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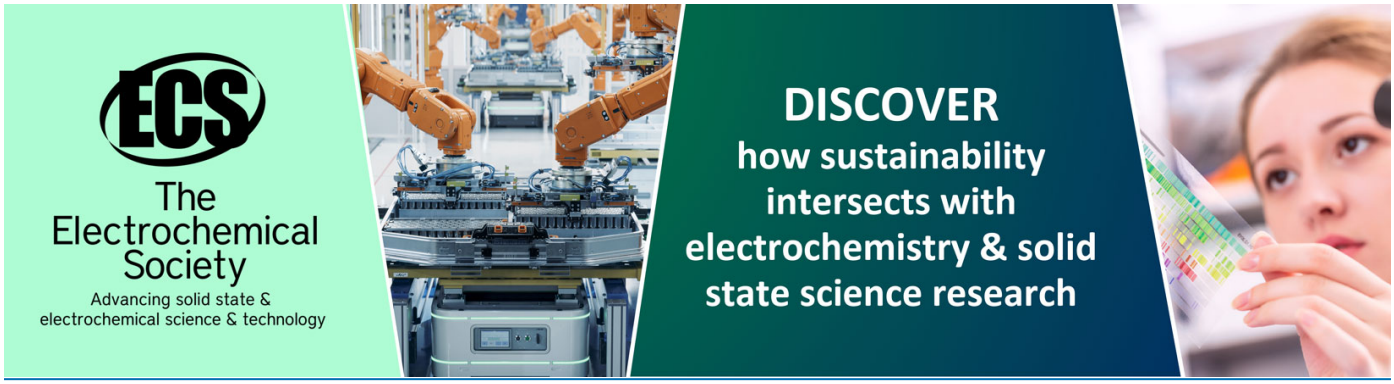

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Pulse laser deposition of $BiFeO_3$ films by polished targets

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Abstract. $BiFeO_3$ films had been deposited by pulse laser deposition using $BiFeO_3$ target. Nearly pure phase $BiFeO_3$ film had been grown simply by polishing target before deposition. X-ray diffraction (XRD), UV-visible-near infrared transmission spectrum and magnetization-magnetic field intensity (M-H) hysteresis loops had been measured. It was found that $BiFeO_3$ film deposited at 700^o C by polishing target had (010) preferred growth orientation. The bandgap is determined to be 2.25eV. The magnetic domain distribution had been investigated by magnetic force atomic force microcopy.

1. Introduction

Multiferroics, which exhibit both electric and magnetic orders in single phase, provide new opportunity to design multifunctional magneto electronic devices. In recent years, $BiFeO_3$ has been intensively researched for its potential application in multifunctional devices due to the coexistence of ferroelectricity (Curie temperature 830ºC) and antiferromagnetisim (Neel temperature 370 ºC) [1-3]. As a new lead free ferroelectric material, the polarization value of $BiFeO_3$ film can be as high as 110μc/cm^2, which is close to the polarization value of PbTiZrO3[4].

$BiFeO_3$ film has been successfully deposited by many researchers using sputtering [5, 6], pulse laser ablation [7-9], sol-gel [4, 10], etc. Pure phase and well crystalline film is very essential for multiferroics application. Unfortunately, impurity phases, usually $Bi_2Fe_4O_9$ and Fe_2O_3, often emerge in the films and deteriorate the multiferroic properties. In vacuum deposition, bismuth rich targets are often used to overcome bismuth loss in order to deposit stoichiometric $BiFeO_3$ film [6]. Other growth parameters, such as oxygen pressure [5] and growth rate should also be carefully controlled. In our research, we find pure phase $BiFeO_3$ films could be deposited by polishing target before deposition. This method is much easier and time consuming for deposition of $BiFeO_3$ films.

2. Experimental details

Sintered Stoichiometric $BiFeO_3$ target were used to deposit $BiFeO_3$ films on quartz substrate by pulse laser deposition (excitation wavelength 193nm). Quartz substrates were carefully cleaned by distilled water and then by anhydrous ethanol. The films with thickness about 80nm were deposited at different growth temperature from ambient temperature to 700 ºC. In

order to control the phase of film, $BiFeO_3$ film was deposited at 700 ºC with polished target. $BiFeO_3$ film was also deposited at 700 ºC without polishing target. Crystal structure of the films was characterized by XRD from 15º to 70º. The transmission spectra of the films were measured from 320nm to 2500nm.Optical constants (*n* and *k*) were calculated by Lorentz oscillator model. Bandgap of the film were obtained by the cross point of tangential line of plot α^2-$h\gamma$ with energy axis. Magnetic domain distribution was investigated by magnetic force model of atomic force microscopy. Magnetization-magnetic field intensity (M-H) hysteresis loops were measured at low temperature at 298K, 273K 150K and 80K.

3. Results and discussions

As is depicted previously, bismuth loss is inevitable during vacuum deposition of $BiFeO_3$ films. Loss of bismuth always leads to the growth of $Bi_2Fe_4O_9$ and other impurity phase, which had also happened in our experiment. As is shown in figure.1, $Bi_2Fe_4O_9$ phase is predominant for the films deposited at all temperature without polishing the target. However, $BiFeO_3$ becomes the predominant phase if the target is polished before deposition, which is shown as curve E in figure.1. The $BiFeO_3$ phase shows (010) preferred growth orientation. It is thought that the loss of bismuth only occurs at the surface of target. Polishing process could make a stoichiometric fresh surface. Therefore the pulse laser ablation could make a stoichiometric $BiFeO_3$ phase and reduce the growth of impurity f $Bi_2Fe_4O_9$ phase.

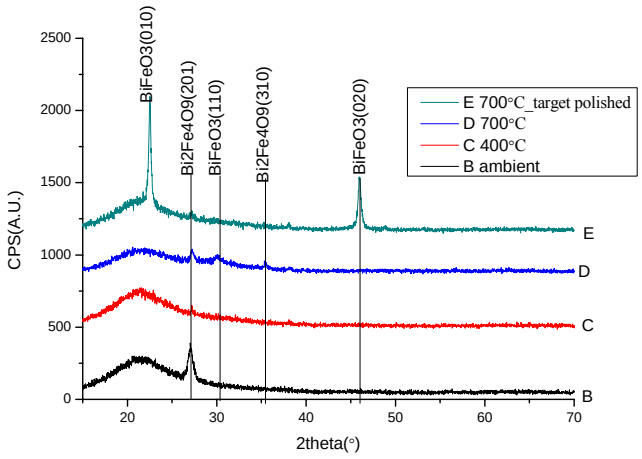

Figure 1. XRD spectrum of different samples deposited at different growth condition.

Optical transmission property in visible light range is very important for its potential application in magnetic-optic storage. In this experiment, UV-visible-near infrared transmission spectra of the samples have been measured and shown in figure.2 It is found the film deposited at different condition have good transmission from about 700nm to the near infrared. It is also found that $BiFeO_3$ films deposited at the center and rim of the glass substrate have different transmission properties, which indicates non-uniform of film thickness and even the different absorption. It is reasonable if the non-uniform intrinsic property of pulse laser ablation is considered. The cutoff wavelength of $BiFeO_3$ film at the center ($BiFeO_3$_700center in fig.2) and rim ($BiFeO_3$_700rim in fig.2) of substrate does not

change very much, which indicates they have nearly the same band gap. It is much interesting that Bi2Fe4O9 phase have larger band gap than $BiFeO_3$, which is also shown in figure.2

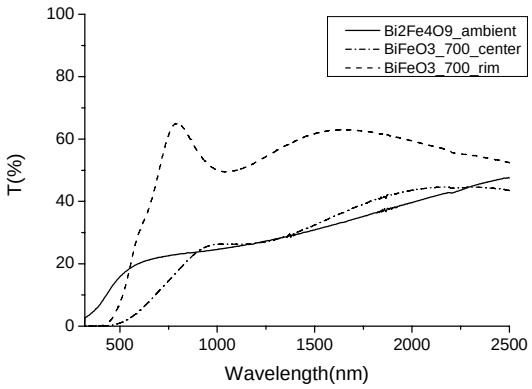

Figure 2. UV-visible-near infrared transmission spectrum of BiFeO3 films

The optical constants of $BiFeO_3$ films are also calculated by fitting the transmission spectra ($BiFeO_3$_700rim curve in fig.2) using Lorentz oscillator model. The refractive index (n) and extinction coefficient (k) is about 2.6 and 0.32 at 635nm, respectively, which is shown in figure.3. The absorption coefficient can be calculated by $\alpha=4\pi k/\lambda$, where λ is the wavelength. The optical band gap can be obtained by intersect of tangential line at energy axis in the plot α^2-hv, where hv is optical energy in eV. The optical band gap of $BiFeO_3$ films is about 2.25eV, which is shown in figure.4.

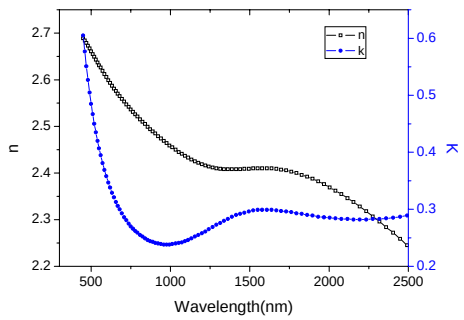

Figure 3. Calculated optical constants of $BiFeO_3$ films

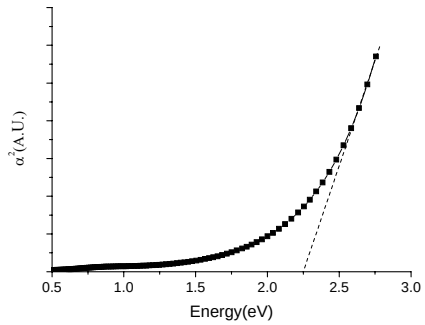

Figure 4. Band gap plot of $BiFeO_3$ films

Magnetic force mode of atomic force microscopy was used to investigate magnetic domain distribution of $BiFeO_3$ films. In first step, the top morphology image was recorded by constant-current mode, which is shown in figure.5. The surface is quite smooth and covered with homogeneous crystal particles. The crystal particle size of $BiFeO_3$ film is about 200nm. In second step, the tip was lift 80nm higher than the surface of film. Far field force image was recorded as magnetic force image, which is shown in figure.6. The size of irregular magnetic domain is estimated to be about 400-500nm.

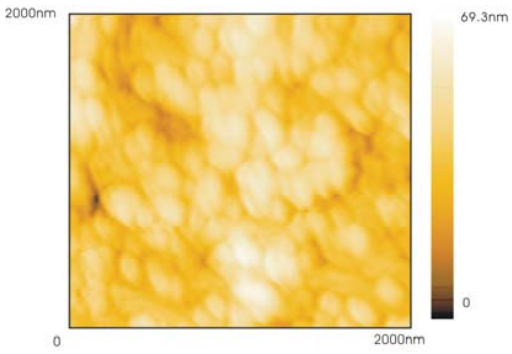

Figure 5 AFM top morphology picture of $BiFeO_3$ films deposited at 700ºC with polished target.

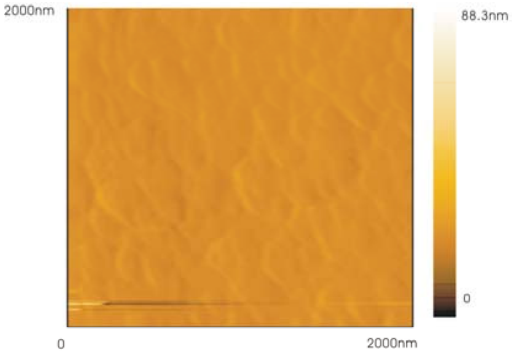

Figure 6. MFM morphology picture of $BiFeO_3$ films deposited at 700ºC with polished target.

Magnetization (M) vs. magnetic field intensity hysteresis loops of $BiFeO_3$ films at different temperature are shown in figure.7. In this experiment, the magnetization of quartz substrate could not be neglected because magnetization of $BiFeO_3$ is very weak for its thin film thickness. Magnetization of $BiFeO_3$ film overlaps with quartz substrate, which leads to the rotation of magnetic hysteresis loops. The area of hysteresis loop increases with decreasing of measurement temperature. The rotation of magnetic hysteresis loop makes the determination of saturated (remnant) magnetization much difficult. However, one can find that the saturated magnetization increase qualitatively with decrease of temperature.

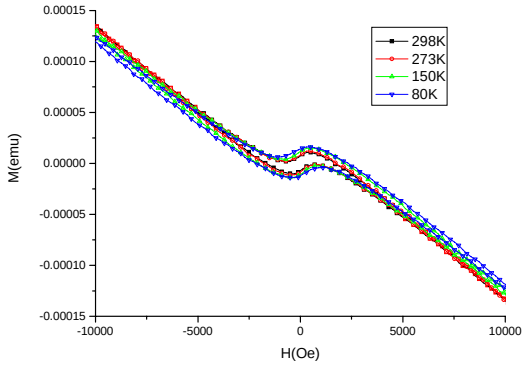

Figure 7. Magnetization hysteresis loops of $BiFeO_3$ film on quartz substrate at different temperature.

4. Conclusions

$BiFeO_3$ films had been deposited on quartz substrate at different deposition temperature. It was found that nearly pure phase $BiFeO_3$ films could be easily deposited by polishing the targets before deposition. The films deposited at 700 ºC by polishing targets have (010) growth orientation. The transmission spectrum were measured and fitted to get absorption coefficients. The bandgap of $BiFeO_3$ film was calculated to be 2.25eV. The magnetic domain distribution was measured by magnetic force microscopy. It was found the saturated magnetization of $BiFeO_3$ film was increased at low temperature.

Acknowledgements

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