Impedance measurements of epitaxial and polycrystalline LSMO thin films

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Abstract. The results are presented of measurements of the complex surface impedance dependence on the frequency in the 1 – 100 kHz range. La$_{2/3}$Sr$_{1/3}$MnO$_3$ (LSMO) single- and polycrystalline films were investigated at temperatures below the Curie temperature. The two types of layers show opposite behaviour – inductive and capacitive, correspondingly. Independently, information on the bulk resistance of the grains in the polycrystalline material and the intergrain complex resistance was obtained.

1. Introduction

The perovskite manganites have been the object of experimental and theoretical studies due to several reasons [1]: colossal-magnetoresistance applications [2, 3], bolometric purposes and applications as a cathode material in solid oxide fuel cells [4].

The typical usage of the perovskite magnetoresistive materials is based on DC measurements. However, the same measurements can be done in AC (above 1 kHz) regime. AC measurements can be more efficient because the 1/f noise is strongly reduced, which makes the study of the AC complex resistance interesting in view of practical applications. At the same time, such a study may yield valuable knowledge on the individual contribution of grains and grain boundaries to the low-field magnetoresistance [5, 6].

We present the results of a study of the complex impedance frequency dependence in the range 1 – 100 kHz.

2. Experimental

The La$_{2/3}$Sr$_{1/3}$MnO$_3$ (LSMO) system was chosen as the film material because its Curie temperature is well above room temperature. Two types of substrates were used: monocrystalline – SrTiO$_3$ (STO) and sapphire Al$_2$O$_3$ (ALO) substrates. RF magnetron sputtering of a solid LSMO target at 4 Pa 50% Ar and 50% O$_2$ gas mixture was used for film deposition on 5×10 mm substrates. The substrate temperature was 780 °C and the deposition was followed by an annealing step in O$_2$ at 80 kPa and 500 °C. The LSMO layer grown on the STO substrate was epitaxial and 30 nm thick. The pure sapphire substrate (without a buffer layer) does not stimulate the epitaxial growth of LSMO layers. Thus, the LSMO films (with a thickness of 40 nm) grown on the ALO substrate were polycrystalline and consisted of grains of submicron size.

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The four probe-method was used to measure the complex surface impedance of the LSMO films. The complex value of the voltage between the test probes were obtained by a SR530 lock-in amplifier in differential mode and was divided by the complex value of the voltage drop on a referent resistor measured in single-ended mode. The results are the modulus and phase angle of the complex surface impedance, as shown in figures 1 and 2. The measurements were conducted at 50 mV input voltage: the measurements at 500 mV input voltage yielded the same result.

The variation of the modulus of the complex surface impedance on STO is negligible, while its mean value is 450 Ω/sq.

![Figure 1. Frequency dependence of the complex surface impedance phase of LSMO films.](image1)

![Figure 2. Complex surface impedance modulus of the LSMO film on a sapphire substrate versus frequency.](image2)

3. Results and discussion

The complex surface impedance of the LSMO film grown on a STO substrate shows slightly inductive behavior (the phase of the complex impedance is positive). The impedance of the film grown on the sapphire substrate, on the contrary, shows a well-expressed capacitive behavior.

In framework of the classical electrodynamics, the skin depth $\delta$ of a conducting plate, where the main part of an AC current is flowing, is expressed as $\delta = (2\rho^{-1}\mu_r\mu_0\omega)^{-1/2}$, where $\mu_0$ is magnetic permeability of vacuum, $\mu_r$ is the relative magnetic permeability of the material, $\rho$ is the resistivity of the material, $\omega$ is the angular frequency of AC current. The estimations show that the skin depth of the LSMO material ($\rho \geq 1$ mΩ cm) at low frequencies ($\leq 100$ kHz) exceeds by far the thickness of the films used in our experiments. This implies a uniform distribution of AC current across our films.

Rahmouni et al [4] proposed an equivalent circuit for the case shown in figure 3A. Here, $R_g$ represents the contribution to the impedance of the grains bulk of the polycrystalline material, $R_{gb}$ is the contribution to the impedance of the grain boundaries and $C_{gb}$ is the contribution of the capacitance between the grains (grain boundary capacitance). Using the data shown in figures 1 and 2, one can calculate the active and reactive parts of the impedance for different frequencies and fit them with the formulae for the active and reactive parts of the impedance corresponding to the equivalent circuit shown in figure 3A with fitting parameters $R_g$, $R_{gb}$ and $C_{gb}$:

$$Z_{compl} = R_g + R_{gb}/(1 + i\omega R_{gb}C_{gb}).$$

The result is shown in figure 4. The optimum values of the fitting parameters are:

$$R_g = 3.435 \text{ kΩ}, R_{gb} = 16.524 \text{ kΩ} \text{ and } C_{gb} = 35.54 \text{ pF}.$$

The mean square error of the complex impedance modulus is about 1% of the mean value of the complex impedance modulus.
It should be pointed out that the circuit shown in figure 3A is a rather rough approximation. The real situation is better represented by the circuit shown in figure 3B. These two cases cannot be distinguished by complex impedance measurements. Nevertheless, two parameters obtained from the circuit 3A are valid for the circuit 3B, too: the ratio \( R_g/R_{gb} = 0.2079 \) shows that 17.2\% of the DC surface resistance is contribution of the bulk of the grains and 82.8\% is contribution of the grain boundaries. Thus, only the bulk part of the surface resistance alone is greater than the total surface resistance of the single crystal layer. The time constant \( R_{gb}C_{gb} = 587.32 \) ps is the mean time constant for all grain boundaries. If the grain boundaries are treated as a homogeneous medium between two highly conductive electrodes, the time constant equals \( \rho \varepsilon \), where \( \rho \) is the volume resistivity of the material and \( \varepsilon \) is its dielectric permittivity.

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\begin{array}{c}
\text{A} \\
\begin{array}{c}
R_{gb} \\
C_{gb} \\
R_g \\
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\[
\begin{array}{c}
\text{B} \\
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**Figure 3.** Simplified equivalent circuits for calculation of the polycrystalline film complex impedance.

**Conclusions**

The complex surface impedance of the single crystal layer exhibits a slightly inductive behavior (the phase of the complex impedance is positive), while the impedance of the polycrystalline layer shows a capacitive one. The time constant of the intergrain transition is \( \tau = 587.32 \) ps. The contribution of the grain volume to the DC resistance of the polycrystalline layer (3.435 kΩ) is substantially higher than the DC resistance of the single crystal layer. The contribution of the grain boundaries the DC resistance of the polycrystalline layer is predominant. It can be reduced (and as a result, the total resistance) using a film with larger grains.

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