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## Room-temperature resistance switching and temperature hysteresis of $\text{Pr}_{0.7}\text{Ca}_{0.3}\text{MnO}_3$ junctions

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Current–voltage ( $I$ – $V$ ) characteristics of  $\text{Ag}/\text{Pr}_{0.7}\text{Ca}_{0.3}\text{MnO}_3(\text{PCMO})/\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}(\text{YBCO})$  junctions fabricated on  $\text{LaAlO}_3$  (001) substrates were measured. Nonlinear, asymmetric, and hysteretic  $I$ – $V$  curves, that are considered to be the nature of the resistance memory effect previously reported, were observed. In some junctions the  $I$ – $V$  characteristics were switched between linear and nonlinear by its thermal hysteresis. Room-temperature  $I$ – $V$  characteristics, originally being linear, turned to be nonlinear after cooling to 100 K, and returned to be linear after heating to 400 K. © 2005 American Institute of Physics. [DOI: 10.1063/1.1851852]

Perovskite-type Mn oxides are attracting much interest in both physical and industrial fields because of their unique properties. For example,  $\text{Pr}_{1-x}\text{Ca}_x\text{MnO}_3$  (PCMO;  $0.3 \leq x \leq 0.75$ ) is known to show a charge-ordered (CO) state in a temperature region lower than  $T_{\text{CO}} \sim 230$  K.<sup>1</sup> An external stimulus such as an electric field can induce the collapse of such CO insulating state, causing a steep insulator-metal (IM) transition.<sup>2</sup> The electric-field-induced IM transition with several orders change of its resistance is expected to be applied for logic or memory devices. As the basic experiments for a device application, we have fabricated  $\text{Au}/\text{PCMO}(x=0.5)/\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}(\text{YBCO})$  layered-type junctions on  $\text{MgO}$  (001) substrates, and observed a seven-order drop of its resistance induced by a voltage less than 5 V at low temperature.<sup>3</sup> On the other hand, a nonvolatile pulsed-voltage (current)-induced high-/low-resistance (HR/LR) switching, which cannot be theoretically explained at present, has been reported in a  $\text{Ag}/\text{PCMO}(x=0.3)/\text{YBCO}$  junction.<sup>4</sup> This resistance-switching effect is different from the IM transition caused by the collapse of the CO state in several points: (1) it occurs even at room temperature, (2) the resistance switches to the opposite state when a reversed bias is applied, and (3) the resistance state is retained even after the voltage is removed (nonvolatility). A resistance random-access-memory (RRAM) device applying this resistance switching effect has been proposed,<sup>5</sup> and is expected as a good candidate that would be superior to the present RAM devices. However, the physical mechanism of this resistance switching effect is quite unclear at present, and thus one has no strategy to improve the device properties or uniformity in order to achieve the practical application.

Being measured by the two-probe method, the resistance of the  $\text{Ag}/\text{PCMO}/\text{YBCO}$  junctions in Ref. 4 ( $\sim 200$  and  $\sim 3500$   $\Omega$  in LR and HR states, respectively) contains the series resistance of electrodes and wires. Supposing that the resistivity of the PCMO layer in this junction is identical to that of the PCMO ( $x=0.3$ ) single crystal previously reported ( $\sim 0.1$   $\Omega$  cm at 300 K),<sup>1</sup> the contribution of PCMO layer to

the junction resistance should be less than  $10^{-2}$   $\Omega$  considering the junction size (0.4 mm in diameter and 600 nm in thickness). It seems to be relevant to suppose that all the other part in the LR value of the junction ( $\sim 200$   $\Omega$ ) corresponds to the “series resistance.” On the other hand, provided the same series resistance exists in the HR state also, then the resistance of PCMO in the HR state should exceed 3 k $\Omega$ , five orders larger than the expected value. Therefore, the question is “what is the nature of a HR state?”

In order to understand the mechanism of the effect and to improve the junction properties as the memory devices, we are studying this electric-field (or current) -induced switching effect. So far, we have reported that the nonstoichiometric (Mn-poor) PCMO films show the switching effect easier than near-stoichiometric ones do.<sup>6</sup> In the present study we have observed the current–voltage ( $I$ – $V$ ) properties of  $\text{Ag}/\text{PCMO}(x=0.3)/\text{YBCO}$  layered-type junctions. As the results, nonlinear asymmetric  $I$ – $V$  properties with hysteresis loops, which explain the resistance-memory effect, were observed. Moreover, some junctions were found to switch their  $I$ – $V$  properties from linear to nonlinear and vice versa according to their thermal hysteresis.

$\text{PCMO}(x=0.3)/\text{YBCO}$  bilayered films were deposited on  $\text{LaAlO}_3$  (001) single-crystal substrates by a pulsed laser deposition method. The YBCO layer was partly covered by a

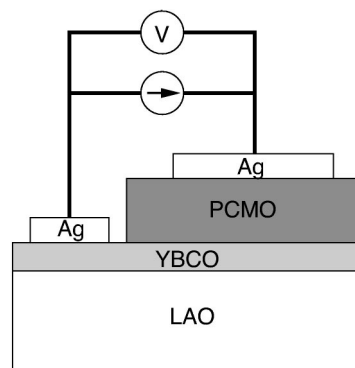


FIG. 1. Schematic diagram of the junction structure and the measurement system.

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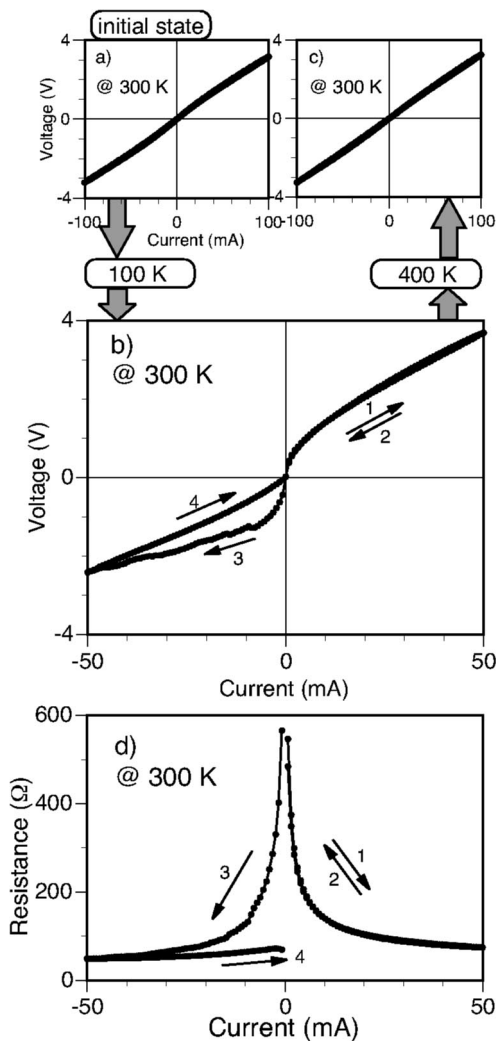


FIG. 2. (a)–(c)  $I$ – $V$  properties of a Ag/PCMO( $x=0.3$ )/YBCO junction at 300 K. The junction area (=Ag pad area) is  $0.5 \times 0.5 \text{ mm}^2$  and the PCMO thickness is 600 nm. This junction, initially showing a linear  $I$ – $V$  property (a), changes its property to nonlinear by being cooled to 100 K and then heated to 300 K (b), and recovers to the initial state by being heated to 400 K and cooled to 300 K again (c). (d)  $R$ – $I$  plot of the data in Fig. 2(b).

metal mask *in situ* before the PCMO layer was deposited, so that the contact to the bottom YBCO layer would be easily done (see Fig. 1). The substrate temperature was 750 °C and the oxygen ( $\text{O}_3 + \text{O}_2$ ) pressure was 150 mTorr during the deposition of both PCMO and YBCO. The thickness of YBCO and PCMO was 200 and 600 nm, respectively. A Ag layer was deposited by a sputtering method to form the contact pads.

Figure 1 shows the schematic diagram of the junction and the measurement system. The  $I$ – $V$  properties were measured in a two-probe configuration, using either physical properties measurement system (PPMS: Quantum Design Co.) or a simple combination of a current source and a voltmeter. In PPMS the current was swept and the voltage was measured. In the simple system a constant current was manually set and the voltage was recorded.

Figure 2(b) shows  $I$ – $V$  curves at 300 K of a Ag/PCMO/YBCO junction. The nonlinear, asymmetric, and hysteretic behavior is important for understanding the phenomenon of resistance memory effect, because the result reported in Ref.

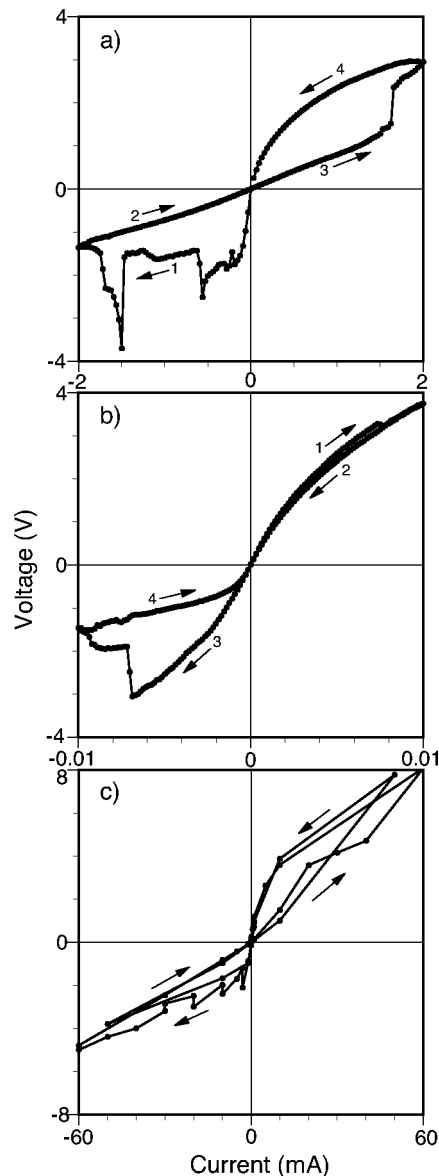


FIG. 3. Several typical  $I$ – $V$  curves of Ag/PCMO( $x=0.3$ )/YBCO junctions. (a) measured in order of negative  $\Rightarrow$  positive bias, (b) positive  $\Rightarrow$  negative bias, and (c) the result obtained with a simple measurement system.

4 that “a positive bias causes HR, and a negative bias causes LR state” is considered to be the description of just one part of the asymmetric and hysteretic  $I$ – $V$  property. We should note that the  $I$ – $V$  properties of our PCMO/YBCO junctions are basically linear at the first stage (just deposited), as shown in Fig. 2(a). The nonlinear  $I$ – $V$  curve in Fig. 2(b) appeared after this junction was cooled to 100 K and then heated to 300 K. Afterwards, the  $I$ – $V$  of this junction was recovered to the initial linear state after being heated to 400 K and cooled to 300 K again. Therefore, there is a temperature hysteresis on the nonlinearity of this junction.

Figure 2(d) shows the current dependence of the junction resistance  $R$  ( $=V/I$ ) based on the data points in Fig. 2(b).  $R$  significantly depends upon  $I$ , and can be changed between  $\sim 70$  and  $>550 \Omega$  during sweeping. Therefore we note that the measurement current (or voltage) should be declared when one discuss the “resistance” of such a junction. Because of the strong nonlinearity near the origin in the posi-

tive bias side, the smaller the resistance-measurement current is, the larger the resistance in HR state can be.

Figure 3 shows some other typical  $I$ - $V$  curves. All these data were measured at 300 K after cooling. One  $I$ - $V$  sweep takes  $\sim 5$  s in PPMS [Figs. 3(a) and 3(b)] and several minutes in the simple system [Fig. 3(c)]. It suggests that the resistance switching could be induced not only by a pulsed voltage but also by a constant current, in contrast with the previous result.<sup>5</sup> The reproducibility in our measurements are not good yet; sometimes one junction shows different  $I$ - $V$  curves at every measurement, for example. In total, the curve is smooth during  $|I|$  is decreasing, while some irregular shapes are often seen during  $|I|$  is increasing.

A popular explanation of RRAM effect at present is that the HR transition occurs only at the interface between PCMO and (an) electrode(s), not in the whole PCMO bulk.<sup>7</sup> S-shaped  $I$ - $V$  curves observed in the present study also suggest the conduction through a metal-insulator-metal (MIM) structure. A Schottky barrier at the interface between PCMO and an electrode layer can play the role of the insulator in the MIM structure.<sup>8</sup> More generally, however, a grain boundary or a defect in the PCMO film is also considered to be the insulating part that constructs a fine MIM structure. Heating/cooling processes and an application of a positive/negative voltage (current) may possibly cause a deformation of the atomic structure in the PCMO film and thus induce the

change of electric properties. Further investigation should be performed to clarify crystallographic, atomic structural, and electronic structural difference between HR/LR states or between linear/nonlinear states of these junctions.

In conclusion, we have measured the  $I$ - $V$  characteristics of Ag/PCMO( $x=0.3$ )/YBCO junctions fabricated on LaAlO<sub>3</sub> (001) substrates. Nonlinear, asymmetric, and hysteretic  $I$ - $V$  curves, that are considered to be the nature of resistance memory effect, were observed. In some junctions the property of linear or nonlinear  $I$ - $V$  was switched by its thermal hysteresis. The above results could be important keys to understand the mechanism of resistance memory effect.

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