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$BaTiO_3$ Based Relaxor Ferroelectric Epitaxial Thin-films for High-temperature Operational Capacitors

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Abstract

A very high dielectric constant exceeding 400 was achieved in (100) oriented epitaxial thin-films of 0.6[$BaTiO_3$]-0.4[$Bi(Mg_{2/3}Nb_{1/3})O_3$] on $SrTiO_3$ substrates. These relaxor type ferroelectric films showed excellent dielectric constant stability of < 11% in the temperature range of 80 - 400°C. This will enable smaller, high-temperature tolerant, monolithically integrated thin-film capacitors on power semiconductor devices.

1. Introduction

Power semiconductor devices based on SiC, GaN and diamond are needed for compact efficient energy-saving domestic, automotive, aviation, space, and geothermal exploration applications and power transmission. SiC has reached to an advanced manufacturing stage and SiC based transistors and Schottky diodes are capable of performing over 250°C. In parallel to the advancement in these active devices, it is also necessary to develop passive elements such as a capacitor, resistor and inductors that are capable of operating at temperatures over 250°C.

Currently available capacitors can be functional up to 175°C only. Moreover, they are bulky and occupy a significant space in the module. Therefore, it is very important to develop a thin-film capacitor that can withstand high-temperature and can be integrated monolithically to the active device. This will facilitate the reduction of inductive losses and faster switching of the device. Also one can take advantage of the high thermal conductivity of the substrate and develop cooling-free compact device. The core issue is to develop a dielectric medium and requirements of a dielectric material to serve as a capacitor is to have (1) a high dielectric constant > 400 over a wide frequency range, (2) high stability of dielectric constant against temperature typically, <15%, (3) low dielectric loss and leakage current, and (4) free from hazardous elements. Furthermore, a thin-film process technology has to be developed for device integration. Here, we demonstrate the epitaxial growth of a relaxor ferroelectric x[$BaTiO_3$]- (1-x)[$Bi(Mg_{2/3}Nb_{1/3})O_3$] on a $SrTiO_3$ substrate employing pulse laser deposition method and show its potential as a dielectric material for high-temperature tolerant power capacitors.

2. Experiment

Sintered ceramics targets of 0.6[$BaTiO_3$]-0.4[$Bi(Mg_{2/3}Nb_{1/3})O_3$] – (BT-BMN) composition[1] and $SrRuO_3$ were prepared and pre-installed in the deposition chamber. The (100) oriented Nb 0.5 wt.% doped $SrTiO_3$ (STO) was used as a substrate. A KrF excimer laser with a wavelength of 248 nm, repetition rate of 10Hz, and energy density of 1.5 J/cm^2 was employed to ablate the target. The growth was performed at specific temperatures of 490, 510 and 530°C and at specific oxygen partial pressures of 0.2, 2, 20Pa. Prior to BT-BMN growth, a thin $SrRuO_3$ was deposited on some substrates at 570°C with oxygen partial pressure of 0.13Pa. The x-ray diffraction (XRD) analysis was carried out using Bruker AXS D8 discover GADDS system equipped with a large angle 2D detector. A 120nm thick Pt was deposited as top and backside electrodes. Current voltage (I-V) and capacitance-voltage (C-V) measurements were conducted using source-measurement unit (Keithley, 2612B) and LCR meter (Agilent E4980A) respectively in the temperature range of 25°C to 400°C.

2. Results and discussion

The 2D XRD pattern of the films grown on STO at 20 Pa and 510°C showed singular spots (Fig.1a) corresponding to (100) and (200) reflections. Fig. 1b is the pole figure projection of {111} reflection on (100) plane confirming the epitaxial nature of the film. The 2D singular spot intensity decreased in films grown at 2 and 0.2 Pa and exhibited poly crystallinity similar to the films grown at 490 and 530°C.

Fig. 1c shows the dielectric constant (solid lines) and dielectric loss (broken lines) as a function of sweep frequency for the films deposited at temperatures 490, 510 and 530°C at 20 Pa. The dielectric constant, in the as-grown (AG) state, is high for the film grown at 510°C that also exhibited high epitaxial relation with the substrate and the other films were poly crystalline. In the next step, we investigated the post growth annealing effects on the films. The films were annealed at high temperatures in oxygen ambient. Annealing process substantially improved the dielectric constant of the film and the highest value is obtained on the film annealed at 850°C. The dielectric loss is less than 0.2 even at 1MHz. The XRD pattern on the annealed film did not indicate any remarkable change.

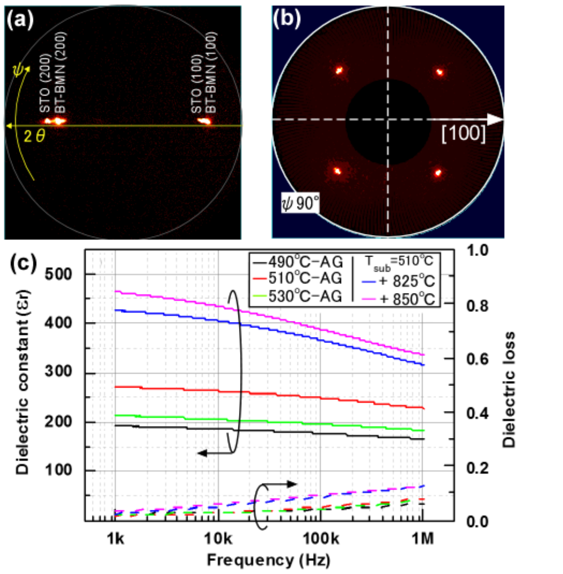

Fig. 1 a) 2D XRD pattern of BT-BMN on STO b) pole figure of {111} reflections on (100) plane and c) dielectric constant as a function of frequency in as-grown (AG) and 825 and 850°C annealed films

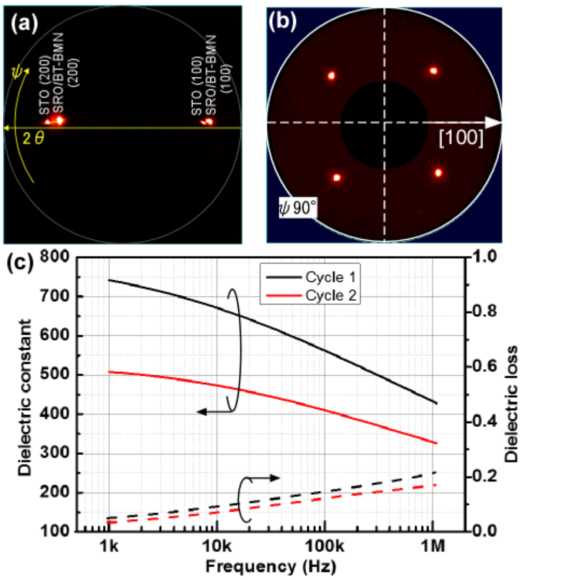

Fig. 2 a) 2D-XRD pattern, b) pole figure projection of {111} reflections on (100) plane and c) frequency dependency of dielectric constant of BT-BMN/SRO/STO structure.

We altered the substrate-film interface by introducing $SrRuO_3$ (SRO), a widely used perovskyte conductive layer, since the interaction of Bi with $SrTiO_3$ has been a subject of interest in other oxide ferroelectric systems [2]. A 2D XRD pattern in Fig. 2a showed singular spots corresponding to (100) and (200) reflections. Fig. 2b also depicts the pole figure projection of {111} reflections on (100) evidencing the epitaxial relationship among BT-BMN, SRO films, and STO. Frequency dispersion of dielectric constant (solid lines) and dielectric loss (broken lines) in 850°C annealed epitaxial films with SRO/Pt top electrode is illustrated in the Fig.2c. The dielectric constant remarkably increased further in the SRO sandwiched BT-BMN film. The films showed a high dielectric constant exceeding 550 at 100 kHz. However, it decreased after one high temperature measurement cycle. This indicates that the interfacial reaction plays a crucial role on the dielectric properties.

Figure 3 shows the dielectric constant (solid symbols) and dielectric loss (open symbols) of the film as a function of temperature in the forward (increasing temperature: black colored) and reverse (decreasing temperature: blue colored) directions. The BT-BMN film showed higher dielectric constant in the forward direction, and better thermal stability in reverse direction. Note that the dielectric constant exceeds 400 and the stability is <11% at temperature in the range of 80-400°C which are more promising while considering recent reports [2,3]. After the first high temperature measurement cycle, the temperature dependence of dielectric constant was stable. The dielectric constant values are similar to the values subjected to reverse measurement. This indicates the interfacial reaction at high-temperature influences the dielectric constant and hence the stability. Therefore, it is possible to achieve higher dielectric constant and better thermal stability of dielectric constant from this material when the interface is properly engineered.

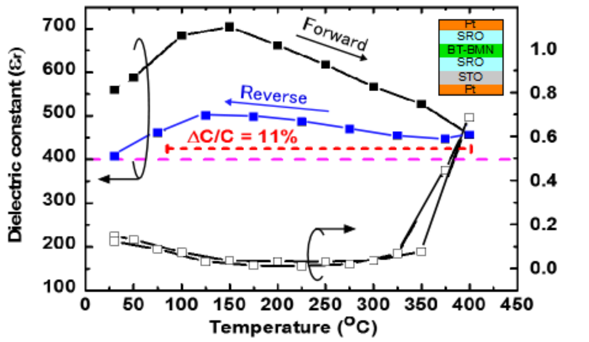

Fig.3 Temperature dependency of dielectric constant and loss (100kHz). The inset shows the sample structure.

4. Conclusions

Epitaxial films of BT-BMN were realized. A high dielectric constant exceeding 400 at 100 kHz and high thermal stability of dielectric constant <11% was accomplished. When the interface is appropriately tuned it is possible to gain higher dielectric constant and stability.

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