N-type current-voltage characteristics of manganites

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Abstract. Experimental data are shown for ceramic samples of La-Sr manganites with substitution of Mn by Zn, containing 0.15, 0.17 or 0.19 f.u. of Mn$^{4+}$ (under the condition that concentration of oxygen is stoichiometric). Bulk manganites were prepared by the traditional solid state reactions in air. All samples were single phase and crystallized in the orthorhombic structure. Dc I-V characteristics at different temperatures were measured without and with magnetic field, the strength of which was 9200 Oe. All I-V characteristics of manganites reveal the regions with negative differential resistance. The samples of La$_{0.885}$Sr$_{0.115}$Mn$_{0.925}$Zn$_{0.075}$O$_3$ exhibit novel multi-peak N-type current-voltage characteristics, particularly regular in the presence of magnetic field.

1. Introduction

Manganites with perovskite-type structure have perspective properties for a wide range of technical applications and are of interest as attractive objects for physical researches. Despite the large number of studies concerned with the electrical properties of manganites, the mechanisms of many transport processes in these materials are not yet fully understood. This is especially true in regard to nonlinear phenomena, such as threshold switching resulting in $S$- or $N$-type current-voltage (I-V) characteristics [1-4], colossal electroresistance and electron instability [5].

The nature of electrical threshold switching in so complex materials is not established unequivocally, and the earlier received results [1,5,6,7] provide some evidence for the connection of this effect with existence of mixed-phase state of manganites, deviation of oxygen content from stoichiometry and with inhomogeneities in conjunction with the theory of electronic phase transition. However, the available information about correlation between the type and the level of doping, structure and nonlinear electrical parameters is meager.

The aim of the study is to examine the influence of Mn$^{4+}$ concentration (dependent on divalent ions content) near the phase boundary “ferromagnetic insulator-ferromagnetic metal” on I-V characteristics of La$_{1-x}$Sr$_x$MnO$_3$ system with small diamagnetic dilution of Mn octahedral sublattice. substitution of Mn by Zn. The choice of Zn$^{2+}$ as a dopant was stimulated by the interesting and discrepant results reported previously [8,9].
2. Experimental

The experiments were performed on polycrystalline samples of La$_{1-x}$Sr$_x$Mn$_{0.925}$Zn$_{0.075}$O$_3$ system ($x=0.075; 0.095; 0.115$). Under the condition that concentration of oxygen is stoichiometric, these manganites contained 0.15, 0.17 and 0.19 f.u. of Mn$^{4+}$, accordingly. The samples were synthesized by traditional ceramic processing. The starting components (dried La$_2$O$_3$, SrCO$_3$, MnCO$_3$, ZnO powders) were mixed in stoichiometric proportions and ground in a ball mill with addition of alcohol. Pellets compacted of the obtained charge mixture were then preliminarily burned at 1273 K for 4 h. This operation was followed by grinding, introducing a binder (an aqueous solution of polyvinyl alcohol), pressing the samples, and burning out the binder (573 K, 40 min). The final sintering step was performed at 1473 K for 10 h, and the samples were cooled together with the furnace.

Phase purity, space group, homogeneity, and cell dimensions were determined by powder X-ray diffraction at room temperature (diffractometer using Cu$K_\alpha$ radiation). Measurements of dc $I$-$V$ characteristics were made using copper electrodes sputter-deposited onto opposite planes of pellets (thickness $\approx 4$ mm). Magnetic field of 9200 Oe was parallel to the current direction.

3. Results and discussion

All samples were single phase and crystallized in the orthorhombic structure (in La$_{0.885}$Sr$_{0.115}$Mn$_{0.925}$Zn$_{0.075}$O$_3$ the presence of very small amount of rhombohedral phase was possible). So, there is a qualitative agreement between our results and the classical data established earlier for La$_{1-x}$Sr$_x$MnO$_3$ system [10].

Manganite La$_{0.925}$Sr$_{0.075}$Mn$_{0.925}$Zn$_{0.075}$O$_3$ displays almost linear $I$-$V$ characteristic at the temperature 193 K, and N-type one at 128-129 K with magnetic field (Figure 1 (a)). Without magnetic field the current oscillates when voltage rises (Figure 1 (b)).

![Figure 1. I-V characteristics of manganite La$_{0.925}$Sr$_{0.075}$Mn$_{0.925}$Zn$_{0.075}$O$_3$ at different temperatures for (a) H=9.2 kOe, (b) H= 0](image-url)

All $I$-$V$ characteristics of manganites La$_{0.905}$Sr$_{0.095}$Mn$_{0.925}$Zn$_{0.075}$O$_3$ and La$_{0.885}$Sr$_{0.115}$Mn$_{0.925}$Zn$_{0.075}$O$_3$ (Figures 2, 3) reveal the regions with negative differential resistance. This regions are most pronounced at relative high temperatures (173-163 K) rather then at lower temperatures (133-129 K). Magnetic field has significant effect on the form of $I$-$V$ characteristics. The sign and the absolute value of magnetoresistance depend on the temperature and the applied electrical voltage. Meanwhile, there are the regions on the $I$-$V$ characteristics, where little dependence of the current on the voltage takes place (Figures 2).
The samples of La$_{0.885}$Sr$_{0.115}$Mn$_{0.925}$Zn$_{0.075}$O$_3$ exhibit novel multi-peak $N$-type current-voltage characteristics, particularly regular in the presence of magnetic field (Figure 3 (a)).

![Figure 2. $I$-$V$ characteristics of manganite La$_{0.905}$Sr$_{0.095}$Mn$_{0.925}$Zn$_{0.075}$O$_3$ at different temperatures for (a) $H=9.2$ kOe, (b) $H=0$](image1)

![Figure 3. $I$-$V$ characteristics of manganite La$_{0.885}$Sr$_{0.115}$Mn$_{0.925}$Zn$_{0.075}$O$_3$ at different temperatures for (a) $H=9.2$ kOe, (b) $H=0$](image2)

Obviously, the mechanism of insulator-metal transition in phase separation model, percolation, electrical field or current-driven transition cannot explain the phenomenon of $N$-type $I$-$V$ characteristics of manganites.

The $N$-type negative-resistance behavior of manganites is associated with the effect of enhancement of inelastic scattering of charge carrier in threshold field [4], charge/spin tunneling limited transport between competing coexisting phases, domains and grains in polycrystals, or can be ascribed to transformation of energetic level structure and carrier concentration in energy bands.
Further experimental and theoretical investigations are needed to more reliable and profound understanding of the mechanisms and processes leading to phenomenon electrical switching with $N$-type current-voltage characteristics of manganites.

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