Room-temperature multiferroic $\text{Bi}_4\text{YFeTi}_3\text{O}_{15}$ thin films of four-layered perovskite

Y. Li^a, C.J. Lu^{a,b,*}, J. Su^{a,c}, Y.C. Zhang^a, C. Zhang^a, S.F. Zhao^a, X.X. Wang^a, D.J. Zhang^a, H.M. Yin^b

^aCollege of Physics Science and Key Laboratory of Photonics Materials and Technology in Universities of Shandong, Qingdao University, Qingdao 266071, China

^bXinjiang Laboratory of Phase Transitions and Microstructures in Condensed Matters, College of Physical Science and Technology, Yili Normal University, Yining 835000, China

^cNational Laboratory of Solid State Microstructures, Physics School, Nanjing University, Nanjing 210093, China

*Author to whom correspondence should be addressed; Electronic mail: cjlu@qdu.edu.cn

Abstract Polycrystalline $\text{Bi}_4\text{YFeTi}_3\text{O}_{15}$ thin films were prepared on (111)Pt/Ti/SiO₂/Si through a sol-gel process. X-ray diffraction analysis shows the films have a single-phase four-layered Aurivillius structure with the space group of *Fmm*2. As expected, the $\text{Bi}_4\text{YFeTi}_3\text{O}_{15}$ films exhibit the coexistence of ferroelectric and weak ferromagnetic properties with the remanent polarization ($2P_r$) $\sim 53.62 \mu\text{C}/\text{cm}^2$ and the saturated magnetization (M_s) $\sim 0.50 \text{ emu}/\text{cm}^3$ at room temperature. More interestingly, both dielectric relaxation behavior and room-temperature magneto-dielectric effect were observed in the $\text{Bi}_4\text{YFeTi}_3\text{O}_{15}$ films, probably resulting from the coexistence of Fe^{2+} and Fe^{3+} . The conduction mechanism of the films from 10 to 300 kV/cm is dominated by Ohmic mechanism, space-charge-limited current (SCLC), and

trap-filled-limit (TFL) current, respectively. The leakage current density remains lower than 1.03×10^{-5} A/cm² under the poling field below 300 kV/cm at 300 K.

Keywords: Bi₄YFeTi₃O₁₅; sol-gel process; Multiferroic; Magneto-dielectric effect

1. Introduction

Multiferroic materials, which simultaneously exhibit ferroelectric and magnetic orders, have been widely studied due to their potential applications in spintronics, multi-states memory, etc [1-3]. In addition, the coupling between magnetic and electrical orders leads to extra versatility for relevant devices, such as magnetic data storage controlled by electric field [4]. Unfortunately, because of the natural exclusion between ferroelectricity and magnetism, there are few room-temperature multiferroics in the nature [5]. Consequently, researchers never stop searching for new-style multiferroic materials to construct ever smaller and multifunctional electronic devices [6-8].

BiFeO_3 is the most frequently studied room-temperature multiferroic material. BiFeO_3 thin films offer relatively larger ferroelectric polarization ($60 \mu\text{C}/\text{cm}^2$) at room temperature [9]. Unfortunately, BiFeO_3 suffers from high leakage current and its magnetization is supposed to be near zero [9-11]. To overcome these obstacles, many attempts have been explored [12-15]. In order to reduce the leakage of the multiferroic material, one of the routes is using the $(\text{Bi}_2\text{O}_2)^{2+}$ layer as a layer of space charge compensation and insulation [13]. Aurivillius phase $\text{Bi}_5\text{FeTi}_3\text{O}_{15}$ is built from the widely studied antiferromagnetic BiFeO_3 and ferroelectric $\text{Bi}_4\text{Ti}_3\text{O}_{12}$ of three-layered perovskites. In $\text{Bi}_5\text{FeTi}_3\text{O}_{15}$ lattice, each four-layered perovskite unit of $(\text{Bi}_3\text{FeTi}_3\text{O}_{13})^{2-}$ is sandwiched by two insulative $(\text{Bi}_2\text{O}_2)^{2+}$ layers along c -axis [15]. The $\text{Bi}_5\text{FeTi}_3\text{O}_{15}$ films experienced a structural transition from $A2_1am$ to the $I4/mmm$ at the ferroelectric Curie temperature of 730°C [14]. However, they are not suitable for device

applications because the $\text{Bi}_5\text{FeTi}_3\text{O}_{15}$ films have antiferromagnetic order with a low Néel temperature of 80 K [12]. Obviously, the antiferromagnetic order of $\text{Bi}_5\text{FeTi}_3\text{O}_{15}$ results from the antiferromagnetic BiFeO_3 unit. If a magnetic perovskite unit with room-temperature ferromagnetic behavior is inserted into the three-layered ferroelectric $\text{Bi}_4\text{Ti}_3\text{O}_{12}$ lattice, a new kind of room-temperature multiferroic material with four-layered perovskite would be obtained.

As we know, orthoferrite YFeO_3 with perovskite structure (*Pnma*) exhibits simultaneously ferroelectric and ferromagnetic behavior at room temperature with a ferroelectric Curie temperature of 460 K and a high Néel temperature of 655 K [16,17]. The remanent magnetization of YFeO_3 crystals is approximately 0.85 emu/g at 300 K, which is superior to that of BiFeO_3 (near zero) [17]. Moreover, Y^{3+} (0.09 nm) has a smaller ionic radius than Bi^{3+} (0.103 nm), which can improve oxygen ion stability in the lattice and thus improved fatigue resistance, and maintained a more significant structural distortion [18]. When the magnetic unit BiFeO_3 in $\text{Bi}_5\text{FeTi}_3\text{O}_{15}$ is replaced by YFeO_3 , thus, the Aurivillius phase $\text{Bi}_4\text{YFeTi}_3\text{O}_{15}$ (BYFTO) of four-layered perovskite is expected to exhibit room-temperature multiferroic behavior. To our knowledge, there is no report on either the synthesis or characterization of BYFTO in the literature.

Recently, we prepared polycrystalline BYFTO thin films on (111)Pt/Ti/SiO₂/Si through a sol-gel process. Their microstructure was characterized and their electric/magnetic properties were measured. As expected, the BYFTO films perform both multiferroic properties and magneto-dielectric effect at room temperature. In this article, the microstructure, multiferroic properties and magneto-dielectric effect of the

BYFTO films are presented.

2. Experimental

BYFTO films were prepared on (111)Pt/Ti/SiO/Si substrates by a sol-gel process. Bismuth nitrate $[\text{Bi}(\text{NO}_3)_3 \cdot 5\text{H}_2\text{O}]$, yttrium oxide (Y_2O_3), iron acetylacetonate ($\text{C}_{12}\text{H}_{21}\text{FeO}_6$), and titanium n-butoxide $[\text{Ti}(\text{C}_4\text{H}_9\text{O})_4]$ were used as the precursors materials for Bi, Y, Fe, and Ti, respectively. Acetic acid ($\text{CH}_3\text{CO}_2\text{H}$) and n-propanol ($\text{C}_3\text{H}_8\text{O}$) were used as solvent, and acetylacetone ($\text{C}_5\text{H}_8\text{O}_2$) was used as a stable agent. A 4% excess of bismuth nitrate was added to compensate for volatilization of Bi during the annealing process. The films were spin coated at 3500 rpm for 30 s. Subsequently, the wet films were dried at 200 °C and pyrolyzed at 450 °C for 300 s. The deposited films were then followed by the rapid thermal annealing at 690 °C for 480 s in oxygen atmosphere. The above steps were repeated five times to obtain the ultimate thickness of 300 nm.

The crystal structures of the BYFTO films were analyzed by X-ray diffraction (XRD, Bruker D8) with Cu $\text{K}\alpha$ radiation and scanning electron microscopy (SEM, Hitachi S-4800). The Premier II ferroelectric test unit was used to measure the ferroelectric responses and leakage current. The dielectric properties at different temperature were measured using the impedance analyzer (Agilent 4294A) and Probe Station (Lake Shore CRX-6.5K). The alternating gradient magnetometer (AGM, MicroMag 2900/3900) was used to examine the magnetization.

3. Results and discussion

Fig. 1(a) shows the XRD pattern of the BYFTO thin films. All the diffraction peaks are indexed on the basis of an orthorhombic cell [19]. The BYFTO films exhibit a single-phase Aurivillius perovskite structure of space group $Fmm2$ and no other impurity diffraction peaks are detected. Moreover, the detected XRD pattern of the BYFTO films is consistent with earlier reports in $\text{Bi}_5\text{FeTi}_3\text{O}_{15}$ films, which indicate that the magnetic unit YFeO_3 are successfully inserted into the ferroelectric unit $\text{Bi}_4\text{Ti}_3\text{O}_{12}$ [12,15]. In addition, the FWHM (full-width at half-maximum) of the main diffraction peak (119) of the BYFTO films is $\sim 0.45^\circ$, which implies good crystallization. Fig. 1(b) shows a cross-section SEM image of the BYFTO films annealed at 690°C and it exhibits a clear boundary between the BYFTO films and the Pt electrodes. It can be seen that the films have a unified thickness of $\sim 300\text{ nm}$.

Fig. 2(a) displays the hysteresis loops of the BYFTO thin films at various frequencies. The films exhibit a P - E loop with remanent polarization $2P_r$ of about $53.62\text{ }\mu\text{C}/\text{cm}^2$ and coercive field E_c of about $328\text{ kV}/\text{cm}$ under an applied electric field of $1300\text{ kV}/\text{cm}$ at 1 kHz . The $2P_r$ of the BYFTO films is comparable to the value of $2P_r = 31.94\text{ }\mu\text{C}/\text{cm}^2$ for the $\text{Bi}_5\text{FeTi}_3\text{O}_{15}$ thin films [15]. The measured polarization of the BYFTO films might be related to the deposition technology and leakage current density.

In order to investigate the ferroelectric dynamics of BYFTO thin films, the scaling behavior for saturation loops were studied as shown in Fig. 2(b). Based on the scaling law $\langle A \rangle \propto f^a E_0^b$ theoretical proposed by Rao *et al.* [20], the P - E loops data of BYFTO thin films can be well fitted with the scaling relations of $\langle A \rangle \propto f^{-0.17} E_0^{2.41}$. Here, the

area $\langle A \rangle$ of ferroelectric hysteresis represents the energy dissipation within one cycle of domain reversal, and the scaling exponents a and b reflect the changing rules of $\langle A \rangle$ with electric field frequency f and electric field amplitude E_0 . Many experimental works were focused on the scaling law $\langle A \rangle \propto f^a E_0^b$ in polarization systems, such as ferroelectric films, ceramics, relaxor ferroelectrics and antiferroelectrics, and meaningful conclusions were obtained [21-24]. However, there has been no report about the dynamic scaling behavior of BYFTO thin films with multiferroic properties as demonstrated later. Table 1 summarized the scaling relations of the theoretically predicted, ferroelectric thin films, and the BYFTO films in this work. One can see that, all the exponents a in the scaling laws of different films are negative, indicating $\langle A \rangle$ decays with f . However, the absolute value of exponent a in the BYFTO films is the smallest one, which implies that the hysteresis area $\langle A \rangle$ decreases more slowly with f compared with other ferroelectric films. By this taken, the BYFTO films have good frequency stability and better application prospect in multi-states memory. On the other side, all the exponents b of various films are positive, which indicates that $\langle A \rangle$ increases with E_0 . This is because, with increasing electric field, the pinned domains in the ferroelectric films began to participate in polarization reversion. Furthermore, the absolute value of exponent b in the BYFTO films is approximate with those of other ferroelectric films, which illustrates that the BYFTO films have similar electric field dependence as other ferroelectric films.

To exclude the effect of leakage current, temperature dependence of leakage current of the BYFTO films was measured as a function of electric field in the range of 50 K-300 K as shown in Figs. 3(a). It is seen that the leakage current shows an

expected exponential increase with electric field and is dependent on temperature. The leakage current density of the BYFTO films increases with increasing temperature. At 300 K, the BYFTO films exhibit a much lower J of $1.03 \times 10^{-5} \text{ A/cm}^2$ at 300 kV/cm than that (10^{-2} – 10^{-1} A/cm^2 at 300 kV/cm) measured from BiFeO₃ films by sputtering [27]. We can see that, compared with the BiFeO₃ films, the leakage current of the BYFTO films decreased by three orders of magnitude. As we predicted, the improved leakage properties of the BYFTO films might be resulted from the reduction of oxygen vacancies and the insertion of the insulative (Bi₂O₂)²⁺ layers mentioned above. To further elucidate the conduction mechanisms at different ranges of applied electric field in BYFTO films at 300 K, we tried space-charge-limited current (SCLC), Schottky emission, and Poole-Frenkel emission model to check the conduction mechanisms of the films [28-30]. For BYFTO thin films [Fig. 3(b)], the slope of the ln-ln curves is from 1.05 to 6.71. The data can be well fitted by three straight lines, implying that three different transport mechanisms can be distinguished in the J - E characteristic curve [30]. The fitting value of slope in the low electric fields (<102 kV/cm) is around 1, which agrees with the Ohmic mechanism. In the intermediate electric fields, the fitting slope is around 2 indicating the dominant SCLC conduction mechanism. In the low electric fields, the conduction mechanism is dominated by the thermal excitation electrons in the films. With the electric fields increasing, the number of electrons injected from the cathode is comparable to the density of the thermal excitation electrons and the injected electrons is attracted by the oxygen vacancies of the films, so the conduction mechanism will transit from Ohmic mechanism to SCLC mechanism at the intermediate electric-field region as discussed by Lampert [28]. In the high electric

fields (>184 kV/cm), larger slope than 2 can be understood by the trap-filled-limit (TFL) model. Under the higher electric fields, the traps of the oxygen vacancies in the films have all been filled and there are unneutralized charges in the traps which will prevent the redundant electrons injected from the cathode [31].

The temperature dependence of the dielectric permittivity ϵ_r and dielectric loss $\tan\delta$ of the BYFTO films at different frequencies are shown in Fig. 4(a) and Fig. 4(b), respectively. The plot of ϵ_r - T exhibits a monotonous increasing trend with certain frequency dispersion and abnormal dielectric loss peaks with strong dispersion are clearly displayed. Actually, the strong frequency dispersion of the dielectric permittivity ϵ_r and the frequency dependence of the dielectric loss peak are features of dielectric relaxation and can be described to a thermally activated process [32]. It is found that T_m vs. f_r (peak in the dielectric loss spectra) can well be ascribed by the Arrhenius law: $f_r = f_0 \exp(-E_a/k_B T_m)$, where f_0 and E_a are the relaxation frequency at infinite temperature and the activation energy respectively, k_B is the Boltzmann constant, and T_m is the peak temperature, a linear relation between $\ln f_r$ and the inverse of peak $10^3/T$ are shown in Fig. 4(c) with the fitting parameter $E_a = 0.29$ eV. The value of activation energy in the BYFTO films is much lower than the value $E_a = 0.87$ eV for Aurivillius $\text{Bi}_4\text{Ti}_3\text{O}_{12}$ thin films [33]. This inconsistency suggests that the relaxation mechanism for the dielectric loss peak in BYFTO may be not associated with the thermal motion of oxygen vacancies in the films. Moreover, in view of the oxygen vacancies and other ion defects in the BYFTO films inevitably make the valence transfer between Fe^{3+} and Fe^{2+} to keep the electrical neutrality of the system, as similarly discussed in BiFeO_3 [34]. We noted that $E_a = 0.29$ eV in BYFTO is consistent

with the value of $E_a = 0.29$ eV in LuFe_2O_4 , $E_a = 0.3$ eV in BiFeO_3 [34,35]. In addition, our previous study showed that the dielectric relaxation effect were weakened by the reduction in Fe^{2+} content after oxygen treatment in $\text{Yb}_3\text{Fe}_5\text{O}_{12}$ ceramics [36]. Thus, we deduced the dielectric relaxation behavior of the BYFTO films is possibly associated with the electron hopping between Fe^{2+} and Fe^{3+} [34].

Magnetization M verse magnetic field H curves were measured by AGM at room temperature as shown in Fig. 5. Interestingly, a weak ferromagnetic hysteresis loop with a small saturation magnetization M_s of 0.50 emu/cm^3 under magnetic field $H = 1.5$ T is observed for the BYFTO films, which is comparable to the value of $M_s = 0.21 \text{ emu/cm}^3$ for the $\text{Bi}_5\text{FeTi}_3\text{O}_{15}$ films [15]. As we assumed that, the replacement of BiFeO_3 by YFeO_3 is helpful to improve ferromagnetic behavior. First, the remanent magnetization of orthoferrites YFeO_3 is superior to that of BiFeO_3 at room temperature. Second, the insertion of YFeO_3 might lead to a more significant lattice distortion in the BYFTO lattice than that in the $\text{Bi}_5\text{FeTi}_3\text{O}_{15}$ lattice. Therefore, we deduced that the improvement of the ferromagnetic behavior is primarily due to the magnetization of the YFeO_3 itself and the lattice distortion [17].

Under the different applied magnetic fields (from 0 to 0.45 T), the frequency dependencies of the dielectric constant ε_r of the BYFTO films at room temperature are shown in Fig. 6. The dielectric constant increases with increasing magnetic field, which indicates that the magnetic ordering promotes the dielectric constant [37]. Under the applied magnetic fields, the spin reorientation can affect the interaction between dipoles which comes from the coexistence of Fe^{2+} and Fe^{3+} in the BYFTO films. Therefore the coupling between the dielectric constant and the magnetic field will take

place, as observed magneto-dielectric effect. Although the magneto-dielectric coefficient (MDC) value, defined as $MDC = [\epsilon_r(0.45T) - \epsilon_r(0T)] \times 100 / \epsilon_r(0T)$, is relatively small $\sim 0.78\%$, presented in the inset of Fig. 6, the magneto-dielectric effect can clearly illustrate the coupling between the electric and magnetic orders. Compared with 0.7% MDC at 5 K under a large magnetic field 5 T in the BFTO thin film, the magneto-dielectric effect in our BYFTO films at room temperature shows more valuable [38].

4. Conclusion

In summary, single-phase Aurivillius structure $\text{Bi}_4\text{YFeTi}_3\text{O}_{15}$ (BYFTO) thin films of four-layered perovskite were prepared using a sol-gel process. The films exhibit room-temperature multiferroic properties with $2P_r \sim 53.62 \mu\text{C}/\text{cm}^2$ and $M_s \sim 0.50 \text{ emu}/\text{cm}^3$. Both the dielectric permittivity and dielectric loss of the BYFTO films have the features of relaxation around $T = 72 \text{ K}$ and the activation energy $E_a = 0.29 \text{ eV}$. More interestingly, the magneto-dielectric effect of the films was exhibited with a magneto-dielectric coefficient of $\sim 0.78\%$ at the frequency of 1 MHz at room temperature. The dielectric relaxation behavior and magneto-dielectric effect of the films might originate from the coexistence of the Fe^{2+} and Fe^{3+} . The leakage current density of the BYFTO films remains lower than $1.03 \times 10^{-5} \text{ A}/\text{cm}^2$ under the electric field below 300 kV/cm at 300 K. Further investigation of the ferroelectric dynamics shows that the films have good frequency stability and better application prospect in multi-states memory.

Acknowledgements This work was supported by the National Science Foundation of China (51472131, 11374169), the Distinguished Middle-aged and Young Scientist Encourage and Reward Foundation of Shandong Province, China (No. BS2013CL006), open project of National Laboratory of Solid State Microstructures (M27003), and China Postdoctoral Science Foundation (2016M590617). H.M. Yin thanks the project supported by the Joint Funds of Xinjiang Natural Science Foundation, China (No. 2015211C296).

References:

- [1] W. Eerenstein, N.D. Mathur, J.F. Scott, *Nature* 442 (2006) 759–765.
- [2] T. Li, J.F. Shen, N. Li, M.X. Ye, *J. Alloys Comp.* 548 (2013) 89–95.
- [3] Y. Sui, C. Xin, X.Q. Zhang, Y. Wang, Y. Wang, X.J. Wang, Z.G. Liu, B.S. Li, X.Y. Liu, *J. Alloys Comp.* 645 (2015) 78–84.
- [4] V.E. Wood, A.E. Austin, A.J. Freeman, H. Schmid, Gordon and Breach, New York (1975).
- [5] F. Jona, G. Shirane, *Ferroelectric Crystals* Dover, New York (1993).
- [6] G. Catalan, J.F. Scott, *Adv. Mater.* 21 (2009) 2463–2485.
- [7] G.H. Jaffari, A. Samad, A.M. Iqbal, S. Hussain, A. Mumtaz, M.S. Awan, M. Siddique, S. I. Shah, *J. Alloys Comp.* 644 (2015) 893–899.
- [8] M.K. Sharif, M.A. Khan, A. Hussain, F. Iqbal, I. Shakir, G. Murtaza, M.N. Akhtar, M. Ahmad, M.F. Warsi, *J. Alloys Comp.* 667 (2016) 329–340.
- [9] J. Wang, J.B. Neaton, H. Zheng, V. Nagarajan, S.B. Ogale, B. Liu, D. Viehland, V. Vaithyanathan, D.G. Schlom, U.V. Waghmare, N.A. Spaldin, K.M. Rabe, M. Wuttig, R. Ramesh, *Science* 299 (2003) 1719–1722.
- [10] D. Lebeugle, D. Colson, A. Forget, M. Viret, P. Bonville, J.F. Marucco, S. Fusil, *Phys. Rev. B* 76 (2007) 024116.
- [11] V.V. Shvartsman, W. Kleemann, *Appl. Phys. Lett.* 90 (2007) 172115.
- [12] R.S. Singh, T. Bhimasankaram, G.S. Kumar, S.V. Suryanarayana, *Solid State Commun.* 91 (1994) 567–569.
- [13] S.K. Kim, M. Miyayama, H. Yanagida, *Mater. Res. Bull.* 31 (1996) 121–131.

- [14] A. Snedden, C.H. Hervoches, P. Lightfoot, *Phys. Rev. B* 67 (2003) 092102.
- [15] W. Bai, Y.Q. Gao, J.Y. Zhu, X.J. Meng, T. Lin, J. Yang, Z.Q. Zhu, J.H. Chu, J. *Appl. Phys.* 109 (2011) 064901.
- [16] S.J. Ahn, J.H. Lee, Y.K. Jeong, E.H. Na, Y.M. Koo, H.M. Jang, *Mater. Chem. and Phys.* 138 (2013) 929–936.
- [17] M.Y. Shang, C.Y. Zhang, T.S. Zhang, L. Yuan, L. Ge, H.M. Yuan, S.H. Feng, *Appl. Phys. Lett.* 102 (2013) 062903.
- [18] J.H. Li, Y. Qiao, X.L. Liu, C.J. Nie, C.J. Lu, *Appl. Phys. Lett.* 85 (2004) 15.
- [19] F. Kubel, H. Schmid, *Ferroelectrics* 129 (1992) 101–112.
- [20] M. Rao, H.R. Krishnamurthy, R. Pandit, *Phys. Rev. B* 42 (1990) 856–884.
- [21] Y.Y. Guo, T. Wei, Q.Y. He, J.M. Liu, *J. Phys. Condens. Matter.* 21 (2009) 485901.
- [22] X.F. Chen, X.L. Dong, Z.Y. Zhou, J.X. Wang, F. Cao, G.S. Wang, H.L. Zhang, J. *Appl. Phys.* 115 (2014) 124103.
- [23] Y.C. Zhang, C.J. Lu, Z.Z. Yang, W.N. Ye, L.H. Xia, *J. Appl. Phys.* 111 (2012) 084104.
- [24] J. Ge, Y. Chen, X.L. Dong, D. Rémiens, X. Guo, F. Cao, G.S. Wang, *Thin Solid Films* 584 (2015) 108–111.
- [25] J.M. Liu, H.L. Chan, C.L. Choy, *Mater. Lett.* 52 (2002) 213-219.
- [26] J.M. Liu, B. Pan, H. Yu, S.T. Zhang, *J. Phys: Condens. Matter.* 16 (2004) 1189-1195.
- [27] Y.H. Lee, J.M. Wu, Y.L. Chueh, L.J. Chou, *Appl. Phys. Lett.* 87 (2005) 172901.
- [28] M.A. Lampert, *Rep. Prog. Phys.* 27 (1964) 329-367.
- [29] J.F. Scott, Springer New York (2000).

- [30] D.P. Song, J. Yang, B. Yuan, X.Z. Zuo, X.W. Tang, L. Chen, W.H. Song, X.B. Zhu, Y.P. Sun, J. Appl. Phys. 117 (2015) 244105.
- [31] M.A. Lampert, Phys. Rev. Vol. 103 (1956) 1648-1656.
- [32] B. Yuan, J. Yang, X.Z. Zuo, D.P. Song, X.W. Tang, X.B. Zhu, J.M. Dai, W.H. Song, Y.P. Sun, J. Appl. Phys. 117 (2015) 233906.
- [33] H.S. Shulman, D. Damjanovic, N. Setter, J. Am. Ceram. Soc. 83 (2000) 528-532.
- [34] M. Maglione, M.A. Subramanian, Appl. Phys. Lett. 93 (2008) 032902.
- [35] N. Ikeda, H. Ohsumi, K. Ohwada, K. Ishii, T. Inami, K. Kakurai, Y. Murakami, K. Yoshii, S. Mori, Y. Horibe, H. Kito, Nature 436 (2005) 1136-1138.
- [36] J. Su, X.M. Lu, J.T. Zhang, H. Sun, C. Zhang, Z.H. Jiang, C.C. Ju, Z.J. Wang, F.Z. Huang, J.S. Zhu, J. Appl. Phys. 111 (2012) 014112.
- [37] W. Bai, C. Chen, J. Yang, Y.Y. Zhang, R.J. Qi, R. Huang, X.D. Tang, C.G. Duan & J.H. Chu, Sci. Rep. 5 (2015) 17846.
- [38] X.W. Dong, K.F. Wang, J.G. Wan, J.S. Zhu, J.M. Liu, J. Appl. Phys. 103 (2008) 094101.

Figure Captions

Fig. 1. (a) XRD pattern and (b) cross-section SEM image of the BYFTO thin films deposited on (111)Pt/Ti/SiO₂/Si.

Fig. 2. (a) P - E hysteresis loops of the BYFTO films. (b) The scaling relations for the saturated loops of the BYFTO films. The solid line represents the linear fitting, and the goodness of R^2 is 0.992.

Fig. 3. (a) Temperature dependence of leakage current for the BYFTO films as a function of electric field in the range of 50 K-300 K. (b) Analysis of leakage current of BYFTO films for Ohmic/SCLC/TFL mechanism at 300 K. The Slopes of linear fit plots are shown.

Fig. 4. Temperature dependence of (a) the dielectric constant ϵ_r and (b) dielectric loss $\tan\delta$ at various frequencies of the BYFTO films. The inset shows the Arrhenius plot of relaxation frequency vs. temperature.

Fig. 5. The M - H curve of the BYFTO thin films measured at room temperature. The inset shows the detail of the low magnetic field region.

Fig. 6. The frequency dependence of dielectric constant under zero, 0.25 T and 0.45 T in the BYFTO films at room-temperature. The inset is the MDC value as a function of the measurement frequency.

Table 1

The comparison of the scaling relation of the BYFTO films with the theoretically predicted relation and those for other ferroelectric thin films.

system	a	b	References
$(\Phi^2)^2$ and $(\Phi^2)^3$ models	-1	2	[20]
PZT thin films	-1/3	3	[25]
BNT thin films	-2/3	2	[26]
PBZT thin films	-0.2	2.2	[21]
BYFTO thin films	-0.17	2.41	This work

Figure 1

Figure 2

Figure3

Figure 4

Figure 5

Figure 6