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Dielectric properties and electrical behaviors of tunable $Bi_{1.5}MgNb_{1.5}O_7$ thin films

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Abstract

The $Bi_{1.5}MgNb_{1.5}O_7$ (BMN) thin films were prepared on Au-coated Si substrates by rf magnetron sputtering. We systematically investigated the structure, dielectric properties and voltage tunable property of the films with different annealing temperatures. The relationships of leakage current and breakdown bias field with annealing temperature were firstly studied and a possible explanation was proposed. The deposited BMN thin films had a cubic pyrochlore phase when annealed at 550 °C or higher. With the increasing of annealing temperature, the dielectric constant and tunability also went up. BMN thin films annealed at 750 °C exhibited moderate dielectric constant of 106 and low dielectric loss of 0.003–0.007 between 10 kHz and 10 MHz. The maximum tunability of 50% was achieved at a bias field of 2 MV/cm. However, thin films annealed at 750 °C had lower breakdown bias field and higher leakage current density than films annealed below 750 °C. The excellent physical and electrical properties make BMN thin films promising for potential tunable capacitor applications.

Keywords: $Bi_{1.5}MgNb_{1.5}O_7$; Thin films; Leakage current; Tunability

1. Introduction

Recently, tunable microwave devices of miniaturization, low cost, high power, and multifunction are urgently demanded by the dramatic development of wireless communication system, and much attention has been paid to tunable dielectric thin films for this application. Ferroelcctric $(Ba_xSr_{1-x})TiO_3$ (BST) thin films have been extensively investigated because of their large tunability and they have been successfully adapted to tunable microwave devices [1–4]. However, high dielectric loss of BST thin films strongly inhibits their industrial applications. The bismuth based cubic pyrochlore $Bi_{1.5}ZnNb_{1.5}O_7$ thin films have been paid close attention as a promising fungible tunable material for their adequate tunability and low dielectric loss in the microwave region [5,6]. Although BZN thin films have above superiorities, they need high bias field to achieve high tunability, which makes integrating them with silicon technology difficult.

In order to achieve high tunability at low bias field, plenty of work has been carried out [7,8]. Recently, $Bi_{1.5}MgNb_{1.5}O_7$ (BMN) thin films, which replace Zn^{2+} with Mg^{2+} on A sites in cubic pyrochlore structure, were prepared and demonstrated to possess even more superior properties than BZN thin films. This is because Mg^{2+} ion, which is smaller than Zn^{2+}, is more flexible for hopping [9]. However, in order to apply BMN thin films to tunable devices, a lot of systematic work needs to be done. The post-annealing process is very crucial to obtain high quality thin films [7,10]. During this process, one can reduce defects, improve crystallization, increase grain size and thereby optimize the properties of thin films. So finding the optimal annealing temperature is necessary to be investigated. Moreover, the leakage current and breakdown bias field are two important parameters which influence the reliability of tunable devices. However, there are few reports discussing the breakdown bias field and leakage current behaviors of the BMN thin films. Such studies are necessary to predict the behaviors of tunable thin films under device operating conditions. In this study, we systematically investigated the dielectric properties, tunability, leakage current and breakdown bias field of the BMN thin films annealed at different temperatures and corresponding explanations were also proposed.

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2. Experiment procedure

The sputtering method was used for fabricating bismuth-based pyrochlore $Bi_{1.5}MgNb_{1.5}O_7$ thin films. The $Bi_{1.5}MgNb_{1.5}O_7$ ceramic targets were prepared by solid state reaction process with Bi_2O_3, MgO, and Nb_2O_5 as starting materials. The mixtures of starting materials in stoichiometry were sintered into disks at 1180 °C for 4 h. The detailed deposition conditions are summarized in Table 1. For electrical measurement, Au top electrodes with 0.2 mm in diameter were patterned by lift-off process to form the metal-insulator-metal-type capacitors.

The crystal structure of the films was characterized by X-ray diffraction (Rigaku D/MAX-RB, Akishima, Tokyo, Japan). Film morphologies were investigated by scanning electron microscopy (JEOL JSM-7600F, Akishima, Tokyo, Japan). The dielectric properties, tunability and breakdown bias field of BMN films were measured at room temperature by Aglient 4285A precision LCR meter (Santa Clara, California, USA). The surface roughness of the films was measured by Alpha-Step D-100 profilometer (KLA-Tencor, California, USA) and the leakage current of the films was measured by Aglient 4339B High Resistance Meter (Santa Clara, CA, USA) at room temperature.

3. Results and discussion

3.1. Crystallization

The XRD patterns of the BMN thin films with various annealing temperatures are shown in Fig. 1. The films were amorphous after annealing at 500 °C. Films annealed at 550 °C were crystalline and showed a cubic pyrochlore structure. With the increasing of annealing temperature, the intensity of diffraction peaks strengthened. The lattice constant of the BMN thin films, which was calculated from the XRD patterns and shown in Table 2, is similar to the one of BZN thin films prepared on Si substrates [11]. This result indicates that the replacement of Zn^{2+} (of ionic radius 0.74 Å) by smaller Mg^{2+} (of ionic radius 0.66 Å) leads to no shrinkage in unit cell dimension of the cubic pyrochlore structure.

Table 1
Deposition conditions of the BMN thin films by rf-magnetron sputtering.

Sputter parameter	Experimental condition
Target	3 in. $Bi_{1.5}MgNb_{1.5}O_7$
Substrate	$Au/Ti/SiO_2/Si$
Base pressure in process chamber	7×10^{-6} Torr
Substrate temperature	450 °C
Working pressure	10 mTorr
rf power	150 W
Ar/O_2 flow rate	4:1 SCCM
Thickness of films	~200 nm
Annealing temperatures	500 °C, 550 °C, 650 °C and 750 °C

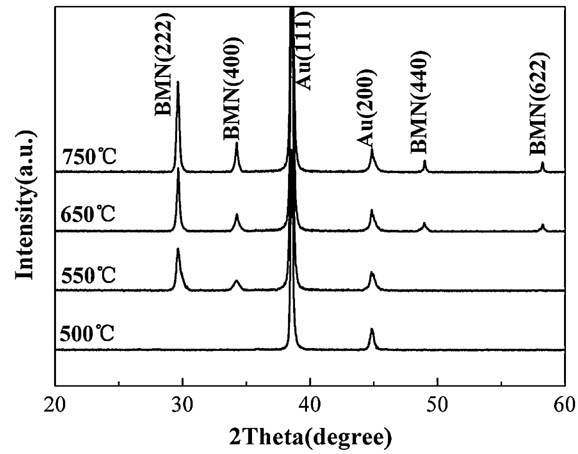

Fig. 1. XRD patterns of BMN thin films annealed at various temperatures.

3.2. Morphology

Fig. 2 illustrates the surface morphology of the BMN thin films annealed at different temperatures. By increasing the annealing temperature from 500 °C to 750 °C, the average surface grain size (the results are shown in Table 2) rose and the crystallization was promoted. Compared with lower temperatures, the films annealed at 750 °C had denser surface and larger grain size.

3.3. Dielectric properties

It is illustrated in Fig. 3 that the dielectric constant and loss tangent of the BMN thin films varied as a function of post-annealing temperatures. The maximum value of dielectric constant was 106 while the film was post-annealed at 750 °C. It was observed that the dielectric constant of BMN thin films went up with the increasing of annealing temperature. When the annealing temperature increased from 500 °C to 550 °C, the dielectric constant increased from 51 to 82 due to the crystallization of thin films. The dielectric constant of the BMN thin films prepared on Si substrates in our experiment is higher than the films prepared on sapphire substrates [9]. This is because films grown on Si substrates exhibit a higher hydrostatic tensile strain than films grown on sapphire substrates [11,12], and larger tensile strains increased the permittivity due to easier polarization [5,12]. Films annealed above 500 °C also exhibited low dielectric losses (0.0039–0.003). With the increasing of annealing temperature, the loss tangent slightly decreased.

Table 2
Lattice constant, grain size, surface roughness and breakdown bias field of BMN films annealed at different temperatures.

Annealing temperature (°C)	Lattice constant (Å)	Grain size (nm)	Surface roughness (nm)	Breakdown bias field (MV/cm)
750	10.54	130	8.65	2
650	10.54	70	4.34	2.4
550	10.51	45	2.13	2.7
500	–	10	0.88	3.5

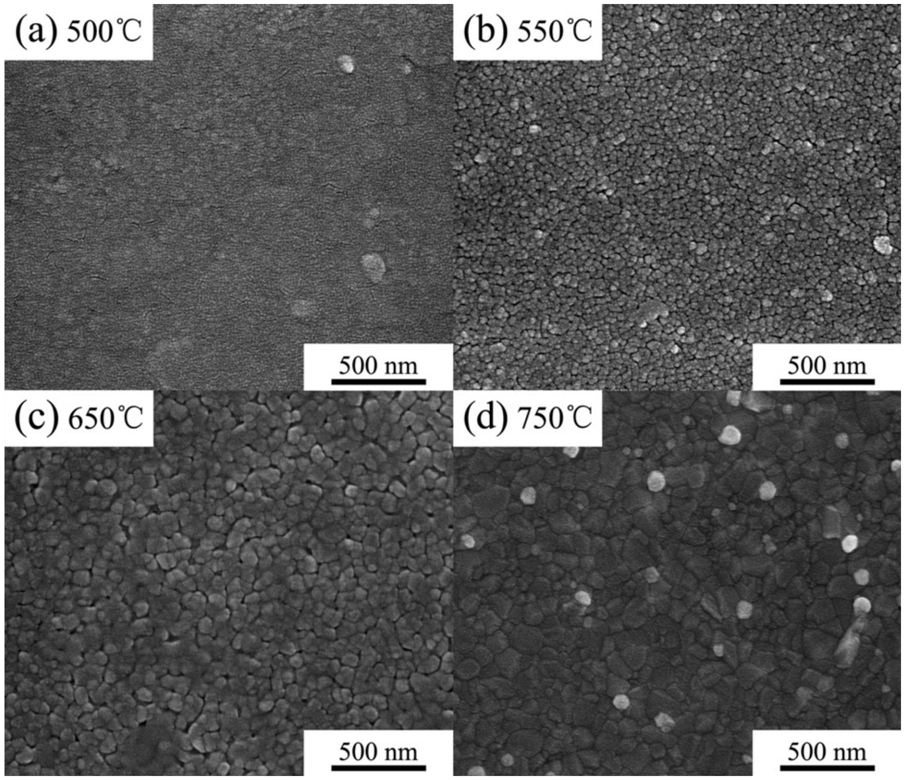

Fig. 2. SEM surface morphology of BMN films annealed at (a) 500 °C, (b) 550 °C, (c) 650 °C and (d) 750 °C.

Fig. 4 shows how the frequency variation between 10 kHz and 10 MHz affects the dielectric constant and loss tangent of the BMN thin films annealed at 750 °C. Between these measurement ranges, the dielectric constant was 106 and exhibited a constant value at room temperature. The loss tangent was 0.003 and remained stable between 1 kHz and 1 MHz. However, after the frequency exceeded 1 MHz, the loss tangent increased with frequency. This is because the total device loss is increasingly dominated by the conductor losses of the electrode [4]. To reduce conductor losses at high frequency (>1 MHz), thick metal bottom electrodes with low electric resistivity are desired, in order to reduce electrode series resistance. Au has a lower electrical resistivity (2 μΩ cm) than Pt (9.7 μΩ cm), which indicates that Au would be a more ideal electrode material [6]. In our experiments, Au bottom electrodes were used and the loss tangent was less than 0.007 at 10 MHz.

3.4. Tunability

Fig. 5 shows the normalized dielectric constant and loss tangent vs. bias-field at the measurement frequency of 1 MHz for BMN films annealed at different temperatures. The tunability of thin films is defined as [5,10]:

$$\eta = \left[\frac{C_{\max} - C_{\min}}{C_{\max}}\right] \times 100\% \quad (1)$$

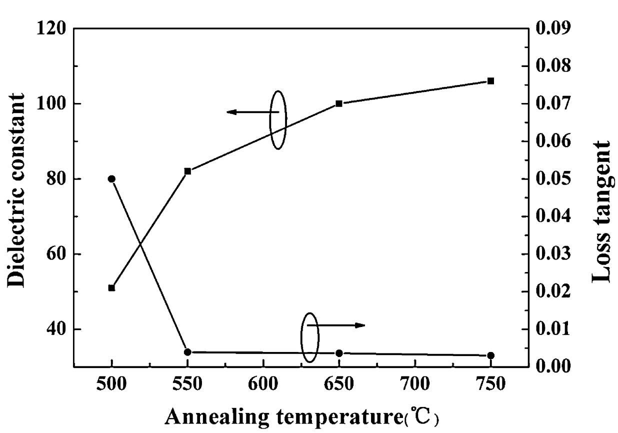

Fig. 3. Annealing temperature dependence of dielectric constant and loss tangent for BMN thin films measured at 1 MHz.

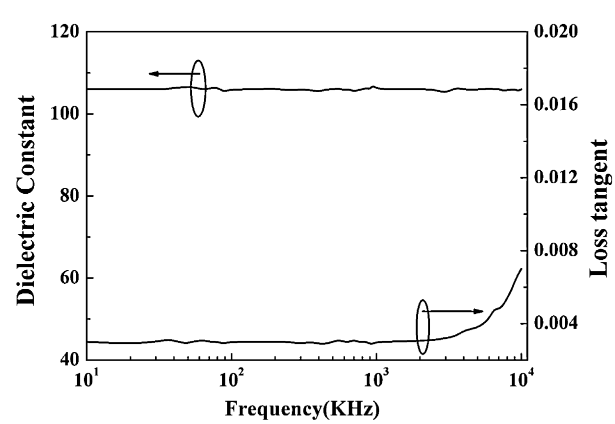

Fig. 4. Dielectric constant and loss tangent of BMN thin films as a function of frequency ranging from 10 kHz to 10 MHz.

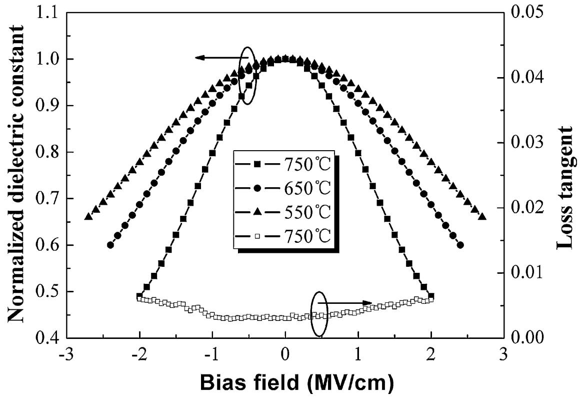

Fig. 5. Normalized dielectric constant and loss tangent vs. dc bias field for BMN films annealed at different temperatures.

where C_{max} is the maximum capacitance value (at zero bias field) and C_{min} is the minimum (at maximum bias field). The tunability of the films increased with annealing temperature due to the enhancing crystallization of the films. The highest value of tunability, 50%, was achieved at the bias field of 2 MV/cm in BMN thin film with annealing temperature of 750 °C. Because the BMN thin films annealed at 500 °C were amorphous, the films did not show tunability and the curve was not included in Fig. 5. The loss tangent of the films was independent of bias field when the applied bias field was low (<1.0 MV/cm). With the increasing of the applied voltage, the loss tangent went up but remained below 0.006 until the bias field of 2 MV/cm. Because the loss tangent of the films annealed at different temperatures showed the similar tendency with bias field, Fig. 5 only presents the loss tangent of films annealed at 750 °C. The loss tangent as a function of bias field can be explained by the fact that high bias field leads to higher leakage, which caused a higher dielectric loss.

3.5. Leakage current and Breakdown bias field

Fig. 6 shows the relationship between leakage current density and applied voltage for BMN thin films annealed at different temperatures. The leakage current density of BMN films varies exponentially with electric field. This phenomenon is in

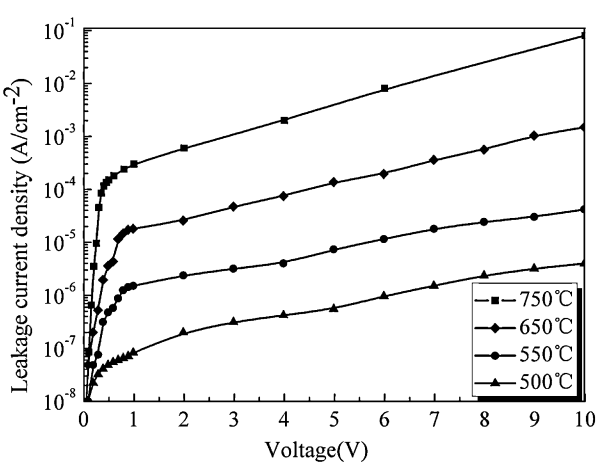

Fig. 6. Dc leakage current density vs. applied voltage of BMN thin films annealed at different temperatures.

accordance with Poole-Frenkel (PF) conduction and Schottky emission theories, two of the most common mechanisms that account for conduction in thin films [13–15]. With the same bias voltage, the leakage current density increased with the rising of annealing temperature. When the films were annealed at 750 °C and the bias voltage was 10 V, the leakage current density reached the highest value. The measuring range (100 μA) of leakage current limited the measurement if applied voltage went higher. The breakdown bias field of BMN films annealed at 750 °C, 650 °C, 550 °C and 500 °C are presented in Table 2. Compared to the leakage current density, the breakdown bias field exhibited the reverse tendency with annealing temperatures. The leakage current and breakdown bias field are thought to be strongly affected by surface roughness (shown in Table 2) of the films. By increasing annealing temperature, the surface roughness goes up due to the larger grain size [16]. The influence of surface roughness on leakage current and breakdown bias field of the films can be explained as follows: for rough films, the local electric field will vary from place to place due to the fluctuation of the surface height. At the valley, the electric field is larger than that at the peak; therefore, the leakage current density at the valley is much higher than the peak and it is easier for the films to be brokendown at the valley of a rough surface. Therefore, the increase in surface roughness will cause an increase in leakage current while a decrease in breakdown bias field in BMN thin films.

4. Conclusions

Cubic pyrochlore BMN thin films were deposited on Au/Ti/SiO_2/Si substrates by rf-magnetron sputtering and annealed at various temperatures. The crystallization, dielectric properties and tunability of the films were promoted by increasing annealing temperature from 500 °C to 750 °C. The films annealed at 750 °C exhibited moderate dielectric constant of 106, low dielectric loss of 0.003 at 1 MHz and the maximum tunability of 50% at bias field of 2 MV/cm. The behaviors of leakage current and breakdown bias field with different annealing temperatures were also studied. With the increasing of annealing temperature, the leakage current went up while the breakdown bias field decreased due to the increase of surface roughness. The medium permittivity, low loss, superior tunability and electrical behaviors guarantee the BMN thin films promising for tunable capacitor applications.

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