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Electric-field-induced phase switching in textured Ba-doped bismuth ferrite lead titanate

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Abstract— The template grain growth technique was used to synthesis textured 60BiFeO₃-PbTiO₃ (60:40BFPT) by using platelets of BaTiO₃ as template. Synchrotron measurement clearly showed textured 60:40BFPT. Moreover, in situ high energy synchrotron radiation was employed to investigate the influence of an external electric field on crystallographic structure of mixed phase 60:40BFPT. Application of an electric field ≥ 1 kV/mm resulted in phase transformation from mixed rhombohedral/tetragonal phases ($\approx 73.5\%$ tetragonal / 26.5% rhombohedral) to predominately tetragonal phase ($\approx 95\%$) at applied field of 6 kV/mm.

Keywords- Texture; Synchrotron; Phase transformation

I. INTRODUCTION

Investigation on materials with a morphotropic phase boundary is of interest due to the possible phase transition between rhombohedral and tetragonal sides. Induced phase transition by an electric field between the ferroelectric tetragonal and rhombohedral phases was highlighted by Park in piezoelectric materials such as single crystals of Pb(Zn_{1/3}Nb_{2/3})O₃-PbTiO₃ and Pb(Mg_{1/3}Nb_{2/3})O₃-PbTiO₃ [1, 2]. He showed how large d_{33} can be obtained for compositions near the MPB.

To have a successful electric field induced phase transition, alignment of the dipoles is critical. A material with an entirely random dipole direction (in the unpoled state) has a high potential energy to transform, which precludes a phase transition. Hence, investigations on electric field induced transitions in ferroelectric materials tend to occur in either single crystals or textured materials. However, phase transition in polycrystalline materials have been reported before, for example in PbZr_xTi_(1-x)O₃ (PZT) as well as K_{1/2}Bi_{1/2}TiO₃-Na_{1/2}Bi_{1/2}TiO₃ (KNBT) during the poling [3].

Due to some difficulties to grow single crystals such as expensive techniques, control of crystal stoichiometry etc.,

there is in textured structures, of which templated grain growth is one possible synthetic route.

The solid solution between bismuth ferrite and lead titanate (BFPT) possesses a morphotropic phase boundary (MPB) between the rhombohedral and tetragonal, with a spontaneous strain of 18% on the tetragonal side of the boundary. This amount of strain is the highest reported in a polycrystalline bulk ferroelectric [4]. It is possible that this crystallographic distortion could be harnessed during transformation, generating unprecedented electric field induced strains. In addition, in BFPT the antiferromagnetic Néel temperature drops by approximately 300K on crossing the MPB from the rhombohedral to the tetragonal side [5]. It is of interest to investigate the influence of field-driven rhombohedral-tetragonal phase transitions across the MPB, to determine whether correctly oriented BFPT can provide both giant piezoelectric properties and significant magnetoelectric coupling, ultimately, turning antiferromagnetic ordering off and on.

In the current investigation, we used template grain growth method to synthesis a texture 60:40BFPT by employing BaTiO₃ as template. Furthermore, the synchrotron analysis was employed for texture investigation. Moreover, the magnetic properties of the specimen were studied.

II. EXPERIMENTAL

60:40BFPT was made by using template grain growth method as detailed in [6]. Platelets of BaTiO₃ were supplied by Penn State and Anadolu Universities which were synthesized via the molten salt method [7].

Sintered pellets were cut into small bars (1 x 0.5 x 6 mm) and polished, followed by electroding for in-situ synchrotron measurements. Synchrotron diffraction was carried out at Beamline I15 at the Diamond Light Source facility (Oxfordshire, UK). Sample bars were placed in a thin polypropylene oil bath and submerged in silicone oil, to reduce the chance of electrical breakdown. A monochromatic high energy (57 keV) x-ray source, 100 x 100 μm in cross section,

was incident on the unelectroded 1 x 6 mm² bar surface, while a voltage was applied across the 1 mm dimension, in 1 kVmm⁻¹ steps. Such energies provide a high level of transmission through such dimensions of this material. Diffraction data was collected using a 2D detector (MAR 3450). The Debye rings were “caked” into individual 2 θ -intensity diffraction patterns, at +/- 5° between $\psi = 0^\circ$ and 355° as reported by Royles and Jones [3, 8]. By comparing 2 θ - I diffraction patterns collected at $\psi=0$ and 90°, where $\psi=0^\circ$ is parallel to the sample thickness, and $\psi=90^\circ$ is parallel to the casting direction, the texture can be gauged.

III. RESULTS

Figure 1 shows synchrotron data for 60:40BFPT made using 10% BaTiO₃, sintered at 1100 °C for 1 hour, revealing mixed rhombohedral/tetragonal phases, with no secondary phase. Increase in intensity of the (001), (002) tetragonal peaks and decrease in (100), (200) tetragonal peaks’ intensity are obtained by changing ψ from 90° to 0°. Comparing the data at normal to the cast direction ($\psi = 0^\circ$) and parallel to the cast direction (90°) shows a strong degree of texture. This is in agreement with what observed earlier by using synchrotron diffraction for texture analysis of 60:40BFPT [6].

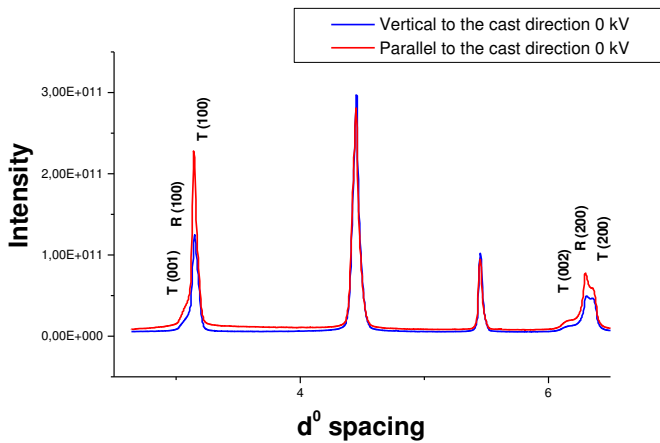


Figure 1. Synchrotron analysis data for 60:40BFPT made via TGG method by using 10% BaTiO₃ as template and sintered at 1100 °C for 1 hour. No external electric field applied on the sample.

Figure 2 shows the effect of the application of an electric field on the diffraction patterns of 60:40BFPT. It shows that upon the application of an electric field a significant change to the phase-contributions occurs, as indicated by disappearing of splitted 200 peak at approximately 6.35 Å, showing a transition from mixed phase to single phase. Moreover, 002 tetragonal peak appears by increasing the external field, indicating the gradual formation of a tetragonal single phase. Sample experienced breakdown at field more than 6 kV/mm. Starting

the phase transition even after applying low electric field at E=1 kV/mm could suggest the existence of textured structure.

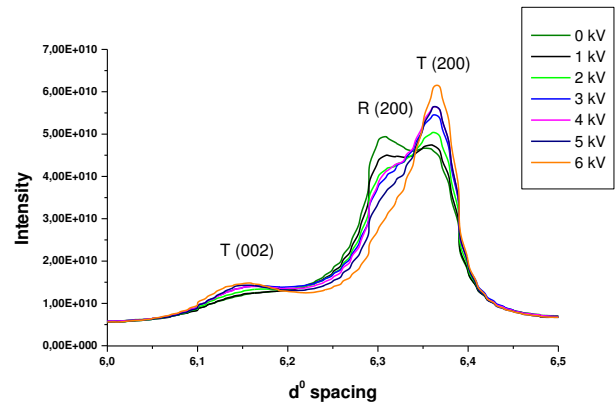


Figure 2. Diffraction patterns for 60:40 BFPT, after the application of electric field.

WinplotR software was used for profile fitting and also calculating the proportion of rhombohedral and tetragonal phases. The data revealed that before applying the external electric field the proportion of tetragonal phase was $\approx 73.5\%$ while at E= 6 kV/mm the proportion of tetragonal phase changed to $\approx 95\%$. A tetragonal c/a ratio of ≈ 1.04 was calculated, somewhat lower than that reported by Comyn et al. for 60:40BFPT pellets [9]. This could suggest reaction of the BaTiO₃ template with the matrix.

IV. CONCLUSION

In conclusion, synchrotron radiation experiments highlight significant differences in crystallographic orientation between perpendicular and parallel to the cast directions which suggests the existence of the textured structure in BFPT. We show that an electric field induced phase transition occurs for textured 60:40BFPT in the system from mixed phase rhombohedral and tetragonal to predominately tetragonal phase.

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