Magnetic Properties of Multiferroic $BiFeO_3/BaTiO_3$ Bi-layer Thin Films

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Abstract : In this article, magnetic properties of multiferroic bi-layer $BiFeO_3$ (BFO)/$BaTiO_3$ (BTO) thin films were studied. It was found that the magnetization increased by the insertion of BTO buffer layer even though the interfacial stress was slightly relaxed, which indicated a coupling between the ferroelectric and ferromagnetic orders. Furthermore, with slightly increase of BFO film thickness, both BFO and BFO/BTO bi-layer films showed anisotropic magnetic properties with higher in-plane magnetization than the values measured out-of-plane. These are attributable to strain constraint effect at the interface.

Key Words : magnetic, multiferroic, thin film

1. Introduction

Multiferroic effects in materials provide a great opportunity to use an electric field to control ferromagnetism or vice versa. Magnetoelectric coupling between electric and magnetic order parameters has been theoretically predicted [1], and induced interest in its implementation in device architectures taking advantage of these properties [2]. Due to the requirement of small device, the search of single-phase multiferroics - materials that show spontaneous magnetization and polarization simultaneously at ambient conditions - continues.

One multiferroic material, however, has played a key role in rejuvenating the field after a report of large ferroelectric polarization combined with interesting magnetic properties - $BiFeO_3$ (BFO) [3]. Central to the interest in BFO is that it is a room-temperature multiferroic material with both a high ferroelectric Curie temperature [4] and a high antiferromagnetic Néel temperature [5]. Thus it offers exciting potential for room temperature device integration, if there is coupling between the order parameters.

Prepared in thin film, it has been reported to have a much larger remnant polarization than bulk, which should be attributed to its sensitivity of the small lattice constraint. The slight change of the structure gives birth to an avenue to increase the polarization and the magnetism.

In this paper, multiferroic $BiFeO_3/BaTiO_3$ bi-layer was prepared on (111) $Pt/Ti/SiO_2/Si$ substrate via pulsed laser deposition (PLD). The magnetization of BFO thin film showed an interesting dependence on size and was enhanced even by buffering a ferroelectric layer, which demonstrates a multiferroic coupling.

2. Experiment

$BiFeO_3$ target has been prepared by the conventional mixed oxide method. Bi_2O_3 and Fe_2O_3 raw powders of 99.9% purity were weighed in excess 10 mol% for Bi, and mixed by ball milling in ethanol for 24 hrs. The ball-milled powders was pressed into disks of one inch diameter and sintered at 850℃ The BFO ceramic target was ablated by an excimer Nd: YAG pulsed laser (wavelength of 355 nm.). The $Pt(111)/TiO_2/SiO_2/Si$ substrates were attached on a heater block. The details of the PLD conditions are summarized in Table 1. The phases of BFO thin films were identified by an X-ray diffractometer. The surface and cross sectional microstructures of the BFO thin films were investigated by a field emission scanning electron microscopy. The magnetic properties were measured via vibrating-sample magnetometer (VSM).

Table 1 PLD conditions of BiFeO3 thin films

Parameters	Deposited conditions
Target	10 % Bi-excess $BiFeO_3$
Substrate	Pt (111)/TiO_2/SiO_2/Si (100)
Substrate temperature	550℃ ~ 700℃
Deposition pressure	$P(O_2)$=1 mTorr
Target to substrate distance	60 mm
Laser type	Nd:YAG (λ=355 nm)
Energy/shot	125 mJ/shot
Pulse repetition rate	5 Hz

3. Results and discussion

Figure 1 shows the $\theta-2\theta$ scan XRD patterns of the $BiFeO_3$ thin films prepared at various temperatures. The result shows all the films consist mainly of polycrystalline. The film deposited at 550℃ and 650℃ showed a second phase of $Bi_{25}FeO_{40}$ due to the 10% excessive Bi, while the film deposited at 700℃showed undetectable Bi-rich phase. Figure

2 shows the SEM images of $BiFeO_3$ thin films deposited at different temperatures. It was found that the films deposited at 700℃ was best crystallized.

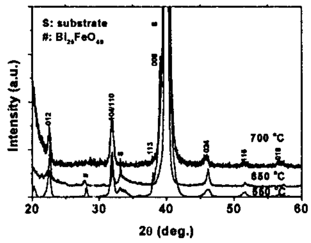
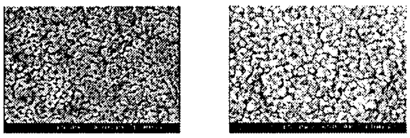


Fig. 1 XRD Patterns of BFO thin films deposited at different temperatures.



550 ℃ 700 ℃

Fig. 2 SEM images of BFO thin films

As aforementioned before, the optimized temperature for BFO deposition is 700℃. In this work, 140nm and 220nm thick BFO thin films were deposited at 700℃ on platinum coated silicon substrate and $BaTiO_3$ layers, respectively. Their magnetic hysteresis loops were measured at room temperature.

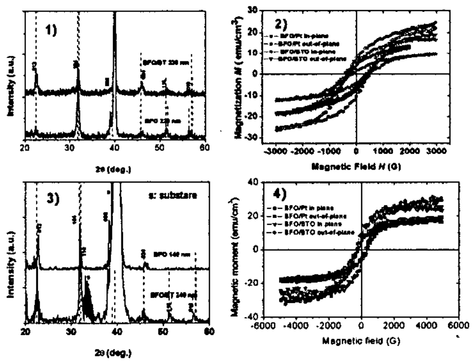


Fig. 3 1) X-ray patterns of 220 nm-thick BFO films deposited on Pt and $BaTiO_3$ (BTO); 2) in plane and out-of-plane magnetization of 220 nm-thick BFO films deposited on Pt and BTO; 3) X-ray patterns of 140 nm-thick BFO films deposited on Pt and BTO; 4) in plane and out-of-plane magnetization of 140 nm-thick BFO films deposited on Pt and BTO.

Figure 3 shows the x-ray patterns and magnetic properties of 220 nm and 140 nm thick BFO films deposited on Pt and BTO, respectively. It is obviously seen that 140 nm-thick BFO films have almost the same value of the in pane and out-of-plane magnetization, which 220 nm-thick BFO films have a higher in plane magnetization than that of out-of-plane, furthermore, 220 nm-thick films have a smaller magnetization than 140 nm- thick films [shown in figures 3(2) and 3(4)]. These should ascribed to the Pt substrate constraint which gives a in plane stress to BFO films whereas gives a stress relax with increasing the thickness. From figures 3(1) and 3(3) we can see that BFO deposited on Pt showed a (104) preferred growth while random oriented growth on BTO, which can be explained by the larger lattice match of BFO and Pt. however, by buffering a BTO layer the magnetization was surprisingly enhanced. This might be attributable to the interface diffusion beside the lattice constraint where the strong hybridization of Ti-O and Bi-O leads to the corresponding mechanisms for stabilizing the distorted structure. It demonstrated a multiferroic coupling between the ferroelectric and magnetic orders, which deserves further study.

4. Conclusion

In this work, BFO thin films and BFO/BTO bi-layer films were prepared by PLD. Films with 140 nm thickness showed larger isotropic magnetization while those with 220nm thickness showed anisotropy with reduced magnetization values compared to thinner films. It was also found the deposition of BFO films on top of 100nm-thick BTO layer helped increase both in-plane and out-of-plane magnetization values, which indicated not only the constraint between the interface but also the multiferroic coupling between them.

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