Investigation of preferred crystal orientation of sol-gel derived Bi$_{4-x}$La$_x$Ti$_3$O$_{12}$ thin-films on silicon substrates

Atsushi KOHNO*,1, Takayuki TAJIRI1, Kazushi SUMITANI2, Rie HARUKI1

1 Department of Applied Physics, Fukuoka University, Fukuoka 814-0180, Japan
2 Saga Light Source, Tosu, Saga 841-0005, Japan
3 Center for Research and Advancement in Higher Education, Kyushu University, Fukuoka 810-8560, Japan

Introduction

Metal-ferroelectric-semiconductor (MFS) structures have attracted attention for ferroelectric memory application. Thin film of Bi$_{4-x}$La$_x$Ti$_3$O$_{12}$ (BLT) is a good candidate for memory application due to their fatigue- and lead-free natures. The structural and electrical properties of BLT thin films formed directly on Si substrates by chemical solution decomposition method have been investigated. The previous work preliminarily suggested that a potential of the control of the crystal orientation in BLT thin films by such a low-cost method.

In this work, we have investigated the crystal orientation characteristics of the BLT thin films formed directly on Si by using sol-gel and spin-coat methods and the relationship to the electrical characteristics of the film.

Experimental

The BLT thin films were formed by the following procedure. After the diluted HF-treatment of chemically cleaned p-type Si (100) wafers, a sol-gel precursor solution was spin-coated on the substrates. The solution-coated wafers were dried on a hot plate at 150°C for 30 min, and the films were crystallized in a furnace at 550 or 600°C. Pole-figure X-ray diffraction measurements were carried out using synchrotron-radiation source at beamline BL-4C of the Photon Factory (Proposal No. 2006G280), KEK in Japan, because the diffraction signal detected by the laboratory equipment was so weak due to the small crystallites in the thin film. The incident X-ray was tuned in 12 KeV. We also measured the capacitance-voltage characteristics of the Au/BLT/p-Si structures.

Results and Discussions

The deposited BLT films have been successfully crystallized to Bi-layered structure at 550-600°C for 1-2 hours. The BLT layer thicknesses were evaluated to be ~66nm for 550°C and ~64nm for 600°C by X-ray reflectivity analysis. The capacitance-voltage (C-V) characteristics of Au/BLT/p-Si structures showed the clockwise hysteresis due to ferroelectric properties of the BLT films in both the cases of crystallization at 550 °C and 600°C. The hysteresis voltage-width ΔV for the sample crystallized at 550°C was larger than that at 600°C. One of the possible explanations for the difference in ΔV is that the crystal orientation in BLT film depends on the crystallization temperature.

Figure 1 shows the intensity distribution for the 008 diffraction pole figure. It is clearly shown that the intensity around Ψ = 0° (the normal to the surface) for 600°C was much higher than that for 550°C. The spontaneous polarization of BLT along c-axis is much smaller than that along a- and b- axes. The results suggest that the difference in the C-V hysteresis width ΔV between the samples crystallized at 550 and 600°C resulted from the difference in the c-axis orientation.

Fig. 1 The pole figures of 008 diffraction in the Ψ range of 0-30° for the BLT films crystallized at 550°C (a) and at 600°C (b).

References


* kohno@fukuoka-u.ac.jp