Threshold Switching and Hysteresis in Transport Characteristics of Mesoscopic Ag-LCMO Junctions

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Introduction

Applications of manganites in nanoelectronic devices with extremely short working times need more knowledge about their non-equilibrium and near-equilibrium properties. Particular interest is given to such processes in contacts of manganites with metals which can be used in memory devices [1,2]. In manganites, oxygen ions are weakly bound to the lattice and easily go out of the crystal bulk [3]. This feature is of decisive importance for the formation of near-surface layers, as well as for the structure of an interface formed between the manganite and a normal metal. Due to the presence of oxygen vacancies on the manganite surface, the concentration of holes in the near-surface layers decreases and, in the limiting case, the surface can transform into a dielectric phase with magnetic ordering at low temperatures [1,4,5]. As a result, a high-resistive several atomic-layer region appears at the manganite surface.

In this report, we present our experimental results concerning the threshold switching effects and hysteretic behavior in the current-voltage characteristics of mesoscopic manganite-normal metal heterostructures.

Experimental results and discussion

The studied samples $La_{0.6}Ca_{0.3}MnO_3$ (LCMO) were prepared by the solid-phase synthesis method from oxides La_2O_3 and carbonate $CaCO_3$. $0.1 \times 1 \times 10$ mm plates were prepared using ceramics fabricated by high uniaxial pressures $P = 6$ GPa, and annealed at $T = 1050°C$ for six hours.

The plates were pressed again and re-annealed at $T = 1100°C$ to get a well-developed texture. As a result, the samples consisting of oriented 10–15 μm sized microcrystals were fabricated. The current and potential contacts for measurements of transport characteristics were obtained by burning colloidal silver into the surface of the plates. The contact resistance of the contacts was less than 10^{-8} Ohm·cm^2. Ag-LCMO contacts for tunneling experiments were prepared by rubbing colloidal silver into the plate surface. The resistance of the tunnel junctions was $50 - 200$ Ohm.

Current-voltage recording was performed by both an automated measuring system and with an ordinary two coordinate recorder to exclude instrument errors. The results of the two measuring techniques coincided completely. In Figure 1, a typical current-voltage characteristic (CVC) of an Ag-LCMO contact is shown as well as the temperature effect on it. Anomalous sharp vertical growth of the tunnel current was observed at voltages $\sim \pm 1$ V. For decreasing voltage, the current decreased smoothly and the contact resistance followed the initial, equilibrium way. The CVCs studied did not exhibit a jump-like transition from the high-resistive to the low-resistive state. There was no memory effect, usually observed in metal-manganite contacts, the contact resistance (at $V \approx 0$) does not change for a long time after switching to a new state. The absence of significant hysteresis and memory effects are apparently due to the fact that the mechanism causing the observed specific CVC features is realized only in a narrow region of the 2D tunneling barrier.

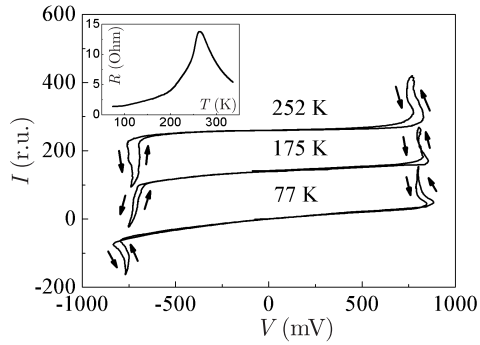

Figure 1: Temperature effect on current-voltage characteristics of Ag-LCMO contact; the inset demonstrates the resistance-vs-temperature dependence of a manganite plate

Some of the measurements were carried out in the pulsed mode to avoid effects caused by possible sample heating during the recording procedure. The experiment showed that in the pulsed regime, the current also increased at the same voltage values ~ 1 V as during the continuous recording mode. In addition, the current increase in the CVC occurred at the same voltage applied to the contact. Thus, the thermal effects did not determine the shape of the CVC.

Since the injector in the Ag-LCMO contact is a noble metal, it is reasonable to assume that the tunnel barrier is due to the oxygen deficiency at the manganite surface. The escape of oxygen ions from the barrier and near-barrier regions generates a system of localized levels capable to trap tunneling electrons. Such behavior is typical for barriers based on perovskites [5] where the tunneling electrons with the energy of junction bias eV can be captured by excited states (the so-called F-centers). The relaxation of the excited state is similar to the usual process of an electron-hole pair recombination that is trapped into the defect.

Typically, the excess energy of the F-center $E = eV$ is partly transferred to the vibrational degrees of freedom of the center and is scattered as a result of the emission of phonons. However, there is another possibility, when the entire vibrational energy E is localized on one of the oxygen ions surrounding the vacancy. Then, when the

condition $eV = E_a$ is satisfied (here E_a is the activation energy of the oxygen ion), the ion can jump to another vacancy. Thus, due to the capture of high-energy electrons by the excited localized state of the vacancy, it is possible for the oxygen vacancy to jump over.

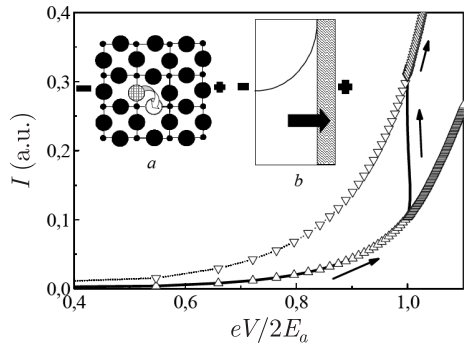

Figure 2: Expected current-voltage characteristic of tunnel junction as a result of transition (at $V = V_c$) of barrier surface layer into metallic-like phase; triangles correspond to the "equilibrium" characteristics; the insets show schematically an oxygen ion jumping to a vacancy (a) and a partial metallization of the tunnel barrier due to di usion of oxygen ions

As is known, the preferred direction of the jump of the activated ion is given by both the direction of the electric field and the electron flow. In our case, the direction of the tunneling electron flow plays a significant role. For a practically fixed voltage $V = V_c \sim 1$ V, the current increases above the intensity of the oxygen ion hopping processes which enhances the inhomogeneous metallization process (see the inset (a) in Figure 2) and accordingly the ohmic resistance $R = V/I$ of the contact decreases. When the voltage decreases the hopping processes in the direction of the electron flow are slowed down, but the activation of the oxygen ions is maintained. As a result, the direction of the jumps becomes more and more random, and the distribution of the oxygen vacancies along the barrier cross section gradually returns to the initial state. As a result of directed activated diffusion of vacancies, a non-equilibrium state with a more metallic nature of conductivity is formed in the region adjacent to the interface, with the effective number of carriers in this region growing monotonically with the increasing transport current. It leads to the appearance of a vertical current section in the $I(V)$ dependence (see Figure 2).

According to [1], the activation energy of oxygen ions is 450–500 meV, it agrees with the observed value of $eV_c \approx 2E_a \approx 1$ eV.

Conclusions

In summary, the current-voltage characteristics of Ag-LCMO contacts with tunneling conductivity exhibit anomalously nonlinear effects, in the absence of "remembering" of the reconstructed state. It is shown that these nonlinearities cannot be explained by thermal effects and are, in all probability, related to the motion of oxygen ions in the region of the tunnel barrier which are stimulated by the tunnel current and the electric field applied to the contact.

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