the actual operating mechanism behind the effect is still under debate.[9,10] It has been suggested that the effect is caused by reversible formation and disruption of conducting paths (filaments) in the film, as a result of the application of high electric field across the material.[11,12] Others have attributed the effect to the film/electrode interface, in which Schottky barriers with trapped charges and space-charge-limited current are involved.[13,14] Recently, high resolution transmission electron microscopy has confirmed the existence of metal oxide layers in between the metal electrodes and the perovskite film.[15,16] It was argued that the RS effect was due to the formation and dissociation of such interfacial metal oxide layer.

While most of the RS studies are focused in the electrical aspects,[17,18] in the present work we conduct an investigation from a thermal point of view by monitoring the temperature variations of the metal electrodes. Temperature of the electrodes at both high resistance state (HRS) and low resistance state (LRS) at the same current bias levels are measured. Such *in situ* thermal measurements allow us to compare the temperature variations at the HRS and LRS directly. Then one can locate the origin of the heat source and understand the power dissipation mechanism inside the devices.

II. EXPERIMENT

The structure under test is Al/PCMO/Ni with a planar geometry. 250-nm-thick PCMO perovskite thin films were epitaxially grown by pulsed laser deposition (PLD) technique using a KrF excimer laser. $LaAlO_3$ (LAO) single-crystal substrates were maintained at 650 °C and an oxygen pressure of 150 mTorr was applied during the deposition process. Metal electrodes (800 μm diameter) were prepared on PCMO by thermal evaporation (Al) or PLD (Ni) through photolithography-prepared lift-off patterns and the minimum separation between pairs of electrodes was 30 μm (Fig. 1). Electrical measurements (*I-V* characteristics and resistance switching measurements) were performed with a Keithley 2400 source meter. For temperature measurements, two 30-μm-diameter type-*T* microthermocouples were placed on the Al and Ni electrodes directly and the temperature data were real-time monitored and recorded by a LABVIEW program at a rate of 5 Hz. All measurements were performed at room temperature. The schematic diagram of electrical and thermal measurement setups are shown in Fig. 1.

The *I-V* characteristic of an Al/PCMO/Ni device measured by two-point probe method is shown in Fig. 2. In the present work, positive bias is defined as electric current flowing from Al to Ni. When the applied voltage bias increases from 0 to +10 V (path 1), the slope of the *I-V* curve is gentle and the device is at HRS. It remains at HRS along path 2 and path 3, until the applied voltage is over −6.4 V at path 3. The current across the sample increases suddenly from this point and the device is switched into LRS. After that, the device remains at LRS as the voltage returns to zero along path 4 and switch again to HRS. It is worthy to point out that

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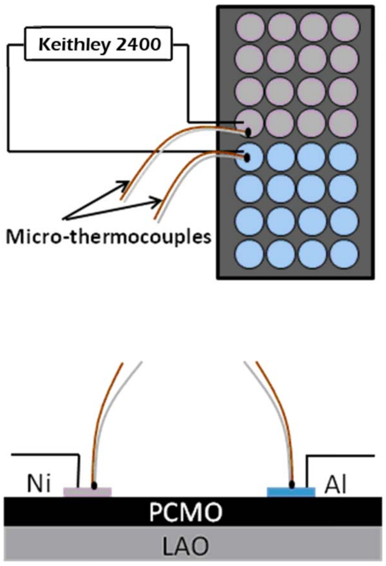

FIG. 1. (Color online) Schematic diagram of electrical and thermal measurement setups.

for a device having the same structure and stored under ambient for 6 months, the RS effect is reduced significantly (inset of Fig. 2) as compared with a fresh device.

It has been shown that the interface resistance between electrodes and PCMO is much larger than the intrinsic resistance of PCMO thin films, and it is believed that the transport properties are dominated by the interfaces.[13] Besides, devices with PCMO films and metal electrodes of similar work functions have shown drastically different *I-V* characteristics and resistance switching phenomena. These suggested that carrier transport at interfaces cannot be simply explained by Schottky barrier effect.[13,19] Indeed, Li *et al.*[15] have observed an AlO_x layer at Al/PCMO interface by transmission electron microscopy, and they attributed the RS effect to the formation and dissociation of the alumina layer. However, due to the experimental difficulties on direct profiling the electrical properties the interface between the metal electrodes and the PCMO film, it remains challenging to investigate the interface effect of resistance switching device *in situ*. Hence we have adapted the thermal measurement technique to investigate such effect.

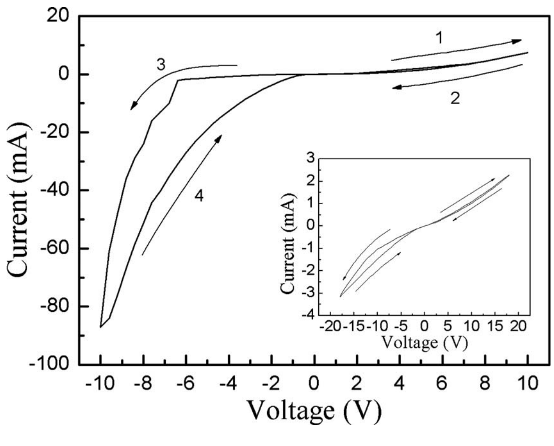

FIG. 2. *I-V* characteristics of a Al/PCMO/Ni device obtained by two-point probe measurement. Inset: *I-V* characteristic of the device having the same structure stored in ambient for 6 months.

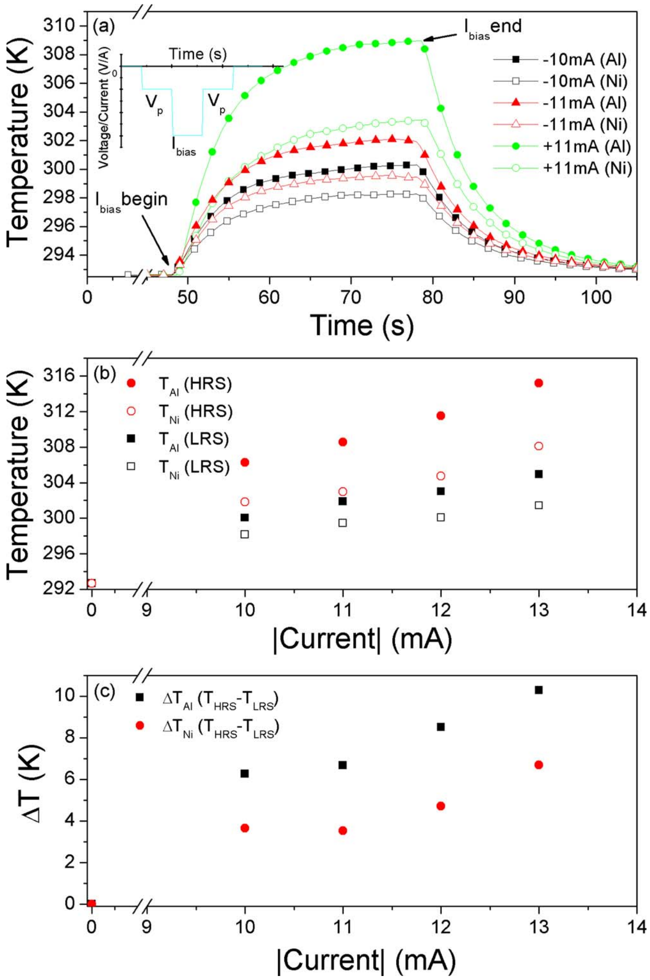

FIG. 3. (Color online) (a) Thermal response of a Al/PCMO/Ni device subjected to external electrical signals, as a function of varying I_{bias}. Inset: the bias current/voltage profile during electrical measurements. (b) The thermal responses of Al and Ni electrodes under HRS and LRS. (c) The temperature differences within the electrodes between HRS and LRS.

III. RESULTS AND DISCUSSION

For thermal measurements at LRS, a −2 V dc probing voltage (V_p) was applied across the sample for 30 s at the beginning, during which the initial temperature of the sample was recorded. It can be seen from Fig. 3 that no observable temperature differences between the two electrodes were detected. Subsequently, a (negative) dc current (I_{bias}) was applied for another 30 s, switching the sample to LRS. A large temperature difference between Al and Ni was measured during the application of I_{bias}, which suggests uneven heating inside the device. Afterwards, V_p was applied to the device for 60 s, and the measured electrode temperatures decrease gradually back to the initial values. The thermal response of the device as a function of time is plotted in Fig. 3(a). Similarly, for HRS measurements I_{bias} of the same positive magnitudes are applied to the device and the thermal responses at LRS and HRS were compared directly. It is worth mentioning that the thermocouples used for measuring Al and Ni electrode temperatures have been swapped during the experi-

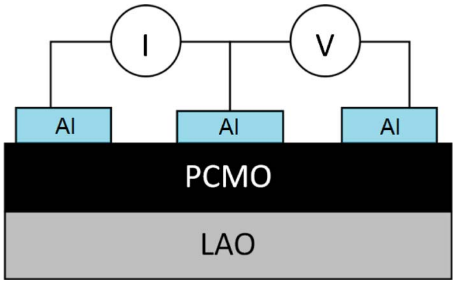

FIG. 4. (Color online) Geometry for contact end resistance measurement of Al/PCMO contacts.

ment, to ensure the measured temperature difference is not due to the intrinsic error from the thermocouples.

The temperature response of Al and Ni electrodes at LRS and HRS as a function of the I_{bias} magnitude is plotted in Fig. 3(b). It can be clearly observed that, for the same magnitude of I_{bias}, both electrodes have higher temperatures at the HRS than the LRS. In order to verify the origin of the heat source, the temperature change within the electrodes under HRS and LRS are plotted in Fig. 3(c). It can be observed that when $|I_{\text{bias}}|$ equals 13 mA, the Al electrode is at 315 and 305 K for HRS and LRS, respectively (i.e., $\Delta T_{\text{Al}} = 10$ K), whereas ΔT_{Ni} is just 6 K. The result suggests that a stronger local heat source exists under the Al electrode. This agrees well with the finding for the existence of AlO_x layer between Al and PCMO thin film, which causes additional heating.[15] Furthermore, the electrode temperatures of the 6 month old device show a smaller temperature increase than the fresh device under the same bias voltages (data not shown) and it can be explained by the smaller current flowing inside the aged device under the same voltage bias as the oxide layer is thicker.

Indirect proof for thicker oxides in aged Al electrodes was obtained by measuring the contact end resistance R_e (which is closely related to the actual contact resistance[20]) at the Al electrodes in both samples, using a geometry shown in Fig. 4. Identical electrode geometries in both new and aged samples automatically eliminated issues related to current distributions.[21] Besides, measurements were performed at parts of samples where switching has *not* been conducted; this prevents any alteration to the devices arising from switching-related mechanisms. With a small direct bias current (10 μA), the measured R_e at Al electrodes in the fresh device and the 6 month old devices were 6 and 17 kΩ, respectively. We also measured the resistance of PCMO between two adjacent electrodes by the standard four-point probe geometry, and similar results ($\sim$0.5 kΩ) obtained from both new and the aged samples. These results suggest the stronger oxidation in the Al electrodes in the aged device.

It is also important to notice that the increase in aluminum electrode temperature shows a relatively linear relationship with I_{bias} and it cannot be simply explained by Joule heating, in which the dissipated power is proportional to the square of I_{bias}. The linear relationship between the temperature variation and I_{bias} suggests that the heating effect is proportional to injected carriers from the electrode, and energy is dissipated similar to the form of trapped states. Such trap state-induced heating have been observed and discussed in laser diode devices.[22,23] In our samples, trap states can be generated by various mechanisms including: (1) intrinsic defects during the fabrication of the PCMO layer and the metal electrodes[24] and (2) defects due to deficiency of oxygen atoms in the PCMO layer.[15] As a result, the trap states at the interface between the PCMO and the metal layers causes localized heating effect. Future work will focus on a detailed thermal model to simulate the temperature distribution within the device for understanding the energy dissipation and resistance switching mechanism.

IV. CONCLUSIONS

To summarize, we proposed a direct and nondestructive method to study resistance switching effect in planar Al/PCMO/Ni structure. Thermal measurements show a significant temperature difference between aluminum electrodes at HRS and LRS. It agrees well with recent finding that aluminum oxide layer exists between the Al and PCMO film and contributes to RS mechanism. *In situ* thermal measurements can be very useful to characterize resistive switching devices, and it can provide useful information on how input power is dissipated inside the devices.

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