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COMMUNICATIONS

CHARACTERIZATION OF ELECTRICAL TRANSPORT IN LSMO WITH ENHANCED TEMPERATURE OF METAL–INSULATOR TRANSITION

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We have studied $La_{0.67}Sr_{0.33}MnO_3$ (LSMO) thin films with temperature of metal-insulator (T_{MI}) transition enhanced to above 400 K, and we estimated characteristic electrical transport mechanisms for these films. We have fitted the measured resistivity vs. temperature $\rho(T)$ dependence in a wide temperature range 4–500 K using different mechanisms of the electrical transport in different parts of $\rho(T)$. In addition to the narrow temperature range around T_{MI} very well agreement was found. We found out that the Debye's temperature was also increased ($\Theta_D \approx 840$ K) probably due to the change in crystallization of LSMO films.

K e y w o r d s: LSMO thin film, electrical transport, Debye temperature

1 INTRODUCTION

Perovskite manganites exhibit strongly correlated magnetic and electrical transport properties. At a certain temperature T_{MI} manganites undergo a metal-insulator transition and a ferromagnetic (FM) to paramagnetic (PM) transition as well. At temperatures higher than T_{MI} the resistance temperature dependence behaves like insulators and magnetically they are in a PM state. For temperatures below T_{MI} they behave like metals and are ferromagnetic (ferromagnetic-metal state). The metal-insulator transition is very simply observable as a resistance peak in the $\rho(T)$ dependence and T_{MI} corresponds to the temperature of the maximal resistance. Among the manganites the $La_{0.67}Sr_{0.33}MnO_3$ (LSMO) is the most attractive material because of its high Curie temperature. For high quality $La_{1-x}Sr_xMnO_3$ ($x = 0.3$ and 0.4) single crystals the Curie temperature $T_C \approx 370$ K was referred [1]. Recently Sadock *et al* [2] published their results that the Curie temperature can reach a value up to 650 K for a multilayer LSMO/$BaTiO_3$ heterostructure.

We have prepared the LSMO thin films with the enhanced value of $T_{MI} > 400$ K whereby detailed X-ray analyzes showed changes in crystallization of these films [3]. While the films with $T_{MI} < 370$ K crystallize in a pseudo-cubic form, the films with $T_{MI} > 370$ K have a proper microstructure with distorted orthorhombic unit cells. Therefore, it was interesting to examine whether the new crystallization has an influence on the LSMO electrical transport properties.

2 EXPERIMENTAL

The LSMO films were deposited using on-axis dc magnetron sputtering from a stoichiometric $La0_{.67}Sr_{0.33}MnO_3$ ceramic target onto one-side polished MgO (001) substrates. The deposition was performed in an $Ar(80\%) + O_2(20\%)$ atmosphere at a total pressure of 65 Pa. The substrate was heated to 800 °C during the deposition. The thickness of the LSMO films was close to 100 nm and the film growth rate was around 12 nm/min. The LSMO films were subsequently in-situ annealed in $O_2(10^4\,\mathrm{Pa})$ at 800 °C for an hour [3]. For the $\rho(T)$ measurement a standard four-point method was applied. The electrodes on the samples were formed with a silver paint and the sample was heated in a calibrated furnace.

3 RESULTS AND DISCUSSION

Structural and compositional analyses (Auger electron spectroscopy, X-ray diffraction, Transmission electron microscopy) indicate that the LSMO films are stoichiometric, single phase and epitaxial [3, 4], in spite of some non-ideal crystallographic matching of the LSMO film with the MgO substrate. The main attribute of the LSMO films — the enhanced T_{MI} — we were able to prepare reproducibly. The experimental resistivity vs. temperature $\rho(T)$ dependence of the LSMO film on the MgO substrate is shown in Fig. 1. The resistivity peak is relatively broad and the maximal ρ is at $T_{MI} = 400$ K. The $\rho(T)$ dependence measured in the temperature range 4–500 K can be divided into several parts. The approximate boundaries between the parts are denoted by black points with corresponding temperatures T_0–T_3 (in Fig. 1). Each

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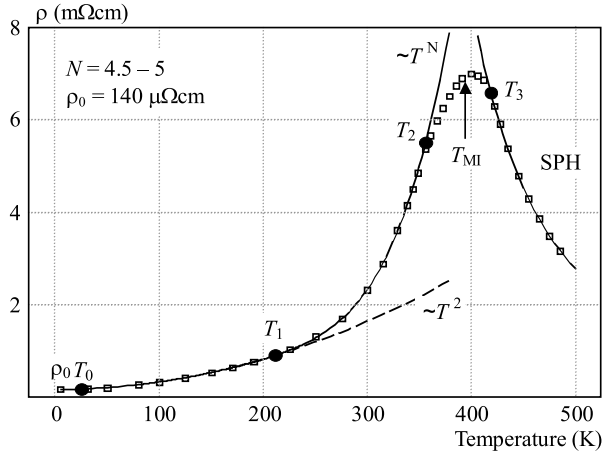

Fig. 1. The measured resistivity vs. temperature dependence of LSMO film on MgO substrate. The black points determinate the parts of dependence with own characteristic scattering mechanism (more in text)

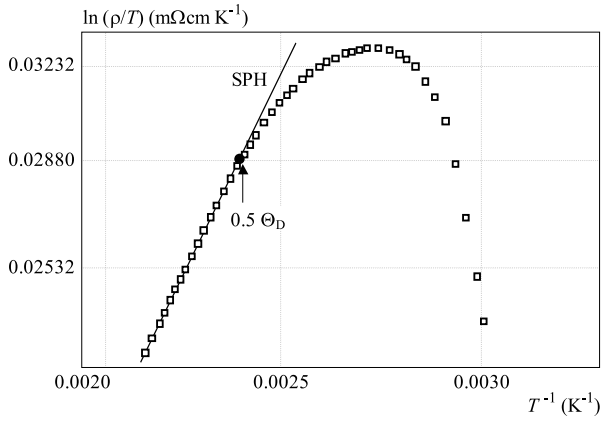

Fig. 2. The plot of $\ln(\rho/T)$ *vs* T^{-1} dependence. Full line represents the SPH model

part of the $\rho(T)$ is characterized by a specific scattering mechanism.

The resistivity in the lowest temperature range (below $T_0 \approx 20$ K) exhibits essentially constant value of the residual resistivity ρ_0, which is determined mainly by temperature independent scattering on impurities, defects, grain boundaries or domain walls. The typical value of ρ_0 is in the range of 100–200 $\mu\Omega$cm and it is about two times higher than ρ_0 of the single crystal [1] or of the high quality LSMO thin films [5]. In the temperature range $T_0 - T_1$ (range 20-220 K) the resistivity data follows the T^2 dependence. Such a dependence of the resistivity is frequently observed and it is ascribed to the intrinsic property of La-based manganites [6] to electron-electron scattering [1] or to one-magnon scattering [7].

Above the temperature $T_1 (\approx 220$ K) the resistivity increases much more than T^2 dependence (see Fig. 1) and the temperature dependence of higher power is evident. The double exchange theory which describes the ferromagnetic ground state offers the $T^{4.5}$ dependence of the resistivity due to an the electron-magnon scattering [8] and some supporting role in this temperature range plays the electron-phonon scattering with the T^5 dependence [5]. Recently we have introduced the modified polynomial fit formula $\rho = \rho_0 + \rho_2 T^2 + \rho_N (T - T_N)^N$, where ρ_0, ρ_2, ρ_N and T_N are fitting parameters and $4.5 \leq N \leq 5$ [9]. This formula fits well the experimental dependence from 4 K up to the temperature $T_2 (\approx 355$ K) (Fig. 1, full line for $T < \mathrm{T_{MI}}$) and has significantly increased temperature range of validity in comparison to previous approaches [1, 5–7], where authors fitted their experimental dependences only to temperatures 250–300 K. The temperature T_2 appears significant because at this temperature the LSMO starts to undergo from the ferromagnetic to the paramagnetic state and simultaneously to the metal-insulator transition. This transition can be described only qualitatively. As it was mentioned above the electron-magnom and electron-phonon scatterings are responsible for the rapid increase of the resistivity in the temperature range T_1– T_2. For further temperature increase (above T_2) the phonon scattering becomes stronger and a polaron conductivity is created. Just above T_2 the polarons are heavy free carriers scattered by phonons and give positive $\mathrm{d}\rho/\mathrm{d}T$ [6]. At the temperature $\mathrm{T_{MI}}$ the phonon scattering is so strong that the polaron carriers are localized and further electrical transport for the temperature above $\mathrm{T_{MI}}$ is possible only through a thermally activated hopping mechanism with negative $\mathrm{d}\rho/\mathrm{d}T$ [6]. The transport in high-temperature range (above $T_3 \approx 420$ K) is well described by small polaron hopping (SPH) theory $\rho = \rho_h \mathrm{T} \exp(E_h/k_B T)$ [10], where E_h is the hopping energy, k_B is Boltzman's constant and ρ_h is a resistivity coefficient. The full line in the temperature range above $\mathrm{T_{MI}}$ in Fig. 1 represents the SPH model. One can see that the presented transport mechanisms well describe the $\rho(T)$ of the LSMO thin films with the enhanced $\mathrm{T_{MI}}$.

The SPH model is valid for temperatures $T > \Theta_D/2$, where Θ_D is Debye's temperature [11]. Our detailed graph (Fig. 2) indicates that the validity of the SPH model (full line) finishes at the temperature $T \sim 420$ K *eg* $\Theta_D \approx 840$ K. This is significantly higher than the reported one before ~ 500 K [11, 12]. The increased value of Θ_D can be evoked by the change in the crystallization of the LSMO thin films with $\mathrm{T_{MI}} > 370$ K and also the correlation between Θ_D and $\mathrm{T_{MI}}$ should be studied in more details.

4 CONCLUSIONS

We have prepared $\mathrm{La_{0.67}Sr_{0.33}MnO_3}$ (LSMO) thin films with the enhanced temperature of metal-insulator transition ($\mathrm{T_{MI}} \geq 400$ K). The resistivity *vs* temperature $\rho(T)$ dependence in the temperature range 4–500 K was characterized by different transport mechanisms in different parts of $\rho(T)$. The temperature independent scattering, electron-electron, one-magnon, electron-magnon

and electron-phonon scattering mechanisms were considered to describe the $\rho(T)$ dependence in the temperature range $0 < T \leq 355\,\mathrm{K}$. In the temperature interval $355\,\mathrm{K} < T < 420\,\mathrm{K}$ the LSMO overcomes ferromagnetic-paramagnetic and metal-insulator transitions via polarons conductivity. For temperatures higher than T_{MI} the polarons become localized and further transport is performed only by thermal activated hopping. The transport above 420 K is described by the model of small polaron hopping and from this dependence we have estimated the Debye's temperature $\Theta_D \approx 840\,\mathrm{K}$.

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