

# Strain effect on resistivity of $\text{La}_{0.7}\text{Ba}_{0.3}\text{MnO}_3$ thin film

Timur A. Shaikhulov<sup>1</sup>, Valery A. Shakhunov<sup>1</sup>, Victor V. Demidov<sup>1</sup>, Gennady A. Ovsyannikov<sup>1a</sup>, Nikolay V. Andreev<sup>2</sup>, Anna E. Pestun<sup>2</sup>, Vladimir L. Preobrazhensky<sup>3</sup>

<sup>1</sup> Kotelnikov Institute of Radio Engineering and Electronics, RAS, Moscow, 125009, Russia

<sup>2</sup> Moscow Technological University, MISIS, Moscow, 119936, Russia

<sup>3</sup> The Wave Research Center, Prokhorov General Physics Institute RAS, Moscow, 119991, Russia

**Abstract.** A complex study of the dc and magnetic characteristics of epitaxial manganite films  $\text{La}_{0.7}\text{Ba}_{0.3}\text{MnO}_3$  (LBMO) was carried out under conditions of the crystal structure tension caused by a mismatch between the parameters of the LBMO crystal and the substrate. The epitaxial thin films with the thickness 150 nm were grown by laser ablation at  $T=700\text{-}800\text{C}$  under pure oxygen pressure 0.3-1 mbar. The substrates (001) $\text{LaAlO}_3$ , (001) $\text{SrTiO}_3$ , (110) $\text{NdGaO}_3$ , (001) $[(\text{LaAlO}_3)_{0.3}+(\text{Sr}_2\text{AlTaO}_6)_{0.7}]$  (LSAT) are used. It is shown that the temperature dependence of the film resistance in the low-temperature region does not depend on the film strain and it is in good agreement with the calculation that takes into account the interaction of carriers with magnetic excitations in the presence of strongly correlated electron states. The study of the of ferromagnetic resonance indicated the inhomogeneity of the ferromagnetic phase in LBMO films and an increase in the width of the FMR line with temperature decreasing.

## 1 Introduction

The parameters of epitaxial films of rare-earth manganite perovskites with the structure  $\text{Re}_{1-x}\text{A}_x\text{MnO}_3$ , where Re are the rare-earth materials (La or Nd) and A are the alkaline earth metals (Sr, Ca, Ba), very often differ substantially from the parameters of the crystals of the same composition. As it was shown in the references [1 - 3] the reason of the change in electric and magnetic properties is the strain in the films, caused by the lattices mismatch of the film with the substrate on which the films are deposited. A correlation was observed between the level of mechanical stresses in manganite films and the temperature of meta-insulator transition ( $T_M$ ) and the magnitude of the magnetoresistance. It was shown that the three-dimensional compression of the crystal lattice increases the hopping probability amplitude in the double exchange model, which leads to an increase in the Curie temperature ( $T_C$ ), while the biaxial distortions of the Jahn-Teller type cause an increase the localization of electrons and decrease  $T_C$  [4-8].

The ferromagnetic phase transition for a single-phase  $\text{La}_{0.7}\text{Ba}_{0.3}\text{MnO}_3$  (LBMO) crystal occurs at  $T_C = 345\text{ K}$  [9]. In the LBMO single crystal the giant magnetostriction (up to  $4 \cdot 10^{-4}$ ) was observed at a temperature equal to Curie temperature [10]. In the LBMO films when the film tension changes multiple resistive states arise. They are modulated by the magnetic field over a wide temperature range [11]. This phenomenon is caused by the strong coupling of spin and charge degrees of freedom [10-13].

This paper presents the results of an experimental studies of the electronic transport and microwave magnetic parameters of epitaxial LBMO films deposited on various substrates.

## 2 Samples and experimental technique

Epitaxial LBMO films 150 nm thick were deposited on (001) $\text{LaAlO}_3$  (LAO), (001) $\text{SrTiO}_3$  (STO), (110) $\text{NdGaO}_3$  (NGO), (001)  $[(\text{LaAlO}_3)_{0.3}+(\text{Sr}_2\text{AlTaO}_6)_{0.7}]$  (LSAT) with a size of  $5 \times 5 \times 0.5\text{ mm}^3$  using laser ablation in off-axis geometry at the temperature of 700-800 C and the oxygen pressure of 0.2-0.5 mbar. The targets were made from a mixture of  $\text{La}_2\text{O}_3$ ,  $\text{BaCO}_3$  и  $\text{MnCO}_3$  powders according to the ceramic technology. The stoichiometric composition of the target was used. The resistance of the films was studied by a four-point technique. Contact areas were produced by sputtering of Pt through a metal mask. The temperature of the metal-insulator transition  $T_{MI}$ , determined from the maximum resistance, varied in proportion to  $T_C$ .

The crystallographic parameters of the films and substrates were determined by means of a 4-circle x-ray diffractometer, by measuring in  $2\theta/\omega$  and  $\phi$  scan mode. The magnetic characteristics were measured by magnetic resonance. The magnetic resonance spectra were recorded using ER-200 EPR Bruker spectrometer operating in a 3-cm electromagnetic wave band (frequency 9.76 GHz). When recording the signals a synchronous detection was used at the modulation frequency 100 kHz of an external magnetic field. The investigated samples were located in the microwave

<sup>a</sup>Corresponding author: [shaikhulov@hitech.cplire.ru](mailto:shaikhulov@hitech.cplire.ru)

cavity of the spectrometer. The plane of the sample was always parallel to the direction of the constant external magnetic field and the magnetic component of the microwave field (parallel orientation). The parallelism of the plane of the samples to a constant magnetic field was controlled by getting minimum of the resonance value of the ferromagnetic resonance field (FMR). This arrangement of the samples eliminated the contribution in the magnetic resonance spectra due form-factor of the sample.

### 3 Lattice parameters

The c-axis of the grown LBMO films are oriented along the normal of the plane of the substrate. The parameter of the interplanar distance in LBMO films along the normal to the substrate  $a_{\perp}$  and the substrate plane  $a_s$  were determined from x-ray diffraction  $2\Theta/\omega$  scan.

The  $2\Theta/\omega$  scan for LBMO/LAO film is presented on Fig.1. The out of plane parameter  $a_{\perp}$  for LBMO films strongly depends on  $a_s$  for the substrate. The in plane film parameters  $a_{\parallel}$  determined from asymmetric diffraction peaks are close to the  $a_s$  for the substrate crystals. These measurements indicate that a fast relaxation of the LBMO parameter of the film occurs at the substrate/film interface. Consequently, the grown manganite films are under the influence of mechanical stresses caused by the substrate during the film growth.

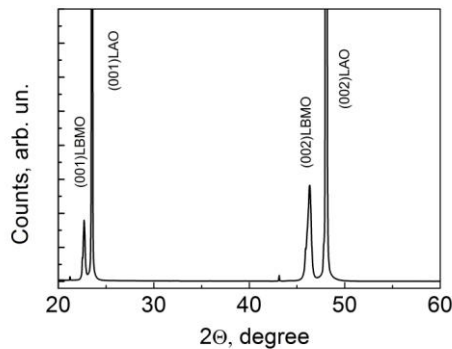


Fig.1.  $2\Theta/\omega$  scan for LBMO film deposited on LAO substrate

The values of the strains are arisen in the plane of substrate  $\varepsilon_{\parallel}=(a_{\parallel}-a_{LBMO})/a_{LBMO}$  and in the perpendicular direction  $\varepsilon_{\perp}=(a_{\perp}-a_{LBMO})/a_{LBMO}$  are presented in Table 1.  $a_{LBMO}=0.390$  nm obtained from XRD data for target is used. The temperatures of metal-insulator transition  $T_{MI}$  measured at max of temperature dependence of resistance  $R(T)$  (see Fig.2) are also shown. It can be seen that the compressive stresses in the plane of the substrate for LSMO films deposited on the NGO, LAO, and LSAT substrates reduce  $T_{MI}$ , and hence the Curie temperature.

We note that the unit cell volume of the crystal lattice of the LBMO film ( $V_{eff}$ ) is not conserved when the substrate is changed. At small lattice distortions the Curie temperature ( $T_C$ ) and magnetic transport

parameters of the manganites depend on two parameters: the relative change in the  $V_{eff}$  :  $\varepsilon_b=2\varepsilon_{\parallel}+\varepsilon_{\perp}$ , and the biaxial distortion  $\varepsilon^*=(\varepsilon_{\perp}-\varepsilon_{\parallel})\sqrt{2/3}$  [7, 14]. The distortions of unit cell volume lead to an increase or decrease in  $T_C$  dependently on the sign of the distortion, but the biaxial ones always decrease  $T_C$  [7, 13, 14].

**Table 1. Crystallographic parameters and the temperature of metal-insulator transition**

Subst	$a_s$ , nm	$a_{\perp}$ , nm	$V_{eff}$ , nm <sup>3</sup>	$\varepsilon_b$ , %	$\varepsilon^*$ , %	$T_{MI}$ , K
LSAT	0.387	0.396	0.0593	0.37	1.87	256
NGO	0.386	0.391	0.0583	-1.88	0.52	259
LAO	0.379	0.393	0.0564	-4.89	1.71	278
STO	0.391	0.393	0.060	3.02	-0.28	287

### 4 Conductivity

A temperature peak of resistance typical for manganites was observed at  $T = T_{MI}$  for all the films studied (Fig. 2). The decrease in resistance at  $T \leq T_M$  is most likely caused by an increase in the area of the conducting ferromagnetic regions. At low temperatures ( $T < 100K$ ), the resistance of all film tends to the asymptotic value which is determined by electron scattering on impurities, defects, grain boundaries, spin waves and domain walls [12, 15, 16].

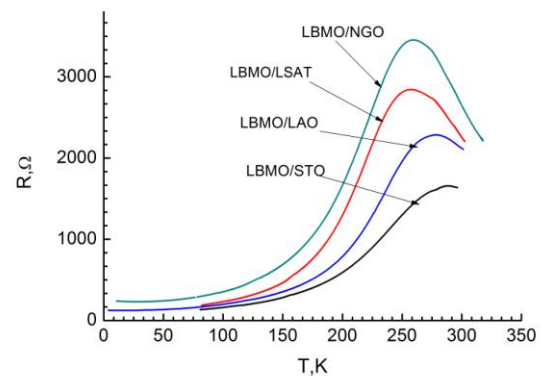


Fig. 2. Temperature dependences of the resistances of 150 nm thick LBMO films deposited on STO, NGO, LSAT, LAO substrates.

The highest value of  $T_{MI} = 283$  K was observed for LBMO grown on STO substrates. However, even in the case of a small misalignment of the substrate and the film crystal parameters (1.12%) the  $T_{MI}$  film is several degrees lower than the  $T_{MI}$  for stoichiometric LBMO single crystal (345K). Perhaps the decrease in  $T_{MI}$  is due to the relatively low concentration of carriers in LBMO films due to a decrease in the Ba content [13]. Indeed, as was shown in the reference [17] for  $La_{1-x}Ba_xMnO_3$  crystal with a  $x \leq 0.2$   $T_{MI}$  is for several degrees lower than in optimal case ( $x=0.3$ ).

Curie temperature  $T_C$  depends on the strain according to the work of Millis as following [7]

$$T_c(\epsilon) = T_c(0)(1 - \alpha\epsilon_b - \delta\epsilon^*/2), \quad (1)$$

where  $\alpha = (1/T_c)dT_c/d\epsilon_b$  and  $\delta = (1/T_c)d^2T_c/d\epsilon^2$ . Assuming that  $T_C$  and  $T_{MI}$  vary in the same way as the change in the film strain, we obtain that for  $T_{MI}(0)=345K$   $\alpha$  changes within tens of units, and the gamma of units is hundreds. For example, for a pair of substrates with close crystallographic parameters NGO and LSAT, substrates we have  $\alpha = 12$ ,  $\delta = 1200$ .

For  $T < T_{MI}$  the resistivity of manganite films depends both on scattering of carriers by impurities and defects, and also on the intensity of electron-electron and electron-magnon interactions. According to [18-23] the temperature dependence of the resistivity of a manganite film can be represented in the the following form

$$\rho = \rho_0 + \rho_1 T^2 + \rho_2 T^{4.5}, \quad (2)$$

where  $\rho_0$  is the contribution to the resistivity of structural defects,  $\rho_1$  and  $\rho_2$  are determined by electron-electron scattering and the scattering of electrons by magnons correspondingly.

The smallest  $\rho_0 = 2,3 \cdot 10^{-3} \Omega \text{ cm}$  is observed for LBMO films grown on STO substrates having the smallest mismatch between the crystal lattice of the substrate and the film. The obtained  $\rho_0$  for the film are several times greater than the resistivity for single crystals (less than  $10^{-3} \Omega \text{ cm}$ ) [21]. In the LBMO film on LAO increasing of the  $\rho_0$  could be due to twinning of the substrate. The dependence  $\rho \propto T^{4.5}$  was indeed observed for manganite LBMO films sputtered on the (001) LSAT substrate [22]. At the same time, the dominance of the  $T^2$  term in  $\rho(T)$  was observed for  $\text{La}_{0.67}\text{Sr}_{0.33}\text{MnO}_3$  (LSMO) films [3]. A small increase in the  $R(T)$  at  $T < 50 \text{ K}$  can be caused by a contribution to the resistance proportional to  $T^{1/2}$  caused by the Coulomb interaction of carriers [12]. Detail analysis of the temperature dependences of the resistivity of the LBMO/NGO films shown that  $\rho(T)$  is described with good accuracy by the expression (2) for  $\rho_1 = 9 \cdot 10^{-9} \Omega \text{ cm K}^{-2}$ ,  $\rho_2 = 4 \cdot 10^{-13} \Omega \text{ cm K}^{-4.5}$  and  $\rho_0 = 2,3 \cdot 10^{-3} \Omega \text{ cm}$  in the temperature range (100-250 K). At  $T = 100 \text{ K}$  the relation  $\rho_0 > \rho_2 T^{4.5} > \rho_1 T^2$  indicates the structural defects dominated over electron-electron and electron – magnon interactions.

## 5 Ferromagnetic resonance

Ferromagnetic resonance spectra (FMR) were obtained both for the purpose of determining the transition temperature of a paramagnet-ferromagnet and for studying the magnetization anisotropy in the plane of the films.

Shapes of all the FMR curves at nearly room temperature ( $T = 280 \text{ K}$ ) can not be described by a single Lorentz line (see Fig.3). This fact unambiguously indicates the inhomogeneity of the ferromagnetic phase in the films. The strain of the films can gradually decrease with increasing of their thickness. The difference in the values of the resonant fields  $H_0$  for

different parts of the inhomogeneous film did not exceed the width of the FMR line  $\Delta_{1/2}$ . The nonmagnetic layer, which according to [24] is at film- substrate interface,

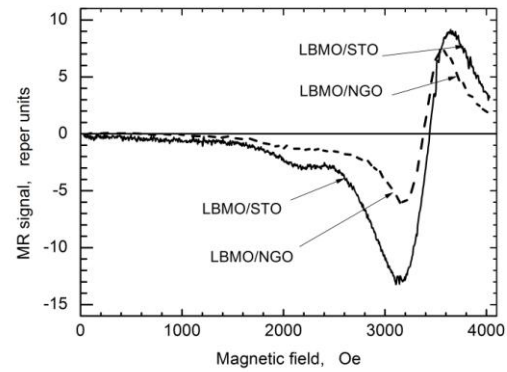


Fig. 3. Ferromagnetic resonance spectra for the two LBMO films measured at nearly room temperature ( $T = 280 \text{ K}$ ) in parallel orientation

does not have a noticeable effect on FMR. The magnetization values  $M_0 = 30 \text{ Oe}$  and uniaxial anisotropy fields  $H_u = 33 \text{ Oe}$  and cubic anisotropy  $H_c = 3.5 \text{ Oe}$

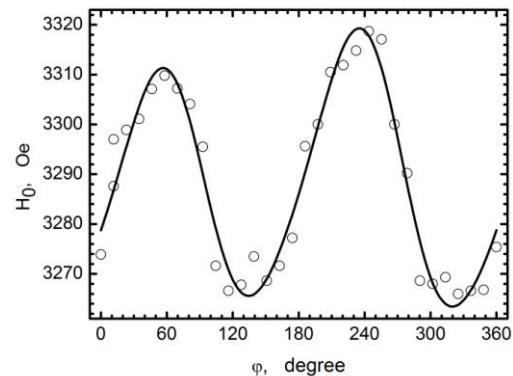


Fig.4. The angular dependence of the resonance magnetic field  $H_0$  for LBMO/NGO for sample rotation around the normal to the film plane. The solid line describes the planar magnetic anisotropy with adjustable parameters given in the text.

were obtained from the fitting the solution of the Landau-Lifshitz equation for the evolution of the magnetization  $M$  in an external constant magnetic field  $H$  (see eq.1 ref.[25])

Fig. 5 shows the temperature dependences of the resistance the values of  $H_0$  for the LBMO/NGO film and the width of the FMR line  $\Delta_{1/2}$ . The value of  $H_0$  decreases due to the increase in the magnetization of the LBMO film. The Curie temperature could be determined from the inflection point of the  $H_0(T)$  dependence. At the same time, as the temperature decreases the width of the FMR line increases. Usually in manganites the width of the FMR line decreases with temperature [25]. Possible cause of increase the line width is strong interaction of carries with magnons, which is absent in LSMO/NGO films [25 - 27].

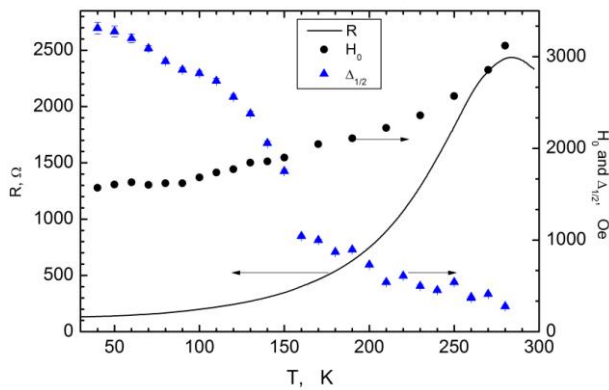


Fig.5. The temperature dependences of the resistance (solid line), the resonance magnetic field (circles) and the width of the FMR line (triangular) for LBMO/NGO film.

## 6 Conclusion

Epitaxial manganite lanthanum-barium films were deposited on 4 types of substrates that provide strain of the crystal lattice of films. The temperature dependence of the resistivity of all films at low temperatures is well described by a power polynomial that takes into account the effect of scattering on defects and impurities as well as electron-electron and electron-magnon interactions. The line of ferromagnetic resonance of the films is not described by a single Lorentz line, which indicates on the inhomogeneity of the ferromagnetic phase in the films. With temperature decreasing the widening of the FMR line is observed.

Supports by the International Associated Laboratory LEMAC-LICS, RFBR grants 17-02-00145 and 16-29-14022 are acknowledged

## References

1. A.-M. Haghiri-Cosnet and J.P. Renard. *J. Phys D: Appl. Phys.* **36**, R127 (2003).
2. Zh. Huang, G.Y. Gao, Zh.Zh. Yin, X.X. Feng, Y.Zh. Chen, X.R. Zhao, J.R. Sun, W.B. Wu, *J. Appl. Phys.* **105**, 113919 (2009).
3. G.A. Ovsyannikov, A.M. Petrzhik, I.V. Borisenko, A.A. Klimov, Yu.A. Ignatov, V.V. Demidov, S.A. Nikitov, *JETP*, **108**, 48 (2009).
4. Y. Lu, J. Klein, C. Hofener, B. Wiedenhorst, J.B. Philipp, F. Herbstritt, A. Marx, L. Alff, R. Gross. *Phys. Rev. B* **62**, 15 806 (2000).
5. Y. Chen, B.G. Ueland, J.W. Lynn, G.L. Bychkov and S.N. Barilo, Y.M. Mukovskii, *Phys. Rev. B* **78**, 212301(2008).
6. F. Tsui, M.C. Smoak, T.K. Nath, C.B. Eom, *Appl. Phys. Lett.* **76**, 2421 (2000).
7. A.J. Millis, T. Darling, A. Migliori, *J. Appl. Phys.* **83**, 1588 (1998).
8. M. Bibes, S. Valencia, L. Balcells, B. Martínez, *Phys. Rev. B* **66**, 134416 (2002).
9. H.L. Ju, J. Gopalakrishnan, J.L. Peng, Qi Li, G.C. Xiong, T. Venkatesan, R.L. Greene, *Phys. Rev. B* **51**, 6143 (1995).
10. R.V. Demin, L.I. Koroleva, A.Z. Muminov, Ya.M. Mukovski, *Physics of Solid States* **48**, 322 (2006).
11. W. Zhou, Y. Xiong, Z. Zhang, D. Wang, W. Tan, Q. Cao, Z. Qian, and Yi Du. *ACS Applied Materials & Interfaces*, **8**, 5424 (2016).
12. P. Orgiani, C. Adamo, C. Barone, A. Galdi, S. Pagano, A. Yu. Petrov, O. Quaranta, C. Aruta, R. Ciancio, M. Polichetti, D. Zola, and L. Maritato, *J. Appl. Phys.* **103**, 093902 (2008).
13. Yu.A. Boikov, T. Claeson, *Technical Physics*, **59**, 1027 (2014).
14. H.Y. Hwang, T.T.M. Palstra, S-W. Cheong, B. Batlogg, *Phys. Rev. B* **52**, 15046, (1995).
15. Y. Suzuki, H.Y. Hwang, S-W. Cheong et al., *J. Appl. Phys.* **83**, 7064 (1998).
16. G.J. Snyder, R. Hiskes, S. DiCarolis, M.R. Beasley, T.H. Geballe. *Phys. Rev. B* **53**, 14434 (1996).
17. J. Zhang, H. Tanaka, T. Kanki, J.-H. Choi, T. Kawai, *Phys. Rev. B*. **64**. 184 404 (2001).
18. K. Kubo, N. Ohata, *J. Phys. Soc. Jpn.* **33**, 21 (1972).
19. Yu.A. Boikov, I.T. Serenkov, V.I. Sakharov T. Claeson, A. Kalabukhov, V.V. Afrosimov, *Physics of Solid States* **55**, 2043 (2013).
20. S. V. Pietambaram, D. Kumar, R. V. Singh, C. B. Lee, and V. S. Kaushik, *J. Appl. Phys.* **86**, 3317 (1999).
21. W. Jiang, X. Z. Zhou, G. Williams, Y. Mukovskii, and K. Glazyrin, *Phys. Rev. B* **77**, 064424 (2008).
22. Yu.A. Boikov, V.A. Danilov, *Physics of Solid State* **50**, 451 (2008).
23. N.G. Bebenin, R.I. Zainullina, V.V. Mashkautsan, V. V. Ustinov, Ya. M. Mukovskii, *Phys. Rev. B*. **69**, 104434 (2004).
24. Yan Wu, Y. Suzuki, U. Rudiger, J. Yu, and A. D. Kent T. K. Nath and C. B. Eom, *Appl. Phys. Lett.* **75**, 2295 (1999).
25. V.V. Demidov, G.A. Ovsyannikov, A.M. Petrzhik, I.V. Borisenko, A.V. Shadrin, and R. Gunnarsson, *J. Appl. Phys.* **113**, 163909 (2013).
26. R. von Helmolt, J. Wecker, B. Holzapfel, L. Schultz, and K. Samwer, *Phys. Rev. Lett.* **71**, 2331 (1993).
27. S. E. Lofland, S. M. Bhagat, H. L. Ju, G. C. Xiong, T. Venkatesan, and R. L. Greene, S. Tyagi. *J. Appl. Phys.* **79**, 5166 (1996).