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Multiferroic and Nanomechanical Properties of $\text{Bi}_{1-x}\text{Gd}_x\text{FeO}_3$ Polycrystalline Films ($x = 0.00\text{--}0.15$)

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Abstract: In this work, we adopted pulsed laser deposition (PLD) with a Nd:YAG laser to develop $\text{Bi}_{1-x}\text{Gd}_x\text{FeO}_3$ (BGFO) films on glass substrates. The phase composition, microstructure, ferroelectric, magnetic, and nanomechanical properties of BGFO films are studied. BGFO films with $x = 0.00\text{--}0.15$ were confirmed to mainly consist of the perovskite phase. The structure is transformed from rhombohedral for $x = 0.00$ to pseudo-cubic for $x = 0.05\text{--}0.10$, and an additional phase, orthorhombic, is coexisted for $x = 0.15$. With increasing Gd content, the microstructure and surface morphology analysis shows a gradual decrease in crystallite size and surface roughness. The hardness of 5.9–8.3 GPa, measured by nanoindenter, is mainly dominated by crystallized structure and grain size. Good ferroelectric properties are found for BGFO films with $x = 0.00\text{--}0.15$, where the largest remanent polarization ($2P_r$) of 133.5 $\mu\text{C}/\text{cm}^2$ is achieved for $x = 0.10$, related to low leakage and high BGFO(110) texture. The improved magnetic properties with the significant enhancement of saturation magnetization from 4.9 emu/cm^3 for $x = 0$ to 23.9 emu/cm^3 for $x = 0.15$ by Gd substitution is found and related to large magnetic moment of Gd^{3+} and suppressed spiral spin structure of G-type antiferromagnetism. Furthermore, we also discuss the mechanisms of leakage behavior as well as nanomechanical characterizations as a function of the Gd content.

Keywords: multiferroics; Gd-doped BFO; nanomechanical properties; microstructure

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1. Introduction

BiFeO_3 (BFO), recognized as a room-temperature (RT) multiferroic (MF) material, has two main ferro-ordering parameters, including ferroelectricity (FE) at temperature (T) below 830 °C [1] and antiferromagnetism (AF) at T below 370 °C [2]. However, it exhibits weak ferromagnetism at RT due to antisymmetric exchange or the Dzyaloshinskii–Moriya interaction [3]. Because of the exceptional FE properties, multiferroic nature, potential magnetoelectric (ME) coupling, and lead-free character, the single-phase MF BFO has received considerable attention last decade. The above properties present the potential applications in non-volatile ferroelectric data storage, the emerging area of spintronics, and ME sensors.

High FE appears in BFO due to the presence of stereo-chemically active Bi^{3+} ions with 6s lone pair electrons. Li et al. [4] reported that the epitaxial pseudo-cubic BFO(001) thin films, grown on $\text{SrRuO}_3/\text{SrTiO}_3$ (100) substrates by a pulse laser deposition (PLD), exhibit a surprisingly large remnant polarization ($2P_r$) response of up to $2P_r \sim 120 \mu\text{C}/\text{cm}^2$, which is one order of magnitude higher than that of bulk BFO ($\sim 6.1 \mu\text{C}/\text{cm}^2$) at that

time [5]. Shvartsman et al. [6] showed that large FE polarization $\sim 40 \mu\text{C}/\text{cm}^2$ was attained in ceramic BFO, polycrystalline, and epitaxial films. The above results indicate that larger polarization comes from the intrinsic nature of BFO, which could also be supported by the first-principles calculations [7]. On the other hand, the magnetic nature of BFO is a G-type AF order, where Fe^{3+} ions are surrounded by six neighboring Fe^{3+} ions with spin antiparallel to the central ion [8]. However, BFO exhibits weak FM properties due to the spiral magnetic spin cycloid. Therefore, the cross-coupling between these ferroic parameters in BFO is weak and is poorly understood due to weak FM.

Although FE properties are prominent, the major drawbacks for the BFO-related devices are high leakage, a tendency towards fatigue [9], and weak FM. Ion modification with rare-earth cations has been demonstrated to be a successful method for the reduction in leakage and the improvement of magnetic properties at once [10]. FE and FM enhancements were also found in A-doped BFO ceramics and films (A = Sm, La, Pr, Gd, Dy, and Ca), due to the suppressed oxygen vacancies in BFO caused by the proper A substitution [11–14]. Among rare earth elements, Gd has higher exchange interaction and large magnetic moment [15]. Gd addition into BFO ceramics could improve FE and FM properties [16], but the MF properties are too low for practical applications up to now. Although the investigations related to A-site-doped BFO were reported, Gd-doped BFO thin films are still lacking. Besides, it is desirable and important to understand mechanical properties of the films, since most practical applications of functional devices are fabricated with thin films.

Therefore, $\text{Bi}_{1-x}\text{Gd}_x\text{FeO}_3$ ($x = 0.00, 0.05, 0.10$, and 0.15) (BGFO) polycrystalline films are prepared on the glass substrates by pulsed laser deposition in this work. A systematic investigation of the structural evolution, surface morphology, microstructure, leakage mechanisms, ferroelectric, and magnetic properties of BGFO films is reported.

2. Experiment

The solid-state reaction method was used to prepare $\text{Bi}_{1-x}\text{Gd}_x\text{FeO}_3$ targets ($x = 0.00$ – 0.15), as described elsewhere [17]. A PLD method was employed to deposit BGFO thin films on Pt buffered Corning's code 1737 glass substrates. A 20-nm-thick Pt layer was grown on a glass substrate as a bottom electrode by sputtering with a power of 60 W within Ar atmosphere of 10 mTorr at RT. A pulsed Nd:YAG laser with a laser wavelength of 355 nm was used to deposit 300-nm-thick BGFO films on the Pt layer at a substrate temperature of 300–500 °C and an O_2 pressure of 30 mTorr. Pulsed laser energy of 2.5 mJ with a repetition rate of 5 Hz was used for BGFO deposition.

The chemical composition of the BGFO thin films was determined using X-ray fluorescence analysis, and double-checked by an energy-dispersive X-ray spectroscopy analysis. X-ray diffractometry (XRD; PANalytical X'Pert PRO, Almelo, The Netherlands) with copper $\text{K}\alpha$ radiation was used to determine the phase and crystallographic orientations. The XRD pattern was refined and structural parameters were determined using the High-Score Plus version 3.0 software. The inorganic crystal structure database was used to infer pseudo-cubic (space group: $Pm3m$), rhombohedral (space group: $R3c$), orthorhombic (space group: $Pnma$), and orthorhombic $\text{Bi}_2\text{Fe}_4\text{O}_9$ (space group: $Pbam$) structures [18–21]. The surface and grain morphologies were observed by atomic force microscopy (AFM; Force Genie AFM, Taiwan, China). The microstructure was directly observed by transmission electron microscopy (TEM; JEOL JEM-2100, Tokyo, Japan). Sputtering was used to deposit a circular-shaped Pt layer with a diameter of 200 μm as a top electrode for following ferroelectric measurements. FE behavior and leakage test were analyzed using an TF-2000 Analyzer FE-Module ferroelectric test device (axiACCT Co., Aachen, Germany). Magnetization–magnetic field (M - H) curves of BGFO films were measured using a vibrating sample magnetometer.

The nanomechanical properties of BGFO films were measured using a nanoindenter with a Berkovich tip, and the hardness was defined as the applied indentation load divided by the projected contact area as follows:

$$H = \frac{P_{max}}{A_p} \quad (1)$$

where A_p is the projected contact area between the indenter and the sample surface at the maximum indentation load, P_{max} . For a perfectly sharp Berkovich indenter, the projected area A_p was given by $A_p = 24.56h_c^2$ with h_c being the true contact depth. In general, the indentation depth should never exceed 30% of the film thickness to avoid the substrate effect on hardness measurements [22].

3. Results and Discussion

At first, the phase composition of BGFO films ($x = 0.00$ – 0.15) at 400°C is analyzed and the XRD patterns are shown in Figure 1. A strong Pt(111) diffraction peak found in all samples reveals a flat and well-crystallized Pt(111) underlayer, which could help to grow following BGFO films. The BFO film consists of a rhombohedral structure, $R3c$ space group, and no other phase is detected. For Gd substitution for Bi in BGFO films, the phase transformation is observed. When x is increased from 0 to 0.05, BFO(104) and BFO(110) peaks merge into a single peak of BGFO(110). It indicates that the rhombohedral structure is transformed into a pseudo-cubic structure with Gd substitution, in good accordance with previous literature [23]. With increasing x from 0.05 to 0.10, the texture of pseudo-cubic structure changes from isotropic orientation to (110). For the sample with a higher $x = 0.15$, the orthorhombic structure with a space group of $Pnma$ is found to coexist with the pseudo-cubic structure. The presence of the orthorhombic phase for higher Gd content is consistent with the result of BGFO bulks [24]. Additionally, the shift of diffraction peaks for the perovskite phase toward a higher angle with the increase in Gd content x in Figure 1 reveals the entry of substituted Gd^{3+} ion into the perovskite BGFO phase to occupy A site due to smaller ionic radius of Gd^{3+} than Bi^{3+} .

Figure 1. XRD patterns of $\text{Bi}_{1-x}\text{Gd}_x\text{FeO}_3$ ($x = 0.00, 0.05, 0.10$, and 0.15) films.

TEM images of $\text{Bi}_{1-x}\text{Gd}_x\text{FeO}_3$ with $x = 0.00, 0.05, 0.10$, and 0.15 are shown in Figure 2a–d. The average grain size, estimated through Image J software, is refined from 63 to 26 nm when Gd content x is increased from 0.00 to 0.15. The size distribution of the grains is also analyzed, and the distribution histograms by fitting with Gaussian distribution are

shown in Figure 2e–h. The singlet distribution for $x = 0.00$ – 0.10 and dual size distribution for $x = 0.15$ are found. When x is increased from 0.00 to 0.10, the size and distribution of the grain becomes gradually small and narrow, shown in Figure 2e–g. As x is further increased to 0.15, there are two distributions for grain size in Figure 2h. According to XRD analysis, smaller grains belong to the orthorhombic phase for the sample with $x = 0.15$. Change in grain size with Gd substitution and the content x might be related to the evolution and texture of the structure.

Figure 2. TEM images of $\text{Bi}_{1-x}\text{Gd}_x\text{FeO}_3$ films: (a) $x = 0.00$, (b) 0.05 , (c) 0.10 , and (d) 0.15 . Grain size distribution of the films: (e) $x = 0$, (f) 0.05 , (g) 0.10 , and (h) 0.15 .

AFM is used to observe the surface morphology and their AFM images are shown in Figure 3. Flat surface with the root-mean-square-roughness (R_{rms}) in the range of 6.7–13.4 nm is found. R_{rms} is decreased from 13.4 to 6.7 nm as x is increased from 0.00 to 0.15. The flattened surface with Gd substitution is possibly related to grain refinement, shown in Figure 2.

Figure 3. AFM images of $\text{Bi}_{1-x}\text{Gd}_x\text{FeO}_3$ films: (a) $x = 0.00$, (b) 0.05, (c) 0.10, and (d) 0.15.

For $\text{Bi}_{1-x}\text{Gd}_x\text{FeO}_3$ films with $x = 0.00, 0.05, 0.10$, and 0.15 films, polarization electric (P - E) field curves are shown in Figure 4a–d. All films exhibit a typical FE behavior with good FE properties. With increasing x , the remanent polarization ($2P_r$) increases from $41 \mu\text{C}/\text{cm}^2$ for $x = 0.00$ to $72 \mu\text{C}/\text{cm}^2$ for $x = 0.05$, and $2P_r$ enhancement may be due to the suppressed vacancies [25]. As x is increased to 0.10, $2P_r$ further increases up to a maximum value of $120 \mu\text{C}/\text{cm}^2$, related to the texture transformation, from isotropy to (110). Finally, $2P_r$ decreases to $38 \mu\text{C}/\text{cm}^2$ for higher Gd content of $x = 0.15$, related to the appearance of the paraelectric orthorhombic phase. On the other hand, with increasing Gd content, the coercive field (E_c) monotonically decreases from $320 \text{ kV}/\text{cm}$ for $x = 0.00$ to $248 \text{ kV}/\text{cm}$ for $x = 0.15$. The reduction in E_c with x may result from two facts: phase transformation and grain refinement, which decrease the energy barrier for the switching of FE polarization [26].

Figure 5a plots the leakage current density (J) versus the external electric field (E) for $\text{Bi}_{1-x}\text{Gd}_x\text{FeO}_3$ films. Large J is found for BFO film, which might be related to the existence of oxygen vacancies and rougher interfaces. For Gd-substituted films, a dramatic decrease in more than one order of magnitude in leakage is found. The suppressed leakage, possibly related to phase transformation and flattened surface, improves FE properties. However, due to the appearance of the paraelectric orthorhombic phase, the FE properties is decreased even though the leakage is further reduced at $x = 0.15$ in this study.

Figure 4. P - E curves of $\text{Bi}_{1-x}\text{Gd}_x\text{FeO}_3$ films: (a) $x = 0.00$, (b) 0.05 , (c) 0.10 , and (d) 0.15 .

Figure 5. (a) Leakage current density and (b) leakage mechanism of $\text{Bi}_{1-x}\text{Gd}_x\text{FeO}_3$ ($x = 0.00, 0.05, 0.10$, and 0.15) films.

In order to further understand the leakage mechanisms of this studied films, five conduction mechanisms adopted to fit are as follows [27,28]:

For Ohmic Conduction

$$J_{Ohmic} = nq\mu E \Rightarrow \log(J) \propto \log(E), \quad (2)$$

where n is the number of charge carriers, q is the electronic charge, and μ is the carrier mobility.

For Space Charge Limited Conduction (SCLC)

$$J_{SCLC} = \frac{9\mu\epsilon E^2}{8d} \Rightarrow \log(J) \propto 2\log(E) \quad (3)$$

where ϵ is the dielectric constant, and d is the film thickness.

For Poole–Frenkel Emission (PF)

$$J_{PF} = A E \exp \left[-q \left(\frac{\varphi_T - \sqrt{\frac{qE}{\pi\epsilon_r\epsilon_0}}}{kT} \right) \right] \Rightarrow \log \left(\frac{J}{E} \right) \propto \frac{\sqrt{\frac{q^3}{4\pi\epsilon_r\epsilon_0}}}{kT} E^{\frac{1}{2}} \quad (4)$$

where A is a constant, φ_T is the trap ionization energy, ϵ_r is the dynamic (or optical) dielectric permittivity, ϵ_0 is the permittivity of free space, k is the Boltzmann constant and T is the thermodynamic temperature.

For Schottky Emission

$$J_{Schottky} = B T^2 \exp \left[-q \left(\frac{\varphi_B - \sqrt{\frac{qE}{4\pi\epsilon_r\epsilon_0}}}{kT} \right) \right] \Rightarrow \log(J) \propto \frac{\sqrt{\frac{q^3}{4\pi\epsilon_r\epsilon_0}}}{kT} E^{\frac{1}{2}} \quad (5)$$

where B is a constant, and φ_B is the interface potential barrier.

For Fowler–Nordheim (FN) Tunneling

$$J_{FN} = C \frac{E^2}{\varphi_B} \left[\frac{D}{E} \varphi_B^{\frac{3}{2}} \right] \Rightarrow \log \left(\frac{J}{E^2} \right) \propto \frac{1}{E} \quad (6)$$

where C is a constant.

The analyses will make assess if the leakage current follows Ohmic, SCLC, Schottky, PF emission, or FN tunneling, as described in Equations (2)–(6). Whether ϵ_r , obtained from the slopes, is within the range of 6.25–9.61 [29] for the developed BFO films is adopted to determine the mechanism dominated by Schottky or PF emission.

The fitting results and the evolution of leakage mechanism with E are summarized in Figure 5b. For all samples, Ohmic mechanism dominates at low E , and PF mechanism, which electrons can thermally emit into the conduction band from trapping centers, governs at larger E . In addition to Ohmic and PF mechanisms, SCLC mechanism is also found for BFO film, but disappears for BGFO films. The disappearance of the SCLC mechanism with Gd substitution, which may be a result of the reduced oxygen vacancy, is helpful in improving FE properties. Besides, for higher Gd content, FN tunneling occurs at larger E . FN tunneling currents which generally occur in the relatively insulating situation, for instance, from a solid surface into a vacuum or any weakly conducting dielectric. In this case, the exhibited FN tunneling mechanism proves the improved insulating ability with Gd substitution.

Especially near the morphotropic phase boundaries (MPBs), the structure in BFO could be easily affected by external electric field, stress, and growth temperature [30]. To optimize the FE characteristics and to explore the relationship between structures and growth temperature, structure and FE properties of the films with $x = 0.10$ at various growth temperatures (T_g) in range of 300–500 °C are also studied. XRD patterns are shown in Figure 6. At lower T_g , no diffraction peak belonging to the perovskite structure is found because heat energy is too low to form the perovskite phase. With increasing T_g from 350

to 450 °C, the peaks belonging to pseudo-cubic structure appear to become stronger, and besides, the orientation of crystal is changed from almost isotropy to BGFO(110) texture. At higher $T_g = 500$ °C, the texture of BGFO films is changed to (001), and an extra weak peak, belonging to the $\text{Bi}_2\text{Fe}_4\text{O}_9$ phase, appears possibly related to the volatilization of Bi at higher T_g . For the film grown at $T_g = 500$ °C, (110) and (002) diffraction peaks are split into (110), (121)_o, (002)_o and (002), (202)_o, (040)_o, respectively, where (121)_o, (002)_o, (202)_o, and (040)_o belong to the diffraction peaks of the orthorhombic phase. The appearance and coexistence of the orthorhombic phase for BGFO with $x = 0.10$ at higher T_g is consistent with phase diagram reported in the previous research of Kan et al. [30].

Figure 6. XRD patterns of $\text{Bi}_{0.9}\text{Gd}_{0.1}\text{FeO}_3$ ($T_g = 300, 350, 400, 450$, and 500 °C) films.

For $\text{Bi}_{0.9}\text{Gd}_{0.1}\text{FeO}_3$ films with $T_g = 350$ – 500 °C, electrical polarization electric (P - E) field curves are shown in Figure 7a–d. At lower T_g , $= 300$ °C, no FE behavior is found due to the absence of the perovskite phase. Good P - E properties are attained at $T_g = 350$ – 500 °C. As T_g is increased from 350 to 500 °C, $2P_r$ is increased from $69.6 \mu\text{C}/\text{cm}^2$ at $T_g = 350$ °C to a maximum $2P_r = 133.5 \mu\text{C}/\text{cm}^2$ at $T_g = 450$ °C, and finally decreased to $58.8 \mu\text{C}/\text{cm}^2$ at $T_g = 500$ °C. The improved FE properties with increasing T_g is related to the promoted crystallinity and texture constitution, and importantly, the highest $2P_r$ could be mainly attributed to BGFO (110) texture at 450 °C. At higher $T_g = 500$ °C, due to low breakdown voltage caused by high leakage, the saturation hysteresis curve cannot be obtained.

The curves of the leakage current density (J) versus the external electric field (E) of $\text{Bi}_{0.9}\text{Gd}_{0.1}\text{FeO}_3$ films at $T_g = 350$ – 500 °C are shown in Figure 8. Low leakage, observed for BGFO at $T_g = 350$ – 500 °C, contributes to good FE properties. The slight increase in leakage with increasing T_g is presumably related to texture evolution, and the appearance of 2:4:9 phase at higher $T_g = 500$ °C.

Figure 7. P - E curves of $\text{Bi}_{0.9}\text{Gd}_{0.1}\text{FeO}_3$ films: (a) $T_g = 350^\circ\text{C}$, (b) 400°C , (c) 450°C , and (d) 500°C .

Figure 8. Leakage current density of $\text{Bi}_{0.9}\text{Gd}_{0.1}\text{FeO}_3$ ($T_g = 350, 400, 450$, and 500°C) films.

Additionally, the magnetic properties of $\text{Bi}_{1-x}\text{Gd}_x\text{FeO}_3$ films are investigated, too. Their M - H curves are depicted in Figure 9. All BGFO films exhibit ferromagnetic behavior and exhibit enhanced magnetic properties. The saturation magnetization (M_s) increases dramatically from 4.9 to 23.9 emu/cm^3 , but the magnetic coercive field (H_c) drops from 502 to 350 Oe as the Gd content increases from 0.00 to 0.15. The magnetization enhancement due to Gd substitution is presumably related to the distorted Fe-O-Fe bond, influencing the Fe-O-Fe exchange way, and the magnetic moment of Gd^{3+} ion [31].

Figure 9. M - H curves of $\text{Bi}_{1-x}\text{Gd}_x\text{FeO}_3$ ($x = 0.00, 0.05, 0.10$, and 0.15) films.

The curves of hardness versus load penetration depth are illustrated in Figure 10a. Above a depth of 120 nm, the hardness of all samples reduces to a constant value. Generally, the relatively high hardness at a small depth results from the transition from fully elastic to elastic/plastic contact. The hardness subsequently decreases to a stable value. At this stage, the hardness value obtained is considered to be the inherent hardness for BGFO films. The hardness is therefore determined based on average values of hardness at depths of 60 to 90 nm. This range of penetration depth was chosen intentionally to be deep enough for observing plastic deformation during indentation, yet shallow enough to avoid the complications arising from the effects of surface roughness [22] and substrate [32]. With increasing Gd content, the hardness increases from 5.9 GPa for $x = 0.00$ to 8.3 GPa for $x = 0.10$, and then decreases to 8.0 GPa for $x = 0.15$. The change in hardness with x might be related to the phase and texture constitutions and grain refinement. With increasing x from 0.00 to 0.05, the denser pseudo-cubic structure forms and replaces rhombohedral, where the densities of rhombohedral and pseudo-cubic structure, estimated from XRD, are 8.33 and 8.47 g/cm³, respectively. With further increasing x to 0.10, the change in texture for pseudo-cubic structure from isotropy to (110) might also help the increase in hardness. In addition to evolutions of the phase and texture, grain size refinement with x , demonstrating the grain boundary's effectiveness in impeding dislocation movement, also contributes to hardness enhancement. When x is further increased to 0.15, the slight decrease in hardness might be related to the presence of the orthorhombic phase.

MF and mechanical characteristics of the Gd-doped BFO bulks and films, which have been developed up to now [33–37], are listed in Table 1. First of all, BGFO bulks exhibit poor ferroelectric and weak ferromagnetic properties. Besides, the significant improvement in ferroelectric properties is found for BGFO polycrystalline films, especially for $\text{Bi}_{0.95}\text{Gd}_{0.05}\text{FeO}_3$ with the increased $2P_r$ of 90 $\mu\text{C}/\text{cm}^2$ [36]. In general, simultaneously improving ferroelectric and ferromagnetic properties is extremely difficult. In this study, BGFO films deposited on the refined Pt(111) underlayer at the reduced temperature of 400–450 °C show superior FE and FM properties. Furthermore, $2P_r$ of 133.5 $\mu\text{C}/\text{cm}^2$ and M_s of 19.1 emu/cm³ attained for the $\text{Bi}_{0.9}\text{Gd}_{0.1}\text{FeO}_3$ thin film suggests that Gd-substituted BFO thin films on a Pt electrode buffered glass substrate at a low deposition temperature become a useful multiferroic material.

Figure 10. (a) Hardness versus depth curves and (b) Hardness versus grain size $^{-1/2}$ of $\text{Bi}_{1-x}\text{Gd}_x\text{FeO}_3$ films.

Table 1. Comparison of the developed Gd-doped BFO bulks and films: the multiferroic and nanomechanical characteristics.

Composition	Sample Form	$2P_r$ ($\mu\text{C}/\text{cm}^2$)	E_c (kV/cm)	M_s (emu/cm^3)	H_c (kOe)	Hardness (GPa)	Ref.
0.05	Bulk	25.6	107.0	~ 0.13	1.5	-	[33]
0.15	Bulk	-	-	~ 1.4	~ 5.0	-	[23]
0.10	Bulk	15	50	0.05	~ 2.0	-	[34]
0.10	Polycrystalline Films	72.6	196.5	-	-	-	[35]
0.05	Polycrystalline Films	~ 90	~ 300	-	-	-	[36]
0.10	Polycrystalline Films	80	5.0	8.33	< 10.0	-	[37]
0.05	Polycrystalline Films	71.5	289.6	11.6	502	7.5	This work
0.10		133.5	275.6	19.1	395	8.3	
0.15		34.7	247.5	23.9	350	8.0	

4. Conclusions

The effects of Gd content on the structure, microstructure, MF, and nanomechanical properties of BGFO thin films deposited by PLD on 20-nm-thick Pt underlayer buffered glass substrates at reduced substrate temperatures of 400–450 °C were reported in this work. The perovskite phase in BGFO films with $x = 0.00$ – 0.15 was verified. The phase transition from a rhombohedral for $x = 0.00$ to a pseudo-cubic for $x = 0.05$ – 0.10 , and an additional orthorhombic phase for higher $x = 0.15$ was found. The surface roughness and grain size of BGFO decreased as the Gd content increases. The hardness of BGFO thin films ranged from 5.9 to 8.3 GPa, and nanomechanical properties were strongly dependent on the phase and texture constitutions and grain size. Furthermore, it was found that microstructure, surface morphology, and Gd content all have a strong influence on ferroelectric properties. For BGFO films with $x = 0.05$ – 0.15 , ferroelectric properties improved to $2P_r = 72$ – $133.5 \text{ C}/\text{cm}^2$ and E_c of 248–312 kV/m, possibly because of the suppressed leakage current which resulted from the suppressed oxygen vacancy, the flattened interface, and microstructure refinement. The enhanced M_s of 11.6–23.9 emu/cm^3 seen in BGFO films due to Gd substitution was caused by the magnetic moment of the Gd^{3+} ion and distortion of the Fe-O-Fe bond angle. This study indicates that Gd-substituted BFO thin films on a Pt electrode buffered glass substrate at the reduced deposition temperature may be a useful multiferroic material.

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