Characterization of epitaxial $La_{0.7}Ba_{0.3}MnO_3$ structures using ferromagnetic resonance

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We have compared a single layer of $La_{0.7}Ba_{0.3}MnO_3$ and a trilayer structure of $SrTiO_3/La_{0.7}Ba_{0.3}MnO_3/SrTiO_3$, both grown epitaxially on a $LaAlO_3$ substrate, using information obtained by ferromagnetic resonance (FMR). The trilayer samples have a more uniform magnetization and are not susceptible to environmental degradation. This may be due to the strain relief that the buffer $SrTiO_3$ layer provides for the $La_{0.7}Ba_{0.3}MnO_3$ layer. We have also studied the magnetic homogeneity of the trilayer structure as a function of the deposition temperature. The perpendicular FMR linewidth, $\Gamma_\perp$, shows a clear window in the deposition temperature where the linewidth is <50 Oe. However, the parallel linewidth, $\Gamma_\parallel$, is nearly ten times larger than $\Gamma_\perp$ with only a weak dependence on the deposition temperature. This broadening of the parallel linewidth compared to the perpendicular linewidth can be explained by invoking a local unidirectional anisotropy in the plane of the film. © *1996 American Institute of Physics.* [S0021-8979(96)01416-8]

I. INTRODUCTION

Recently, the Mn oxide perovskite compounds of the form $(R_{1-x}A_xMnO_3)$ where R is (La, Nd, Pr) and A is (Ba, Ca, Sr) have generated much attention due to the colossal magnetoresistance observed in them.[1–5] These compounds have been given so much attention since they could possibly be used for sensor applications,[6,7] especially to increase data storage by increasing the sensitivity of hard disk drive read heads.[8] With this in mind, many studies are being performed on the transport and magnetic properties of these materials. Magnetoresistance (MR) of $>10^6$% in $Nd_{0.7}Sr_{0.3}MnO_\delta$ has been observed at 60 K and 8 T (Ref. 4) and even larger MR has been reported in semiconducting $La_{1-x}Ca_xMnO_3$.[5] However, to be technologically applicable, sensitivities of this level are needed at room temperature and low magnetic fields.[8] As a characterization technique, ferromagnetic resonance (FMR) is proving to be a very useful tool in these studies. FMR is characterized by two parameters, the resonance field, H_R, and the linewidth, Γ. Measurement of these two parameters yields information on the magnetization and high frequency losses, respectively. The advantage of this method is that FMR can reveal inhomogeneous effects not uncovered by other means of examination.[9]

In this paper, we make a comparison between a single layer of $La_{0.7}Ba_{0.3}MnO_3$ (LBMO) and a trilayer structure of $SrTiO_3/LBMO/SrTiO_3$ using information obtained by FMR. We also study the magnetic homogeneity of the trilayer structure as a function of the deposition temperature. The trilayer samples have better uniformity in the magnetization and their microwave response does not change with time. X-ray diffraction spectra of the trilayer samples show a single peak from (00l) LBMO and $SrTiO_3$ (STO) layers indicating that the strain relief that comes from using the buffer STO layer is important for uniform magnetization. For the trilayer structure, we observe a range of deposition temperatures where the perpendicular (magnetic field applied perpendicular to the surface of the thin film) linewidth, $\Gamma_\perp$, is less than 50 Oe, and we observe spin wave resonances obeying Kittel's equation.[10] This suggests that the magnetization is uniform and that this uniformity is dependent on deposition temperature. However, even in the best films the parallel (magnetic field applied parallel to the surface of the thin film) linewidth, $\Gamma_\parallel$, is nearly 10 times larger than $\Gamma_\perp$ and does not appear to depend on the structure of the sample or on the deposition temperature. We propose a model of local unidirectional anisotropy in the plane of the film, presumably caused by the twinning of $LaAlO_3$ (LAO) and the strain-induced effects due to differential thermal expansion, to explain the broadening of $\Gamma_\parallel$ compared to $\Gamma_\perp$.

II. EXPERIMENT

The fabrication of samples was accomplished using a pulsed laser deposition technique. The system included a

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multiple target holder in order to achieve *in situ* deposition. The laser energy density was ~2 J/cm^2. The oxygen pressure during deposition was 400 mTorr and was increased to 300 Torr for cool down after deposition. Two types of samples were examined in the course of this study. The first was a single layer of LBMO and the other was a single layer of LBMO deposited between two layers of STO, creating a trilayer structure. The deposition temperature was then varied between 650 and 800 °C for both the trilayer and the single layer structures. The single layer film was 1100 Å and the trilayer structure consisted of a 1600 Å STO base followed by a 1300 Å layer of LBMO and a 500 Å STO cap. All films were grown on (100) single crystal LAO substrate and no post anneals were performed on the films.

X-ray diffraction was performed on a Siemens four circle x-ray diffractometer. The ion channeling minimum yield measurement was performed using Rutherford backscattering spectroscopy (RBS) with a 1.5 MeV He beam obtained from a 1.7 MV tandem accelerator. The FMR measurements were made using conventional microwave cavity techniques and field modulation at 10 GHz from 77 K to room temperature. The sample was actually placed inside the cavity for the measurements. The magnetic field was applied both perpendicular and parallel to the surface of the sample. The measurement setup in this study is described in an earlier publication.[9]

III. RESULTS/DISCUSSION

Figure 1 shows typical x-ray diffraction spectra for the trilayer samples. X-ray diffraction spectra of the single layer and trilayer samples display an absence of other phase peaks in the θ–2θ scan and the ϕ scan around the (202) LBMO peak, indicating that these samples exhibit single phase and epitaxial growth. In the θ–2θ x-ray spectrum of a trilayer structure, the STO and LBMO (00l) peaks coincide almost exactly indicating an almost perfect lattice match between the STO and the LBMO. The (004) peaks split enough to help determine that the lattice constants of the STO and LBMO layers are what we expect, i.e., d_{STO}=3.92 Å and d_{LBMO}=3.91 Å. The crystallinity of the single layer and trilayer samples has been characterized by RBS channeling as shown in Fig. 2. The channeling minimum yields, $\chi_{\min}$, are in the range of 2.5%–7% at room temperature, indicating high crystalline quality.

Figure 3 shows an example of the FMR spectra for a single layer of LBMO (a) and for a STO/LBMO/STO trilayer (b) at room temperature. It is plotted as the derivative of the power absorption with respect to magnetic field (dP/dH) as a function of applied dc magnetic field. The perpendicular linewidth (the field separation between points of maximum slope), $\Gamma_{\perp}$, for both samples is approximately 50 Oe. In both kinds of structures, a single resonance can sometimes be observed as shown in Fig. 3(a). However, most films show spin wave resonances (SWR) where multiple modes appear, as indicated by arrows in Fig. 3(b) for a trilayer structure. SWR occur in a thin film when a dc field is applied normal to the surface and there is spin pinning at the

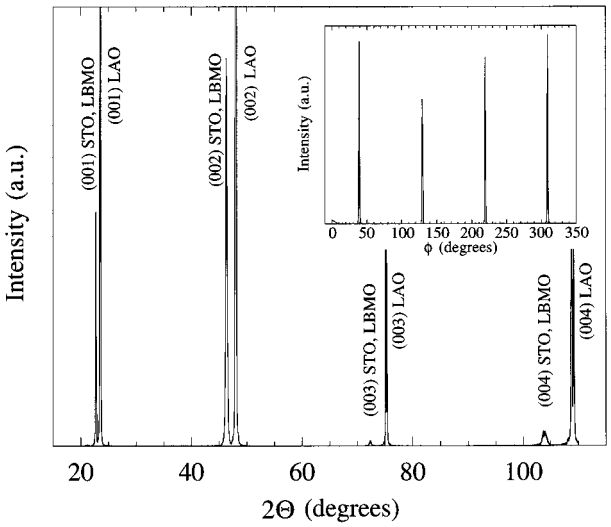

FIG. 1. (a) The x-ray diffraction θ–2θ spectrum of a trilayer sample showing no peaks other than the (00l) of LBMO and STO. (Inset) The ϕ scan around (202) LBMO.

interfaces. Samples which show SWR display either Portis modes (a linear dependence of the resonance field of the nth mode, H_n, on the mode number n) or Kittel modes (H_n linear in n^2). The Portis modes, indicative of a parabolic spatial dependence of the magnetization, are found in both the single layer and trilayer samples. On the other hand, Kittel modes, seen when the magnetization is uniform, are observed only in the trilayer structure. According to the Kittel formula,

$$H_n=\frac{\omega}{\gamma}+4\pi M-\frac{D}{2\pi\gamma h}\left[\frac{n}{\pi L}\right]^2 \quad n \text{ odd},$$

where γ is the gyromagnetic ratio, h is Planck's constant, and D is the spin wave stiffness parameter. By measuring SWR at various temperatures, it is possible to calculate that $D(0)=(150\pm 8)$ meV Å^2.[10] Also, with the value of D computed from the Kittel modes, we estimate the spatial variation of M to be ~20% for the films exhibiting Portis modes.

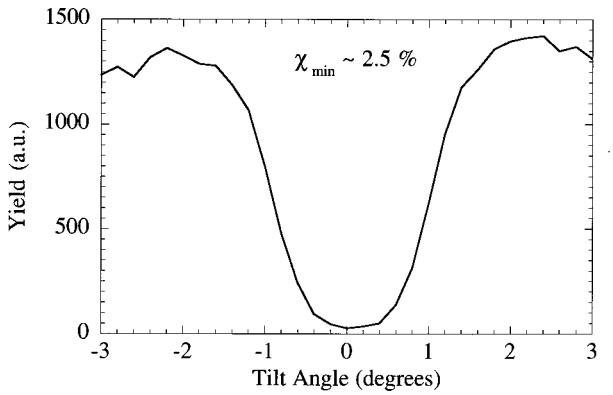

FIG. 2. The angular channeling scan of a trilayer structure at room temperature done by Rutherford backscattering spectroscopy showing a minimum yield of 2.5%.

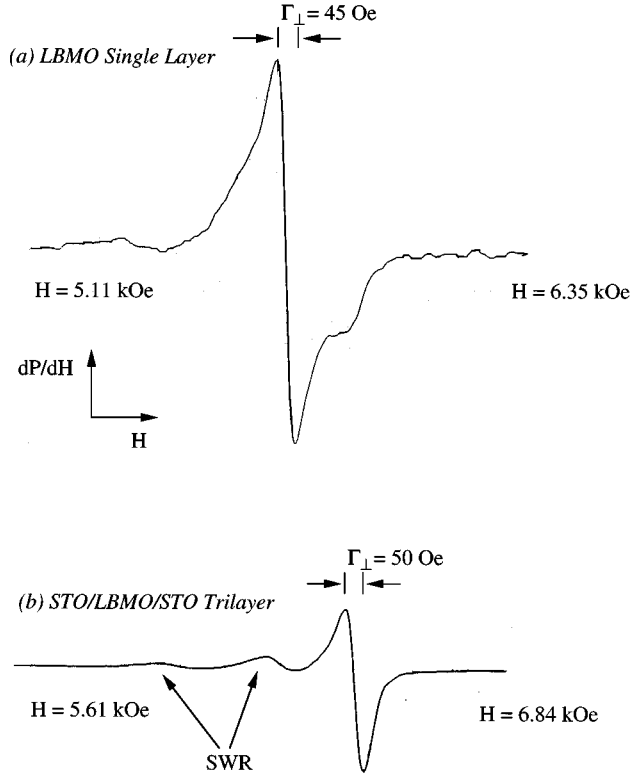

FIG. 3. The output signal for FMR measurements as dP/dH vs H: (a) for a single layer of LBMO and (b) for a STO/LBMO/STO trilayer. The measurements were performed at room temperature and 10 GHz with the field applied perpendicular to the sample surface.

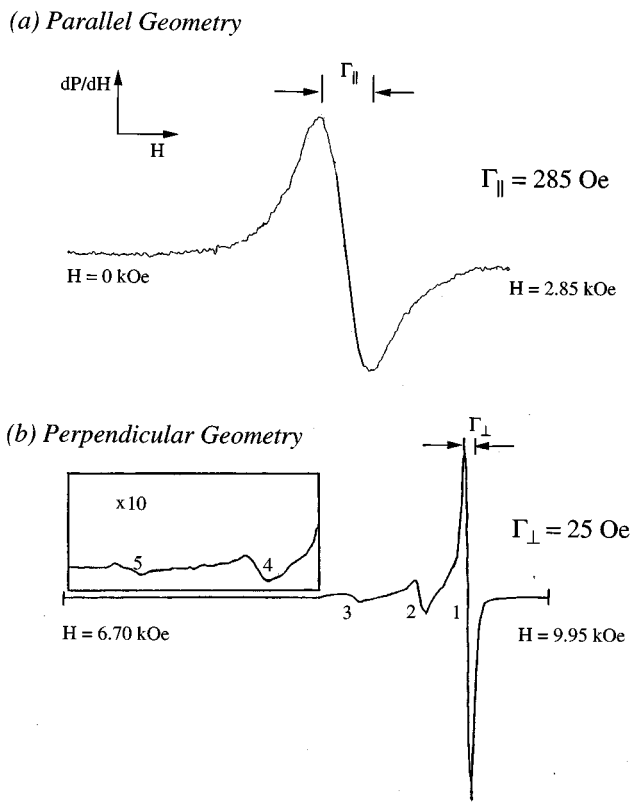

FIG. 4. A plot of FMR signal as dP/dH vs H for STO/LBMO/STO trilayer at 228 K for the magnetic field applied (a) parallel and (b) perpendicular to the sample surface. The spin wave resonance peaks are marked by the mode number for the perpendicular direction.

The constant magnetization, which occurs across the LBMO layer when Kittel modes are seen in the trilayer structure, is the reason for using this trilayer structure rather than a single layer in our studies. As shown in Fig. 1, the LBMO layer grown on the STO buffer has a smaller lattice mismatch (0.3%) compared to that of the single LBMO layer on a LAO substrate (2.8%). Hence, the buffer STO layer eliminates strain in the LBMO layer and, as a consequence, creates a uniform magnetization. Furthermore, the top layer of STO is grown to protect the LBMO layer from environmental degradation. This degradation was observed as a time dependence of the microwave surface resistance in a single LBMO layer without the STO cap. The day after deposition there was a 50% decrease in the microwave surface resistance compared to that measured immediately after deposition. A thin layer of STO deposited on top of the LBMO served to halt this time-varying effect.

As shown in Fig. 4, there is a big difference in the linewidth of the FMR spectra when the magnetic field is applied parallel and perpendicular to the sample surface. The broadening of the FMR linewidth for the parallel direction is observed in all samples independent of the structure. A possible model for this occurrence is given next.

There are several important observations that should be kept in mind. The values of $4\pi M_{\text{eff}}$ determined from the resonance fields, $H_{\parallel,R}$ and $H_{\perp,R(n=1)}$, using $g=2$, agree with one another as well as with the dc superconducting quantum interference device (SQUID) data. Also, $H_{\parallel,R}$ and $\Gamma_{\parallel}$ are isotropic for field rotation in the film plane. Thus, when averaged over the entire film, the anisotropy torques are negligible. However, if the film consists of a number of regions with local anisotropy energies whose symmetry axes are

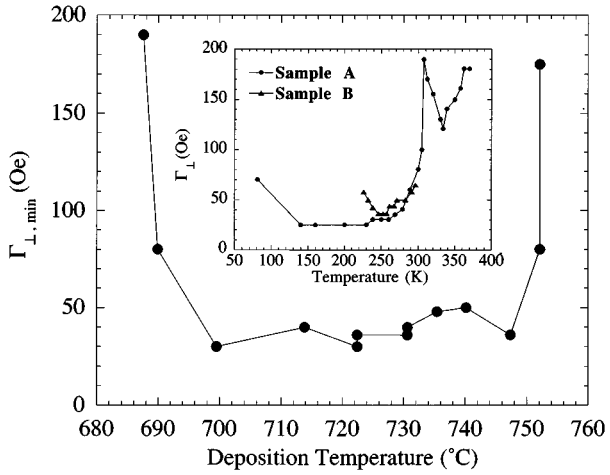

FIG. 5. (a) The deposition temperature dependence of the minimum perpendicular ($\Gamma_{\perp,\min}$) linewidth. (Inset) The temperature dependence of the perpendicular linewidth ($\Gamma_{\perp}$) for two different trilayer samples.

spread uniformly over all possible orientations within the film plane, the parallel linewidth may be greatly enhanced. This will still preserve the anisotropy of $H_{\parallel,R}$ and $\Gamma_{\parallel}$.

Thus, consider a unidirectional anisotropy $K\cos\phi$, where ϕ is the inclination of the local symmetry axis with respect to the applied field, and assume that all values of ϕ are equally likely. This will leave the perpendicular resonance unaffected. To model the parallel spectrum, we also assume that the anisotropy fields, K/M, have a flat distribution between 0 and some maximum value, $(K/M)_{\max}$. Using the Landau–Lifshitz–Gilbert[9] equation of motion we can write for the FMR signal in the parallel configuration:

$$P(H)\approx\int_0^{K_{\max}}\int_0^{2\pi}\frac{\left[\frac{\omega}{\gamma}\right]^2+\left[H+\left(\frac{K'}{M}\right)\cos\phi\right]\left[H+\left(\frac{K'}{M}\right)\cos\phi+4\pi M\right]d\phi dK'}{\left[\frac{\omega}{\gamma}\right]^2-\left[H+\left(\frac{K'}{M}\right)\cos\phi\right]\left[H+\left(\frac{K'}{M}\right)\cos\phi+4\pi M\right]+\Gamma_0^2\left[\frac{\omega}{\gamma}\right]^2},$$

where ω is the angular frequency, γ the gyromagnetic ratio, and Γ_0 the intrinsic linewidth. By using numbers appropriate for LBMO and 10 GHz, i.e., $\omega/\gamma=3500$ Oe, $M=200$ G, and $\Gamma_0=50$ Oe, this equation is sufficient to explain all of the observed features with a relatively small value for $(K/M)_{\max}$ of 100 Oe. Considering the occurrence of twinning in LAO and the strain-induced effects due to differential thermal expansions, the existence of an anisotropy of this magnitude appears reasonable.

Since the films studied here cover a wide variety, that is, some exhibit Kittel modes while others show only single lines, the FMR linewidth becomes the most telling criterion for fixing the quality of a film. However, even in the best films the effects of inhomogeneities still exist. As shown in the inset of Fig. 5 for two trilayer samples, this is revealed by the temperature dependence of the perpendicular ($\Gamma_{\perp}$) linewidth. The peak in $\Gamma_{\perp}$ at around 310 K originates from the fact that the entire film does not have a single transition temperature.[11] The increase in $\Gamma_{\perp}$ at low temperatures is most likely due to a residual nonuniformity in the magnetization.[11] The two samples shown in the inset of Fig. 5 have almost the same minimum $\Gamma_{\perp}$. However, the range of temperature where $\Gamma_{\perp}$ is relatively independent of T is quite different, indicative of the different levels of uniformity.

Figure 5 shows the deposition temperature dependence of the perpendicular ($\Gamma_{\perp}$) linewidth at its minimum value. A definitive window of deposition temperatures where $\Gamma_{\perp,\min}$ is less than 50 Oe has been found. This is the region of interest since the narrower linewidths indicate samples with more uniform magnetization. This direct relationship between the deposition temperature and the perpendicular linewidth suggests that the deposition temperature is a good control factor for obtaining magnetic uniformity in the sample. However, the room temperature parallel linewidth exhibits only a weak dependence on the deposition temperature and is nearly ten times as wide as the perpendicular linewidth. Therefore, something else must be done to align the local anisotropy axes in the plane of the film, resulting in an improvement in the parallel linewidth.

The dependence of the resistivity on temperature was measured using a four probe method for the samples in the deposition temperature sweep. The peak resistivity occurs between 325 and 335 K and the peak temperature has no special relationship with $\Gamma_{\perp}$ and $\Gamma_{\parallel}$. However, the peak resistivity is in the range 10–19 mΩ cm and has a similar relationship as $\Gamma_{\parallel}$ with the deposition temperature.

IV. CONCLUSION

In summary, we have achieved considerable improvement in magnetic homogeneity using a strain relieved, epitaxial STO/LBMO/STO trilayer structure. We observe a range of deposition temperatures for the trilayer structure where $\Gamma_{\perp}$ is less than 50 Oe, suggesting that uniformity in the magnetization is sensitive to the deposition temperature. However, $\Gamma_{\parallel}$ is about ten times larger than $\Gamma_{\perp}$ and exhibits only a weak dependence on deposition temperature, suggesting the important role played by a distribution of strains in the film plane. The broadening of $\Gamma_{\parallel}$ compared to $\Gamma_{\perp}$ has been explained by invoking a local unidirectional anisotropy whose axis is taken to be distributed uniformly in the film plane. As noted previously,[11,12] the FMR linewidth is a more exacting test of inhomogeneity than any of the conventional structural studies, such as x-ray linewidths, RBS channeling, etc. Unfortunately, one cannot use it to obtain unequivocal quantitative information as an inhomogeneously broadened line can arise from a number of sources.

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