Surface and bulk spin-wave resonances in $La_{0.7}Mn_{1.3}O_3$ films

2001 J. Phys.: Condens. Matter 13 4049

(http://iopscience.iop.org/0953-8984/13/18/313)

INSTITUTE OF PHYSICS PUBLISHING JOURNAL OF PHYSICS: CONDENSED MATTER

J. Phys.: Condens. Matter **13** (2001) 4049–4064 www.iop.org/Journals/cm PII: S0953-8984(01)19374-4

Surface and bulk spin-wave resonances in $La_{0.7}Mn_{1.3}O_3$ films

V Dyakonov[1,2], A Prohorov[1], V Shapovalov[1], V Krivoruchko[1], V Pashchenko[1], E Zubov[1], V Mihailov[1], P Aleshkevych[2], M Berkowski[2], S Piechota[2] and H Szymczak[2]

[1] Donetsk Physics and Technology Institute, National Academy of Sciences of Ukraine, 83114 Donetsk, Ukraine
[2] Institute of Physics, Polish Academy of Sciences, 02-668 Warsaw, Poland

Received 27 November 2000, in final form 26 February 2001

Abstract

In this work, we present the measurements of exchange-dominated nonpropagating surface and bulk spin-wave modes in the La-deficient epitaxial $La_{0.7}Mn_{1.3}O_3$ films prepared by dc-magnetron sputtering. The angular and temperature dependences of the modes observed are discussed. The main result obtained is the observation of the spin-wave resonance (SWR) consisting of a series (17) of well resolved standing spin-wave modes in the perpendicular external magnetic field geometry. The surface spin-wave modes have been observed in manganites for the first time. As the magnetization is rotated out of perpendicular to the film surface, a 'critical angle', φ_{cr}, is fixed, at which the surface and first spin-wave modes have been transformed into the uniform mode. It is shown that only the uniform mode exists in the region $0 < \varphi < \varphi_{cr}$. The surface mode data are consistent with the surface-inhomogeneity model in which the surface-anisotropy field acts on the surface spin. Possible origins of the surface anisotropy are discussed. Based on the temperature and angular dependences of SWR spectra, the main microscopic parameters (the spin-wave stiffness, exchange constant and g-factor value) are established.

1. Introduction

During the past years, physical properties of doped manganites with the general formula $La_{1-x}D_xMnO_3$, where D is the divalent ion, have been extensively studied, both from a fundamental point of view and for their potential applications ([1] and references therein). At a fundamental level, new physics associated with such systems arises from a strong correlation between structural, transport and magnetic properties of these materials.

The character of spin ordering at the transition to the ferromagnetic metallic state was shown to influence the mobility of charge carriers and, correspondingly, the electronic transport. Therefore, in studying the magnetic properties of manganites, it is important to investigate the

spin dynamics as it is intimately connected with charge motion between Mn^{3+} and Mn^{4+} ions through the so called 'double-exchange' interaction [2].

There are a few main experimental methods of spin dynamics study: inelastic neutron and Brillouin scattering and magnetic resonance. In this work, the magnetic resonance technique, as a highly precise and effective method, is used for studying the magnetic dynamics in magnetoresistive films as well as for determination of the effective nearest-neighbour Heisenberg interaction or spin-wave stiffness and other fundamental characteristics of the spin system.

It has been shown by Kittel [3] that the SWR spectrum can be excited in a ferromagnetic thin film if it exhibits appropriate boundary conditions. The pinning mechanism, which is responsible for excitation of the SWR spectra, is related either to the intrinsic surface magnetic anisotropy or to the magnetic inhomogeneities of the film [4, 5]. In any case, the magnetic inhomogeneities should be distributed regularly in the thickness of the film (or localized near the film surface).

Measurements of the standing spin-wave resonance in thin manganite films have been carried out recently [6–8]. However, the observation of the nonpropagating surface spin-wave modes has not been reported yet. These exchange-dominated surface modes were a subject of both theoretical and experimental investigations for identification of the SWR spectrum in iron-garnet films [9–11].

The purpose of this work is to study the characteristics of low-lying excitations in the La-deficient epitaxial $La_{0.7}Mn_{1.3}O_3$ films. The parent compound $LaMnO_3$ is known to be an antiferromagnetic insulator. In the $La_{0.7}Mn_{1.3}O_3$ compound the cation deficiency responsible for the existence of mixed-valent Mn^{3+}/Mn^{4+} ions is due to the provision of superfluous Mn content. The substitution of La^{3+} with Mn induces the metal–insulator transition and the charge carriers due to this doping mediate the ferromagnetic interactions between the localized Mn spins. Such samples are self-doping systems and show a colossal magnetoresistance at the manganese content of 1.2–1.3 [12].

In the present work, the temperature and angular dependences of the exchange-dominated spin-wave modes have been investigated. The identification of the acoustic-type surface modes in the SWR spectrum was performed. Peculiarities of the FMR spectra for in-plane and out-of-plane magnetic field geometry have been also studied. Based on a study of the FMR and SWR spectra, the values of the spin-wave stiffness, exchange constant and g-factor have been calculated.

The paper is organized as follows: In section 2 the sample preparation and experimental procedure are briefly discussed. To provide a theoretical background for the experimental results, the main features of the Puszkarski surface-inhomogeneity model of SWR modes are reviewed in section 3. Section 4 contains the experimental results, which confirm the excitation of the nonpropagating space and surface spin-wave modes. Discussion of the experimental data obtained, comparison with the model as well as calculation of microscopic parameters of magnetic state in the film studied are also presented in section 4.

2. Samples and experimental procedure

The samples studied are epitaxial $La_{0.7}Mn_{1.3}O_3$ films. The preparation technology of both films and ceramic pellets as targets has been previously described in detail in [13] and [14]. In brief, the films were deposited onto [001] oriented single-crystalline $LaSrAlO_4$ substrates using dc-magnetron sputtering. During the sputtering of films, the discharge current was 100 mA in a 10 mTorr $Ar:O_2 = 4:1$ atmosphere. The temperature of the substrate surface was 600 °C. After that the films were cooled to room temperature at the rate of 5 K min^{-1}. In order to

achieve a homogeneous film, they were post-deposition annealed at 600 °C for an additional 30 min in oxygen flow. The thickness of the films was estimated to be about 3500 Å.

The chemical composition of the films studied as well as the ceramic target used for film preparation has been discussed in [12] and [15]. The ionic structure of

$$(La^{3+}_{0.66}V^{(c)}_{0.05}Mn^{2+}_{0.29})_A(Mn^{3+}_{0.65}Mn^{4+}_{0.29}V^{(c)}_{0.06})_B O^{2-}_{2.84}V^{(a)}_{0.16} \tag{1}$$

obtained for ceramic samples is conserved for the $La_{0.7}Mn_{1.3}O_3$ films with an uncertainty of about 10%.

The x-ray diffraction spectra (figure 1) obtained with the Bragg–Brentano geometry and Ni $K\alpha_1 + \alpha_2$ radiation show that the films are single phase and oriented along the pseudocubic axis [100]. The distortion of the crystallographic structure from the cubic symmetry with the accuracy 0.025% has not been detected. The lattice constant obtained from the peak position at the (400) reflection is 3.907 Å. Despite the fact that the lattice constants of the film and substrate (3.764 Å in the ab-plane) are slightly different the films have both a strong in-plane and axis texture and quality close to the quality of single crystals. Magnetization, magnetoresistive and thermoelectric (Seebeck coefficient) properties of these films have been studied earlier [12, 13]. The films exhibit a substantial negative magnetoresistance of $\Delta R(H = 1\ \mathrm{T})/R(0) \approx 60\%$ near T_C.

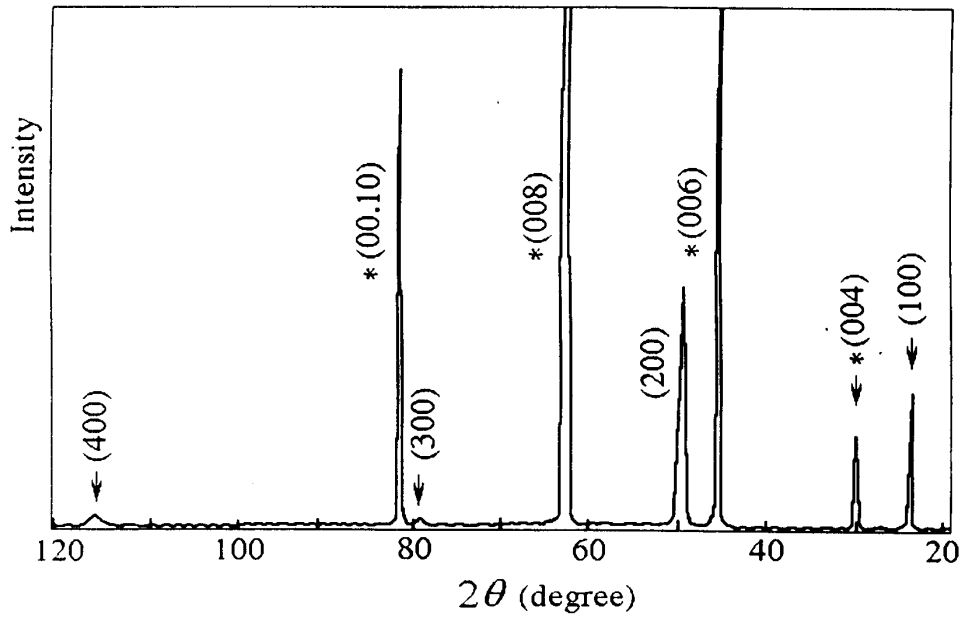

Figure 1. The x-ray diffraction pattern of the epitaxial $La_{0.7}Mn_{1.3}O_3$ films deposited onto [001] oriented $LaSrAlO_4$ substrates. The small reflection at 11.44° belongs to the superstructure along the c-axis. Reflexes of the substrate are indicated by *.

The resonance measurements for in-plane magnetic field (H_{II}) geometry were carried out by conventional electron paramagnetic resonance (EPR) spectrometry (at about 9.47 GHz) over a temperature range from 300 to 77 K. The external magnetic field was also scanned in the film plane.

Measurements of the SWR spectra in out-of-plane magnetic field ($H_\perp$) geometry were performed using a X-band reflection spectrometer operating at fixed frequency (about 9.235 GHz) in conjunction with a variable temperature flowing gas cryostat. A standard

high frequency field (HFF) modulation and phase-sensitive detector techniques were used, so that the detected signal corresponded to the field derivative of the absorbed power. The film samples were positioned in the centre of the microwave cavity. Rotation of the sample was realized in the cavity in plane normal to the film plane with dc magnetic field changing its angle from the film normal to the film plane. The applied magnetic field was changed from 0 to 1.1 T. An HFF field was perpendicular to the external magnetic field and was parallel to the film plane. Sample temperature was varied in steps from room temperature to liquid-helium temperature.

3. Model

When an applied magnetic field is perpendicular to a single-crystal ferromagnetic film, the resonance field positions for long-wavelength modes of spin excitations, including the demagnetization and magnetocrystalline fields and the Heisenberg exchange, are given by the Kittel relation [3]:

$$H_n = \omega/\gamma + 4\pi M_0 + H_A - Dk_n^2. \tag{2}$$

Here ω is the angular frequency, γ is the gyromagnetic ratio, M_0 is the saturation magnetization, D is the spin-wave stiffness constant, $k_n = n\pi/L$ is the wave vector of the nth mode ($n = 1, 2, 3, \ldots$), H_A is the magnetocrystalline anisotropy field and L is the film thickness.

Kittel [3] has shown that the normal spin-wave resonance modes excited in thin magnetic films depend on an effective anisotropy field acting on the surface spins. The major sources of the surface anisotropy are first of all the broken symmetry of the atoms at the surface as well as various extrinsic effects such as inhomogeneous distribution of atoms, epitaxial surface strains etc.

Following the Puszkarski surface-inhomogeneity model [10, 11], the pinning condition at each film surface can be described by an effective parameter

$$A = 1 - (g\mu_B/2SzJ)(\boldsymbol{K}_S\boldsymbol{n}) \tag{3}$$

where g is the spectroscopic-splitting factor, μ_B is the Bohr magneton, S is the atom's spin, J is the Heisenberg exchange interaction parameter of two nearest spins, z is the number of nearest neighbour spins in a crystal lattice, $\boldsymbol{K}_S$ is the effective surface anisotropy field and $\boldsymbol{n}$ is the unit vector parallel to the spin quantization axis. The parameter A measures the strength of pinning of the surface spins. When $A < 1$, only bulk spin-wave modes with real values of the wave vectors k_n can be exited, whereas for $A > 1$, the surface states also appear. If $A = 1$ (absence of surface anisotropy), only one resonance corresponding to a uniform spin-wave mode with $k = 0$ is present.

For $A > 1$, the highest-field modes in the SWR spectrum are acoustic-type surface modes. When the parameters A for both surfaces are greater than one and different, two acoustic surface modes can be exited. These exchange-dominated surface modes are different from magnetostatic surface modes, which generally are long-wavelength modes having negligible exchange energies [16, 17]. As the magnetization is rotated out of perpendicular to the film surface, one should observe a 'critical angle', φ_{cr}, at which the surface and first spin-wave modes transform into the uniform mode of the film.

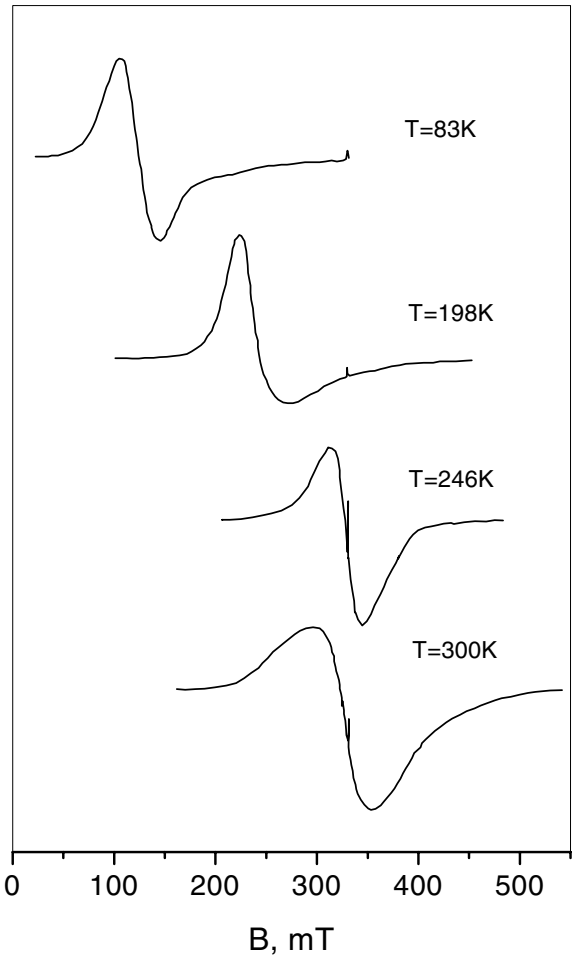

Figure 2. FMR spectrum in the parallel external magnetic field geometry (along the [110] direction) for different temperatures.

4. Experimental results

4.1. In-plane geometry

A resonance spectrum for the parallel field geometry ($H_\parallel$ film plane) in the $La_{0.7}Mn_{1.3}O_3$ film studied is presented in figure 2. It displays one intensive peak identified with the FMR zero mode (the uniform procession mode) only. The higher modes are not excited in accordance with theoretical prediction [3]. An almost symmetric EPR line shape has been observed in the paramagnetic range from 200 up to 300 K. Most changes of shape and linewidth are observed near the transition temperature ($T_C \approx 186$ K) in spite of the fact that conductivity in this temperature range is less than at low temperatures. On the one hand, the non-uniform broading of a magnetic resonance line can be connected with magnetic nonuniformity of the film, which causes a spread of T_C in volume. On the other hand, a change of ratio between the time of current carrier diffusion through a skin layer and the times of spin–lattice and spin–spin relaxation can exert an essential influence on the shape of the magnetic resonance

line in a conducting environment. Below T_C, the shape of FMR signal is asymmetric but the Dysonian-like FMR signal changes gradually to the symmetric one again with decreasing temperature.

The resonance peak-to-peak linewidth, ΔH_{pp}, increases abruptly near the Curie point and then reduces significantly again (figure 3). In the ferromagnetic state the linewidth slightly increases as the temperature is reduced. A large peak in the linewidth just near the transition temperature is a typical feature of manganite samples with magnetoresistive effect [18, 19]. Possible causes of such ΔH_{pp} behaviour could be an existence of AFM clusters near defects or an intrinsic phase separation [20]. The ferromagnetism of the whole sample forms the basic contribution to the linewidth in the paramagnetic range ($T > 240$ K) and at low temperatures ($T < 150$ K), which corresponds to typical behaviour of ΔH_{pp} in FM systems [21]. The ΔH_{pp} behaviour in the temperature interval of $150 < T < 240$ K is presumably due to the fluctuation contribution to the linewidth from AFM areas taking into account that an increase of the EPR linewidth is characteristic for AFM [22]. As seen in figure 3, the resonance field, $H_{res}(T)$, decreasing at $T < 220$ K becomes slightly dependent on temperature at $T < 150$ K. The maximum of the $\mathrm{d}H_{res}/\mathrm{d}T$ dependence takes place at $T \approx 205$ K, which corresponds to a peak in the linewidth. The behaviour of the absorption line is consistent with the expression for the resonance frequency $\omega/\gamma = [H(H + 4\pi M_{eff})]^{1/2}$ (this will be shown below for the case when in-plane magnetic field H is much stronger than the anisotropy field H_A). It should be noted that the value of the effective magnetization $4\pi M_{eff}$ obtained from FMR is in satisfactory agreement with the SQUID data [12].

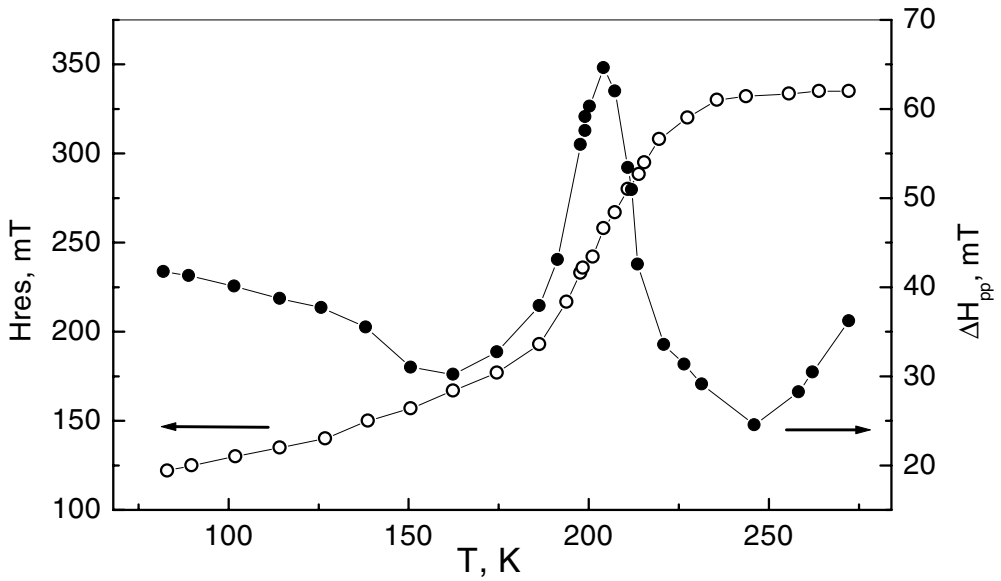

Figure 3. Temperature dependence of the resonance field H_{res} and linewidth in the $La_{0.7}Mn_{1.3}O_3$ film at 9.47 GHz.

The anisotropy of the H_{res} field was detected at $T < T_C$ when the external field was scanned in the film plane. In figure 4, the $H_{res}(\phi)$ dependence at 80 K is presented, where ϕ is the angle between the [100] axis and field direction. The anisotropy of linewidth was also fixed: ΔH_{pp} varies from 35 mT when $H_{\|}$ [110] to 85 mT for $H_{\|}$ [100]. It should be mentioned that epitaxial strain-induced magnetic anisotropy in thin (<3000 Å) films of manganites has been observed recently [23, 24].

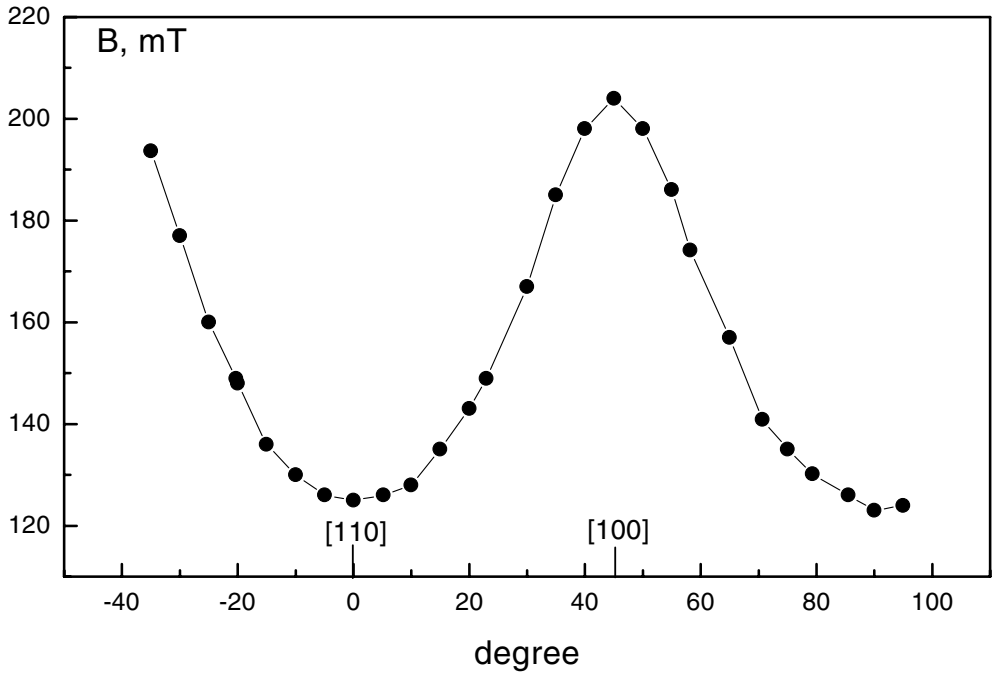

Figure 4. Angular dependence of the in-plane resonance field H_{res} at 9.47 GHz and 80 K.

4.2. Out-of-plane magnetic field geometry. Temperature and angular dependences

Generally, the additional lines observed in the FMR spectrum at the transverse excitation can be attributed to the surface modes [25] or bulk spin-wave modes [26]. Such interpretation correlates with the observed spectra evolution as the sample is rotated in a magnetic field. Indeed, according to the results of measurements for magnetic field perpendicular to the film plane, a resonance spectrum consists of 11 well resolved lines (figure 5). Two of them on the high-field side, SM_1 and SM_2, we identify as the acoustic surface modes, while the others are the bulk nonuniform spin-wave modes. The following experimental facts give evidence for conclusion of the surface character of the SM_1 and SM_2 modes:

(i) the resonance field of the modes is above the theoretical uniform-mode field position;
(ii) the intensity of the modes is smaller than the next-lower-field mode ($n = 1$);
(iii) the angle dependence of these modes (see figure 8) is consistent with the predictions for a surface mode as given by Puszkarski [10, 11].

The theory based on the surface inhomogeneity model predicts that multi-peak SWR occurs only if $A \neq 1$. No peak corresponding to the uniform mode ($k = 0$) appears in this case. According to [10] and [11], the exchange surface modes have an imaginary wave vector $k_z = \mathrm{i}\delta$ normal to the film surface and a zero component in the film plane. Two acoustic-type surface modes observed are the evidence of both the facts that the system is asymmetrical and that both surface A parameters are greater than one and different. The highest-field mode (SM_2 in figure 5) corresponds to a quasisymmetric surface mode and the next mode (SM_1 in figure 5) corresponds to a quasiantisymmetric surface mode. For the quasisymmetric mode, the amplitude of the oscillating dipole moment is greatest at the surface having the stronger surface anisotropy field. For the quasiantisymmetric mode, the amplitude is larger at the surface having the weaker surface anisotropy.

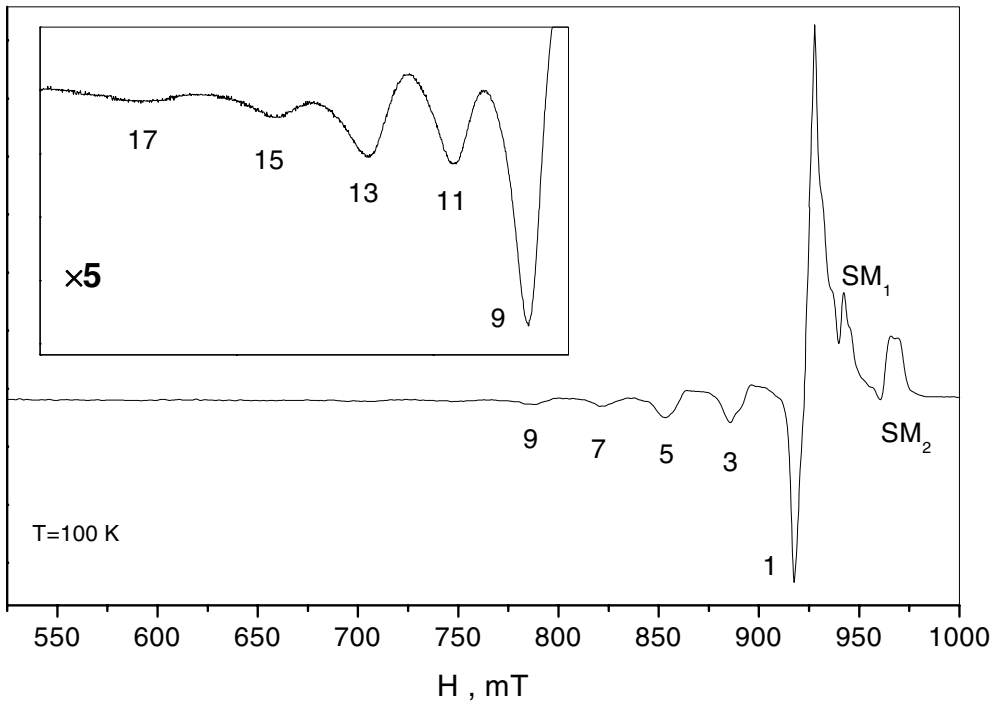

Figure 5. Spin-wave resonance spectrum at 100 K. The inset shows the 9–17 SWR modes increased by a factor of five.

Figure 6 displays the temperature dependence of the SWR spectrum for the $H_\perp$ field. As the temperature approaches T_C from below, the resonant field H_{res} decreases and becomes independent of temperature at $T > 200$ K. The ferromagnetic ordering temperature ($T_C = 186$ K) obtained from FMR differs somewhat from the value of T_C measured by a SQUID [12]. An unusual transformation of the SWR spectrum is observed with decreasing temperature. While nine lines of the SWR, namely, two surface modes and seven bulk modes with $n = 1, 3, 5, 7, 9, 11, 13$, are observed below 165 K, the 15th and 17th modes occur only below $T = 143$ K and disappear at 48 and 87 K, respectively. Possible mechanisms of this transformation are not clear at present and further experiments should be performed. Thus, the maximum number of detected modes is observed over a temperature range of 87–143 K.

Another interesting feature was detected in the temperature dependence of SWR linewidths (figure 7). The peak-to-peak linewidth for the first bulk spin-wave mode ($n = 1$) is equal to about 0.17 kOe at $T = 5$ K and changes negligibly as temperature is increased up to $T = 140$ K. Such behaviour is typical of uniform ferromagnetic samples. In the vicinity of T_C, the ΔH_{pp} value increases sharply due to critical fluctuations reaching the maximum value of 0.55 kOe. It should be noted that the linewidth increases more weakly for H perpendicular to the film plane than for the $H_\parallel$ direction. Above T_C, the $n = 1$ mode linewidth increases smoothly as expected for a paramagnetic state. The $\Delta H_{pp}(T)$ values of SWR modes 3–11 also change smoothly from 0.1 to 0.2 kOe below 165 K.

The fact that the resonance linewidths for both in-plane and out-of-plane magnetic field geometry are relatively narrow and depend slightly on temperature below T_C is indicative of the high quality of films studied taking into account that the FMR linewidths provide a very sensitive probe of the uniformity and magnetic homogeneity of film samples [27].

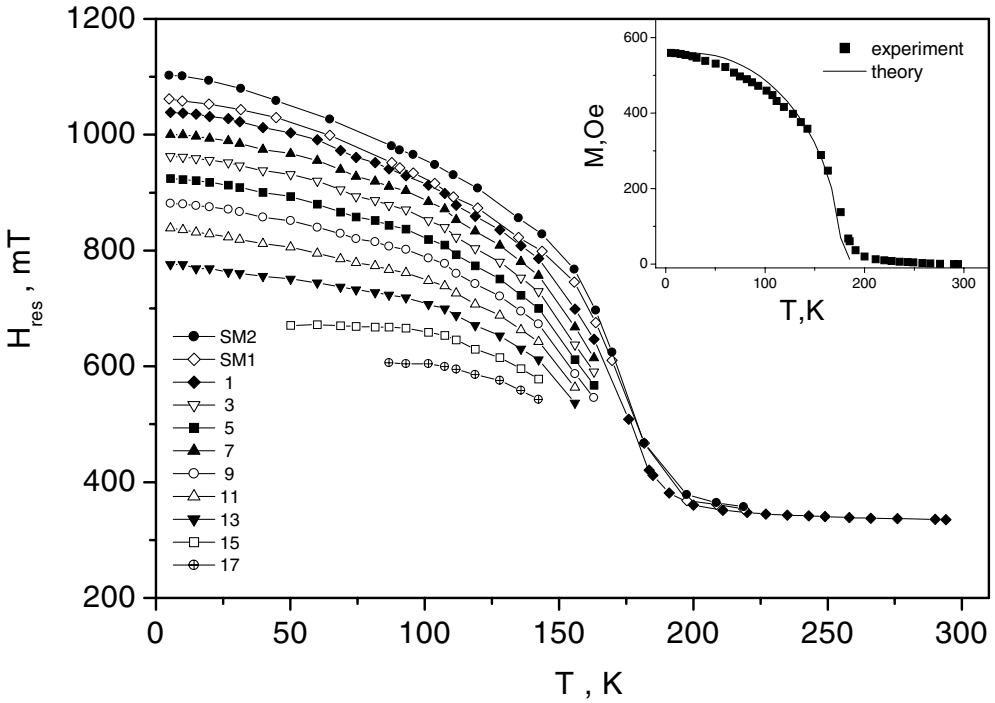

Figure 6. Temperature dependence of resonance field of surface (SM_1 and SM_2) and bulk (1–17) SWR modes. Inset: theoretical $M(T)$ dependence (line) and experimental temperature dependence of resonance field (points) for the $n = 1$ SWR mode in the perpendicular external magnetic field geometry.

The linewidths of the higher-order modes ($n = 13$, 15 and 17) have, however, nonmonotonic low temperature behaviour. As seen, a decrease of the $n = 11$ mode linewidth is accompanied by occurrence of the $n = 13$ mode. Similar behaviour is observed for the $n = 15$ mode. A smooth peak (at 30 K for the 13th mode, at 80 and 110 K for the 15th and 17th modes, respectively) is typical for these excitations. It should be mentioned that decrease of the distance between modes with increasing temperature is due to the spin-wave stiffness constant dependence on temperature in accordance with theoretical calculations [28].

The observed strong increase of dissipation from the 13th mode depends upon the fact that the length of the spin-wave modes is compared with the thin film inhomogeneity size. In [29] the influence of pointed magnetic disorder on the spin-wave spectrum was observed. It was shown that if the spin-wave vector becomes of the order of or more than some value $k_0 = 2/b$, where b is the characteristic size of the inhomogeneity, the dispersion law $\omega(k)$ will be essentially different from quadratic and spin-wave decay is considerably increased. This reflects the fact of strong interactions between spin-wave and scattering defects.

Let us estimate the value of inhomogeneity size in our thin film. The maximum number n of observed modes is 17. It is known that the wave vector of the normal spin-wave mode $k_n = n\pi/L$, where $L = 3500$ Å is the film thickness. From condition $k_{17} \sim k_0$ we have a size of the magnetic inhomogeneity:

$$b \approx \frac{2L}{17\pi} \approx 130 \text{ Å}.$$

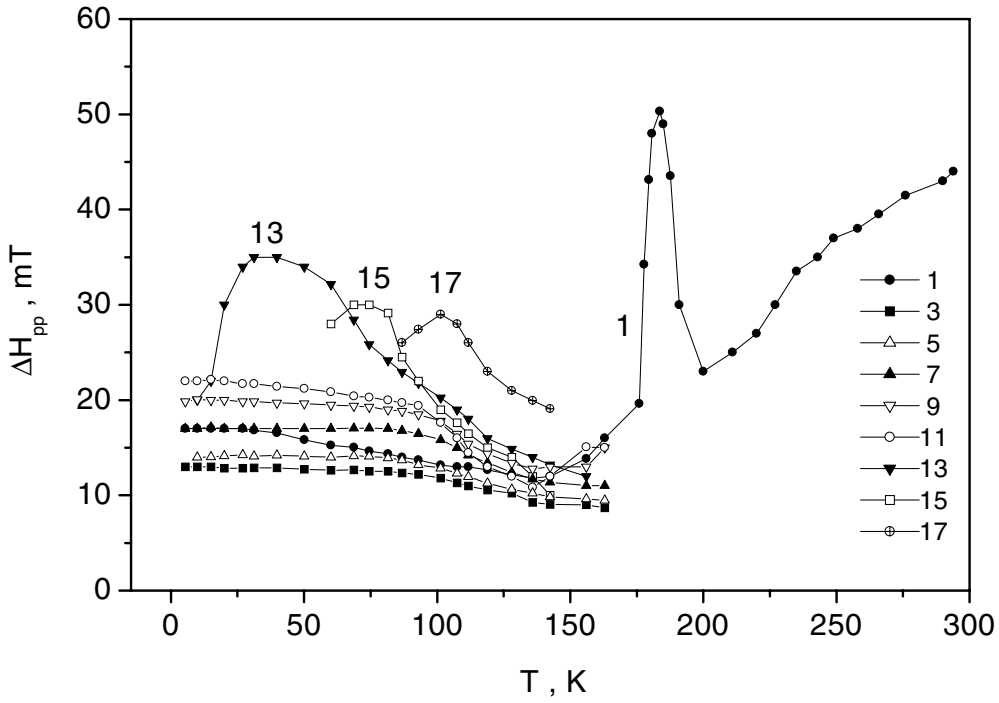

Figure 7. Temperature dependence of the derivative peak-to-peak linewidth, ΔH_{pp}, of SWR modes in out-of-plane magnetic field geometry.

This is a maximum size because the next mode 19 has intensity $I(n) \sim 1/n^2$ in the limit of experimental sensitivity. In [30] the evidence of anisotropic magnetic polarons in $La_{0.94}Sr_{0.06}MnO_3$ observed by neutron scattering is presented. The polaron size and intercluster distance are equal to approximately 17 and 34 Å, respectively. One can suppose that the large increase of spin-wave damping in our case beginning from $n = 13$, 15 or 17 is caused by magnon scattering on the magnetic polarons. Also, in [30] the authors point out the existence of some magnetic disorder on a scale of 130 Å, which corresponds to antiferromagnetic (AFM) short-range correlation within the canted AFM layered structure. Such inhomogeneities may be responsible for increasing of spin-wave decay.

Using the experimental data obtained, we have made some quantitative estimations of the g-factor and interaction parameter. At temperature $T > 200$ K, the film magnetization can be considered as negligible [12]. If it is taken into account that at $T = 4.2$ K the sample is magnetized to saturation, the difference of resonant fields at 4.2 and 294 K (inset to figure 6) determines the effective value of the anisotropy field, $H_{eff} = 4\pi M_{eff} = 4\pi N M_0 - 2H_A =$ 7.04 kOe, where $4\pi N M_0 = H_{demag}$ is the demagnetization field connected with the shape of the sample. The H_{demag} field in the film, as a rule, considerably exceeds the anisotropy field H_A. As in our experiment the resonant fields are about 3 kOe and higher, an influence of H_A on the resonant characteristics of the film studied (in a first approximation) can be ignored. In this case, we do not exclude the surface anisotropy, which should be rather strong to realize a pinning of magnetic moment on a film surface in SWR conditions. Taking into account the measuring frequency $\nu = 9.235$ GHz, the values of the g-factor and saturation magnetization were obtained to be equal to 1.96 and 560 Oe, respectively. The value of $g < 2$ is due to a spin-orbital interaction.

For further calculations, it is necessary to know the value of spin S. We should take for S its effective value S_{eff}, that is equal to the weighted average of the spins for the Mn^{3+} and Mn^{4+} ions. For the effective spin, we obtain $S_{eff} = 2x_1 + (3/2)x_2 = 1.735$. Here we use, in accordance with the ionic composition formula (1), the concentrations of $x_1 = 0.65$ and $x_2 = 0.29$ for Mn^{3+} and Mn^{4+} ions, respectively.

To test the validity of the S_{eff} determination, we assume that the lattice is cubic with one magnetic ion in the unit cell. Than, it is easy to find the lattice constant a from the following expression:

$$a = \left(\frac{\mu_B g S_{eff}}{M_0}\right)^{1/3} \tag{4}$$

where $g = 1.96$, $S_{eff} = 1.735$ and $M_0 \approx 560$ Oe. From the expression (4), we obtain a reasonable value of $a \approx 3.83$ Å, that is comparable with the value of 3.907 Å determined from the x-ray diffraction data.

In the molecular field approximation, the magnetization of a ferromagnet can be described by the formula

$$M = M_0 B_S(x) \tag{5}$$

where $x = 3S_{eff}^2[JzM(T)]/[k_B T M_0]$, $B_S(x)$ is the Brillouin function, $J = 3T_C/2S(S+1)z$ and k_B is the Boltzmann constant. A comparison of the theoretical $M(T)$ curve obtained using the calculated exchange parameter and dependence (5) and the experimental temperature dependence of the resonant field for the $n = 1$ mode (points) is presented in the inset to figure 6. The rather good agreement of experiment and theory is observed.

Since the observed parallel and perpendicular spectra are very different, the angular dependence of SWR has been measured. The angular dependences of the mode positions are shown in figure 8. As seen in figure 8, in the case of $\varphi = 90^\circ$ ($H_\perp$ film plane), the spectrum consists of nine lines belonging to modes 1–17 of SWR. One can see that with deviation of the $H_\perp$ field from perpendicular orientation, the resonance modes of SWR start to disappear. The 15th and 13th modes disappear at the angle of $90 \pm 3^\circ$. Other modes of SWR disappear at higher deviation: the 11th at $90 \pm 6^\circ$; the ninth at $90 \pm 8^\circ$; the seventh at $90 \pm 10^\circ$; the fifth and third at $90 \pm 12^\circ$. There exists a critical angle $\varphi_{cr} \approx 14^\circ$ between the static magnetic field and the normal to the film ($90^\circ \pm \varphi_{cr}$), at which the magnetization of sample is perpendicular to the surface anisotropy field, $\boldsymbol{M} \perp \boldsymbol{K}_S$, and the parameter A is equal to 1. As seen in figure 8, for angles $\varphi < \varphi_{cr}$, a well-resolved multi-peaks SWR spectrum is detected, whereas for $\varphi > \varphi_{cr}$ the spectrum contains in general only one resonance absorption line. At critical angle, the surface and first spin-wave modes transform into the uniform mode. The uniform mode exists only in the region $0 < \varphi < \varphi_{cr}$. These results are consistent with the Puszkarski surface-inhomogeneity model [10, 11].

The strong dependence of number of excited spin-wave modes and their parameters on both temperature (figure 7) and orientation of the film in the magnetic field (figure 8) is indicative of the domination of the dynamic mechanism of spins pinning at the film surfaces [31].

The quantitative and qualitative correlation between experiment and theory is also exhibited in an angular dependence of the SWR modes. As it is known (see, e.g., [32]), the FMR resonant field can be expressed through tensor N_{ij} ($i, j = 1, 2, 3$ or x, y, z) of demagnetization factors of ellipsoid by the formula

$$\begin{aligned}(\omega/\gamma)^2 = {} & [H\cos(\vartheta - \varphi) + (N_{11} - N_{33})M + 2H_A\cos^2\vartheta] \\ & \times [H\cos(\vartheta - \varphi) + (N_{22} - N_{33})M + 2H_A\cos 2\theta] - (N_{12}M)^2\end{aligned} \tag{6}$$

where φ is the angle between the z-axis and the external magnetic field direction and ϑ is the azimuthal angle of magnetization. The axis z is proposed to be perpendicular to the

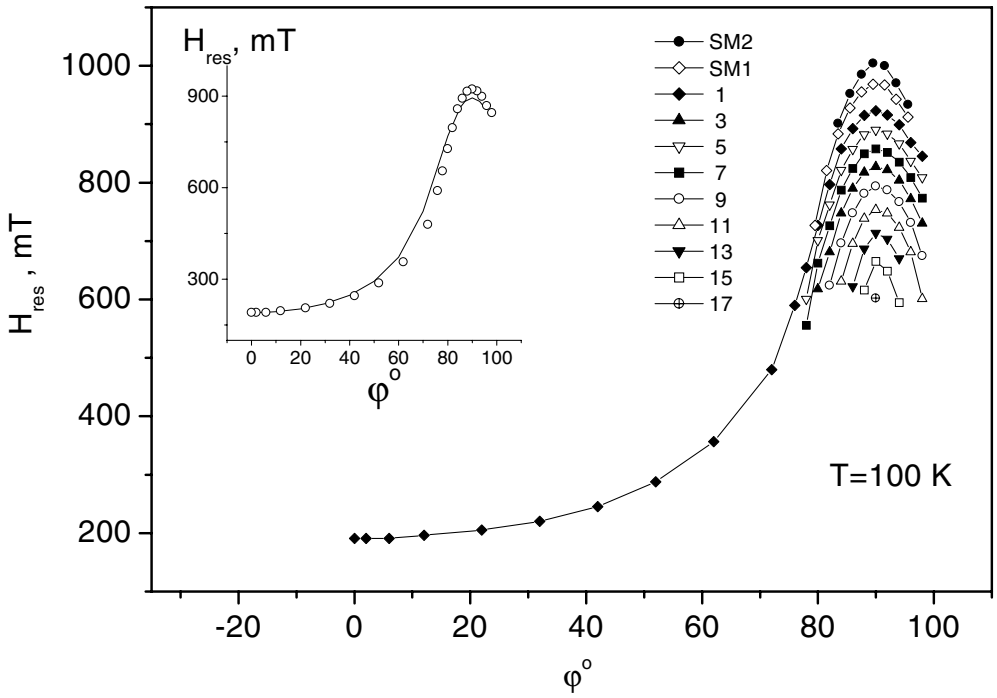

Figure 8. Resonant fields of the SWR modes 1–15 versus the rotation angle of the film relative to the magnetic field (points). Inset: comparison of the angular dependence of resonant field H_{res} calculated using equations (6) and (7) (denoted by line) and the experimental H_{res} dependence for the $n = 1$ SWR mode at 100 K.

film surface, being in the x–y-plane. In the coordinate system with the axis z' parallel to the magnetization direction, the tensor N_{ij} (diagonal in the main axes of ellipsoid) transforms, and its components have the form $N_{11} = 4\pi \cos^2\alpha \sin^2\theta$, $N_{22} = 4\pi \sin^2\theta \sin^2\alpha$, $N_{33} = 4\pi \cos^2\theta$, $N_{12} = -2\pi \sin(2\alpha) \sin^2\theta$ where α is the polar angle of the vector $\boldsymbol{M}$ in an initial spherical coordinate system.

From the condition of the free energy minimum

$$E = -H_S M \sin\vartheta \sin\alpha - H_0 M \cos\vartheta - H_A M \cos^2\vartheta + 1/2(4\pi M^2)\cos^2\vartheta$$

where $H_S = H \sin\vartheta$, $H_0 = H \cos\varphi$, the equilibrium angle is equal to $\alpha = \pi/2$, and the angle ϑ is determined from the equation

$$H \sin(\vartheta - \varphi) - H_{eff} \sin\vartheta \cos\theta = 0 \tag{7}$$

where $H_{eff} = 4\pi M - 2H_A$.

From the equations (6) and (7), for two limiting cases we have

(i) if $\theta = \pi/2$ and $\varphi = \pi/2$ (a field H is perpendicular to the film plane),

$$\omega = \gamma(H_\perp - H_{eff});$$

(ii) if $\vartheta = 0$ and $\varphi = \pi/2$ (for field $H_\parallel$ direction),

$$\omega = \gamma\{H_\parallel(H_\parallel + H_{eff})\}^{1/2}.$$

From experiment we have $H_\perp > H_\parallel$. That is why in our case the anisotropy of easy-plane type is realized. Unfortunately, we cannot find the value of H_A because it is necessary to have

additional experimental data. However, our estimations of lattice constant a using equation (4), which is comparable with one determined from x-ray diffraction data, support our suggestion of a small value of crystal anisotropy field H_A. We suppose that both magnetocrystalline anisotropy and the film–substrate relaxation of strain may be the source of the weak magnetic anisotropy observed in our film.

The comparison of the angular dependence of resonant field H_{res} for the $n = 1$ SWR mode calculated using equations (6) and (7) with experimental data is presented in the inset to figure 8. Taking into account a rather approximate character of the calculations, the agreement between theory and experiment may be considered to be quite satisfactory.

Following equation (2), the distance between the resonance fields of the n_ith and n_jth modes is described by

$$\Delta H = H_{ni} - H_{nj} = D(\pi/L)^2(n_j^2 - n_i^2).$$

Figure 9 indicates the dependence of $\Delta H_n = H_1 - H_n$ (where H_1 and H_n are the resonance fields for first and nth modes) on the square of the bulk SWR mode number, n^2, obtained at $T = 82$ K. It is seen that at $n > 5$ we have a linear spectrum ω from k_n. The SWR modes with small n deviate from the linear $\Delta H_n(n^2)$ dependence determined by the higher-order ones. Such a deviation of the first modes from the lines determined by high-order modes was observed earlier [33] on evaporated Ni–Fe films. It is in qualitative agreement with the surface anisotropy and inhomogeneity model, which predicts a deviation of H_n from the quadratic law for the first modes [10, 11]. The deviation of the $\Delta H_n(n^2)$ dependence from linearity also shows that the spin pinning at the boundaries of the film becomes weaker with increasing spin-wave number. The Kittel condition [3] of spin pinning on a film surface is disrupted. It may be related to the existence of an inhomogeneity in the internal distribution of either the saturation magnetization, fluctuations of exchange interaction parameter and magnetocrystalline anisotropy or stress [29, 34]. Since magnetic anisotropy in the investigated films was shown to be small one should suppose that the nonuniform distribution of the saturation magnetization is responsible for observed deviation from the n^2 law. One can say that pointed peculiarities substantially depend on conditions of specimen preparation, film thickness and tension arising from interaction with the substrate.

From figure 9, we obtain the tangent of the line slope angle of $R = \pi^2/h\gamma L^2 = 10.3$ Oe. The saturation magnetization ($M_0 = 450$ Oe) determined using the expression (2) and figure 9 is close to the analogous value obtained from the analysis of the temperature dependence of the demagnetization field.

Using the linear $\Delta H(n^2)$ dependence we have calculated the value of the spin-wave stiffness constant $D = 150$ meV Å^2 . The value of D determined by the SWR method for the $La_{0.7}Mn_{1.3}O_3$ film is in satisfactory agreement with those measured in other ferromagnetic manganites, for example, 134 meV Å^2 for $La_{0.7}Pb_{0.3}MnO_3$ [35], 170 meV Å^2 for $La_{0.7}Ca_{0.3}MnO_3$ [36], 188 meV Å^2 for $La_{0.7}Sr_{0.3}MnO_3$ [37].

According to spin-wave theory, the spin-wave stiffness constant $D(0) = 2JSa^2$. Using equation (2) and the parameter D, it is easy to obtain the expression for the exchange parameter J:

$$J = \frac{Rg\mu_B}{2S\pi^2 k_B}\left(\frac{L}{a}\right)^2. \tag{8}$$

From expression (8), we obtain the exchange interaction parameter of $J \approx 32$ K for $L = 3500$ Å and $a = 3.9$ Å. This value agrees with one obtained in [38]. The accuracy of the J parameter determination depends strongly on the accuracy of the ratio of the film thickness to the lattice constant.

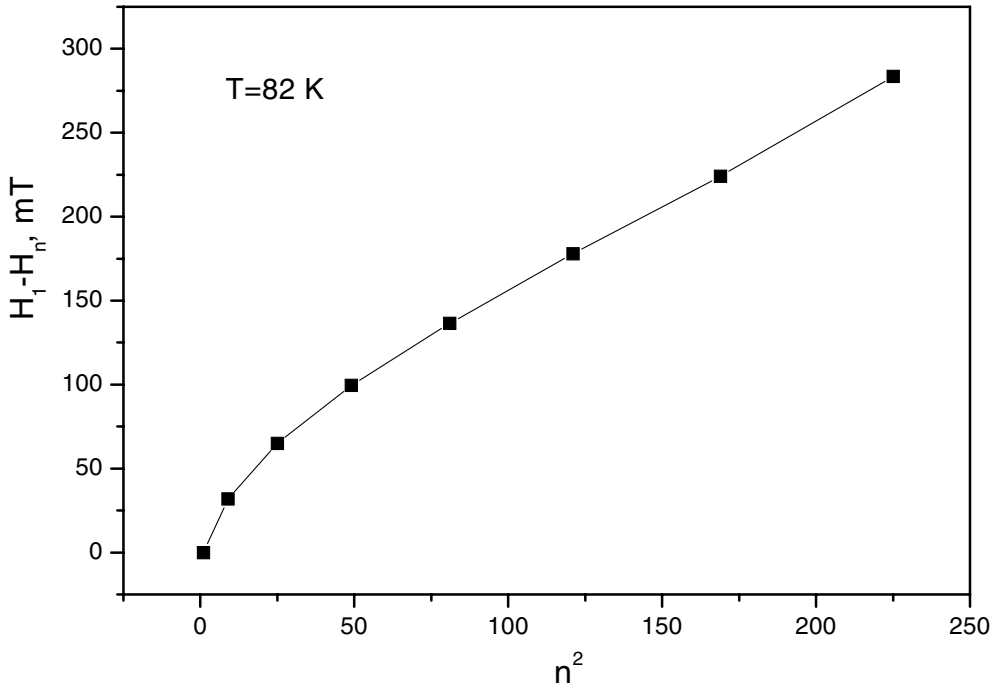

Figure 9. Dependence of $\Delta H_n = H_1 - H_n$ on the square of the corresponding mode number, n^2, at $T = 82$ K.

5. Conclusions

We report the first measurements of surface and bulk SWR modes in La-deficient epitaxial films prepared by magnetron sputtering.

The spin-wave resonance consisting of a series (17) of well resolved standing spin-wave modes in the perpendicular external magnetic field geometry has been observed.

The analysis of the magnetoexchange-branch dispersion relation in the $La_{0.7}Mn_{1.3}O_3$ films has indicated that the internal fields in these films are very homogeneous and that the spin-wave modes depend on the surface pinning conditions rather than variations in the internal field. Assuming pinned surface spins, the spin-wave resonance data have been shown to fit a quadratic dispersion relation. To date, nearly all experimental results on spin-wave excitation in manganite films have been limited to bulk resonances. As shown in this paper, it is an incomplete description of the spin-wave properties of films with the colossal magnetoresistance effect.

The mode separation with angle observed in this study suggests that the boundary conditions are angular dependent. By varying the orientation of an applied magnetic field from perpendicular to parallel to the film surface, the highest field mode wave vector changes from an imaginary value at perpendicular to zero at the critical angle. Its real value ($\sim\pi/L$) is between the critical angle and parallel resonance.

The observation of two acoustic-type surface modes is consistent with Puszkarski's theory if asymmetric boundary conditions are assumed. The lack of symmetry in the boundary conditions is understandable experimentally, since the 'bottom' surface layers can be affected by the substrate, while the top surface is exposed to the atmosphere.

We found an unusual transformation of the spin-wave spectrum, namely, the disappearance of the highest resonance modes with decreasing temperature. However, the origin of the observed transformation is not clear yet.

Based on the temperature and angular dependences data of SWR spectra, the main microscopic parameters (the spin-wave stiffness, exchange constant and g-factor value) were established.

Acknowledgments

This work was partially supported by the Polish Government Agency KBN under contracts 2 P03B 139 18 and 7 TO8A 011 15. The help offered by Dr S Khartsev during realization of this work is gratefully acknowledged. E Zubov is grateful to the Mianowski Foundation (Poland) for a scholarship.

References

[1] Coey J M D, Viret M and von Molnar S 1999 *Adv. Phys.* **48** 167
[2] Zener C 1951 *Phys. Rev.* **82** 403
[3] Kittel C 1958 *Phys. Rev.* B **110** 1295
[4] Portis A M 1963 *Appl. Phys. Lett.* **2** 69
[5] Wigen P E, Kooi C F and Shanabarger M 1964 *J. Appl. Phys.* **35** 3302
[6] Lofland S E, Bhagat S M, Kwon C, Robson M C, Sharma R P, Ramesh R and Venkatesan T 1995 *Phys. Lett.* A **209** 246
[7] Yin J, Sui Y X, Du J H, Zhang Y X, Liu X Y and Liu Z G 1999 *Phys. Status Solidi* a **174** 499
[8] Dyakonov V *et al* 2000 *Phys. Lett.* A **268** 202
[9] Yu J T, Turk R A and Wigen P E 1975 *Phys. Rev.* B **11** 420
[10] Puszkarski H 1970 *Acta Phys. Polonica* A **38** 217
[11] Puszkarski H 1970 *Acta Phys. Polonica* A **38** 899
[12] Krivoruchko V N, Khartsev S I, Prohorov A D, Kamenev V I, Szymczak R, Baran M and Berkowski M 1999 *J. Magn. Magn. Mater.* **207** 168
[13] Krivoruchko V N and Khartsev S I 1998 *Low Temp. Phys.* **24** 803
[14] Pashchenko V P, Andreev A A, Shemyakov A A, Prokopenko V K, Darovskikh E G, Cherenkov O P and Loiko A D 1998 *Inorg. Mater.* **34** 62 (in Russian)
[15] Abramov V S, Pashchenko V P, Khartsev S I and Cherenkov O P 1999 *Funct. Mater.* **6** 64
[16] Eshbach J R and Damon R W 1960 *Phys. Rev.* **118** 1208
[17] Damon R W and Eshbach J R 1961 *J. Phys. Chem. Solids* **19** 308
[18] Oseroff S B, Torikachvili M, Singley J, Ali S, Cheong S-W and Shultz S 1996 *Phys. Rev.* B **53** 6521
[19] Domingues M, Lofland S E, Bhagat S M, Raychudhauri A K, Ju H L, Venkatesan T and Greene R L 1996 *Solid State Commun.* **97** 193
[20] Nagaev E L 1996 *UFN* **166** 833 (in Russian)
[21] Searle C W and Wang S T 1969 *Can. J. Phys.* **47** 2703
[22] Taylor R H and Coles B R 1975 *J. Phys. F: Met. Phys.* **5** 121
[23] Suzuki Y, Hwang H Y, Cheong S-W and Dover R B 1997 *Appl. Phys. Lett.* **71** 140
[24] O'Donnel J, Rzchowski M S, Eckstein J N and Bozovic I 1998 *Appl. Phys. Lett.* **72** 1775
[25] Zhang Z, Wigen P E and Suzuki T 1993 *J. Magn. Soc. Japan* **17** (Suppl. 51) 119
[26] Feng J W, Pam F M, Jin G J, Kang S S, Hu A and Jiang S S 1996 *J. Magn. Magn. Mater.* **156** 241
[27] Lofland S E, Bhagat S M, Ju H L, Xiong G C, Venkatesan T and Greene R L 1995 *Phys. Rev.* B **52** 15 058
[28] Akhiezer A I, Baryakhtar V G and Peletminskii S V 1968 *Spin Waves* (Amsterdam: North-Holland)
[29] Ignatchenko V A and Iskhakov R S 1977 *Zh. Eksp. Teor. Fiz.* **72** 1005 (in Russian)
[30] Hennion M, Moussa F, Biotteau G and Rodriguez-Carvajal J 2000 *Phys. Rev.* B **61** 9513
[31] Hoekstra B, van Stapele R P and Robertson J M 1977 *J. Appl. Phys.* **48** 382
[32] Gurevich A G 1973 *Magnetic Resonance in Ferrites and Antiferromagnets* (Moscow: Nauka) p 591
[33] Bajorek C H and Wilts C H 1971 *J. Appl. Phys.* **42** 4324
[34] Schreiber F and Frait Z 1997 *Phys. Rev.* B **54** 6473
[35] Perrin T G, Aeppli G, Hayden S M, Carter S A, Remeika J P and Cheong S-W 1996 *Phys. Rev. Lett.* **77** 711

[36] Lynn J W, Erwin R W, Bochers J A, Huang Q, Santoro A, Peng J-L and Li Z Y 1996 *Phys. Rev.* B **76** 4046
[37] Martin M C, Shirane G, Endoh Y, Hirota K, Moritomo Y and Tokura Y 1996 *Phys. Rev.* B **53** 14 285
[38] Huber D L 1998 *J. Appl. Phys.* **83** 6949