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<tr>
<td>Citation</td>
<td>IEEE Transactions on Magnetics, 2014, v. 50 n. 7, article no. 3000804</td>
</tr>
<tr>
<td>Issued Date</td>
<td>2014</td>
</tr>
<tr>
<td>URL</td>
<td><a href="http://hdl.handle.net/10722/202995">http://hdl.handle.net/10722/202995</a></td>
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Investigating the Uneven Current Injection in Perovskite-Based Thin Film Bipolar Resistance Switching Devices by Thermal Imaging

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Bipolar resistive switching (RS) phenomenon in planar Al/Pr0.7Ca0.3MnO3 (PCMO)/Ti devices was investigated by thermoreflectance method. Thermal images of devices undergoing switching were used to quantify the unevenness of injected current under different voltage bias. At low resistance state, the injected current at the current crowding area of the Al/PCMO interface was twice the current at the high-temperature region. This unevenness was quantified using the thermoreflectance method. The distribution of the resistance, and hence the uniformity of current injection into the RS devices, has not been completely studied.

Index Terms—Nonvolatile memories, Pr0.7Ca0.3MnO3 (PCMO), resistive switching (RS), thermal imaging.

I. INTRODUCTION

As the demand on the data storage capacities for nonvolatile memories increases continuously, memory devices based on resistance switching (RS) have been studied vigorously since the last decade [1], [2]. RS devices incorporating solid electrolytes, organic molecules, or transition-metal oxides have been successfully demonstrated [3]–[5]. Among different RS devices, those with metal/oxide/metal planar and capacitor geometries have several outstanding advantages, including the low production cost, high chemical stability, and compatibility with flexible substrates [6].

For the oxide layer in such class of RS devices, both binary and perovskite oxides have been employed [7]–[10], and it has been shown that the RS mechanisms are different for devices with distinct structures and materials [11], [12]. Sawa et al. [13] proposed that the RS effect in capacitor-geometry RS devices originates from the Schottky barriers, with trapped charges present at the interface between the film material and the electrodes [14]. Using transverse electromagnetic, Li et al. [15] showed that a thin layer of AlOx exists at the interface between the Al and Pr0.7Ca0.3MnO3 (PCMO), and they proposed that the formation and dissociation of the AlOx layer contributed to the RS effect in the perovskite oxide-based devices. On the other hand, the unipolar RS effect in binary oxide-based devices (such as nickel oxide) is believed to be related to the formation of conductive filamentary paths within the device [16], [17]. In particular, two types of unipolar RS effect (memory and threshold RS) have been reported, of which the threshold type is unsuitable for nonvolatile memory applications [16]. The threshold-type RS has not been reported so far in perovskite oxide-based bipolar RS devices.

Recently, we reported single-point (size \( \sim 30 \mu m \)) thermal measurements in Al/PCMO/Ni devices to investigate the RS phenomenon [18]. Our measurements showed an unequal heating at Al and Ni electrodes, and a larger temperature increase was observed at the Al electrodes when the device was switched from the high resistive state (HRS) to the low resistive state (LRS). At LRS, a larger current passes through the device, resulting in a stronger heating than HRS. The larger temperature rise at the Al electrode than the Ni electrode represents a larger series resistance exists at the Al side, given the identical current passing through the Al and Ni electrodes. Both the single-point thermal and electrical measurements suffer from the constraint of limited spatial resolution of the technique. The distribution of the resistance, and hence the uniformity of current injection into the RS devices, has not been completely studied.

In this paper, we take a step further by investigating the uniformity of the series resistance in between the PCMO and the metal electrodes of the planar RS device, using the nondestructive charge-coupled device (CCD)-based thermoreflectance method. Based on the thermal images, we quantified the current injected into the high temperature region during LRS is 60% more than the surrounding region. It provides direct evidence that localized resistance, hence uneven current injection, exists in the Al/PCMO interface of the RS device.

II. MATERIALS AND METHOD

2-D thermal images were obtained by CCD-based thermoreflectance; the technique has been previously applied for characterizing various electronic devices including semiconductor lasers, transistors, and amplifiers [19]–[21]. The schematic diagram of the thermoreflectance experimental setup is shown in Fig. 1(a). A 10× objective was used, yielding a spatial resolution of approximately 2 \( \mu m \). By assuming a linear relationship between the changes of the reflectivity and the temperature, the temperature variation \( \Delta T \) can be obtained by the thermoreflectance equation

\[
\Delta T = \kappa \frac{\Delta R}{R}
\]
In the above equation, $\Delta R/R$ is the normalized variation of reflectivity, and $\kappa$ is the proportionality constant known as thermoreflectance coefficient, which is dependent on the illuminating wavelength and the material [22]. The measured thermoreflectance signal was transferred to a personal computer equipped with LabView for data analysis.

The devices under test contained 250 nm PCMO perovskite thin films epitaxially grown on LaAlO$_3$ (001) single crystal substrates, prepared by pulsed laser deposition (PLD) [23]. The substrates temperature and oxygen pressure during deposition were maintained at 650 °C and 150 mtorr, respectively.

II. RESULTS AND DISCUSSION

The $I$–$V$ characteristic of the device is shown in Fig. 1(b). When the applied voltage bias is increased from zero to +5V (path 1), the slope of the $I$–$V$ curve is gentle. The same trend is followed along paths 2 and 3, until a $-3.8$ V bias is applied across the electrodes, which is accompanied by a sharp increase in current and the large current trend remains along path 4. Similar asymmetric $I$–$V$ behavior is commonly observed in bipolar RS devices [24], [25]. By comparing the current values under HRS and LRS in Fig. 1(b), a resistance ratio between the HRS and LRS equals to 27 for the device under a $-2$ V probing voltage was obtained.

Stability and repeatability of the LRS and HRS in the devices have been confirmed, and the results are shown in Fig. 1(c). The resistance of the device was obtained by a probing voltage of $-2$ V. In the stability test, the device was first driven from LRS (HRS) to HRS (LRS) through the application of +5 V ($-5$ V) voltage pulse of 100 ms. The device was then disconnected from the measurement circuit for 10 min, before the resistance was probed again. No drastic change in the resistance value can be observed, demonstrating the nonvolatile nature of the resistance states. Repeated pulsing between the HRS and the LRS inset of Fig. 1(c) with +5 and $-5$ V pulses indicated that such states can be achieved for many times.

The optical image of an Al/PCMO/Ti device is shown in Fig. 2(a), and the corresponding thermoreflectance images under different bias conditions are shown in Fig. 2(b)–(e). The thermal images are taken while modulating the voltage bias across RS device at 10 Hz. The thermoreflectance coefficients of Al, Ti, and PCMO calibrated by microthermocouple are $3 \times 10^{-4}$, $8 \times 10^{-4}$, and $1.3 \times 10^{-3}$ K$^{-1}$, respectively. The images can be converted into temperature
is further increased to at the PCMO edge next to the Al electrode. If the bias voltage increased to no heating is observed. However, when the applied voltage is remains nearly the same as in the positive bias regime, and two localized hot spots can be clearly observed, and the heated heating occurs and the hot spots grow in size. From Fig. 2(e), The temperature gradient across the hot spot is about channel at various voltage biases are compared in Fig. 3(a).

The horizontal averaged temperature variation of the PCMO [25]. The observed hot spots can arise from the uneven formation and deformation of the AlO
x
layer has shown to be related to the chemical reaction between the Al electrode and O
2
in the capacitor-like structure RS devices, where AlO
x
layer is formed underneath the Al electrode. The formation and deformation of the AlO
x
layer is believed to be related to the RS mechanisms of the device [24], [25], [27]. At HRS, AlO
x
layer is formed at the Al/PCMO interface and the current passing though the device is small. When the device switches to LRS, deformation of the AlO
x
layer starts to occur, hence a larger current can pass though. The formation and deformation of the AlO
x
is believed to be related to the chemical reaction between the Al electrode and O
2
from the PCMO [25]. The observed hot spots can arise from the uneven dissolution of the AlO
x
layer while the device is switched from the HRS to LRS, resulting in the current crowding at the region without AlO
x
. The nondestructive, in-situ current profiling method by thermoreflectance measurement supplements the electrical measurements to provide a more complete picture of the switching effect.

IV. CONCLUSION

In conclusion, planar perovskite-based bipolar RS devices with Al and Ti electrodes have been fabricated by PLD and investigated by thermoreflectance measurement. The nonuniform current injection in the planar PCMO RS device has been quantified by the high-resolution thermal imaging method. For the Al/PCMO/Ti planar device under test, it has been shown that the uneven current injection at the Al/PCMO interface is related to the RS mechanism of the device. The unevenness of the injected current from Al to the PCMO can be as large as 1.6 at −5 V bias. As the uneven current injection is difficult to
be characterized by electrical measurements, thermal imaging provides a versatile method to characterize the performance of the RS devices. The technique should be applicable to both bipolar and unipolar switching device structures.

ACKNOWLEDGMENT

This work was supported in part by UGC and in part by HKSAR under Grant PolyU 5013/08P, Grant PolyU 5112/08E, and Grant PolyU A-PM21/G-YN08/H-ZDAC. The work of Z. Luo was supported in part by the Fundamental Research Funds for the Central Universities under Grant 21612416 and in part by the National Science Foundation of China under Grant 11204105. P. K. L. Chan would like to thank Dr. D. Lüerßen for the fruitful discussion and help at the beginning of developing the thermoreflectance setup.

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