Electrical transport in epitaxial and polycrystalline thin LSMO films

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Abstract. We report studies on the electrical transport in La0.7Sr0.3MnO3 (LSMO) thin films of different quality. The temperature dependence of the resistivity (ρ-T) of high-quality LSMO films (ρ ≲ 10 mΩ cm) can be described by a polynomial approximation based on electron-magnon, electron-phonon, electron-electron and temperature-independent scatterings. On the other hand, the ρ-T dependence of polycrystalline LSMO films prepared under less than optimal conditions (ρ ≳ 1 Ω cm) can be described by a two-conductance-channel model. The first one is a spin-polarized tunneling between neighboring ferromagnetic grains with good contacts, while the other is implemented via a thermally-activated transport through an insulator (semiconductor) phase located mainly at the grain boundaries or between grains.

1. Introduction

In what concerns the family of manganites, a large number of studies have been focused on La0.7Sr0.3MnO3 (LSMO) as it possesses the highest Curie temperature [1-3]. The manganites exhibit a large variety of electrical transport properties depending on their quality. At low-temperatures (T < 10 K), the LSMO resistivity has values from ρ0 ≈ 60 – 80 μΩ cm for high-quality epitaxial films or single crystals [4,5] or up to ρ > 100 Ω cm for very thin films (< 10 nm) [2], non-stoichiometric or not-optimally oxidized LSMO [5,6], or polycrystalline (granular) thin films [7]. A resistivity maximum appears in the temperature dependence of the resistivity (ρ-T) at a certain temperature TMI indicating a metal-insulator transition and a simultaneous ferromagnetic (FM) – paramagnetic (PM) transition. To interpret the ρ-T dependence of high-quality LSMO thin films (ρ ≤ 10 mΩ cm), the electron-magnon interaction, electron-phonon and electron-electron scattering, as well as scattering on impurities and defects, should be taken into account [4,8,9]. The ρ-T dependence of granular (polycrystalline) composite or not-optimally prepared LSMO thin films (ρ ≳ 1 Ω cm) can be described by a model of two intergrain conduction channels [10,11]. The first channel is spin-polarized tunneling between neighboring ferromagnetic grains with a good contact, the other, a thermally-activated transport through an insulator (semiconductor) phase situated at the grain boundary or between grains.

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In this paper we present a possibility to approximate and characterize the transport mechanisms involved in the $\rho$-$T$ dependences of LSMO thin films of different quality. A polynomial fit equation describes more adequately epitaxial LSMO films of high quality, while the two-channel model is better suited to the description of polycrystalline and/or less oxidized films.

2. Experimental

RF magnetron sputtering was used to deposit ferromagnetic La$_{0.7}$Sr$_{0.3}$MnO$_3$ thin films. During the sputtering in Ar:O$_2$ (1:1) atmosphere, with a total pressure of 5.3 Pa, a RF power supply of 30 W and a substrate temperature of 780°C were applied. The thickness of the LSMO layer was about 30 nm. The LSMO films deposited on SrTiO$_3$ single-crystal substrates showed high crystallographic quality, a high $T_{MI}$ value (342 K), ferromagnetic properties at room temperature and low resistivity [12], whereas the LSMO films deposited on $\tau$-cut Al$_2$O$_3$ single-crystal substrates exhibited polycrystalline character, low $T_{MI}$ values (below 250 K) and a high resistivity ($\rho \geq \Omega$ cm) [13].

The patterning of the basic structure was carried out by a photolithographic process and argon-ion-beam etching with cooling the substrate. After the patterning, the DC 4-point measuring method was used.

3. Results and discussions

A typical $\rho$-$T$ dependence of our low-resistivity LSMO films is shown in figure 1. The values of the metal-insulator transition $T_{MI} = 342$ K, $\rho_{\text{max}} \leq 5\Omega$ cm and the low-temperature residual resistivity $\rho_0 \approx 150 \mu\Omega$ cm are comparable with other high-quality epitaxial thin films [4,14]. We approximated the $\rho$-$T$ dependence in the ferromagnetic-metallic state ($0 < T < T_{MI}$) by a polynomial fit equation $\rho(T) = \rho_0 + \rho_2 T^2 + \rho_N (T - T_0)^N$, where $\rho_0$, $\rho_2$, $\rho_N$, $T_0$ and $N$ are fitting parameters [9]. $\rho_0$ represents the residual resistivity at low temperatures (figure 1, inset) and is determined predominantly by temperature-independent scattering on defects and impurities, i.e., $\rho_0$ indicates the quality of the LSMO film. The second term ($\sim T^2$) characterizes the electron-electron [5] or single-magnon scattering [15].

The first two terms of the polynomial equation describe well the $\rho$-$T$ dependence up to a temperature of about 0.5 $T_{MI}$ (figure 1, inset). The third term represents mainly the contribution from the electron-magnon interaction according to the FM double-exchange theory ($\rho \sim T^{4.5}$) [16]. This interaction (figure 1, dotted line) cannot explain the increase of resistivity at temperatures close to $T_{MI}$ alone; thus, additional contributions from electron-phonon ($\rho \sim T^6$) [4,17] or from polaron-phonon scattering [8] should be taken into account. We fitted (figure 1, solid line) the experimental $\rho$-$T$ dependence in a ferromagnetic-metallic state by a polynomial expression with the parameters $\rho_0 = 0.141 \ m\Omega \ cm$, $\rho_2 = 1.15 \times 10^{-5} \ m\Omega \ cm \ K^2$, $T_0 = 117$ K and $\rho_N = 5.97 \times 10^{-13} \ m\Omega \ cm \ K^{-5.5}$ for $N = 5.5$.

![Figure 1. Approximation of the resistivity vs. temperature dependence of a high-quality LSMO thin film by a polynomial expression (solid line) in a ferromagnetic-metallic state ($0 < T < T_{MI}$) and by the small polaron hopping (SPH) model (dot-dashed line) in a paramagnetic-insulating state ($T > T_{MI}$). The dotted line indicates the contribution to resistivity by the double-exchange theory; the $\rho$-$T$ dependence in a log-log scale is shown in the inset.]
The electrical transport in a paramagnetic-insulating state \((T > T_{MI})\) is determined by the small polaron hopping (SPH) mechanism \(\rho(T) = \rho_h T \exp(E_h/k_B T)\), where \(E_h\) is the hopping energy, \(k_B\) is Boltzmann’s constant and \(\rho_h\) is a coefficient \([18,9]\). The SPH approximation with parameters \(\rho_h = 1.538 \times 10^{-3}\) m\(\Omega\) cm K\(^{-1}\) and \(E_h = 1.076 \times 10^{-20}\) J is shown in Fig.1 as dot-dashed line for \(T > T_{MI}\).

The \(\rho\)-\(T\) dependences of polycrystalline LSMO films can be interpreted in the framework of the phenomenological model of two intergrain conduction channels developed for granular or composite ferromagnetic materials \([10,11]\). The first type of channel is related to intrinsic transport properties of the system and is implemented through spin-polarized tunneling between neighboring ferromagnetic grains with good contacts. The other type is realized through thermal activation of the insulator phase arising from the grain boundary, contaminations, crystal disorder and poor connectivity between grains. The total resistivity \(I\) is determined by the sum of the conductances of the two channels:

\[
\frac{1}{\rho} = \frac{A_f}{\rho_f} + \frac{A_i}{\rho_i},
\]

where \(A_f\) and \(A_i\) are effective constants and fit parameters. The dimensionless resistivity \(\rho_i\) of the insulator phase channel is modeled by a semiconductor-like resistivity

\[
\rho_i = e^{W_2/(2k_B T)},
\]

and the resistivity \(\rho_f\) of the channel responsible for spin-polarized tunneling, is

\[
\rho_f = e^{[E_c + W_1(1-m^2)]/(2k_B T)},
\]

where \(k_B\) is Boltzmann’s constant, \(T\) is the absolute temperature, \(W_2\) is the insulator-phase activation energy, \(E_c\) is the charging energy, \(W_1(1-m^2)\) is the magnetic correlation function of two neighboring grains of the same volume and shape, \(W_i\) is a temperature- and magnetic-field-independent constant and \(m\) is the magnetization normalized to the saturation value for each grain.

The numerical fit (solid line) of the experimental \(\rho\)-\(T\) dependence (dots) according to the two-channel model for three different LSMO films is shown in figure 2.

![Figure 2](image)

**Figure 2.** Numerical fit (solid line) of the experimental \(\rho\)-\(T\) dependence according to the two-channel model for three different LSMO films: a) polycrystalline LSMO film covered with non-superconducting YBCO b) thin polycrystalline film c) less-oxidized polycrystalline LSMO film.

The insulator behavior of the \(\rho\)-\(T\) dependence for the polycrystalline LSMO covered with non-superconducting YBCO (figure 2a) can be explained mainly by the transport channel with a thermal activation of carriers in an insulator phase \((\rho)\) because no ferromagnetic contribution was registered. In the other two cases (Fig. 2 b, c), the \(\rho\)-\(T\) dependence is a combination of insulating and ferromagnetic phases. To approximate the experimental \(\rho\)-\(T\) dependences, it is necessary to find adequate fitting parameters for the two channels to match the experimental dependence. As fitting parameters we used \(A_f, A_i, W_1, W_2, E_c\) and \(T_{Curie}\) \([10]\); their values are shown in table 1.
The two-channel model is suitable for approximating the $\rho$-$T$ dependences in the cases when manganite exhibits a low-temperature resistivity upturn.

Table 1. Fitting parameters of the two-channel model for the polycrystalline LSMO films in figure 2.

| Sample  | $A_f$ (Ω⁻¹) | $A_i$ (Ω⁻¹) | $W_1/2k_B$ (K) | $W_2/2k_B$ (K) | $E_C/2k_B$ (K) | $T_{\text{Curie}}$ (K) |
|---------|-------------|-------------|----------------|----------------|----------------|---------------------|
| figure 2a | 60000       | 150000000   | 50             | 600            | 50             | 340                 |
| figure 2b | 8500        | 5000000     | 400            | 1180           | 7              | 320                 |
| figure 2c | 527.5       | 18000       | 442            | 1310           | 2.5            | 340                 |

4. Conclusions

We presented approaches to approximating the $\rho$-$T$ dependences of LSMO thin films. A polynomial fit formula describes properly the $\rho$-$T$ dependence of high-quality epitaxial LSMO films ($T_{\text{MI}} > 300$ K, $\rho \leq 0.1 \Omega \text{cm}$, $\rho_0 \approx 150 \mu \Omega \text{cm}$) in a ferromagnetic-metallic state ($T < T_{\text{MI}}$). The polynomial fit formula includes the following transport mechanisms: scattering on impurities and defects, and electron-electron, electron-magnon and electron-phonon scattering. The electrical transport in a paramagnetic-insulating state ($T > T_{\text{MI}}$) can be described by the small polaron hopping model. In the case of the LSMO films of high resistivity ($\rho \geq \Omega \text{cm}$) and with a low-temperature resistivity upturn, the two-channel model describes well the experimental $\rho$-$T$ dependences. One channel characterizes the thermally-activated transport through the insulator at the grain boundary, the other one describes the transport through spin-polarized tunneling between neighboring ferromagnetic grains with good contacts.

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