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Thickness dependent interfacial magnetic coupling in [La_2NiMnO_6/$LaMnO_3$] multilayers

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ABSTRACT

In the present work, interfacial magnetic exchange coupling at FM/AFM interface has been studied by varying the thickness of AFM layer ($LaMnO_3$) in $(La_2NiMnO_6/LaMnO_3)_{15}$ multilayer thin film based system. In multilayer thin films, the thickness of LMO was varied from 30 to 50 Å, while the thickness of LNMO was kept constant at 100 Å. Thin films of LNMO, LMO and LNMO/LMO multilayers have been deposited by pulsed laser deposition technique on (0 0 1) $LaAlO_3$ substrate. The thin films have been studied for their structural and magnetic properties. XRD analysis reveals the c-axis epitaxial growth of LNMO, LMO and their multilayer thin films. Exchange bias (EB) effect has been observed in the field cooled hysteresis loops of multilayer thin films and the interaction between FM and AFM spins at the interface is responsible for the observed effect. EB measurements reveal that the thickness variation influences the interfacial interaction between two layers. The EB increases with increasing AFM film thickness in multilayer thin film samples and maximum EB (~740 Oe) is observed for sample with 50 Å thickness of AFM layer. Temperature dependence of EB and training effect measurements have also been performed to confirm the EB in the sample.

1. Introduction

The interfaces between dissimilar complex oxide materials have been widely studied over the past few decades due to the coupling of the charge, orbital, lattice and spin degrees of freedom of electrons [1,2]. The advancement of techniques for fabricating and characterizing oxide thin films has given a direction for the study of the interfacial effect between perovskite oxides [3,4]. Perovskite based heterostructures composed of magnetically active materials are considering much more attention with discoveries of ferromagnetism at interfaces between two antiferromagnets or even between an antiferromagnet and a paramagnet [5–8]. Thus, it can be realized that epitaxial heterostructures of different complex oxides may offer excellent opportunities to study the rich and fascinating magnetic phenomena at the interface due to the competing interactions. One of the most interesting interfacial phenomenon is the exchange bias (EB) in the heterostructures of ferromagnetic (FM) and antiferromagnetic (AFM) layers [9,10]. When a FM/AFM interface based system is cooled through the Neel temperature (T_N) of antiferromagnetic material ($T_N < T_C$, T_C is the curie temperature of the ferromagnetic material), the hysteresis loop is now shifted away from the origin [11]. The EB effect manifests itself as a shift in field cooled hysteresis loop along the magnetic field axis and the additional unidirectional anisotropy is attributed to the exchange interaction between the FM and AFM spins at the interface [12,13]. A unidirectional pinning or anisotropy of the magnetization of ferromagnet is observed at the FM/AFM interface due to the effect of EB and can be utilized to control the magnetization in a magnetic reference layer in a spin valve structure or a magnetic tunnel junction (MTJ), which consists of two FM layers separated by a nonmagnetic material (spin valve) or an insulator (MTJ)[14,15].

Among perovskites, ABO_3 type $LaMnO_3$ (LMO) compound has attracted much more attention for being a building block in some heterostructures exhibiting fascinating phenomena such as EB in $LaNiO_3$-$LaMnO_3$ superlattices, $LaMnO_3/SrMnO_3$ heterostructures and observation of ferromagnetism (FM) at the interface of $LaMnO_3/SrTiO_3$ [16–19]. Additionally, bulk LMO is a Mott insulator and an antiferromagnet with a Neel temperature of 140 K. Nevertheless, when grow as a thin film, the magnetic behaviour of LMO is rather conflicting as LMO exhibits ferromagnetic behaviour with a curie temperature $T_C \sim 200$ K [20] as well as it shows antiferromagnetic behaviour with a Neel temperature of $T_N \sim 131$ K, depending on the synthesis conditions [21]. It has been found that in oxygen environment, the epitaxial strain stabilizes the

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orthorhombic phase with slightly less Mn^{4+} in LMO and leaves the film insulating but on the AFM side of the AFM/FM boundary [21]. In recent years, double perovskite ($A_2BB'O_6$, A = alkaline earth and B,B' = transition metal oxides) R_2NiMnO_6 (RNMO, R = Rare earth) compounds are of considerable interest due to their rich physics and possible application in spintronics [12,22–27]. Among the double perovskites, La_2NiMnO_6 (LNMO) is well studied due to its near room temperature ferromagnetic transition ($T_C \sim 280$ K) accompanied by magnetoresistance and magnetodielectric effects [22,28,29]. A lot of work on epitaxial growth of LNMO thin film and its magnetic properties is already reported [23,30–32]. Both the LNMO and LMO exhibit perovskite structure with lattice parameters of about 3.88 Å and 3.98 Å. The close lattice parameters of FM (LNMO) and AFM (LMO) layers allow the growth of epitaxial heterostructures with almost atomically perfect interfaces. It has been observed that the EB is inversely proportional to the thickness of FM layer in all the studied systems [11]. However, many discrepancies have been observed in EB behaviour with antiferromagnetic thickness variation. Generally, EB is independent of thickness for thick antiferromagnetic layers and reduces with decreasing antiferromagnetic layer thickness and becomes zero for thin enough AFM layer (a few Å) [11,33]. In some cases, EB decreases with larger AFM thicknesses [34,35]. On the other hand, EB also shows a peak like behaviour with increasing AFM layer thickness in some systems [36,37]. Additionally, the presence of nonmagnetic defects in the AFM layer can enhance the EB [38]. It has also been noticed that AFM domain structure may also affect EB, if the thickness becomes comparable to the AFM domain wall size [11]. Thus, it would be interesting to see the trend and magnitude of EB with thickness variation of antiferromagnetic $LaMnO_3$ layer.

In the present study, we have fabricated the high quality LMO, LNMO and their $[LNMO/LMO]_{15}$ multilayer thin films. We have studied the EB effect in $[LNMO/LMO]_{15}$ multilayers with varying LMO thickness, while the thickness of LNMO was kept constant. This effect is also confirmed by performing the temperature dependence of EB and training effect measurements. To the best of our knowledge, EB effect in LNMO/LMO multilayer thin films have not been reported yet.

2. Experimental

LNMO, LMO and $[LNMO/LMO]_{15}$ multilayer thin films were fabricated on single crystalline (0 0 1) $LaAlO_3$ (LAO) substrate using multitarget pulsed laser deposition (PLD) technique. To ablate sintered pellet targets, a pulsed laser beam generated by a KrF excimer laser at a wavelength of 248 nm and pulse duration of 25 ns was introduced into the deposition chamber and focused onto the target surface. Prior to each deposition, the targets were preablated for 1 min in order to ascertain the same state of the target in every deposition. The substrate temperature, oxygen pressure, laser fluence, repetition rate and target-substrate distance were held constant for all the deposited films. Epitaxial thin films were grown at fixed substrate temperature and pressure of 700 °C and 10 mTorr, respectively. The laser repetition rate of 5 Hz was used for the ablation. The target to substrate distance was fixed at 50 mm for all the samples. The laser energy density on the target surface was $\sim$2 J/cm^2. The thickness of the films was measured using cross-sectional FE-SEM and was kept constant for LNMO and LMO thin films at approx. 190 nm. In case of multilayer, first layer of LMO with different thickness (30, 40 and 50 Å) was deposited on the substrate followed by LNMO layer with constant thickness (100 Å). The fifteen such bi-layers were deposited so the total thickness of multilayers were approx. 195 nm, 210 nm and 225 nm, respectively. The crystallinity and epitaxial quality of thin films were investigated in θ-2θ geometry using X-ray diffractometer (Bruker D8 advance) of Cu K_α (1.54 °A) radiation. Magnetic properties of the samples were characterized using superconducting quantum interference device magnetometer (MPMS XL Evercool, Quantum Design). For all the magnetic measurements, the samples were mounted parallel to the direction of applied magnetic field.

3. Results and discussion

3.1. Structural properties

We have deposited the two single layer thin film samples (LMO and LNMO) and three multilayer samples [LNMO (100 Å)/LMO (x Å)]$_{15}$ with varying thickness of AFM layer LMO (x = 30, 40 and 50 Å). The three multilayer samples with different LMO layer thickness x = 30, 40 and 50 Å, are represented as samples S1, S2, and S3, respectively. For each deposition, the number of laser shots was varied to obtain the LMO layer with different thickness. The cross-sectional FE-SEM images of both single layer thin films of LMO ($\sim$190 nm) and LNMO ($\sim$190 nm) are shown in Fig. 1 (a) and (b), respectively. Using the obtained thickness and the applied number of laser shots, we have determined the growth rate of both the materials and then deposited the multilayer of these materials based on the same deposition parameters. In order to confirm the growth of LNMO on LMO layer, we have taken the cross-sectional FE-SEM image (Fig. 1(c)) for a bilayer (LNMO/LMO) sample, which shows that the LNMO can be grown easily on LMO layer. X-ray diffraction patterns of LMO, LNMO and multilayer thin film S3 [LNMO (100 Å)/LMO (50 Å)]$_{15}$ deposited on LAO substrate at fixed deposition temperature 700 °C and pressure of 10 mTorr are shown in Fig. 2. It is clear from Fig. 2 that all the samples show only the diffraction peaks corresponding to (0 0 *l*) reflections. No other peaks belonging to any impurity phase are present in the XRD pattern which reveals that the films are epitaxial and grown coherently in the pseudocubic 0 0 *l*-direction. This implies that films are highly oriented along c-axis.

3.2. Magnetic properties

We have measured the zero field cooled $M-H$ loop of LNMO thin film at 5 K within the range of −50 kOe to +50 kOe applied magnetic field in a direction parallel to the surface of the film. The $M-H$ curve of LNMO is shown in Fig. 3 after subtracting the substrate magnetization. A clear hysteresis curve suggests the ferromagnetic behaviour of LNMO thin film. LNMO film shows a saturation magnetization of $\sim$4.8 μ_B/f.u., which approaches the theoretical value of 5 μ_B/f.u. as expected for completely ordered Ni^{2+}/Mn^{4+} ferromagnetic configuration [30,31]. As the cation ordering in the double perovskites is the B-site ordering. If a double perovskite is not properly B-site ordered, it means that antisite disorders are present in the system. Experimentally, the presence of the antisite disorders is well evidenced by the fact that the saturation magnetization (M_S) of the LNMO synthesized by different methods is found to be always smaller than the expected M_S of 5 μ_B/f.u. [39–41]. However, in the present case, we found M_S ($\sim$4.8 μ_B/f.u.) close to the M_S of B-site ordered LNMO [30,31]. Additionally, we observed that $M-H$ loop shows a saturation behaviour even at below ±50 kOe. Such type of behaviour of $M-H$ loop in the parallel direction of magnetic field clearly shows the presence of in-plane magnetic anisotropy in LNMO thin film [27]. The temperature dependent magnetization ($M-T$) measurements under zero field cooled (ZFC) and field cooled (FC) conditions were performed in a temperature range of 5–300 K in presence of applied magnetic field of 100 Oe. Prior to the measurements, the sample was cooled from 300 K to 5 K in the absence of magnetic field. Then, the exter-