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Temperature dependence of ferromagnetic resonance in double perovskite La_2NiMnO_6 thin films

Sinan KAZAN*[1]

Abstract

The results of the linewidth analysis of ferromagnetic resonance (FMR) spectra of epitaxial La_2NiMnO_6 (LNMO) thin films on (100) oriented $SrTiO_3$ and (110) oriented $NdGaO_3$ substrates at different temperature have been presented. Observed two well resolved FMR signals have been attributed to the coexistence two magnetic phases with different easy and hard axis in the film plane of LNMO. The line shape and the linewidth of the ferromagnetic resonance spectra were measured as a function of temperature. Asymmetry ratio of field derivative FMR absorption curve shows unusual behavior with decreasing temperature. This unusual temperature dependency of the FMR linewidth confirms the presence of magnetoelectric effect in epitaxial LNMO thin films grown by pulse laser deposition technique.

Keywords: Ferromagnetic Resonance, Double perovskite, Magnetodielectric, La_2NiMnO_6

1. INTRODUCTION

Multiferroic materials have been studied intensively due to their wide applications in the fields of magnetoelectronics and spintronics [1–6]. Recently, the semiconducting materials with double perovskite structure has drawn more attention due to the presence of charge and magnetic order simultaneously at near-room temperature [7]. Even though most oxides doped with transition metal ions are antiferromagnet, La_2NiMnO_6 (LNMO) is a ferromagnet due to the superexchange interaction and the nature of this interaction is explained by some rules [7-8]. In LNMO crystal structure, NiO_6 and MnO_6 octahedrons are organized in order to produce double perovskite with a pseudo cubic structure. In LNMO, 3d ions (Ni^{2+} and Mn^{4+}) are distributed randomly at B site of ABO_3 perovskite unit cell resulting Ni-O-Mn, Ni-O-Ni and Mn-O-Mn chemical bonds with different superexchange interaction. Ni-O-Ni and Mn-O-Mn are expected to result antiferromagnetic phase [8-10]. High temperature ferromagnetic order is caused by only Ni-O-Mn bonds.

Magnetic properties of bulk and thin film structured double perovskite sample have been studied intensively for the past decades to understand the nature of interaction mechanism between the magnetic ions in this materials. It is well known that LNMO is a ferromagnetic at near-room temperature compared to

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antiferromagnetic and paramagnetic properties of perovskites $LaMnO_3$ and $LaNiO_3$ [11-18]. It was recently reported that the LNMO exhibits remarkable magnetodielectric effects, which is very useful for spintronic devices, at room temperature [8]. Furthermore, considerable increase in the electronic properties with the external magnetic field have been observed in LNMO at a temperature of 280 K which is the suitable temperature for the magnetoelectric devices [8].

Ferromagnetic LNMO thin films were fabricated epitaxially on different substrates by the pulsed laser deposition (PLD) technique [19]. On the other hand, the fabrication technique, growth temperature, and substrate related mismatch mechanism cause considerable influence on the magnetic and magnetoelectric properties of the thin films.

It was reported that the ferromagnetic resonance (FMR) is a very informative technique for the investigation of structural, magnetic, and transport properties of perovskite materials [19, 25]. By this point of view the investigation of the temperature dependent FMR spectra, as well as the linewidth analysis of the spectra and subsequent study of magnetic anisotropy in these films gains much interest. This paper presents mainly the results of the linewidth analysis of the temperature dependent FMR spectra of double perovskite LNMO thin films growth on different substrates.

2. MATERIALS AND METHODS

A ferromagnetic semiconductor La_2NiMnO_6 (LNMO) double perovskite thin films were grown on (100)-oriented cubic $SrTiO_3$ (STO) and (110)-oriented $NdGaO_3$ (NGO) substrates using pulsed laser deposition (PLD) technique. The substrate temperature and the background oxygen pressure were maintained 750 ^oC and 28-800 mTorr. A KrF excimer laser (248 nm) source was used. After 5000 laser pulses the thickness of deposited films of about 170 nm were measured by small angle X-Ray reflectometry [9].

Ferromagnetic resonance spectra (FMR) were recorded with Bruker EMX electron spin resonance spectrometer which operates at X band frequency (9.8 GHz) at two conventional measurement geometries. The first geometry is known as the in-plane geometry. In this geometry, external dc magnetic field lies in the plane of the sample and magnetic field component of the microwave is always perpendicular to the surface of the film during rotation of the sample. Second one is known as the out of plane geometry. In this geometry, magnetic field component of microwave always lies in the film plane during the rotation of the sample. The characteristics of the FMR spectra used in this study are illustrated in Figure 1. The details of ferromagnetic resonance measurement at different geometries were provided elsewhere [19]. Low temperature FMR measurements were performed by using the continuous He gas flow cryostat of Oxford Instruments between the temperature of 10 K and 300 K.

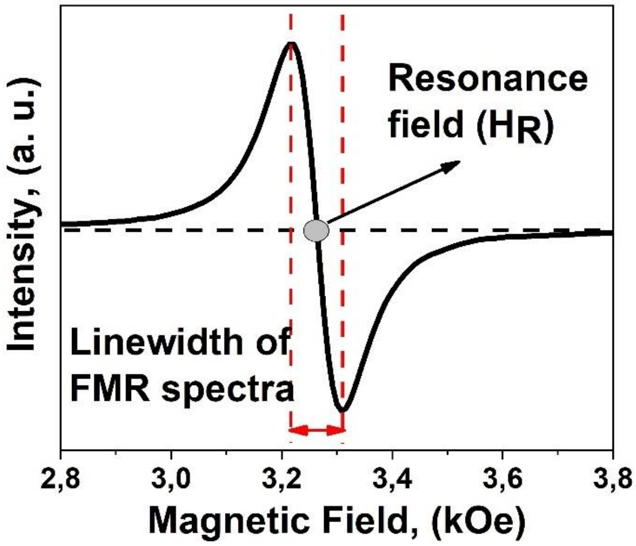

Figure 1 Characteristics of a FMR spectra (Resonance field and linewidth)

3. EXPERIMENTAL RESULTS AND DISCUSSION

In out of plane ferromagnetic resonance (FMR) measurements, two distinct well resolved FMR peaks were observed at room temperature for the La_2NiMnO_6 (LNMO) double perovskite thin films on (100)-oriented $SrTiO_3$ (STO) and (110)-oriented $NdGaO_3$ (NGO) substrates as seen in Figure 2. These two signals have been attributed to the two different magnetic phases [16, 19-20] with different magnetocrystalline anisotropy. Angular variation of these two resonance fields at

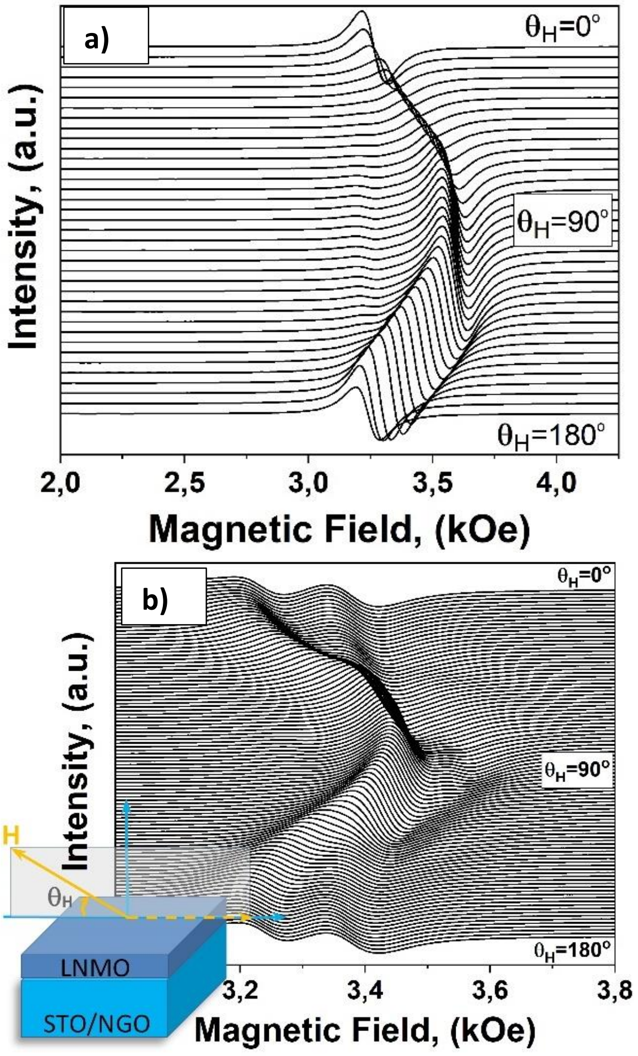

Figure 2 Out of plane FMR spectra of LNMO thin films on a) STO and b) NGO substrate

out of plane FMR spectrum show opposite angular response that is the minimum resonance field of the first mode correspond to the maximum resonance field of second mode for LNMO/NGO in contrast to the LNMO/STO sample. The reason of these two different magnetic phases lies in the nature of magnetic superexchange interaction between different spin configurations of Mn and Ni ions in different compound in LNMO sample. The effective magnetization which is the distance between the maximum and the minimum resonance fields on the spectra for both sample have been considered as low due to the measurement temperature which is close to the Curie temperature. The intensity ratio of the two observed FMR signals which belong to the different phase gives very strong evidence of site ordering and coexistence of two crystallographic form in La_2NiMnO_6 thin film samples. The FMR spectra at in-plane geometry as a mirror of magnetocrystalline anisotropy show four-fold cubic anisotropy for LNMO/STO sample and two-fold uniaxial anisotropy for the LNMO/NGO sample due to the mismatch mechanism between the lattice and the substrate [19]. The peak-to-peak ferromagnetic resonance linewidth denoted as ΔH_{pp} presents the rate of relaxation of the magnetization back to relaxed position when the external static magnetic field is zero. The linewidth of the observed FMR spectra, ΔH_{pp} is affected by two mechanisms. The First one is the intrinsic damping of magnetization (ΔH_{hom}, Gilbert damping) which depends on the frequency of applied microwave and the second one is the magnetic inhomogeneity of the ferromagnetic sample (ΔH_{inhom}) and it is independent from the frequency. The inhomogeneity in ferromagnetic sample may be originated from the distribution of demagnetizing field in the sample, randomly distributed defects, exchange-conductivity mechanism and two magnon scattering from the spatially localized magnetic inhomogeneities [21]. The magnetic inhomogeneities cause FMR line broadening due to the distribution of different local resonance frequencies. The frequency dependent damping term (ΔH_{hom}) manifests itself by the way of dissipation of absorbed microwave energy from the spin system into the lattice generating phonons. Therefore it reflects the anisotropy via spin orbit coupling and generally ΔH_{hom} damping term is explained by the following formula [22],

$$\Delta H_{hom}(\theta,\emptyset) = \frac{2}{\sqrt{3}}\frac{1}{\left|\frac{\partial\omega_R}{\partial H}\right|}\frac{G}{M^2}\left(F_{\theta\theta}+\frac{F_{\emptyset\emptyset}}{sin^2\theta}\right) \approx 1.16\,\frac{G}{\gamma^2 M(T,H)}\omega$$

where θ and $\emptyset$ denotes to the angular position of the magnetization M. $F_{\theta\theta}$ and $F_{\emptyset\emptyset}$ are second derivatives of the total magnetic free energy. G is the Gilbert damping constant at given temperature T. γ is the gyromagnetic ratio. ΔH_{hom} has a minimum (hard axis) at a special direction of magnetization and has a maximum where magnetization varies strongly (easy axis). Also

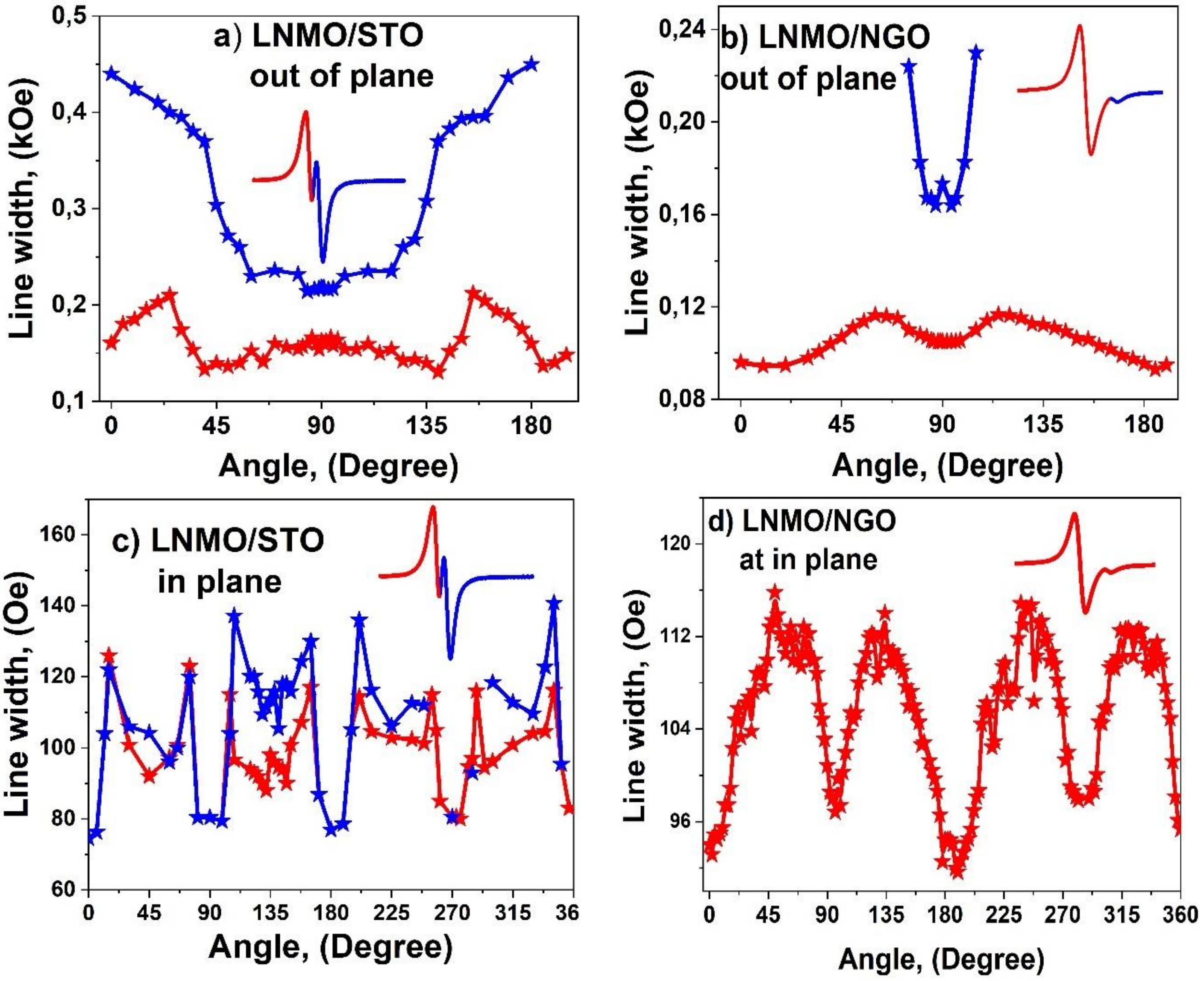

Figure 3 Angular variation of FMR linewidth at out of plane and in-plane geometry at room temperature belong to samples growth on NGO and STO substrate

there is another important contribution to the FMR line broadening due to the exchange conductivity mechanism. Microwave can penetrate a few nm into the surface of conductive ferromagnet producing inhomogeneous dynamic magnetization. The Spatial variation of magnetization exert a torque on M is given as $\left(\frac{2A\gamma}{M^2}\right)\left(\vec{M}x\nabla^2\vec{M}\right)$ [23] where A is the exchange stiffness constant. Therefore this term must be included in equation for the dynamic of magnetization. In conductive ferromagnetic spin system the exchange term causes a net contribution to linewidth. However, if the conductivity of sample is very low or exchange stiffness is very small this contribution is negligible as in our sample system. It is well known that La_2NiMnO_6 (LNMO) double perovskite thin film is ferromagnetic semiconductor and its dielectric constant is very large [8]. The angular variation of the FMR linewidth of the LNMO sample grown on two different substrates at room temperature is shown in Figure 3. Figures 3a and 3b show anisotropic FMR linewidth of two well resolved signals (peak) at out of plane geometry. According to the figure, the linewidth of signals has a minimum at perpendicular orientation where applied static magnetic field is parallel to the film normal as expected. The variation of linewidth near 90

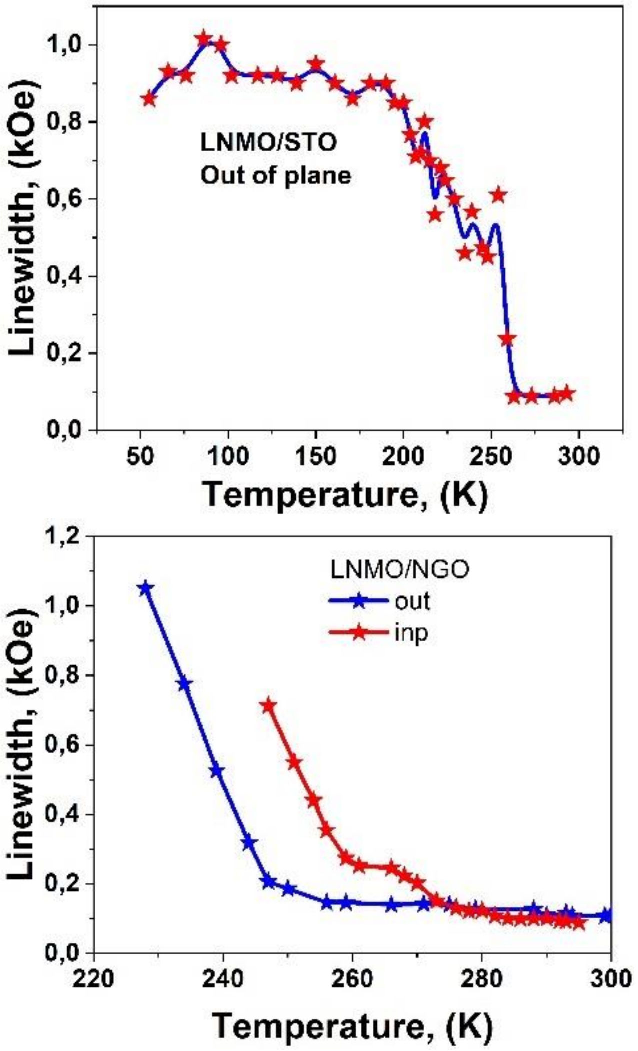

Figure 4 Temperature variation of FMR linewidth at out of plane and in-plane geometry of the sample La_2NiMnO_6 thin film

degree is very large since the measurements have been made at ferromagnetic-paramagnetic phase transition temperature and the magnetic exchange interaction between the cation ions is very small. Figures 3c and 3d show FMR linewidth of two well resolved signals (peak) at in-plane geometry showing four-fold anisotropy. This anisotropic behavior have been attributed to the two magnon scattering [22]. As a result the microscopic origin of the damping mechanism base on the spin orbit coupling and quantitative analyzing to the angular variation of linewidth was very difficult due to the various contribution to the angular dependent linewidth.

The results can be interpreted using the previously reported results of the magnetodielectric effect in La_2NiMnO_6 presented by N. S. Rogada and Co-workes [8] who presented the temperature dependency of the dielectric constant (ε) at the frequency of 10 kHz at external magnetic field. It was observed that the dielectric constant ε has a value of approximately 600 at zero magnetic field and at room temperature. It decreases gradually with decrease of the temperature until it reach 220 K. At this temperature a sharp drop of dielectric constant value exists. When an external magnetic field (0.1 T) was applied, this sharp transition shift to the room temperature region resulting a very large magnetodielectric response of La_2NiMnO_6. This remarkable change in dielectric constant by external magnetic field makes ferromagnetic semiconductor La_2NiMnO_6 (LNMO) a magnetoelectric material suitable for spintronic applications. Analyzing of temperature dependent FMR linewidth of the sample LNMO growth on STO/NGO substrates leads to some interesting results in accordance with the magnetodielectric effect observed in the mentioned temperature dependency of the dielectric constant.

The FMR linewidth of the both samples growth on different substrates at different temperatures are shown in Figure 4. The temperature dependency of linewidths shows similar variation of magnetization with temperature (M-T graph). The asymmetric FMR spectrum which is the field derivative of absorption curve, is shown in Figure 5a. The spectrum was divided into two parts assigned as A and B as shown in Figure 5a for presenting the asymmetry in FMR spectrum. The variation of the parameters of A, B and the asymmetry ratio (A/B) of FMR line shape with the temperature are shown on Figures 5b and 5c. According to the Dyson theory this ratio is related with conductivity of the samples [24].

At in-plane geometry and at room temperature the asymmetry parameter (A/B) is nearly one showing the paramagnetic phase or referring to the charge ordering in LNMO sample. A and B parts of the spectrum show different responses with degrease of the temperature. B part decreases faster than the A part Therefore the ratio of A/B decreases from 1 to 0.6 between the temperatures of 300 K and 265 K. This asymmetric response of the line shape coincides with the magnetodielectric effect given in Ref. [8]. The appearance of asymmetric line shape between the

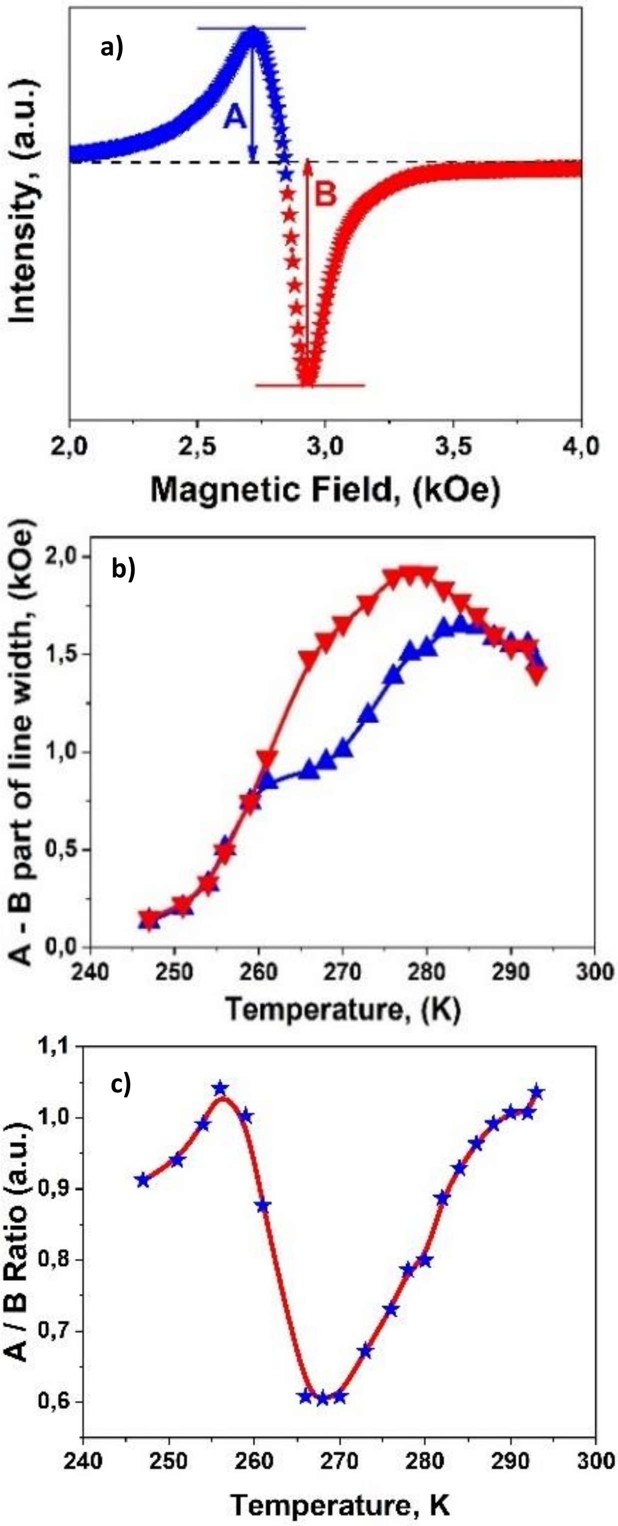

Figure 5 Temperature dependency of the asymmetry parameter at in-plane geometry. a) Asymmetric FMR spectrum of the LNMO/ NGO at 274 K. b) Temperature dependency of A and B part of the spectrum. c) Variation of the A/B ratio with the temperature

temperature of 280 and 250 K in ferromagnetic semiconductor compound La_2NiMnO_6 indicates a coupling between magnetic and electric properties of the films. The magnetoelectric coupling in the form of $\gamma P^2 M^2$ where P and M are the polarization and magnetization respectively and γ is the coupling constant. This coupling causes a net changing in the dielectric susceptibility below the Curie temperature by applying the magnetic field.

4. CONCLUSIONS

In conclusion, the linewidth analysis of the temperature dependent FMR spectra of ferromagnetic La_2NiMnO_6 double perovskite thin films synthesized on STO and NGO substrates by using laser pulse deposition technique has been performed. The observed peculiarities in the angular and temperature dependences of FMR linewidth have been interpreted on the base of the existence of previously reported magnetoelectric effect in this material.

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No conflict of interest or common interest has been declared by the author.

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