MICROWAVE LOSSES IN FERROMAGNETIC THIN LSMO FILMS.

T. Nurgaliev[1], V.V. Demidov[2], A.M. Petrzhik[2], G.A. Ovsyannikov[2], S. Miteva[1], B. Blagoev[1],

[1]*Institute of Electronics, Bulgarian Academy of Sciences, 72 Tsarigradsko Chausse, 1784 Sofia, Bulgaria*

[2]*Institute of Radio Engineering and Electronics RAS, Moscow 125009, Mokhovay 11, bld 7.*

Abstract. Thin films of the $La_{0.7}Sr_{0.3}MnO_3$ (LSMO) manganite material were prepared on LAO substrates by RF magnetron sputtering and their microwave characteristics were investigated. The EPR absorption spectrum of one of the samples showed a double-line character below the Curie temperature. The effect was interpreted as due to a coexistence of the paramagnetic-and ferromagnetic-like phases in the sample and the main microwave parameters of these phases were determined at T=293 K. A possibility for investigation of ferromagnetic resonance (FMR) in thin films using a simple parallel plate resonator was demonstrated.

Keywords: LSMO film; microwave properties; EPR

I. INTRODUCTION

The interest to ferromagnetic (FM) oxides such as $La_{0.7}Sr_{0.3}MnO_3$ (LSMO) has grown rapidly due to their original physical characteristics and possible applications in micro- and microwave electronics and spintronics. The LSMO manganite has a fully spin-polarized conduction band and exhibits ferromagnetic transition around room temperature [1]. The LSMO films grown on $LaAlO_3$ (LAO) substrates are under compressive stress (decreasing in the plane and expanding in the out-of-plane direction) due to the lattice mismatch between the film and substrate [1,2]. At low temperatures the out-of-plane growth direction is the magnetization easy direction for $La_{0.7}Sr_{0.3}MnO_3$ compressive films grown on LAO [1]. At higher temperatures, the easy direction rotates towards the plane [1]. The phase separation in manganites occurs below the Curie temperature where ferromagnetic metallic and non-ferromagnetic (for example, antiferromagnetic) insulating areas of nanometer dimensions (or submicron domains) coexist [3,4]. Together with that, the magnetic domain structure with perpendicular or parallel orientation of the magnetization in the domains with respect to the film plane can exist in thin manganite films [2,4,5].

Microwave measurements may give information about the module, orientation of the magnetization vector and about the magnetic anisotropy of the manganite materials [6-8].

In opinion of the authors, at present there is not a lot of information in literature about the microwave characteristics of thin LSMO films deposited on different substrates. In this paper we report the microwave characteristics of LSMO samples grown on LAO substrates.

2. EXPERIMENTAL

Ferromagnetic $La_{0.7}Sr_{0.3}MnO_3$ thin films were grown in-situ on (100) $LaAlO_3$ substrates (5 mm x 10 mm x 0.5 mm) by RF off-axis magnetron sputtering technique. The substrate temperature T_{dep} and the film annealing temperature T_A were T_{dep} ~700 °C, T_A ~ 600 °C, respectively. The film thickness was about 100 nm. The film resistance was measured by four contact method and the film resistivity has been calculated from the experimental data.

For a rough evaluation of microwave characteristics of LSMO films with a high value of magnetization we have adapted a simple parallel plate resonator (PPR) technique. For this purpose the sample was placed between two metallic electrodes of the PPR and the microwave transmission characteristics of PPR was measured at different values of the applied static magnetic field at the resonance frequency of the PPR. Unfortunately, a simple PPR with the metallic electrodes is not very sensitive and does not “work” when the magnetization of the sample is weak. In our opinion, the sensitivity of the PPR technique for magnetic resonance measurements could be improved drastically if one uses the superconducting electrodes in it.

Two LSMO samples were chosen for experiments. The simple PPR technique was not successful for evaluation of one of the samples (sample 1), while the second samples (sample 2) was evaluated by using of PPR technique. For this reason the Electron Paramagnetic Resonance (EPR) technique was used for characterization of sample 1. The EPR spectrum was obtained with a x-band spectrometer operating at 9.58 GHz.

3. RESULTS AND DISCUSSION

3.1. Formulas for the analysis

The signal of EPR is proportional to the derivative of absorbed (by the sample) microwave power P_a on a field [8]:

$$F(H) = \frac{dP_a(H)}{dH} \qquad (1).$$

The microwave absorption P_a of a thin film sample with a low value of the conductivity can be expressed in terms of the imaginary part of the dynamic magnetic susceptibility χ''. If thin film includes the areas with the paramagnetic (PM) and ferromagnetic (FM) phases, the absorbed power can be presented approximately in the form

$$P_a \sim f(A\chi''_{PM} + B\chi''_{FM})h^2, \qquad (2)$$

where f is the microwave field frequency; h is the microwave field amplitude; A and B are the coefficients characterizing the “concentrations” of the PM and FM phases in the film. The imaginary part of the susceptibility χ'' is expressed as

$$\chi''_{PM} \sim \frac{\omega_H(\Delta\omega_{PM})}{(\Delta\omega_{PM})^2 + (\omega_H - \omega)^2}, \qquad (3)$$

in the PM state of the sample (here $\omega_H = 2\pi\gamma_e H_{ef}$ is the resonance frequency for the PM state; γ_e is the gyromagnetic ratio for an electron; $\Delta\omega_{PM}$ is the half width of EPR peak, H_{ef} is the effective magnetic field, affecting the PM electronic system).

The same formula (3) can be used for describing the imaginary part of the dynamic susceptibility χ''_{FM} of the FM phase of the film if one assumes that the FM phase consists of microparticles, which linear dimensions are comparable with the thickness of the film, and takes into account the appropriate values of the linewidth $\Delta\omega_{FM}$ and the effective field $H_{ef} \approx H - NM$ (M is the saturation magnetization; N is the demagnetization factor). Formula (3) is also valid in the perpendicular geometry (H is applied perpendicularly to the film surface) if the linear dimensions of the FM phase significantly exceeds the film thickness.

The following formula can be used for describing of χ''_{FM} if external field H is applied parallel to the film surface and the linear dimensions of the FM phase significantly exceeds the film thickness:

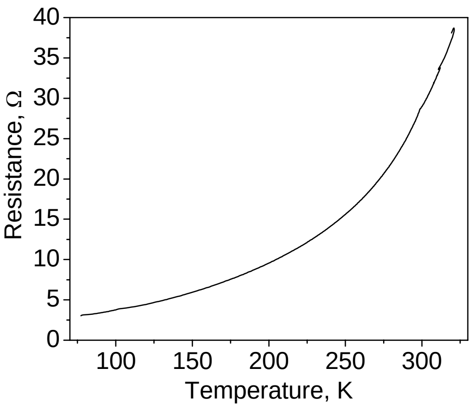
$$\chi''_{FM} \sim \frac{\alpha\omega\omega_M[\omega^2 + (\omega_M + \omega_H)^2]}{(\omega_{0F}^2 - \omega^2)^2 + \alpha^2\omega^2(2\omega_H + \omega_M)^2}, \qquad (4)$$

where $\omega_{0F} = \sqrt{\omega_H(\omega_H + \omega_M)}$ is the FM resonance (FMR) frequency of the tangentially magnetized film; $\omega_M = \gamma_e M$; α is the dimensionless Gilbert (Landau-Lifshitz) damping parameter; M is the saturation magnetization.

3.2. Microwave characteristics obtained by EPR

Temperature dependence of the resistance of the $La_{0.7}Sr_{0.3}MnO_3$ thin film 1 is shown in Fig.1. The film demonstrates a typical decrease of the resistance on decreasing temperature T. This is due to the spontaneous alignment of the Mn spins below the Curie temperature T_C [1] which allows a delocalization of the electrons leading to a low resistivity of the FM phase. It can be noted, that the film resistivity ρ is quite low (ρ<1 mΩ cm at T< 320 K and ρ~70 μΩ cm at T = 77 K) which is typical for the LSMO material of high quality [9].

The EPR spectra of film 1 in the magnetic field H applied parallel to the film plane are shown in Fig.2. The film demonstrates the spectrum in which both position and width of the absorption line strongly depends on temperature. The spectrum is shifted to the lower fields H with decreasing of T (Fig.2 and Fig.3) due to the ferromagnetic transition of the sample at the Curie temperature $T=T_C$ and to increasing of the magnetization vector M at lower temperatures. The Curie temperature of the sample can be evaluated from Fig. 3 as T_C~350 K. The EPR signal with the peak-to-peak linewidth $\mu_0\Delta H_{pp}$=16-19 mT (Fig.3) is observed at 350 K<T<375 K and corresponds to the PM state of the film. A significant increase of the linewidth ΔH_{pp} and a decrease of resonance field H_r occurs at lower temperature (Fig.3 and 4) and at T<T_C the EPR is gradually transformed to the ferromagnetic resonance (FMR). Such a temperature dependence of the parameters of EPR is typical for bulk LSMO samples and are



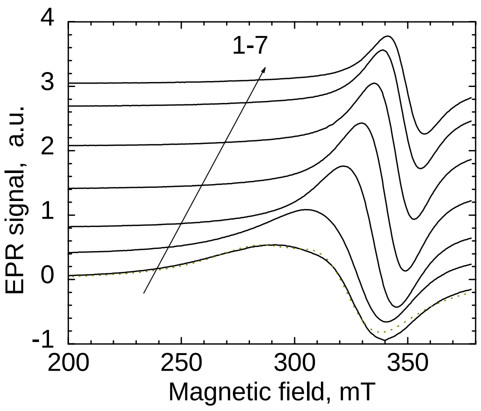


Fig.1. Temperature dependence of the resistance of LSMO film 1.

Fig.2. EPR spectrum of a tangentially magnetized LSMO film 1 (~100 nm) obtained at T=293, 315, 337, 346, 352, 360, 373 K (curves 1-7, respectively). The results of simulation of the spectrum obtained at T=293 K (curve 1) is shown by the dots, the simulation parameters are given in the text.

similar to those observed recently in LSMO thin films grown on STO [8] and STO/Si substrates [10], although the Curie temperature of our sample was slightly higher than that, reported in literature (for example T_C~330 K is reported in [10] for the thin films).

The main difference of the spectrum of sample 1 from the reported ones in [8,10] is that it is transformed to a double-line spectrum at $T<T_C$ (see, for example, Fig.2, curve 1).

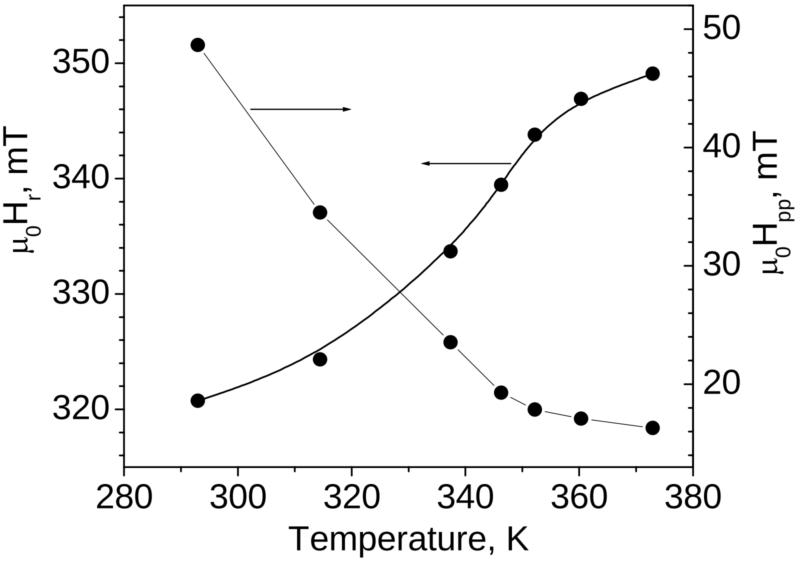


Fig. 3. Temperature dependence of the average resonance value of the field and the peak-to-peak linewidth of sample 1, evaluated directly from Fig.2.

To perform a more accurate analysis, the simulation of the spectrum obtained at T=293 K (Fig. 2, curve1) was made assuming a possible simultaneous presence of the hypothetic PM-like and FM-like phases in sample 1 at $T<T_C$. The simulation was performed by using of formulas (1,2), where the PM and FM parts of susceptibility was described by formulas (3) and (4), respectively. By fitting of the simulated dependence to the experimental one (see Fig.2) the following values of the parameters of the PM-like and FM-like phases for sample 1 were found as results: the resonance field and the peak-to-peak linewidth are μ_0H_r= 324 mT and $\mu_0\Delta H_{pp}$=54 mT for the PM-like phase and μ_0H_r= 295.8 mT, μ_0M = 100 mT, α = 0.095, $\mu_0\Delta H_{pp}$=74.5 mT for the FM-like phase.

In fact, the parameter which is obtained as the saturation magnetization M of FM-like phase from the resonance frequency measurements in EPR can differ from a real value of parameter M. This is due to the fact that the effective field H_a of the crystallographic anisotropy affects the resonance frequency as well. For example, a simple uniaxial anisotropy (with the preferential axis perpendicular to the film surface) of hard-axis type (which is more typical at low temperatures for LSMO thin films deposited on LAO substrates) contributes to increase of the resonance field (and leads to an underestimation of M, determined from the resonance frequency if this anisotropy is ignored) while an opposite situation is observed in the case of “easy-axis” anisotropy. The field H_a can not always be taken into account in the experimental data treatments because of complexity of its determination.

It should be noted that the EPR absorption spectra, indicating a double-layer spectrum below the Curie temperature T_C have recently been observed by the authors of [11] in LSMO nanoparticles synthesized by sol-gel routes. The phenomenon has been explained by a presence of two, the core (FM-like) and the surface (PM-like) states of the nanoparticles. Results of our investigations of the spectrum below T_C are consonant with those of paper [11] and this allow to conclude, that thin LSMO films can reveal a multiphase behavior as well. It is interesting to note, that the resonance field μ_0H_r= 324 mT obtained for the PM-like phase is by about 18 mT is less than the field which is necessary for observation of EPR (the functioning frequency of the spectrometer is ~9.58 GHz) in the conventional PM systems. Magnetic field created by the FM-like phase which can enhance the external magnetic field in the PM phase areas of the tangentially magnetized film could be one of reasons of such a difference.

3.3. Characteristics obtained by PPR

Microwave characteristics of LSMO film 2 were investigated by using a simple parallel plate resonator (PPR) technique. For this purpose the sample was placed between two metallic electrodes (with dimensions 5 mm x 10 mm) of the PPR and the transmission characteristics S_{21} of PPR was measured at the resonance frequency on dependence of the induction of the external field H, applied parallel to the surface of the film and to the wave propagation direction in the resonator. The resonator had a maximal transmission coefficient at f_0 ~ 4.6 GHz if no DC magnetic field is applied. For sample 2 a resonance – like increase of microwave losses of the structures was observed at magnetic field μ_0H = 60 mT (Fig. 4), while for sample 1 the changes of the transmission characteristics were small.

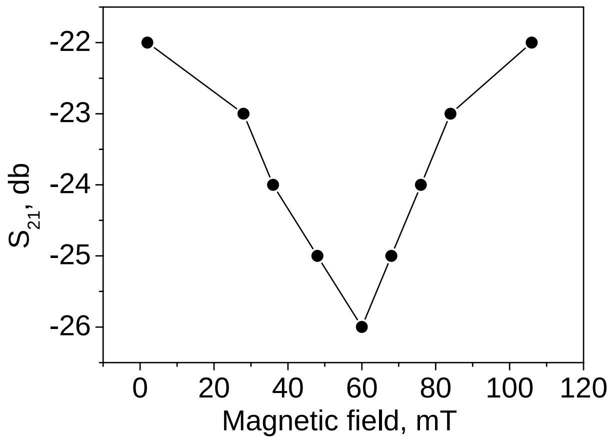


Fig.4. Dependence of the transmission characteristics S_{21} of PPR at with tangentially magnetized LSMO film 2 on the external field induction $\mu_0 H$. The dependence was obtained at 295 K and 4.6 GHz.

Analysis of the behavior of the considered PPR structure with the FM thin film can be made on the basis of an approximation developed in our previous work [12]. It can be shown, that information about the field dependence of the susceptibility of the FM film can be obtained from the field dependence of the transmission coefficient S_{21} of the PPR if the linewidth of FMR exceeds significantly that of the PPR. In this case the minimum of S_{21} is observed when the FMR conditions are satisfied (see formula (4)) for the FM film. The magnetization of the sample 2 and the linewidth of FMR determined from the data presented in Fig.4. were $\mu_0 M$ = 390 mT and $\mu_0 \Delta H_{pp}$~60 mT, respectively. In fact, FMR was successfully registered by a simple PPR technique in thin (~100nm) film 2 because of its several times higher magnetization as compared to sample 1.

4. CONCLUSIONS

Thin films of the manganite LSMO material were prepared on LAO substrates by RF magnetron sputtering and microwave characteristics of these films were investigated. The EPR absorption spectrum of one of the samples showed a double-line character below the Curie temperature. The effect was interpreted as due to the coexistence of the paramagnetic- and ferromagnetic-like phases in the sample being characterized by different values of the magnetic resonance fields and linewidths. The main microwave parameters of these phases were determined at 293 K. A possibility for application of simple parallel plate resonators for investigation of the microwave parameters of FM LSMO thin films at FMR frequency has been demonstrated.

Acknowledgments

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