Tuning of magnetic properties for epitaxial Y_2NiMnO_6 thin film: substrate is crucial

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ABSTRACT

The effects of epitaxial strain induced by lattice mismatch and substrate type on the structure and magnetic properties of Y_2NiMnO_6 thin films have been systematically investigated. Y_2NiMnO_6 thin films grown on (001)-oriented LaAlO_3 (LAO), $(\text{La,Sr})(\text{Al,Ta})\text{O}_3$ (LSAT), and SrTiO_3 (STO) substrates with varying film thickness are obtained by a simple polymer assisted deposition method. X-ray diffraction and Raman scattering observations indicate that the single-phase epitaxial films are successfully obtained. By magnetic measurements, it is found that all the films show an obvious ferromagnetic transition with a lower transition temperatures (T_c) than that of the Y_2NiMnO_6 bulk. With the biaxial tensile strain decreasing or the film thickness increasing, the T_c of the film increases. Besides, due to the different strain states of the films and the different surface migrations of the substrates, the structure and magnetic properties show a strong dependence on the type of substrate. It is suggested that the biaxial tensile strain and substrate type have crucial effects on the structure, magnetic properties and the related T_c of the thin film, which can be utilized to engineer the magnetic properties of the films and the related ferroelectricity.

Keywords: Y_2NiMnO_6 thin film, Polymer assisted deposition, Epitaxial strain, Substrate type

1. INTRODUCTION

Recently, magnetoelectric (ME) multiferroics, simultaneously possess ferromagnetic and ferroelectric order, have attracted much attention due to their potential applications in memory and magnetic storage devices [1,2]. The ME materials with magnetically induced ferroelectric order are particularly interesting due to the electric control of magnetism or vice versa. Su et al have reported magnetism driven ferroelectricity in Y_2NiMnO_6 , which is confirmed by the appearance of ferroelectric polarization accompanying by the magnetic phase transition and the strong magnetoelectric (ME) effect

[3]. Therefore, controlling of the magnetism may be an effective way to tune the ferroelectricity in Y_2NiMnO_6 and the related double perovskites. On the other hand, for potential device applications, the ferroelectric films play an even more important role. It has been reported that the electronic and magnetic properties can be adjusted by structural modifications [4-6]. The modifications in the Ni-O-Mn bond length and bond angle can directly influence the intensity of superexchange interaction. The structural modifications can be easily induced by the film-substrate lattice mismatch. Thus, it is feasible to manipulate physical properties of materials through rigid compression or extension of their chemical bonds via epitaxial strain. Moreover, because of the strong ME in Y_2NiMnO_6 [3], it is plausible to expect that the epitaxial strain is an effective way to tune the magnetism driven ferroelectricity in this system. These have motivated our present study on magnetic property of Y_2NiMnO_6 film under epitaxial strain.

In addition, for further application in industry, development of a simple and low-cost technology to prepare the uniform and thickness-controllable large area film material is very important. In this work, the Y_2NiMnO_6 (YNMO) thin epitaxial films are grown via a simple polymer assisted deposition (PAD) method [7-12]. The PAD technique, as a surfactant assisted sol-gel process, is suitable for film deposition by spin and dip coating. Therefore, it is suitable for coatings over large areas and complex-shaped objects avoiding the use of high-vacuum chambers in the film deposition process [13]. Besides, the desired thickness of the film can be easily controlled by the solution concentration, the molecular weight of the soluble polymer and the speed of coating [14-16]. By X-ray diffraction and Raman scattering observations, it is conformed that the single-phase epitaxial YNMO films are obtained successfully at a related lower annealing temperature compared to the previous reports [17,18]. The effects of film thickness and substrate type on the structure and magnetic properties of YNMO thin films are discussed in details. It is found that the magnetic transition temperature and low-temperature moment are increased with biaxial tensile strain reduced. In addition, the choosing of substrate provides another way to engineer the property of film.

2. EXPERIMENTAL

A precursor solution containing metal ions was prepared as follows. High purity (>99.99%) metal salts $\text{Y}(\text{NO}_3)_3 \cdot 6\text{H}_2\text{O}$ (0.5mmol) and $\text{Ni}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ (0.25mmol) and $\text{Mn}(\text{NO}_3)_2$ (0.25mmol, AR, 50 wt.% in H_2O) were mixed and dissolved in 5 mL of H_2O . Then 0.2922 g of EDTA (Ethylene Diamine

Tetraacetic Acid) and 0.2922 g of PEI (polyethyleneimine) with molecular weight of 70,000 were dissolved in 10 mL of H₂O and stirred at room temperature for 1h. After 1 h chelating-reaction, 10 mL of PEI and EDTA transparent aqueous solution was slowly added into the 5 mL of nitrate solution by a dropper. Finally, the mixed solution was stirred by a magnetic stirring apparatus for 10 h at room temperature, and then concentrated to ~5 mL in a 60 °C oil bath.

The polymeric films were deposited on single-crystal substrates with different lattice parameters, including (001) LaAlO₃ (LAO, $a_{\text{LAO}}=3.792 \text{ \AA}$), (001) (La,Sr)(Al,Ta)O₃ (LSAT, $a_{\text{LSAT}}=3.868 \text{ \AA}$), and (001) SrTiO₃ (STO, $a_{\text{STO}}=3.905 \text{ \AA}$) by a spin coating technique. The precursor solutions were deposited on single-crystal substrate at 3000 rpm (revolutions per minute), 5000 rpm, and 7000 rpm over 50 s to obtain YNMO films with different thickness. Then the coated substrates were placed in a muffle furnace. A low ramp rate of 1 °C min⁻¹ was used from room temperature to 700 °C to make sure the water evaporated and polymers burned up to avoid the formation of voids in the bulk of the films. The samples were then rapidly heated (5 °C min⁻¹) to a temperature of 900 °C. After 2 h heat treatment, the films were cooled down to room temperature at 1 °C min⁻¹. The morphological properties of the deposited YNMO samples were observed by the atomic force microscopy (AFM, Auto Probe Pc; in contact mode, with low stress silicon nitride tip of less than 200 Å radius). All the images were obtained with a scanning area of 2.0µm×2.0µm. A Philip's X'Pert X-ray diffraction system was used to characterize the crystallization and epitaxial quality of YNMO films. Besides, the morphological characteristics and thickness of YNMO films is also obtained by an FEI Sirion 200 field emission scanning electron microscope (SEM). The Raman backward scattering spectrum was measured on a JY LABRAMHR Raman spectroscopy, with an Ar⁺ (514 nm line) laser as an excitation light source. In addition, the temperature-dependent magnetization (M-T) curve under a constant applied magnetic field was measured via a Quantum Design MPMS-7 superconducting quantum interference device (SQUID) magnetometer. The field-cooled (FC) magnetization curve was measured with an external magnetic field of H = 100 Oe applied parallel to the film surface in the temperature range 10 K-300 K.

3. RESULT AND DISCUSSION

The morphological characteristics of the YNMO epitaxial thin films deposited on STO, LSAT, and LAO substrates at 5000 rpm are characterized via a SEM as shown in Fig. 1. It is found that the thickness of YNMO films obtained at 5000 rpm is about 50nm (as a typical example, the

cross-sectional image of YNMO/STO is shown in Fig. 1d). Besides, the surface morphologies and roughness are found to be different for the YNMO films grown on different substrates. To further confirm this result, an atomic force microscope (AFM) is involved to observe the surface morphology, which is a very effective method to explore the surface roughness in nanometer scale and surface morphology of thin films. The two-dimensional (2D) and three-dimensional (3D) surface topography of the 50nm YNMO thin films deposited on LAO, LSAT, and STO substrates are presented in Fig. 2, which show obvious 3D island topography. However, the obvious difference of micrographs can be found for YNMO films with different substrates, which is consistent with the results of SEM observation. Compared with those of the YNMO films on LSAT and STO substrate, larger grains and more holes are shown in the surface of the film on LAO substrate. The average roughness (R_a) of the surface for the YNMO films on LAO, LSAT, and STO substrates are 25.3nm, 20.1nm and 19.0nm, respectively. It is surprised to find that the average R_a of YNMO films on LSAT and STO substrates, which have larger film-substrate lattice mismatch, are smaller than that of YNMO/LAO film. This may indicate that the epitaxial film grown on cubic STO and LSAT substrates is superior to that on orthorhombic LAO substrate. Furthermore, the substrate effect may also be responsible for this result [19,20]. In this work, the different 3D morphology and average roughness indicate that the mode of substrate-surface migration is dependent on the substrate type.

To verify the crystallinity and epitaxial quality of YNMO films, the X-ray diffraction (XRD) $\theta/2\theta$ scans and ω scans (rocking curve) are carried out. The XRD patterns for the 50-nm YNMO films grown on (001)-oriented LAO, LSAT, and STO substrates are shown in Fig. 3. It can be found that only (00 l) peaks for YNMO films are observed along with the (00 l) peaks of the substrates without any undesirable peak from XRD patterns, indicating that the YNMO films have a preferential c-axis orientation. The FWHM (full width at half maximum) of the rocking curves for (002) peaks of the films are less than 0.3 °, which indicates a high degree of crystallite orientation for all the YNMO films.

Figures 4(a)-(c) show the substrate dependence of reciprocal space mappings around the (103) reflection for the YNMO thin films deposited on STO, LSAT, and LAO substrates, respectively. The in-plane lattice constant of the YNMO/LAO film matches with that of LAO substrate, confirming coherent epitaxial growth of the YNMO/LAO film. In contrast, as can be seen from Fig. 4(a) and Fig. 4(b), the peak position of (103) reflection of the YNMO films has been slightly shifted from those of

the STO and LSAT substrates along the Q_{100} axis. In addition, the full width at half maximum of the peak for the YNMO film on LAO is apparently smaller than those of the films on STO and LSAT, which is consistent with the full width at half maximum of rocking curves. Since the similarity between the lattice parameters of the pseudocubic bulk YNMO and LAO substrate, epitaxial YNMO thin films are coherently grown on LAO substrate. Although the YNMO films are also grown epitaxially on the LSAT and STO substrates, the large lattice mismatch between the films and the substrates will cause relaxation in the YNMO structure. The similar results have been reported by Hashisaka et al in the $\text{La}_2\text{NiMnO}_6$ film system [26]. In $\text{La}_2\text{NiMnO}_6$ film system [26], since the lattice parameter of pseudocubic bulk $\text{La}_2\text{NiMnO}_6$ is close to lattice parameters of STO and LSAT substrates, epitaxial $\text{La}_2\text{NiMnO}_6$ thin films are coherently grown on these substrates. However, the large lattice mismatch between the $\text{La}_2\text{NiMnO}_6$ film and LAO substrate causes relaxation in the $\text{La}_2\text{NiMnO}_6/\text{LAO}$ structure, which can be seen from the slight shift between $\text{La}_2\text{NiMnO}_6$ film and LAO substrate [26].

Raman spectroscopy is an excellent tool to obtain the information on the local structure of the different microphases or macrophases present simultaneously in many manganites. To detect the structural characteristics of the YNMO films, the Raman spectra for the YNMO films on different substrates were measured at room temperature, as shown in Fig. 5. It is found that the Raman spectra are dominated by two main peaks at around 512 cm^{-1} and 649 cm^{-1} , which can be assigned to the antisymmetric stretching (AS)/bending and symmetric stretching (S) vibrations of $\text{Ni}(\text{Mn})\text{O}_6$ octahedra, respectively [21]. Moreover, in addition to these two peaks, two high-frequency scattering peaks are observed at around 1180 cm^{-1} and 1295 cm^{-1} . No obvious difference in peak shape is observed from the spectra, suggesting that all the samples have the same composition in essentially. The results are consistent with the previous report [22] and confirm again that the YNMO films with different substrates are obtained successfully by PAD method.

Besides, the structures of different thickness YNMO films grown on (001)-oriented STO are also investigated. The XRD patterns for 20-, 50-, 80-nm-thickness YNMO films grown on (001)-oriented STO substrate are displayed in Fig. 6. According to Bragg formula, the out-of-plane c-axis lattice parameters (c_{film}) for 80-, 50- and 20-nm-thickness YNMO films obtained from the XRD peak positions are $3.787(1)\text{ \AA}$, $3.784(2)\text{ \AA}$ and $3.780(5)\text{ \AA}$, respectively. Based on the previous report [23], the monoclinic structure (P21/n) YNMO bulk with lattice parameters of $a=5.2285\text{ \AA}$, $b=5.5518\text{ \AA}$, $c=7.4872\text{ \AA}$, and $\beta=89.744^\circ$ can be considered as a pseudocubic perovskite structure corresponding to a

bulk with a parameter of $a \approx 3.787(6) \text{ \AA}$. Therefore, the YNMO ($a_{\text{bulk}} \approx 3.787(6) \text{ \AA}$) films grown on STO ($a_{\text{STO}} = 3.905 \text{ \AA}$) are under biaxial tensile strain in ab plane, resulting in the compressive effect along the c-axis and tetragonal distortion of $\text{MnO}_6/\text{NiO}_6$ regular octahedrons. As a result, the c-axis parameter of films is smaller than that of corresponding bulk material. Besides, the c-axis parameter of the YNMO films shows a continuously increasing trend with increasing the thickness of film, which can be attributed to the lattice relaxation effect. With film thickness increasing, the effect of the biaxial tensile strain will be relaxed, which induces the c-axis parameter increasing. It should be noted that the c-axis lattice parameters of 80-nm YNMO film is close to that of the bulk, which indicates that the biaxial tensile strain is nearly fully relaxed.

The temperature dependent magnetization for YNMO films grown on STO with different thickness are shown in Fig. 7. It can be found that all the YNMO films show an obvious ferromagnetic transition. The Curie temperature (T_c , identified from the minimum in dM/dT) of the films for 80 nm, 50 nm, and 20 nm are determined to be 84 K, 81 K, and 77 K, respectively, which are lower than that of the YNMO bulk ($T_{c\text{bulk}} = 85\text{K}$) [24]. In general, as one typical factor, the film-substrate lattice mismatch can induce epitaxial strain states, sample inhomogeneities and phase separation [14,15], which evidently affect the magnetic properties of the thin films in comparison to their bulk materials. In this work, the T_c of YNMO increases from 77 K to 84 K as film thickness increasing, which reveals that lattice mismatch induced strain may be relieved by increasing film thickness. It is worthy to mention that the $T_c = 84\text{K}$ of 80nm-thickness YNMO/STO film is very close to that of bulk material (85K), which provides another evidence to prove the strain in film is nearly fully relaxed. Due to the lattice relaxation effect, the degree of lattice distortion and/or the amount of the inhomogeneities in YNMO films are expected to decrease as the film thickness increasing. Indeed, the FM transition gets narrower with the YNMO films thickness increasing (see the insets of Fig. 7), which indicates the less inhomogeneities in film by film thickness increasing.

Beside the effect of the film thickness, the substrate type can also evidently affect the strain state of the film. In order to distinguish the difference between film thickness effect and substrate effect, the magnetic properties for YNMO films on different substrates are observed. The temperature-dependent magnetization M (T) curves for YNMO films grown on (001)-oriented STO, LSAT, and LAO substrates are shown in Fig. 8. The ferromagnetic transition temperatures (T_c) of YNMO epitaxial films grown on STO, LSAT, and LAO are estimated to be 81 K, 78 K, and 74 K, respectively. Due to the

different lattice parameters of LAO, LSAT and STO substrates, the films on different substrates are speculated to have different strain states, which may be responsible for the different magnetic behaviors. According to previous studies [25-28], the physical property of the film is closely related to the strain state in film and strain relaxation. It should be noticed that although the film-substrate lattice mismatch of YNMO/STO is large than YNMO/LAO and YNMO/LSAT, the absolute value of out-of-plane strain (can be expressed as $\varepsilon_{zz} = (c_{\text{film}} - a_{\text{bulk}}) / a_{\text{bulk}}$, where c_{film} is the out-of-plane lattice parameter of YNMO film and a_{bulk} is the pseudocubic lattice parameter of bulk YNMO) of films is in ascending order of YNMO/LAO > YNMO/LSAT > YNMO/STO, as shown in Table 1. Moreover, the absolute variation of volume Δ_V ($\Delta_V = (V_{\text{film}} - V_{\text{bulk}}) / V_{\text{bulk}}$; $V = a^2 c$) for YNMO/LAO is larger than that for YNMO/STO, which reveals the large strain in YNMO/LAO. Therefore, it is reasonable to believe that the actual strain in YNMO/STO film is smaller than that of YNMO/LSAT and YNMO/LAO. The tensile/compressive strain will be relaxed with film thickness increasing, and it will be fully relaxed when the film thickness exceeds a critical thickness. It has been reported in $\text{La}_2\text{NiMnO}_6$ thin film [29] that the critical thickness for $\text{La}_2\text{NiMnO}_6/\text{LAO}$ (-2.32%) is between 40 and 80 nm. Because of the similar structure of Y_2NiMnO_6 to that of $\text{La}_2\text{NiMnO}_6$, it is plausible to believe that the biaxial strain (3.10%) is partially or fully relaxed in YNMO/STO but fully strained in YNMO/LAO. Besides, the different T_c of YNMO films can also be due to the difference in substrate-surface migration related to substrate type. The effect of the substrate type on the structure, morphology and property of film has been confirmed by previous works [19,20,27]. In this work, the different morphologies of YNMO films on different substrates, as revealed by Fig. 1 and Fig. 2, also gives an indication of the substrate effect, which have been discussed in previous paragraph. Therefore, the different magnetic properties of the YNMO films grown on different substrates may be attributed to the substrate effect.

4. CONCLUSION

In summary, the double perovskite YNMO epitaxial thin films with different strain states induced by substrates and film thickness have been obtained by a simple PAD method. The X-ray diffraction and Raman scattering observations indicate that the YNMO films were obtained successfully. All films show ferromagnetic behavior, and the T_c of the films is lower than that of the YNMO bulk due to the strain in films. The T_c increases as film thickness increasing, which is ascribed to the reducing of the biaxial tensile strain in film. Besides, the magnetic properties of YNMO films show a dependence on

substrate type, which can be associated with the difference in strain relaxation and substrate effect. All the above results suggest that the magnetic properties of the film can be adjusted by the strain state in film, which can be controlled by film thickness and substrate type.

ACKNOWLEDGMENT

This project was financially supported by the National Basic Research Program of China (973 Program, Grant No. 2009CB939901), and the National Science Foundation of China, Grant No. 10874161.

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Table

Table 1 Structure parameter (a_s) of substrate; out-of-plane lattice parameter c_{film} , in-plane lattice parameter a_{film} (based on the Bragg formula, the a_{film} is calculated from XRD in-plane scans for (110) reflection of the 50nm YNMO thin films), biaxial strain (ε_{zz} , out-of-plane strain) and the change of volume (Δ_V , $V=a^2c$) of Y_2NiMnO_6 films with varying substrates.

Sample	$a_s(\text{\AA})$	$c_{film}(\text{\AA})$	$a_{film}(\text{\AA})$	ε_{zz} (%)	Δ_V (%)
YNMO/LAO	3.792	3.754(1)	3.789(0)	-0.884(4)	-0.806(1)
YNMO/LSAT	3.868	3.775(3)	3.804(0)	-0.324(7)	0.545(4)
YNMO/STO	3.905	3.784(2)	3.804(1)	-0.089(7)	0.787(7)

FIGURE CAPTIONS:

Figure 1. The SEM micrographs of the YNMO epitaxial thin films: the surface morphologies of (a) YNMO/LAO, (b) YNMO/LSAT, and (c) YNMO/STO, respectively; (d) the typical cross-sectional area of YNMO/STO.

Figure 2. 2D and 3D AFM micrographs of YNMO thin films deposited on: (a) LAO, (b) LSAT, and STO substrate, respectively.

Figure 3. XRD θ -2 θ scan for 50nm-YNMO films grown on (001)-oriented STO, LSAT, and LAO, and corresponding ω scans for (002) peaks of them, respectively.

Figure 4. Logarithmic intensity mappings around the LNMO (103) peak in reciprocal space for YNMO films grown on (a) STO, (b) LSAT, and (c) LAO substrates.

Figure 5. Raman spectra of YNMO films (~50nm) grown on (001)-oriented STO, LSAT, and LAO substrates at room temperature.

Figure 6. XRD θ -2 θ scan curves for 80-, 50-, and 20-nm-thickness YNMO films grown on (001) STO, respectively. Inset: (a) the variation in c -axis lattice parameter (top left corner); (b) ϕ scans of (101) reflections of YNMO/STO (top right corner).

Figure 7. Temperature dependent magnetizations for 80nm, 50nm, and 20nm YNMO films grown on STO, respectively, under an applied field of 100 Oe parallel to the film surface.

Figure 8. Temperature dependent magnetizations for 50nm-thickness YNMO films grown on STO,

LSAT, and LAO, respectively, under an applied field of 100 Oe parallel to the film surface.

figure 1

Figure 2

Figure 3

Figure 4

Figure 5

Figure 6

Figure 7

Figure 8

YNMO/LAO YNMO/LSAT YNMO/STO

the AFM micrographs of YNMO thin films

the magnetic property of YNMO films