24P012-O

Structural and Electrical Properties of $BiFeO_3$ Thin Films Deposited on Polycrystalline Diamond Substrates

多結晶ダイヤモンド基板上への$BiFeO_3$薄膜の作製と特性評価

H. Kawasaki[1], T. Kawae[1], T. Nakajima[2], N. Tokuda[1], Y. Takano[3], S. Okamura[2] and A. Morimoto[1]
川崎寛樹[1], 川江 健[1], 中嶋宇史[2], 徳田規夫[1], 高野義彦[3], 岡村総一郎[2], 森本章治[1]

[1]*Kanazawa University,* [2]*Tokyo University of Science, and* [3]*National Institute for Materials Science*
[1]金沢大学、[2]東京理科大、[3]物質・材料研究機構

Pr and Mn co-doped $BiFeO_3$ (BPFM)/B-doped diamond layered film structure was fabricated on a polycrystalline diamond substrate. In the X-ray diffraction analysis, we observed the crystallization of BPFM and B-doped diamond without impurity phases in the present structure. In the polarization versus electric field curve, the BPFM/B-doped diamond layered film shows saturated hysteresis behavior at room temperature. The remnant polarization and coercive field for a maximum electric field of 1000 kV/cm were 135 μC/cm^2 and 700 kV/cm, respectively.

1. Introduction

Nonvolatile ferroelectric random access memories (FeRAM), composed of semiconductor and ferroelectrics, have attracted attention because of low power consumption and high-speed operation[1]. However, it is not easy to operate the FeRAM devices using Si and Pr(Zr, Ti)O_3(PZT) at high temperature due to the narrow gap of semiconductor and low Curie temperature T_C of ferroelectrics. Hence, the developing of FeRAM device materials with excellent physical properties for high temperature operation is required. For this requirement, we had proposed the integration of wide-gap semiconductor diamond and high-T_C ferroelectric material $BiFeO_3$[2], and reported the fabrication and characterization of BFO/B-doped diamond layered film capacitor on the single crystal diamond substrate[3]. In that structure, excellent ferroelectric properties were observed at room temperature and the maximum operation temperature was 150 ˚C [4].

On the other hand, due to the development of recent industrial technology for diamond substrate, large sized polycrystalline diamond wafer preparation has come to be established. Hence, in this work, we report on the fabrication of the BFO/B-doped diamond layered film structure on the polycrystalline diamond substrate and the investigation of their structural and electrical properties.

2. Experimental Methods

Vapor-grown polycrystalline diamonds were used as substrates of 36 mm^2 in areas (Element Six Technologies). Conductive B-doped diamond layer was grown on the substrate by microwave plasma-enhanced chemical vapor deposition method. The details of deposition conditions were described in the previous works[3-5]. Then, Pr and Mn co-doped BFO (BPFM)[5] thin film was deposited on the B-doped diamond layer using chemical solution deposition method. Here, Pr and Mn co-doping was used to reduce the leakage current of BFO film. The BPFM precursor solution was spin-coated on the B-doped diamond layer at 3000 rpm for 30 sec. After the coating, the specimen was dried at 240℃ for 5 min, and prefired at 350℃ for 10 min in air. This process was repeated 15 times to obtain the film of desired thickness. Finally, for crystallization, the film was annealed at 550 ℃ for 20 min in nitrogen atmosphere.

For electrical measurement, Au top electrodes of 2.025×10^{-5} cm^2 in areas were deposited on the BPFM film by conventional thermal evaporation system. The crystal structure of the specimen was determined by X-ray diffraction (XRD; Shimadzu XD-D1). The electrical properties were characterized using ferroelectric test system (Toyo FCE-1A).

3. Results & Discussion

Figure 1 shows the XRD θ-2θ scanning pattern of the BPFM/B-doped diamond layered structure on the polycrystalline diamond substrate. It is considered that the B-doped diamond layer is homoepitaxially grown on the polycrystalline diamond substrate since the diffraction peaks of diamond, other than (220), have not been detected in the result. Also, for the BPFM layer, the

polycrystalline growth of BPFM without any impurity phases was also observed in the figure. From the above result, the BPFM/B-doped diamond layered films are found to be crystallized on the polycrystalline diamond substrate in our deposition process.

Figure 2(a) shows the polarization versus electric field (*P-E*) curves of the BPFM thin film on the B-doped diamond layer. The curves were measured at room temperature with a measurement frequency of 10 kHz. As shown in the figure, saturated hysteresis loops were observed without influences of leakage current.

Figure 2(b) shows electric field dependence of remnant polarization $2P_r$ and coercive field $2E_c$ for the present structure. $2P_r$ and $2E_c$ at the maximum applied electric field of 1000 kV/cm were 135 μC/cm^2 and 700 kV/cm, respectively. Then, the observed $2P_r$ and $2E_c$ value were roughly comparable with that of the BPFM thin films on the Pt-coated Si substrate ($2P_r$=125 μC/cm, $2E_c$=600 kV/cm at maximum electric field of 1000 kV/cm). This indicates simultaneous achievement of excellent ferroelectric properties of the BPFM film and good conductivity of the B-doped diamond layer meaning that the dopant activation in the diamond layer was sufficient at room temperature. Hence, it is concluded that the present structure and processing technique are effective for an integration of BFO with diamond for high-temperature FeRAM device applications.

Summary

We have prepared the BPFM/B-doped diamond layered film structure on the polycrystalline diamond substrate and investigated the structural and electrical properties. The BPFM and B-doped diamond were crystallized on the polycrystalline diamond substrate. The BPFM/B-doped diamond layered film showed saturated hysteresis behavior at room temperature in the *P-E* curve. $2P_r$ and $2E_c$ for a maximum electric field of 1000 kV/cm were 135 μC/cm^2 and 700 kV/cm, respectively.

References

[1] J. F. Scott *et al*., Science, **246** (1989) 1400.
[2] Y. N. Venestsev *et al*., Sov. Phys. Dokl., **7** (1963) 742.
[3] T. Kawae *et al*., Electrochem. Solid-State. Lett., **14** (2011) G31.
[4] H. Kawasaki *et al*., Private Communication.
[5] T. Kawae *et al*., J. Ceram. Soc. Jpn. **118** (2010) 652

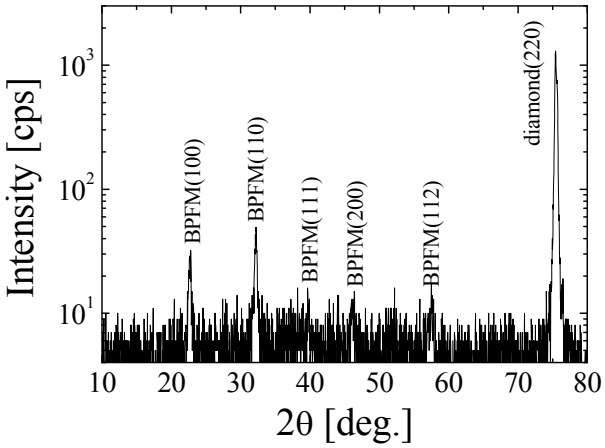

Fig.1 XRD pattern for BPFM/B-doped diamond layered film deposited on a polycrystalline diamond substrate.

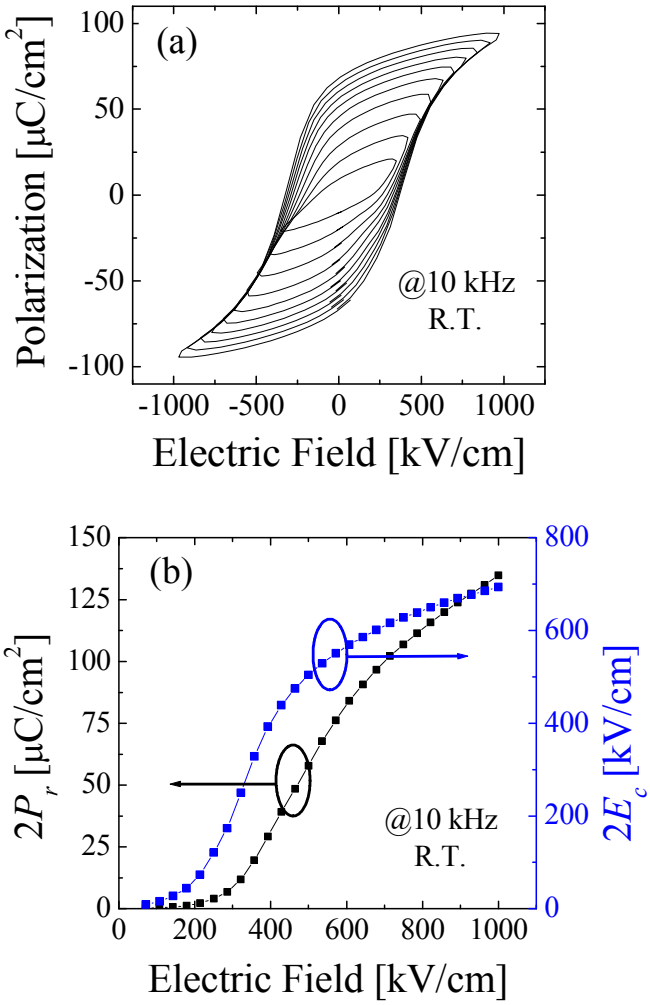

Fig.2 (a) *P-E* curves and (b) electrical field dependences of $2P_r$ and $2E_c$ for BPFM film on heavily B-doped diamond layer.