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Reversible Resistive Switching in Electrically Nonhomogeneous $La_{0.67}Ca_{0.33}MnO_3$ Thin Films by Short Electrical Pulses

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Resistance changes in thin electrically nonhomogeneous $La_{0.67}Ca_{0.33}MnO_3$ films were investigated using electrical pulses of nanosecond duration in the 80–300 K temperature range. Two types of reversible switching to higher resistive states with different starting temperature induced by series of the positive pulses were observed. Possible mechanisms of the resistance switching by short electrical pulses in the vicinity of T_m and at 80–90 K are discussed.

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1. Introduction

Recently thin films of perovskite oxides ($R_{1-x}A_xMnO_3$, where R is a rare-earth element and A stands for alkaline-earth ions) have attracted much interest, because of the resistance changes under the applied electric field (electroresistive effect, ER) [1] and resistance switching phenomenon [2]. The resistance switching phenomenon in thin films could be promising for fabrication of fast non-volatile memory devices.

The $La_{0.67}Ca_{0.33}MnO_3$ films fabricated by pulsed laser deposition (PLD) technique demonstrate transition from the paramagnetic to ferromagnetic phase (PM–FM transition) with the resistance maximum at temperature T_m (just slightly above the Curie temperature — T_C). In Ref. [3] it was shown that series of electrical pulses of 10 ns duration might be used for switching films to high resistive (HR) states at 80 K temperature. Film switching to the HR state in the vicinity

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of T_m has also been observed. The main goal of this work is further investigation of the resistance switching in thin $La_{0.67}Ca_{0.33}MnO_3$ films, in order to understand better the origin of this intriguing phenomenon.

2. Preparation of high resistive samples and experimental setup

Our $La_{0.67}Ca_{0.33}MnO_3$ thin films were grown in situ by the PLD technique on cleaved, (100) MgO single crystals. The film deposition procedure and investigation of the properties of the electrically nonhomogeneous films were described in Ref. [4]. The prepared films demonstrated the PM–FM transition in the temperature range 125–140 K. A pair of Ag pads (width $w = 1$ mm and the gap between them $d = 10 \div 30$ μm) magnetron sputtered onto the film surface through a mask served as electrodes for electrical measurements.

The experimental setup used for dc measurements and investigation of voltage-current characteristics in a nanosecond time scale was described in Ref. [3]. Figure 1 demonstrates the R–T dependences of the sample in the initial state after deposition (curve 1) and after applied series of the pulses creating electric field above 20 kV/cm (curve 2) at 80 K temperature. The HR states of the samples were found to be stable at relatively low electric fields. The resistance changes (demonstrating ER effect) were observed at pulsed electric fields. The observed resistance decrease due to the ER effect [1] at fixed temperature was calculated using the relationship: $\mathrm{ER} = [R(E) - R(0)]/R(0)$ where $R(E)$ is the sample resistance in the electric field $E = V/d$, V is the applied voltage.

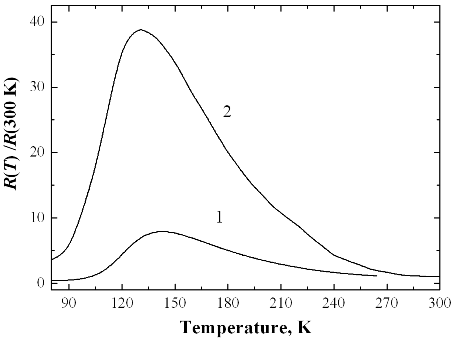

Fig. 1. Typical normalized R–T dependences of the sample after deposition (1) and in high resistive state (2). Average film thickness — 120 nm.

3. Experimental results and discussion

Figure 2 demonstrates resistance changes of the sample with average film thickness 100 nm, $d = 30$ μm (Fig. 2a) and 80 nm, $d = 10$ μm (Fig. 2b). Resistance switching from the initial state (Fig. 2a, curve 1, up arrow) to HR state was observed at 80 K by applying a series of 10 ns pulses, $E = 25$ kV/cm over the sample. Transition to a new HR state (Fig. 2b, curve 1, up arrow) occurred after

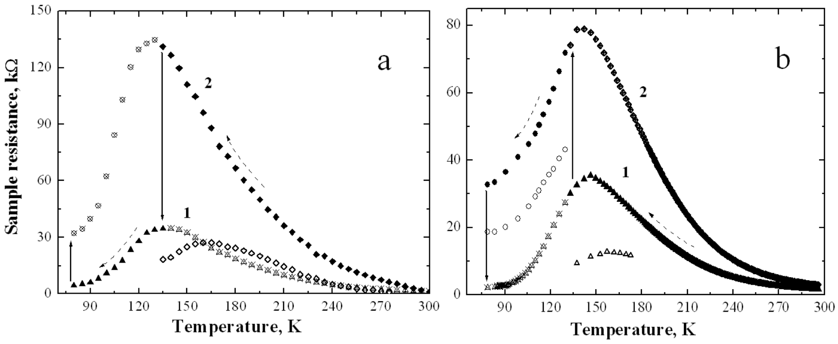

Fig. 2. The resistance changes of the samples with $d = 30$ μm (a) and $d = 10$ μm (b). 1 — R–T dependence of initial state (black and crossed triangles in (a)) and of the high resistive state (crossed and black triangles in (b)), 2 — R–T dependences of a high resistive state (crossed circles and black diamonds in (a)) and of new high resistive state (black circles and crossed diamonds in (b)). Open triangles (b) show EReffect of HR state induced by the pulses at switching field $E = 14$ kV/cm. Open diamonds in (a) show ER effect of HR state induced by the pulses at switching field $E = 26$ kV/cm. Open circles in (b) show ER effect of a new HR state induced by the pulses at switching field $E = 4$ kV/cm. Vertical arrows both at 80 K and in the vicinity of T_{m} indicate switching places.

series of 100 ns pulses at $E = 14$ kV/cm and $T = 130$ K. The R–T dependences in new states (curves 2 in Fig. 2a and b) were obtained at low E values during film cooling from 293 K to 80 K. Further investigation of ER properties of the samples in the HR states have shown that the pulses did not change irreversibly the electric properties of the samples in 80–290 K temperature range (at 300 ns duration up to 26 kV/cm and at 100 ns duration up to 14 kV/cm for the cases presented in Fig. 2a and b, correspondingly). Resistance drop with the electric field due to ER effect is not shown for the initial state, but it is shown for HR state (open triangles in Fig. 2b).

The reverse switching (Fig. 2a, down arrow) from the HR state to the initial state occurred at $T = 134$ K and $E = 26$ kV/cm. The ER value at the switching point was equal to –85.9% (open diamonds in Fig. 2a). After the reverse switching, the sample was cooled down to the starting point at 80 K temperature. The reverse switching from the new HR state to the former state (Fig. 2b, down arrow) occurred in this case at $T = 80$ K. The value of ER effect at $E = 4$ kV/cm near the switching was equal to –42.7% (open circles in Fig. 2b). Then, the sample was heated up to starting 130 K temperature after switching.

To explain the switchings we used a network-of-filaments current flow concept in nonhomogeneous media with high resistance intergrain boundaries [1, 3, 5]. According to this concept, in FM state an array of conducting channels separated from each other by thin intergrain boundaries of high resistance disturbs the current. In the first cycle (Fig. 2a) the local electric field strength at the boundaries should be significantly higher than the average one. The strong electric field regions may form tunnel junctions by extraction of weakly bounded oxygen ions. This causes forming of HR state at 80–95 K. Local thermal cycling from pulse to pulse in the barrier region can also stimulate the resistance increase after another series of the pulses as it was observed during thermal cycling of the whole sample [6].

Near T_{m} temperature electrical inhomogeneity level in the films increases due to phase separation phenomena. Therefore, effective cross-section of conducting channels decreases and density of channels increases. Only some channels will be completely in FM phase, but they contain intergrain boundaries demonstrating the ER effect as well. At reverse switching, for 300 ns duration pulses, the ER effect value was very strong: –85.9% at $E = 26$ kV/cm. To explain the strong ER effect the strict theories assume both the electron hopping preferentially along the applied electric field and the self-optimized conducting channel formation [7]. Therefore, one can expect that a concentrated current flows near the intergrain boundaries under high external electric field. Relatively long (300 ns) electrical pulses provide effective heating of intergrain boundaries leading to thermally-assisted random distribution of weakly bounded oxygen in the vicinity of tunnel junction and melted HR state formation in FM phase.

Discussing the second switching cycle beginning at 130 K temperature (Fig. 2b) it should be noted that despite the HR state formed at 80 K, the $R(80\ \mathrm{K})/R(300)$ value is only slightly higher than 1. The ER effect value of the HR state (open triangles in Fig. 2b)near the switching point was –62% ($E = 14$ kV/cm). Due to above mentioned reasons, the current flows near intergrain boundaries in the presence of high external electric field. Most probably, near T_{m}, local heating in this sample induces stable increase in the PM phase volume leading to sample resistance growth. We assume that the reverse switching occurs due to such resistance increase at a relatively weak (4 kV/cm) electric field.

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