Investigation of the resistive properties of HTS/manganite bilayers

T Nurgaliev¹, B Blagoev², V Štrbik³, Š Chromik³ and M Sojková³

¹Emil Djakov Institute of Electronics, Bulgarian Academy of Sciences, 72 Tzarigradsko Chaussee, 1784 Sofia, Bulgaria
²Georgi Nadjakov Institute of Solid State Physics, Bulgarian Academy of Sciences, 72 Tzarigradsko Chaussee, 1784 Sofia, Bulgaria
³Institute of Electrical Engineering, Slovak Academy of Sciences, 9 Dubravska cesta, 84104 Bratislava, Slovak Republic

E-mail: blago_sb@yahoo.com

Abstract. Temperature dependences of the resistivity of manganite La₇₀Ca₃₀MnO₃ (LCMO) films deposited on LaAlO₃ and SrTiO₃ substrates by RF magnetron sputtering were shown to be successfully simulated in the whole temperature range (covering metal, insulator and metal-insulator transition regions) using a phenomenological phase-coexistence transport model. Quantitative data on the internal parameters of these films were obtained. The possibility was also considered for investigation of individual resistive characteristics and excess conductivity of a high temperature superconducting (HTS) YBa₂Cu₃O₇₋ₓ (YBCO) thin film in the vicinity of Tc included into a YBCO/LCMO bilayer structure. It was shown that the considered YBCO film in the temperature range from 85.5 K to 114.9 K behaves as a two-dimensional system with respect to the fluctuations in the superconducting order parameter, while a three-dimensional regime is observed in a narrow range of temperatures at T < 85.5 K. Such behavior was assumed to be partly due to the FM LCMO component of the bilayer, the spin-polarized charge carriers of which enter into the YBCO film and cause a “breaking” of superconducting pairs in the superconducting gap and pseudogap regimes.

1. Introduction

Manganites La₀.₇Ca₀.₃MnO₃ (LCMO), La₀.₇Sr₀.₃MnO₃ (LSMO) show a broad spectrum of physical properties due to the coexistence of spatially separated different electronic and magnetic states [1, 2]. The conduction mechanism of manganites is very complex and dominated by double-exchange interaction accompanied by strong electron-phonon coupling due to the dynamic Jahn-Teller effect [3-6]. At low temperatures, when the sample is in ferromagnetic (FM) metal state (T < TMI, where TMI is the metal-insulator transition temperature), the origin of the electrical resistivity ρ is related to scattering on the crystal defects, impurities (temperature-independent residual resistance) and to electron-electron, electron-phonon and electron-magnon scatterings [1, 2, 6]. At higher temperatures T > TMI the paramagnetic insulating phase becomes dominant, and ρ(T) can be described as the effect of an adiabatic small polaron hopping process. In our recent works [7, 8], we showed that the ρ -T dependence for high-quality LSMO films, except for the temperature region T ~ TMI, can be described

4 To whom any correspondence should be addressed
in the framework of the above mechanisms, while not-optimally prepared polycrystalline LSMO thin films can be described by a model of two intergrain conduction channels [9-10]. A phenomenological phase-coexistence transport model was developed successfully in the last decade [6, 11-13], which allows the description of the resistivity in broad temperature ranges, including the temperature region $T \sim T_{\text{MI}}$. In this work, we investigated the LCMO film resistive characteristics in the framework of the phase-coexistence transport model. LCMO films were obtained by RF magnetron sputtering.

It is known that the high temperature superconducting (HTS) cuprates with optimal (corresponding to the maximum critical temperature $T_c$) and with lower carrier concentration show some specific peculiarities in the electrical, magnetic and optical properties [14-18] at temperatures $T < T^*$ well above the critical temperature $T_c$. These phenomena are associated with a pseudogap regime opening at $T = T^*$, where onset of fluctuations in the superconductor order parameter set in. The existence of fluctuations leads to an increase of the samples’ normal conductivity (i.e. to the appearance of excess conductivity) [15, 17]. Analysis of the individual resistive characteristics of the HTS YBa$_2$Cu$_3$O$_{7-x}$ (YBCO) thin film in the YBCO/LCMO structure at $T > T_c$ in terms of excess conductivity was another aim of the work.

2. Experimental

The deposition of FM LCMO film on LaAlO$_3$ (LAO) and SrTiO$_3$ (STO) substrates was performed by off-axis RF magnetron sputtering from a stoichiometric ceramic target. To prepare HTS/FM bilayers, HTS YBCO films were deposited by laser ablation on top of the RF-magnetron sputtered manganite films. Some of the deposition details were described in our previous paper [19]. The thickness of the LCMO films deposited on LAO and STO substrates were 75 nm and 90 nm, respectively. The thickness of the LCMO and the YBCO films in the HTS/FM bilayer, deposited on the LAO substrates, were 75 nm and 90 nm. The standard four-probe method was used for measuring the temperature dependence of the resistance of the thin films and bilayers prepared.

3. Results and discussion

3.1. Investigation of La$_{0.7}$Ca$_3$MnO$_3$ thin films resistivity using the phase-coexistence transport model

The temperature dependence of the resistivity $\rho(T)$ of La$_{0.7}$Ca$_3$MnO$_3$ thin films are of metallic behavior ($d\rho/dT > 0$) at low temperatures and of semiconducting (or “insulative”) ($d\rho/dT < 0$) character at higher temperatures (figures 1 and 2). The temperature of maximum resistivity $T_{\text{MI}}$ is significantly lower for the LCMO film with a small thickness deposited on the STO substrate than for the thicker LCMO film deposited on the LAO substrate. As a rule, the thin films demonstrate a decrease in $T_c$ and $T_{\text{MI}}$ as the thickness decreases [2,5]. Moreover, a tensile strain [1], which LCMO film on STO substrate is exposed to, can contribute to a decrease in $T_c$ as well.

A phenomenological phase-coexistence transport model was applied to describe the temperature dependences obtained of the resistivity in the whole temperature range. The model is based on the assumption that the FM metal and paramagnetic insulator (PI) phases coexist at the transition region in doped manganites and nanoregions, with these phases being distributed randomly in the sample. The total resistivity $\rho$ is assumed to be the product of contributions of the resistivities $\rho_{\text{PI}}$ and $\rho_{\text{PM}}$ from the PI and FM regions $\rho = \rho_{\text{PI}}^\nu \rho_{\text{FM}}^\nu$, where $\nu$ is the volume fraction of the FM phase. Assuming that the Curie temperature distribution for a disordered system is Gaussian, the volume fraction $\nu$ is supposed to follow a complementary error function [6,11-13] $\nu(T) = (1/2)\text{erfc}((T - T_{\text{CurieM}})/\Gamma_0)$, where $2^{1/2}\Gamma_0$ is the parameter of standard deviation of the Gaussian distribution, $T_{\text{CurieM}}$ is some mean value of the Curie temperature. The resistivity of the PI phase of LCMO can be described in the framework of the small polaron hopping model $\rho_{\text{PI}}(T) = \rho_a T \exp(E_a/k_BT)$, where $E_a$ is the activation energy for hopping conduction, $\rho_a$ is the resistivity parameter, $k_B$ is the Boltzmann constant. The resistivity of the FM phase can be approximated by a polynomial function $\rho_{\text{FM}}(T) = \rho_0 + \rho_2 T^2 + \rho_3 T^N$, where $\rho_0$ is...
the residual resistivity caused by grain boundaries and defects in the crystal structure, \( \rho_2 T^2 \) is the resistivity related to electron-electron scattering and the third term (with \( N \sim 4.5 \)) represents mainly the contribution to the resistivity from the electron-magnon interaction according to the FM double-exchange theory [1, 2].

Figures 1 and 2 display the simulated results (red solid line) for the resistivity-temperature dependences for the above mentioned LCMO thin films. The parameters used for the simulation are shown in table 1.

![Figure 1](image1.png)  
![Figure 2](image2.png)

**Figure 1.** \( \rho - T \) dependence in a LCMO film with 75 nm thickness prepared on a LAO substrate. Open circles – experiment, solid line – data modelled using the phase-coexistence transport model (fitting parameters – see table 1).

**Figure 2.** \( \rho - T \) dependence in a LCMO film with 20 nm thickness prepared on a STO substrate. Open circles – experiment, solid line – data modelled using the phase-coexistence transport model (fitting parameters – see table 1).

| Sample       | \( \rho_0, \text{\Omega cm} \) | \( \rho_2, \text{\Omega cm/K}^2 \) | \( \rho_4, \text{\Omega cm/K}^4 \) | \( \rho_a, \text{\Omega cm/K} \) | \( T_{\text{CurieM}}, \text{~K} \) | \( T_0, \text{~K} \) | \( E_a, \text{meV} \) |
|--------------|--------------------------------|-------------------------------|---------------------------------|-------------------------------|-----------------|-----------------|----------------|
| LCMO/LAO     | 0.00165                        | 0.1452 \( 10^{-6} \)           | \( 2.211 \times 10^{12} \) \( N = 4.4 \) | 2.366 \( 10^{-6} \)          | 185.5           | 29              | 114.43          |
| LCMO/STO     | 0.1                            | 0.2 \( 10^{-6} \)              | 172 \( 10^{12} \) \( N = 4.635 \) | 5 \( 10^{-6} \)             | 97              | 36.8            | 107             |

It is seen that the results of simulation describe well the experimental \( \rho-T \) dependences. The LCMO film with a small thickness deposited on the STO substrate is characterized by a high value of the residual resistance \( \rho_0 \) and the scattering parameters \( \rho_2, \rho_4, \rho_a \), while the mean transition temperature \( T_{\text{CurieM}} \) of this sample is significantly lower than that of the LCMO/LAO sample. This is due to the fact that thin films, as a rule, contain more local strains, more defects and impurities, which contribute to activation of the scattering processes and to an increase of the scattering parameters.

### 3.2. Investigation of a YBCO thin film conductivity in a YBCO/LCMO bilayer

The conductive properties of HTS/FM bilayers are determined by the contribution of the individual conductivities of both HTS and FM layers and by the interaction peculiarities of the layers in the interface. To obtain more detailed information on the resistivity of the YBCO film in the YBCO/LCMO bilayer, we first measured the resistance temperature dependence \( R_0(T) \) of the bottom LCMO layer before deposition of the top YBCO layer. The temperature dependence of the resistance...
$R_1(T)$ of the top YBCO layer can be determined using the formula $R_1^{-1}(T) \approx R_2^{-1}(T) - kR_B^{-1}(T)$, where $R_2(T)$ is the measured $R$-$T$ dependence of the bilayer and the parameter $k$ takes into account the resistance change of the bottom LCMO layer during deposition of the top YBCO layer. Figure 3 shows the $R$-$T$ dependence measured in our YBCO/LCMO bilayer (dash dot) and the individual $R$-$T$ dependence of the YBCO film (solid line, red) in this bilayer for temperatures $T > T_C = 83.5$ K, determined in the above way with $k = 1.5$. The $R$-$T$ dependence obtained for the YBCO film is nearly linear and shows that a sharper decrease of $R$ with the decrease of $T$ occurs when $T_C$ is approached. The extrapolation of the linear part of the above $R(T)$ dependence to the low temperature range is depicted by dots (blue) in figure 3.

The small nonlinearity in the $R$-$T$ dependence can be associated with the appearance of fluctuation conductivity [15, 17], which causes a more rapid increase in the excess conductivity $\Delta \sigma$ in the sample. This excess conductivity was calculated from the measured $R(T)$ and extrapolated $R_{\text{ex}}(T)$ dependences as $\Delta \sigma(T) = (R^{-1}(T) - R_{\text{ex}}^{-1}(T))/4.53t$, where $t$ is the film thickness. Information about the behavior of the excess conductivity $\Delta \sigma(T)$ was obtained using the Aslamazov-Larkin model [20]. For this purpose, the $\ln(\Delta \sigma)$ versus $\ln(\varepsilon)$ dependence was calculated (with $\varepsilon = (T - T_m)/T_m$, $T_m = 84$ K) and plotted (figure 4). The slope $S$ of this plot can serve as an indicator of the dimensionality of the excess conductivity: $\Delta \sigma$ is three-dimensional if $S \approx -0.5$ and two-dimensional if $S \approx -1$ [15,17].

It is seen in figure 4 that $S \approx -0.5$ at $-6 < \ln(\varepsilon) < -4$ (temperature range from 84.2 K to 85.5 K) and $S \approx -1$ at $-4 < \ln(\varepsilon) < -1$ (temperature range from 85.5 K to 114.9 K). This means that the YBCO film considered in the temperature range from 85.5 K to 114.9 K behaves as a two-dimensional system with respect to the fluctuations in the superconducting order parameter, while a three-dimensional regime is established at $T < 85.5$ K. We assume that a low value of $T_C$ and a narrow range of temperatures, where $\Delta \sigma(T)$ of the YBCO film demonstrates a three-dimensional character, could partly be a consequence of spin-polarized charge carriers which enter the YBCO film from the FM LCMO component of bilayer and cause a “breaking” of superconducting pairs in both the superconducting gap and pseudogap regimes.

Figure 3. $R$-$T$ dependence measured in the YBCO/LCMO bilayer (dash dot) and the individual $R$-$T$ dependence of YBCO film (solid line, red) in this bilayer. Dots (blue) – the extrapolation results in the linear part of $R(T)$ dependence of YBCO film to the low temperature range.

Figure 4. Plot of the $\ln(\Delta \sigma)$ versus $\ln(\varepsilon)$ dependence.
4. Conclusions

Temperature dependences of the resistivity of manganite LCMO films deposited on LAO and STO substrates by RF magnetron sputtering were shown to be successfully simulated in the whole temperature range (covering the metal, the insulator and the metal-insulator transition regions) using a phenomenological phase-coexistence transport model. Quantitative data on the internal parameters of these films were obtained. The LCMO film with a small thickness deposited on a STO substrate is characterized by a high value of the residual resistance $\rho_0$ and scattering parameters $\rho_2$, $\rho_4$, $\rho_a$ (which is due to the presence of more local strains defects and impurities in thin films). The mean Curie temperature $T_{\text{CurieM}}$ of this sample is significantly lower than that of the LCMO/LAO sample. The possibility was also considered for investigating the individual resistive characteristics and excess conductivity of an HTS thin film included into a HTS/FM bilayer structure in the vicinity of $T_C$. It was shown that the YBCO film considered in the temperature range from 85.5 K to 114.9 K behaves as a two-dimensional system with respect to the fluctuations in the superconducting order parameter, while the three-dimensional regime is observed at $T < 85.5$ K. It was assumed that a low value of $T_C$ and a narrow range of temperatures, where $\Delta \sigma(T)$ of the YBCO film demonstrates a three-dimensional character, could partly be a consequence of spin-polarized charge carriers which enter the YBCO film from the FM LCMO component of the bilayer and cause a “breaking” of superconducting pairs in both the superconducting gap and pseudogap regimes.

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