Vol. **105** (2004) *ACTA PHYSICA POLONICA A* No. 1–2

Proceedings of the Symposium on Colossal Magnetoresistance

# Ferromagnetic Resonance Studies of $La_{0.67}Ca_{0.33}MnO_3$ Film under Stress

B. Kundys*, V.P. Dyakonov, M. Baran, P. Gierłowski and R. Szymczak

Institute of Physics of the Polish Academy of Sciences
al. Lotników 32/46, 02-668 Warsaw, Poland

Ferromagnetic resonance spectra of the $La_{0.67}Ca_{0.33}MnO_3$ ferromagnetic film deposited onto the $x$-cut $LiNbO_3$ substrate were investigated in a wide temperature range. The strain was mechanically introduced into the film using a special holder configuration. This leads to a shift of resonance field with respect to that of the as-grown sample. Analysis of the magnetic resonance shift, induced by a mechanical stress, allowed us to determine the magnetostriction constant $\lambda_{100}$. The magnetostriction determined in this way versus saturation magnetization could be well described by the Callen and Callen theory suggesting the domination of the single ion crystal field interaction in the sample being in the ferromagnetic state.

PACS numbers: 73.50.Jt, 75.80.+q, 68.60.Bs, 71.70.Ej

## 1. Introduction

Manganites attract considerable scientific and practical interest due to their colossal magnetoresistance effect (CMR) involving double exchange (DE) interaction [1–3] and can be very useful in technical applications of magnetic devices [4–7]. The DE interaction model [3] is a basic mechanism to explain the paramagnet–ferromagnet and semiconductor–metal transitions as well as CMR. However, it was recently suggested [8] that the double exchange interaction alone is not sufficient to explain the resistivity behavior as a function of temperature and that the strong electron–phonon interaction arising from the Jahn–Teller (JT) splitting can play an important role in the mechanism of the magnetotransport.

The strong electron–crystal lattice interaction suggests that manganites should be extremely sensitive to applied mechanical stresses, which affect the

---

[*] corresponding author; e-mail: kundy@ifpan.edu.pl

magnetic system through magnetoelastic interactions. Experiments performed on manganites up to now imply that there are a lot of mechanisms responsible for magnetoelastic phenomenon in these materials. It was shown that the magnetostriction below the Curie point is strongly anisotropic and takes values typical of 3*d* metals. However, the magnetostriction of manganites in paramagnetic state is related to the charge carriers localization in applied magnetic field. In many cases the electronic phase separation is responsible for unusual properties of these materials [9, 10].

The fact that mechanical deformation has a strong influence on magnetic properties of manganites is responsible for significant difference in magnetic and transport properties of thin films comparing to their bulk counterparts.

Magnetic properties of manganite thin films are strongly dependent on a lattice constant mismatch between a thin film and its substrate. Strain arising from this mismatch could be responsible for the magnetic state of a manganite sample.

It is shown [11] that $La_{0.5}Ca_{0.5}MnO_3$ thin films are metallic and ferromagnetic, while bulk samples are antiferromagnetic insulators. Probably some mismatch and resulting strain led here to the elimination of the cooperative JT effect, which determines the ground state of the system. It was also shown that about 1% of lattice misfit results in lattice relaxation, which occurs in manganite films thicker than 100 nm. That gives a possibility to change film properties by changing its thickness. The experimental verification of this hypothesis would permit "manganite films engineering". Conductivity measurements of manganite films with different thickness [12] confirm the above suggestion.

Since the magnetoelastic tensor components of thin films differ from their bulk counterparts (because of surface contribution, mismatch stresses, as well as possible texturing effects) the magnetic properties and magnetostriction of thin films could be also different. High magnetostriction is not welcomed for CMR development due to its direct connection with magnetic anisotropy and with the magnetic softness of the material.

It is therefore important that one should obtain maximum information on magnetoelastic properties of manganite films in order to understand macroscopic mechanisms responsible for magnetoelastic interactions.

A strain applied to a ferromagnetic sample induces a magnetoelastic anisotropy, which strongly influences spin configuration and can change magnetic parameters such as resonance field, coercive force, and magnetic susceptibility.

In this work we present the ferromagnetic resonance (FMR) investigation of $La_{0.67}Ca_{0.33}MnO_3$ (LCMO) film under stress. The stress gives a magnetoelastic contribution to magnetic energy of the system. The analysis of the results of these investigations can be treated as an indirect method of magnetostriction measurement in thin films, allowing temperature dependent studies.

## 2. Experimental

Using the $La_{0.67}Ca_{0.33}MnO_3$ target the LCMO 3000 Å thick film was deposited onto $x$-cut thick $LiNbO_3$ substrate having temperature of 730°C by pulsed laser deposition without postannealing. X-ray diffraction investigation showed that the film is single phase, epitaxial, (001) oriented with the pseudocubic lattice parameter $a_0 = 0.3853$ nm. The magnetic properties of the film were studied using SQUID magnetometer.

There are many mechanical methods of introducing stress into films [13–16]. In this work, the controlled strain was introduced to the film using the special holders with known radii of curvature [17]. The sample holders were machined from a block of PTFE, which was appropriate because it did not give any contribution to FMR spectrum. The FMR studies were performed using the X-band spectrometer with the reflection resonance cavity (operating at the fixed frequency about 9.25 GHz), equipped with a variable temperature flowing gas cryostat. FMR measurements, performed in the magnetic field applied in the plane of the sample, have allowed observing the changes in the resonance field when the constant stress was introduced mechanically into the film.

## 3. Results and analysis

It was the most suitable to express the stress effect as a shift $\delta H$ of the resonance field at a fixed resonance frequency. In order to calculate magnetoelastic parameters we have used similar approach as for stress modulated ferromagnetic resonance (SMFMR) technique [18], taking into account the analysis used in Ref. [19]. However, one should note that SMFMR involves dynamic stress applied to the film in contrast to static one used in this work.

As it could be seen in Fig. 1, the stress results in the shift of in-plane resonance field towards higher field values corresponding to non-zero magnetostriction in our sample. The shift of the resonance line contains information on magnetoelastic tensor components and can be estimated by evaluating the resonance field for the stressed and as-grown states from well known FMR conditions [20]:

$$\frac{\omega_0}{\gamma} = (M_\mathrm{s}\sin\theta)^{-1}\left[\frac{\partial^2 E}{\partial\theta^2}\frac{\partial^2 E}{\partial\phi^2} - \left(\frac{\partial E^2}{\partial\theta\partial\phi}\right)^2\right]^{\frac{1}{2}}, \tag{1}$$

where $E$ is a free energy of the sample evaluated for the condition $\partial E/\partial\theta = \partial E/\partial\phi = 0$, $\omega_0$ is a ferromagnetic resonance frequency, $\gamma$ is a gyromagnetic ratio, $M_\mathrm{s}$ is a saturation magnetization, $\theta$ and $\phi$ are polar and azimuthal angles representing the orientation of magnetization. The introduction of the dimensionless parameters allows the task to be simplified. Taking $\Omega = \omega/\gamma\mu_0 M_\mathrm{s}$ and $F = E/\mu_0 M_\mathrm{s}^2$ the dimensionless resonance condition is obtained [20]. The $F$ includes all kinds of energies taken into account: magnetostatic, magnetocrystalline, and magnetoelastic, likewise an energy of Zeeman splitting, as well as of the uniaxial anisotropy.

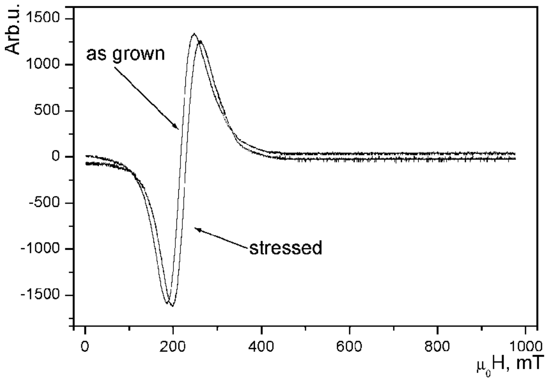

Fig. 1. FMR spectra of $La_{0.67}Ca_{0.33}MnO_3$ film in stressed and in as-grown states at 161 K with in-plane magnetic field geometry.

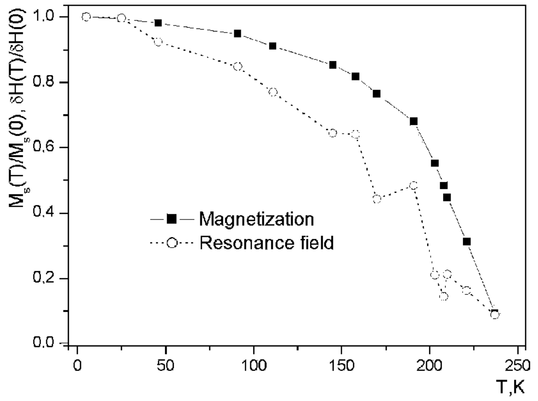

Fig. 2. Reduced saturation magnetization and reduced shift in resonance magnetic field as a function of temperature. Magnetic field is in the film plane.

Figure 2 shows how the reduced shift of resonance field and the reduced saturation magnetization of the sample change with the temperature. The measurements of FMR under stress, with the in-plane dc magnetic field, allow a $\lambda_{100}$ magnetostriction constant to be determined [20].

$$\mu_0 \delta H = [-6 + (9 + 3\alpha)\sin\varphi]\frac{\sigma\lambda_{100}}{2M_s}, \tag{2}$$

where $\varphi$ is the angle between $H$ and applied stress $\sigma$ defined as $\sigma = \varepsilon Y_m/(1-\nu^2)$ [14]. Here $\varepsilon = t/(2R) = 250$ ppm is the strain induced by film bending over radius $R$, $t$ is the thickness of the film and substrate, $Y_m$ and $\nu$ are the Young modulus and Poisson ratio equal to 232 GPa and 0.26, respectively [21]. The dimensionless parameter $\alpha$ is determined by the following expression:

$$\alpha = \frac{m_e}{2\omega}, \tag{3}$$

where $m_{\mathrm{e}} = 1 - 2K_{\mathrm{u}}/\mu_0 M_{\mathrm{s}}$, $K_{\mathrm{u}}$ — the uniaxial anisotropy constant, $\omega = \sqrt{\Omega^2 + m_{\mathrm{e}}^2/4}$. In the case when $\varphi = 90°$, the corresponding dimensionless quantities of $\Omega = 0.5614$, $\omega = 0.72411$, $m_{\mathrm{e}} = 0.9145$ give at 46 K $\alpha = 0.6314$. The magnetostriction constant is calculated to be $(7.9 \pm 0.34) \times 10^{-5}$ at 46 K. This is comparable with previously reported data for the $\mathrm{La_{0.67}Ca_{0.33}MnO_3}$ film on $\mathrm{SrTiO_3}$ substrate [22].

Figure 3 presents the $\lambda_{100}$ magnetostriction constant as a function of temperature. To get insight into the origin of magnetostriction the reduced magnetostriction constant $\lambda_{100}$ is plotted against reduced saturation magnetization (Fig. 4) and one can see that experimental data are well described by Callen and Callen theory [23, 24]:

$$\lambda_{100}(T) = \lambda_{100}(0)\hat{I}_{l+1/2}\{L^{-1}[m(T)]\}, \tag{4}$$

where $m = M_{\mathrm{s}}(T)/M_{\mathrm{s}}(0)$ is the reduced saturation magnetization of the film, $\hat{I}_{l+1/2}$ is the reduced hyperbolic Bessel function, and $L^{-1}$ — the inverse Langevin function. Equation (4) can be well fitted to the experimental points (within the experimental errors) using hyperbolic Bessel function of order $l = 2$ that corresponds to the domination of single ion magnetostriction mechanism in our sample.

Manganites are well known materials characterized by a cooperative JT contribution to the magnetocrystalline energy associated with $\mathrm{Mn^{3+}}$ $(3d^4)$ configuration. The $e_{\mathrm{g}}$ electron of Mn-ion interacts with the lattice due to the JT distortion and the effective hopping is determined by the nearest neighbor spin–spin coupling of $t_{2\mathrm{g}}$ electrons. It is reasonable to make an assumption about a close relation in the origin of JT effect in ferromagnetic manganites and single ion magnetostriction reported here. Both effects are determined by $d$-shells of ions in the crystalline lattice and both effects are accompanied with existence of the orbital moments

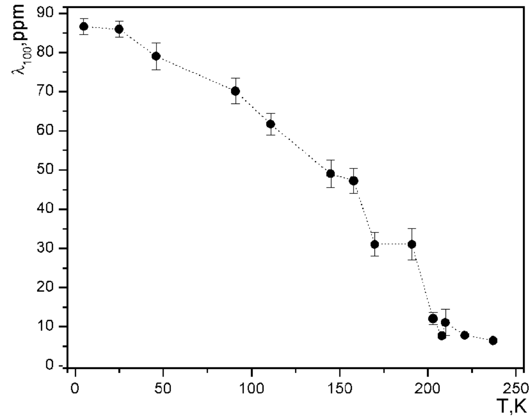

Fig. 3. Magnetostriction constant $\lambda_{100}$ of the $\mathrm{La_{0.67}Ca_{0.33}MnO_3}$ film versus temperature.

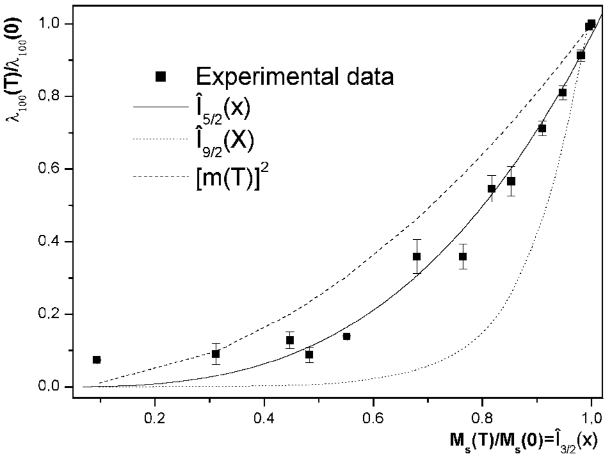

Fig. 4. Reduced magnetostriction constant $\lambda_{100}$ versus reduced saturation magnetization of $La_{0.67}Ca_{0.33}MnO_3$ film (points). Theoretical calculations with $l = 2$ in Eq. (4) (solid line), $\hat{I}_{9/2}(x)$ (dotted line) and $[m(T)]^2$ (dashed line) respectively.

degeneracy. Both effects also involve the strong electron–phonon coupling as well as elastic interactions [25].

## 4. Conclusions

The magnetostriction of the $La_{0.67}Ca_{0.33}MnO_3$ film was determined from the strain dependence of ferromagnetic resonance. This technique for measuring magnetoelastic properties in thin films is straightforward and highly sensitive. The temperature dependence of the magnetostrictive constant $\lambda_{100}$ has been determined over the wide temperature range. Single ion mechanism of crystal field interaction in $La_{0.67}Ca_{0.33}MnO_3$ film was found to be dominating in magnetostriction phenomenon. It is assumed that the Jahn–Teller effect in ferromagnetic manganites and the single ion mechanism of magnetostriction could be originally associated.

## Acknowledgment

This work was supported by the State Committee for Scientific Research grant No. 2P03B04924.

## References

[1] J.H. van Santen, G.H. Jonker, *Physica* **16,** 599 (1950).
[2] C. Zener, *Phys. Rev.* **82,** 403 (1951).
[3] P.G. de Gennes, *Phys. Rev.* **118,** 141 (1960).

[4] R. Von Helmolt, J. Wecker, B. Holzapfel, L. Schultz, K. Samwer, *Phys. Rev. Lett.* **71,** 2331 (1993).

[5] S. Jin, T.H. Tiefel, M. McCormack, R.A. Fastnacht, R. Ramesh, L.H. Chen, *Science* **264,** 413 (1994).

[6] G. Zhao, K. Conder, H. Keller, K.A. Mueller, *Nature* **380,** 676 (1996).

[7] Y. Moritomo, A. Asamitsu, H. Kuwahara, Y. Tokura, *Nature* **380,** 141 (1996).

[8] A.J. Millis, P.B. Littlewood, B.I. Shraiman, *Phys. Rev. Lett.* **74,** 5144 (1995).

[9] E. Dagotto, T. Hotta, A. Moreo, *Phys. Rep.* **344,** 1 (2001).

[10] M. Uehara, S. Mori, C.H. Chen, S.-W. Cheong, *Nature* **399,** 560 (1999).

[11] E.B. Nyeanchi, I.P. Krylov, X.-M. Zhu, N. Jacobs, *Europhys. Lett.* **48,** 228 (1999).

[12] H.-U. Habermeier, F. Razavi, R. Praus, G.M. Gross, *Physica C* **341-348,** 777 (2000).

[13] M. Ali, R. Watts, *J. Magn. Magn. Mater.* **202,** 85 (1999).

[14] B. Lydia, B. Gurney, D. Wilhoit, V. Speriosu, *J. Appl. Phys.* **85,** 5139 (1999).

[15] D. Garcia, J.L. Munoz, F.J. Castano, C. Parados, A. Asenjo, J.M. Garcia, M. Vazquez, *J. Appl. Phys.* **85,** 4809 (1999).

[16] H. Ratajczak, I. Gociańska, *Phys. Status Solidi A* **152,** 523 (1995).

[17] B. Kundys, Yu. Bukhantsev, H. Szymczak, M.R.J. Gibbs, R. Żuberek, *J. Magn. Magn. Mater.* **258-259,** 551 (2003).

[18] R. Zuberek, K. Fronc, A. Szewczyk, H. Szymczak, *J. Magn. Magn. Mater.* **260,** 386 (2003).

[19] E. du Tremolet de Lacheisserie, *Phys. Rev. B* **51,** 15925 (1995).

[20] L. Pust, Z. Frait, *Phys. Status Solidi A* **85,** 179 (1984).

[21] Z. Galazka, *Cryst. Res. Technol.* **34,** 635 (1999).

[22] J. O'Donnell, M.S. Rzchowski, J.N. Eckstein, I. Bozovic, *Appl. Phys. Lett.* **72,** 1775 (1998).

[23] E. Callen, H.B. Callen, *Phys. Rev. A* **139,** 455 (1965).

[24] E. Callen, *J. Appl. Phys.* **39,** 519 (1968).

[25] D.I. Khomskii, K.I. Kugel, *J. Magn. Magn. Mater.* **258,** 251 (2003).