Peculiar Features of Magnetic and Resistive Transitions in Partially Crystallized La_{0.84}Na_{0.16}MnO₃ Films

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Abstract. The present work aims to study magnetic, electric, magnetoresistance and resonance properties of partially crystallized La_{0.84}Na_{0.16}MnO₃ thin films. Peculiar behavior resulting from the wide-range coexistence of highly conductive conductive ferromagnetic and weakly paramagnetic phases has been analyzed. Based on the results of magnetic measurements and theoretical calculations, the dependence of the volume fraction of ferromagnetic phase, V_{FM} , on temperature is calculated. The whereby electric values resistance and V_{FM} magnetoresistance display maxima are determined.

Key words: doped manganites, ultrathin films, paramagnetic and ferromagnetic phases, magnetic resonance, magnetic and electric parameters.

I. INTRODUCTION

Doped perovskite manganites $R_{1-x}A_xMnO_3$ (A = Ca, Sr, Ba, Na, K) have attracted great attention due to the rich variety of unusual physical properties, especially, due to the effect of colossal magnetoresistance [1]. For this reason, the thin films of doped manganites are considered as prospective materials for development of a new generation of magnetic sensors and magnetic information readout devices [1-2].

The researches show that in doped manganites the character of conductivity is closely related to their magnetic state [1-3]. The transition from metallic to activated conductivity, which is usually observed near a transition temperature from paramagnetic (PM) to ferromagnetic (FM) state, is of especial interest. In these materials, the presence of manganese ions in various oxidation states (Mn³⁺ and Mn⁴⁺) and charge transfer between these ions (double a simultaneous occurrence exchange) favors of ferromagnetism and metallic conductivity. At the same time, a number of other effects (electron-phonon interaction, antiferromagnetic indirect exchange, charge and orbital ordering) hinder FM ordering and favor localization of charge carriers. If the double exchange prevails over the other interactions, the PM to FM transition and semiconductor to metal one occur practically simultaneously [1, 2]. This leads to the formation of electric resistance peak near the magnetic transition temperature $(T_{\rm C})$ and is the reason for the strong influence of magnetic field on the conductivity.

In single crystalline samples the temperature of the

electric resistance peak (T_R) practically coincides with Curie temperature T_C [1-3]. In polycrystalline, defective, amorphous and partially crystallized samples such situation is not observed and the difference between T_R and T_C may reach tens of degrees [3-5]. It should be noted that for the manganites with a broadened magnetic transition, the data on the behavior of electric resistance and magnetization in the transitional region are inconsistent and need refinement [2-4].

The aim of this work is to establish the features of magnetic and resistive transitions in partially crystallized $La_{0.84}Na_{0.16}MnO_3$ films, the peculiarity of which is a wide-temperature transition region from PM to FM state.

II. EXPERIMENTAL DETAILS

The 250-nm-thick La_{0.84}Na_{0.16}MnO₃ (LNMO) films were chosen for research. The samples were deposited on polycrystalline Al₂O₃ substrates by magnetron sputtering. The target for the film preparation was synthesized by a standard solid phase reaction [6]. The films were obtained at the substrate temperature $T_s = 300$ °C in the argon (30 %) and oxygen (70 %) mixture. The pressure of gas medium at the sputtering was 2×10^{-2} Torr. After the preparation, the films were annealed for 4 hours at $T_{ann} = 750$ °C.

The electric resistance of the films was measured as a function of temperature and magnetic field [6-7]. Magnetoresistance was measured in the fields of up to 15 KOe and defined as $MR = (R_0 - R_H)/R_0$, where R_0 is the electric resistance in zero magnetic field, and R_H – in the external field H. The static magnetization was measured in vacuum by a Faraday balance. The temperature dependences of the magnetization were measured in the range of 120 – 320 K in the field H = 3 kOe, which is remarkably higher than the magnetization saturation field [7].

III. RESULTS AND DISCUSSION

Fig. 1(a) shows the temperature dependence of magnetization, M(T), measured in the field H = 3 kOe for the LNMO film. The sample is found to be in paramagnetic state at high temperatures. Magnetization increase with the temperature lowering becomes noticeable as temperature crosses 290 K. The temperature dependence of dM/dT is plotted in the inset of Fig. 1(a). It is seen that the sharpest change of magnetization ($|dM/dT| = \max$) is observed at $T_M \cong$ 227 K. In the low temperature region the magnetization change considerably decelerates.

Fig. 1(b) shows the temperature dependence of electric resistance R_0 . The appearance of a wide maximum on $R_0(T)$

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curve is caused by the formation of highly conductive FM regions [6-7]. The maximum electric resistance value is reached at $T_R \cong 166$ K.

Fig. 1(c) presents the magnetoresistance, MR, measured in the field H = 15 kOe, as a function of temperature. In the high temperature region the influence of magnetic field on electric resistance is negligibly small ($MR \cong 0$), which is characteristic of the PM phase of doped manganites [1]. With the temperature lowering, MR increases, reaches a maximum and then slowly decreases with further temperature lowering. The temperature, at which MRreaches the maximum, T_{MR} , is equal to 190 K. The T_{MR} value exceeds T_R . It is, however, far lower than T_M , which is not typical of single crystals of doped manganites [1, 2].



Fig. 1. Temperature dependences of magnetization in the field H = 3 kOe (a), electric resistance (b) and magnetoresistance in 15 kOe field (c) for the LNMO film. The inset shows dM/dT as a function of temperature.

The results of the electron spin resonance investigation in the LNMO films were given in Ref. [7]. It was demonstrated that the resonance signal from FM phase becomes evident as T crosses 280 K (denoted as T_0 in Fig. 1(a)). Above this temperature the sample is completely paramagnetic; below the PM phase coexists with the FM one in quite a wide temperature range (the presence of the PM phase down to 110 K has been confirmed experimentally).

For the film under consideration we have calculated the temperature dependence of the volume fraction of the FM phase and found the V_{FM} values, which correspond to the characteristic temperatures T_R , T_{MR} and T_M . Assuming that the magnetization of the FM phase significantly exceeds that of the PM phase, the V_{FM} value may be calculated by finding the ratio of the experimentally obtained magnetization to the theoretically calculated one.

In the mean field approximation the behavior of the magnetization is described by Brillouin's formula [1, 7],

which makes it possible to calculate the temperature dependence of the reduced magnetization (M_{theor}/M_S) for the FM phase of the LNMO film.

The temperature dependence of the volume fraction of FM phase was found and the values of V_{FM} , which correspond to the characteristic temperatures T_R , T_{MR} and T_M , were established for the film under investigation. It has been found that the maximum value of electrical resistance is observed at $V_{FM} \cong 67\%$, while the magnetoresistance peak is observed at $V_{FM} \cong 63\%$. A noticeable magnetoresistance occurs only when the value of V_{FM} exceeds 15%.

IV. CONCLUSIONS

The magnetic, electric and magnetoresistive measurements on $La_{0.84}Na_{0.16}MnO_3$ films have shown that the temperatures of electric resistance and magnetoresistance maxima are significantly different from each other and from the temperature at which the anomaly of magnetic property is observed. The V_{FM} values, when the maxima on the $R_0(T)$ and MR(T) dependences are observed, have been determined. The conclusions about the dynamics of the change of the magnetic and resistive parameters for the manganite systems with a broadened magnetic transition have been drawn.

The results obtained in this work provide new insights into the dynamics of the phase transformations in doped manganites and verify the possibility of using the materials studied for fabrication of new functional elements of nanoand spin electronics.

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