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Investigation of Magnetoresistance and Its Anisotropy of Thin Polycrystalline $La_{0.83}Sr_{0.17}MnO_3$ Films in High Pulsed Magnetic Fields

N. Žurauskienė[a,b,*], S. Keršulis[a], L. Medišauskas[a] and S. Tolvaišienė[b]

[a]Semiconductor Physics Institute, Center for Physical Sciences and Technology
A. Goštauto 11, Vilnius, Lithuania

[b]Vilnius Gediminas Technical University, Sauletekio al. 11, Vilnius, Lithuania

The results on the study of grain boundary effects and influence of film deposition conditions on the magnetoresistance and its anisotropy in polycrystalline $La_{0.83}Sr_{0.17}MnO_3$ films are presented. The magnetoresistance was measured in high pulsed magnetic fields up to 25 T (pulse duration $\approx$ 0.6 ms) in the temperature range of 120–300 K. A modified Mott hopping model was applied to describe the main behavior of high-field magnetoresistance for both ferromagnetic and paramagnetic phases of the polycrystalline films by taking into account the demagnetization field of the films measured in low magnetic fields perpendicular to film plane. It was also found that to obtain the higher magnetoresistance saturation field at room temperature it is necessary to use the films with smaller crystallites ($D \approx 100$ nm). Such films could be used for design of megagauss pulsed magnetic field sensors.

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1. Introduction

The renewed interest in doped polycrystalline perovskite manganites during last decade has been motivated by fundamental understanding as well as potential device applications [1]. It was demonstrated that colossal magnetoresistance (CMR) phenomenon in thin manganite films can be successfully used for the development of B-scalar sensors, which are able to measure magnitude of high pulsed magnetic fields up to 40 T in the volume $\approx 5 \times 10^{-2}$ mm^3 [2]. However, in low fields (< 1 T) the CMR depends on the direction of magnetic field in respect of the film plane and current direction. The anisotropy of magnetoresistance is the main reason of B measurement error in low fields. At high magnetic fields the CMR effect tends to saturation which limits application of manganite films for megagauss magnetic field measurement. Therefore, the investigations of magnetoresistance and its anisotropy of polycrystalline films are of great importance. It was demonstrated that grain boundaries (GBs) naturally formed between randomly oriented crystallites during growth of polycrystalline manganite films are mostly responsible for transport properties of such films. Various models suggesting different origins of the transport process were reported in literature to explain experimental results: spin-polarized tunnelling of free carriers through grain boundaries; spin-dependent scattering of polarized free carriers, and models based on "activated carrier transport", e.g. variable range hopping (VRH) or activated carrier transport within the disordered mesoscopic region adjacent to a GB with depressed magnetization and *Curie* temperature T_C [3–5].

In this paper, the results on the study of grain boundary effects and influence of film deposition conditions on the magnetoresistance and its anisotropy in polycrystalline $La_{0.83}Sr_{0.17}MnO_3$ (LSMO) films are presented and discussed.

2. Experimental

The LSMO films were deposited by a pulsed injection metal organic chemical vapour deposition (PI MOCVD) technique on a special polycrystalline lucalox substrate (99.9% Al_2O_3 + 01% MgO). Such substrate was used to obtain polycrystalline structures of the films with randomly oriented crystallites separated by structurally and magnetically disordered GB. To clarify the role of GBs in the magnetoresistance (MR) behaviour, we investigated films grown at various deposition temperatures T_d resulting in different average sizes D of crystallites in the films. The thicknesses of the films were 400 nm. The morphology and the dimensions of the crystallites were studied using atomic force microscopy (AFM). It was found that dimensions of crystallites decrease with a decrease of T_d. For films grown at $T_d = 750\,^\circ$C, $700\,^\circ$C

* corresponding author; e-mail: zurausk@pfi.lt

and 650 °C (f-750, f-700 and f-650) the obtained D was 170 nm, 140 and 110 nm, respectively. The samples were fabricated using a conventional integrated circuit processing technique. They had a co-planar shape with two 0.4 mm × 0.4 mm square Ag electrodes deposited by thermal evaporation and separated by a distance of 50 μm. The contacts were annealed in argon atmosphere at 420 °C for 40 min. Therefore, the active volume of each sample was only 400 μm × 50 μm × 0.4 μm. Then the samples were soldered to wires bifilarly twisted in the direction perpendicular to the surface of the films. The resistivity ρ dependence on temperature T was investigated using a low dc electric field in a closed cycle helium gas cryocooler in the range 4–300 K. The low field magnetoresistance measurements were performed in the cryocooler using an electromagnet with maximum amplitude of 0.8 T. High field magnetoresistance was measured in the range of 100–320 K using a pulsed magnetic field generator equipped with a multi-shot coil that produced sine waveform 0.6 ms long magnetic field pulses with amplitudes up to 40 T. The magnetoresistance anisotropy MRA was studied applying magnetic fields in two different configurations: (1) in the film plane parallel to the current direction, and (2) perpendicular to the plane.

3. Results and discussion

The resistivity dependence on temperature of the investigated LSMO films showed metal–insulator transition at a certain temperature T_m: 165 K, 205 K and 230 K for films f-650, f-700 and f-750, respectively. The resistivity maximum values ρ_m at this temperature for the same films were the following: 54 Ω cm, 6.5 Ω cm and 1.6 Ω cm. The decrease in film T_m and increase in ρ_m with decrease of the deposition temperature is a result of increased grain boundary material which is more structurally and magnetically disordered in comparison with crystallites interior. The study of magnetoresistance $\mathrm{MR} = \{[\rho(B) - \rho(0)]/\rho(0)\} \cdot 100\%$ showed that it also depends on the preparation conditions of the films and magnetic field direction in respect of the film surface. Figure 1 presents the MR at low magnetic field for sample f-700. The observed anisotropy remains up to high magnetic fields (see Fig. 2). The main reason of the MRA might be the demagnetization effect due to high aspect ratio of the sample.

It was shown [5] that grain boundaries naturally formed due to polycrystalline lucalox substrate in LSMO films might be assumed as ferromagnetic regions with suppressed magnetic order, and the MR properties of these polycrystalline films could be analyzed using modified Mott's hopping model [6]. In this work, we studied magnetoresistance anisotropy taking into account the demagnetization field DM of the films. In such a case the MR in ferromagnetic state is supposed to scale the Brillouin function $\mathcal{B}$:

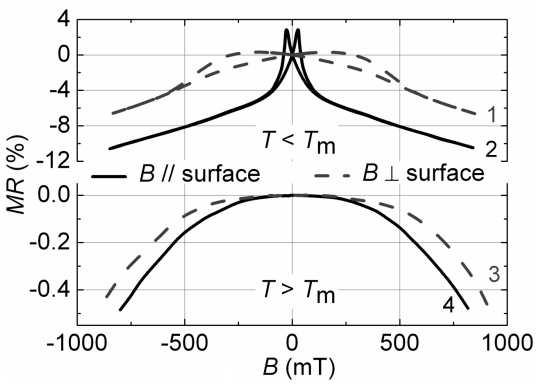

Fig. 1. Low field magnetoresistance in ferromagnetic (1, 2) and paramagnetic (3, 4) state. $T_\mathrm{d} = 700\,^\circ\mathrm{C}$.

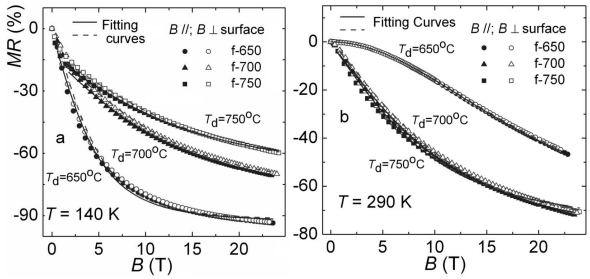

Fig. 2. Experimental (symbols) and fitted (lines) high field magnetoresistance dependences for LSMO films grown at various deposition temperatures: (a) ferromagnetic, (b) paramagnetic state.

$$\mathrm{MR}(B) = A \cdot \mathcal{B}_{jg\mu_\mathrm{B}k_\mathrm{B}}(T, J, B - \mathrm{DM}) + \mathrm{LFMR}\,, \quad (1)$$

in paramagnetic state to scale $\mathcal{B}^2$:

$$\mathrm{MR}(B) = A \cdot \mathcal{B}^2_{jg\mu_\mathrm{B}k_\mathrm{B}}(T, J, B - \mathrm{DM})\,, \quad (2)$$

while at intermediate temperatures between T_m and T_C the results were fitted by sum of both contributions

$$\mathrm{MR}(B) = A \cdot \big[(1-f)\mathcal{B}_{jg\mu_\mathrm{B}k_\mathrm{B}}(T, J, B - \mathrm{DM}) + f\mathcal{B}^2_{jg\mu_\mathrm{B}k_\mathrm{B}}(T, J, B - \mathrm{DM})\big] + \mathrm{LFMR}\,. \quad (3)$$

Ferromagnetic phase fraction f, spin moment J and MR amplitude A are fitting parameters. Low-field magnetoresistance LFMR and demagnetization field DM were obtained from low-field measurements performed in parallel and perpendicular magnetic field configurations, respectively. The J values were found to be similar for all investigated films. The maximal $J \approx 18$–20 values were obtained in the vicinity of T_m of each film and decreased with the decrease of temperature. Such J values indicate that the GBs in polycrystalline films behave like a superparamagnet of magnetically aligned clusters with extension of ≈ 9–10 manganese ions. The J values were found to be similar for both parallel and perpendicular magnetic field configurations. This means that the MR

anisotropy of the films at high magnetic fields could be explained taking into account only the demagnetization field of the films.

The parameter A shows the largest possible MR values. Therefore, the product $A\rho_0$ indicates the largest possible resistivity change $\Delta\rho$ due to action of magnetic field. Figure 3 presents resistivity vs. temperature dependences for two LSMO films. The residual resistivity ρ_r is obtained after subtraction of fitted $\Delta\rho = A\rho_0$ from zero field resistivity ρ_0. It presents the resistivity in the limit of magnetic field saturation. The largest ρ_r values were obtained for films grown at lowest deposition temperature (with smallest crystallites).

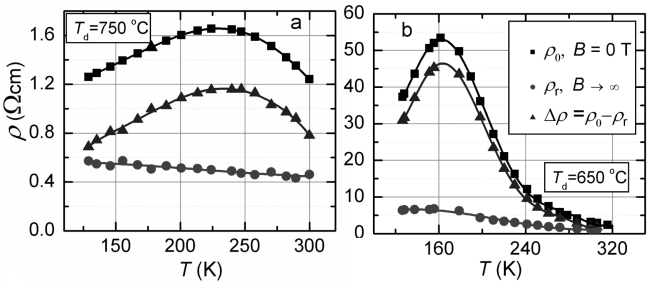

Fig. 3. Resistivity vs. temperature dependences for two LSMO films: ρ_0 — zero field resistivity; ρ_r — residual resistivity; $\Delta\rho$ — maximal resistivity change obtained as fitting parameter $A\rho_0$.

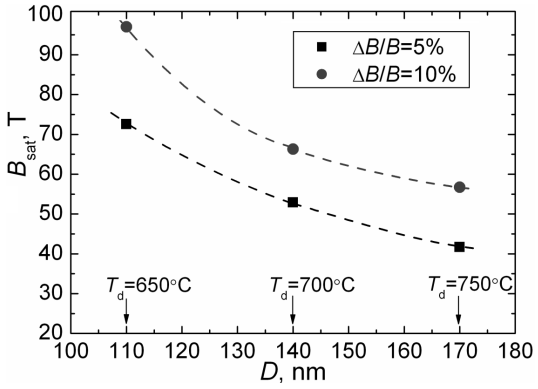

Fig. 4. Dependence of saturation magnetic field vs. average dimensions D of crystallites in LSMO films grown at different deposition temperatures T_d. The dashed lines are guides to the eye.

At very high magnetic fields the MR tends to saturation. For epitaxial films the saturation starts at 7–10 T, while for polycrystalline films at 40–60 T [4, 6]. The saturation of MR makes it difficult to apply manganite films for the development of very high (up to megagauss) magnetic fields sensors. We defined saturation magnetic field B_{sat} as a technical parameter which depends on magnetic field measurement error $\delta_B = (\Delta B/B) \cdot 100\%$ and accuracy of recording equipment $\delta_U = (\Delta U/U) \cdot 100\%$. For a given δ_B value, the change of magnetic field ΔB results in the change of manganite sensor's resistance ΔR which in turn corresponds to a change of voltage drop ΔU across this resistance. Therefore, the magnetic field at which measurement of ΔU is limited by accuracy of recording equipment δ_U, was defined as saturation magnetic field B_{sat}. Figure 4 presents dependences of B_{sat} values vs. average dimensions of crystallites obtained for $\delta_B = 5\%$ and 10%, when $\delta_U = 0.3\%$. B_{sat} was found from approximation of MR(B) dependences at room temperature by the Brillouin function (Eq. (2)) extended up to 100 T. One can see that B_{sat} increases with the decrease of crystallite's dimensions in the polycrystalline films. It has to be noted that with decrease of D the amount of grain boundary material in the films increases. On the other hand, B_{sat} of epitaxial films ($D \to \infty$) of the same composition was found to be much smaller: ≈ 15 T, for given $\delta_B = 5\%$. Therefore, the polycrystalline films grown at lowest deposition temperature could be used for the development of megagauss magnetic field sensors.

4. Conclusions

In conclusion, the modified Mott hopping model could be applied to describe the main behaviour of high-field MR of polycrystalline LSMO films and its anisotropy taking into account the demagnetization field of the films.

The higher MR saturation field could be obtained by growing films with smaller crystallites ($D \approx 100$ nm). Such films could be used for design of megagauss pulsed magnetic field sensors.

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