Transport and magnetic properties of epitaxial LSMO thin films grown on MgO single-crystal substrates

V Štrbík1,3, M Španková1, M Reiffers2, J Kováč2 and Š Beňačka1

1Institute of Electrical Engineering, Slovak Academy of Sciences, 9 Dúbravská cesta, 841 04 Bratislava, Slovak Republic
2Institute of Experimental Physics, Slovak Academy of Sciences, 47 Watsonova, 040 01 Košice, Slovak Republic
E-mail: elekstrb@savba.sk

Abstract. Thin epitaxial La0.67Sr0.33MnO3 (LSMO) films were deposited on single crystal MgO substrates. The electrical transport and magnetic properties of the films in the temperature range 4 - 350 K were investigated and strong correlation between them was registered. Magnetoresistance up to 52 % at temperature T = 256 K and magnetic field B = 5 T was achieved. The results obtained indicate that LSMO films with such properties are suitable for application as “barrier” layers in superconducting-ferromagnetic-superconducting heterostructures, but optimization of LSMO film thickness is needed.

1. Introduction
The perovskite manganite La0.67Sr0.33MnO3 (LSMO) is an interesting material from several points of view. It is a material for colossal-magnetoresistance applications [1], for bolometric purposes [2] and is also used as a barrier material in the heterostructures superconductor (S) – ferromagnet (F) – superconductor (S) [3]. Structures consisting of oxide high-temperature superconducting (HTS) and ferromagnetic films (thickness < 100 nm) constitute an attractive model for investigating the interplay (proximity effect) between the two fundamental condensed-matter phenomena of superconductivity and ferromagnetism [4]. SFS layered heterostructures are promising for applications in devices manipulated by the spin-state of charge carriers; in particular, the S/F based Josephson technology holds great promise in nanoelectronics, e.g. as a physical realization of the qubit of quantum computation [5]. In this paper we report the results of the preparation and investigation (electrical and magnetic properties) of epitaxial LSMO thin films fabricated on MgO single-crystal substrates.

2. Experimental
The LSMO films were deposited by means of an on-axis dc magnetron sputtering system (Torus 2C, LESKER) onto single-crystal MgO (001) polished substrates at a temperature of 800 °C. The deposition was performed in an Ar (80%) + O2 (20%) gas mixture at a total pressure of 30 Pa. The thickness of the LSMO films was about 500 nm. In order to increase the oxygen content, the LSMO...
films were annealed in-situ in O₂ (10⁴ Pa) at 800 °C for an hour. After annealing, the temperature was lowered down to room temperature at a rate of 4 K/min [6].

The crystallographic orientation perpendicular to the film surface was determined by recording X-ray diffraction (XRD) patterns in the θ - 2θ configuration. The degree of the preferred orientation, perpendicular to the substrate, was derived from the full-width at half maximum (FWHM) of the rocking curve. The microstructure of the LSMO film was investigated by transmission electron microscopy (TEM) (JEOL 1200EX).

The electrical transport properties were measured by the 4-point dc method in a cryostat in temperature range 4 - 350 K. The magneto-resistive and magnetic properties of the films were measured on PPMS (physical properties measurement system) and MPMS (magnetic properties measurement system) equipment (Quantum Design) in temperature range 150 - 350 K and magnetic field up to 9 T.

3. Results

The XRD analyses showed (00l) oriented growth of the LSMO films with FWHM value of about 1 degree measured on the (001) LSMO peak due to the 8% mismatch of the lattice parameters of the LSMO film and the MgO substrate. The TEM investigations confirmed the epitaxial growth of the LSMO films and a 20 nm thin film surface of a polycrystalline character (only in the case of annealed samples).

The electrical resistivity ρ of the LSMO thin films was investigated in the temperature range 4 - 330 K. For the high-quality films at a temperature of \( T_p \gtrsim 300 \) K a resistivity peak (transition metal-insulator) was observed. For temperatures higher than \( T_p \) (insulating character) the resistivity vs. temperature dependence (\( ρ(T) \)) can be described by the variable range hopping (VRH) model (figure 1). The rapid decrease of the resistance for \( T < T_p \) (metal character) can be interpreted mainly by the electron-electron scattering, electron-magnon scattering and impurity (defects) grain-boundary scattering.

The non-monotonic dependence of the resistivity on the temperature can be characterized by a temperature coefficient of resistivity \( (TCR) = [(1/ρ)×dρ/dT]×100\% \) and its maximal value \( TCR_{max} \) indicates the most suitable temperature region for bolometric applications. Our best LSMO films reached \( TCR_{max} \approx 3\% K^{-1} \) at 290 K.

The magnetoresistivity (MR) of the LSMO films is shown in figure 2. The magnetoresistivity is defined by the relation \( MR = [(ρ_B - ρ_0)/ρ_0]×100\% \), where \( ρ_0 \) and \( ρ_B \) are the resistivities at zero and \( B \) magnetic field applied, respectively. The MR maximum was registered at the temperature \( T = 256 \) K; the MR depends nearly linearly on the parallel magnetic field applied. MR

![Figure 1. Resistivity vs. temperature dependence of LSMO films. The inset represents the fit of the variable range hopping (VRH) model to the experimental data for \( T > T_p \) (paramagnetic state).](image1)

![Figure 2. Magnetoresistivity of LSMO thin film. The MR maximum was achieved at \( B = 5 \) T and \( T = 256 \) K. The resistivity was changed by 52% compared to value at 0 T.](image2)
dependences taken at other temperatures are also shown in figure 2.

The further study showed the strong correlation between the magnetic and transport properties of the LSMO films. The MR maximum was achieved at \( T = 256 \) K, the minimum of \( \frac{d\chi'}{dT} \) (figure 3) was obtained at \( T = 250 \) K (\( \chi' \) being the real part of the susceptibility) and the \( TCR_{\text{max}} \) was found at \( T = 248 \) K. The correlation between \( T_p \) and \( T_{\text{Curie}} \) was mentioned above. The width of the ferromagnetic transition is about 100 K, as is indicated by \( \frac{d\chi'}{dT} \) (T) dependence in figure 3.

Thus, the LSMO films are ferromagnetic below 200 K, so that it is possible to use such films as barrier layers in superconductor-ferromagnetic-superconductor heterostructures operating at temperatures 77 K or lower.

4. Conclusion

Relatively thick (about 500 nm) epitaxial LSMO films were grown on single crystal MgO substrates. The electrical (resistivity peak at \( T_p \approx 290 \) K, \( TCR \)) properties indicate good results on such type of substrate. The magnetic measurements yielded maximal magneto resistivity at a temperature \( T = 256 \) K and magnetic field \( B = 5 \) T, ferromagnetic transition in the temperature range 200 - 300 K, Curie temperature \( T_{Curie} = 293 \) K. For temperatures below \( T_p \) (or \( T_{Curie} \)) the LSMO films behave as a metal (from electric point of view) and as a ferromagnet (from magnetic point of view). We believe that such properties allow us to use the LSMO thin films as “barrier” layers in HTS SFS structures in a regime of long-range proximity effects with the LSMO films thickness being in the range of 5 - 20 nm.

Acknowledgments

This work was supported by the VEGA Slovak Grant Agency under projects 2/7125/27 2/0007/09; the Slovak Research and Development Agency under contract No. VVCE-0058-07; the the Centre of Excellence of the Slovak Academy of Sciences and P.J. Šafárik University. The liquid nitrogen for the experiment was sponsored by U.S. Steel Košice, s.r.o.

References

[1] Jin S, Tiefel T H, McCormack M, Fastnacht R A, Ramesh R and Chen L H 1994 Science 264 413
[2] Goyal A, Rajeswari M, Shreekala R, Lofland S E, Bhagat S M, Boettcher T, Kwon C, Ramesh R and Venkatesan T 1997 Appl. Phys. Lett. 71 2535
[3] Alvarez G A, Wang X L, Peleckis G, Shi D Q and Dou S X 2007 Physica C 460-462 438
[4] Buzdin A I 2005 Rev. Mod. Phys. 77 935
[5] Ortlepp T, Ariando, Mielke O, Verwijs C J M, Foo K F K, Rogalla H, Uhlmann F H and Hilgenkamp H 2006 Science 312 1495
[6] Španková M, Chromík Š, Vávra I, Sedláčková K, Lobotka P, Lucas S and Stanček S 2007 Appl. Surf. Sci. 253 7599