

Ferroelectric polarization and resistive switching characteristics of ion beam assisted sputter deposited BaTiO₃ thin films

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ABSTRACT

In this work, 150 nm thick polycrystalline BaTiO₃ (BTO) films were deposited on Pt/TiO₂/SiO₂/Si substrate by ion beam assisted sputter deposition technique. The bias voltage dependent resistive switching (RS) and ferroelectric polarization characteristics of Au/BTO/Pt devices are investigated. The devices display the stable bipolar RS characteristics without an initial electroforming process. Fittings to current–voltage (I–V) curves suggest that low and high resistance states are governed, respectively, by filamentary model and trap controlled space charge limited conduction mechanism, where the oxygen vacancies act as traps. Presence of oxygen vacancies is evidenced from the photoluminescence spectrum. The devices also display P–V loops with remnant polarization (P_r) of 5.7 $\mu\text{C}/\text{cm}^2$ and a coercive electric field (E_c) of 173.0 kV/cm. The coupling between the ferroelectric polarization and RS effect in BTO films is demonstrated.

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1. Introduction

Resistive random access memory (RRAM) based on resistive switching (RS) effect has been considered as one of the most leading candidates for next-generation nonvolatile memories because of its remarkable characteristics such as low power consumption, high operation speed, nondestructive readout, etc... [1,2]. Usually, RS behavior has been observed in transition metal oxides (TMO) like ZnO, TiO₂, Cu₂O, MgO, etc... [3,4]. However, a different type of RS effect has been reported in ferroelectric materials due to their polarization reversal and high leakage currents, which has fundamental merit over the TMO [5]. Several mechanisms are proposed to explain the RS behavior in metal/ferroelectric/metal (MFM) structures including the formation/rupture of conductive filaments [6], defect-induced modulation of depletion layer width [7], the charge trapping/detrapping [5], the modulation of Schottky-like barriers, which might be related to ferroelectric polarization and the polarization direction dependent

tunneling effect [8]. However, the detailed understanding on the relation between the polarization reversal and the resistive switching in ferroelectric thin films is still lacking.

Barium titanate (BTO, BaTiO₃) is a well-known ferroelectric material; It has been widely investigated for many electronic applications due to its excellent spontaneous polarization, as large as 26 $\mu\text{C}/\text{cm}^2$, large dielectric constant, moderate coercive field and ferroelectric transition temperature above the room temperature [9,10]. Recently, the resistive switching has been observed in epitaxial BaTiO₃ thin films and Ag nanoparticles doped BTO films [11,12]. However, the studies on RS effect in polycrystalline films are not published yet. In the case of polycrystalline films, RS seems to be more complicated than epitaxial films since the electric conduction is quite different due to presence of grain boundaries. Moreover, the contribution of the ferroelectric polarization or defects, or both, to RS effect in these films needs to be understood.

In this work, polycrystalline BTO films were deposited on Pt/TiO₂/SiO₂/Si substrates by ion beam assisted sputter deposition technique. The bias voltage dependent resistive switching (RS) and the ferroelectric polarization characteristics of Au/BTO/Pt devices were investigated. The devices display the stable bipolar resistive switching characteristics. The conduction mechanism of RS effect and its coexistence with ferroelectric polarization has also been analyzed.

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2. Experimental procedures

BTO films with thickness of 150 nm were grown on Pt/TiO₂/SiO₂/Si substrates using a commercially available BaTiO₃ target (Neyco, 99.9%) by ion beam assisted sputter deposition technique. The chamber was first evacuated down to a low pressure of 1×10^{-6} mbar prior to the deposition. During the deposition, the substrate was kept at a temperature of 330 °C and at a distance of 87.3 mm from the target. The gas pressure inside the chamber was kept constant at 3.4×10^{-4} mbar. A gas flow of 8.0 ml/min of Ar was introduced into the ion beam gun and the Ar atoms were ionized in the ion source with an rf-power of 120 W. The argon ions beam further accelerated to 900 V and the argon ion beam current was regulated to be 30 mA. Subsequently, an annealing was performed in vacuum ($=2.0 \times 10^{-5}$ mbar) at 650 °C for 30 min in order to improve the crystalline structure. The crystal structure of the thin films was investigated by x-ray diffraction (XRD) using CuK_α radiation. Photoluminescence (PL) spectrum was recorded on a Spex Fluorolog spectrometer in front-face geometry in spectral range from 350 to 600 nm under a 330 nm excitation from a Xenon lamp. Gold (Au) electrodes having the diameter of 1 mm were deposited by thermal evaporation on the upper surface of BTO films. Electrical properties of BTO films were obtained by investigating the capacitors of Au/BTO/Pt with MFM configuration. Polarization–voltage (P–V) hysteresis loops were recorded using a modified Sawyer–Tower circuit (schematic diagram is shown in inset of Fig. 4(a)), at 1 kHz. Current–voltage (I–V) characteristics were measured using a Keithley 617 programmable electrometer.

3. Results and discussion

Fig. 1 reveals representative XRD pattern of BTO film and Pt/TiO₂/SiO₂/Si. The peaks corresponding to BTO film were indexed in Fig. 1 according to standard powder diffraction data (JCPDS Card No. 5-0626). The standard peaks position was also marked as red lines in Fig. 1. The XRD pattern exhibited the planes such as (100)/(001), (101), (111), (200)/(002), (201) and (211) related to

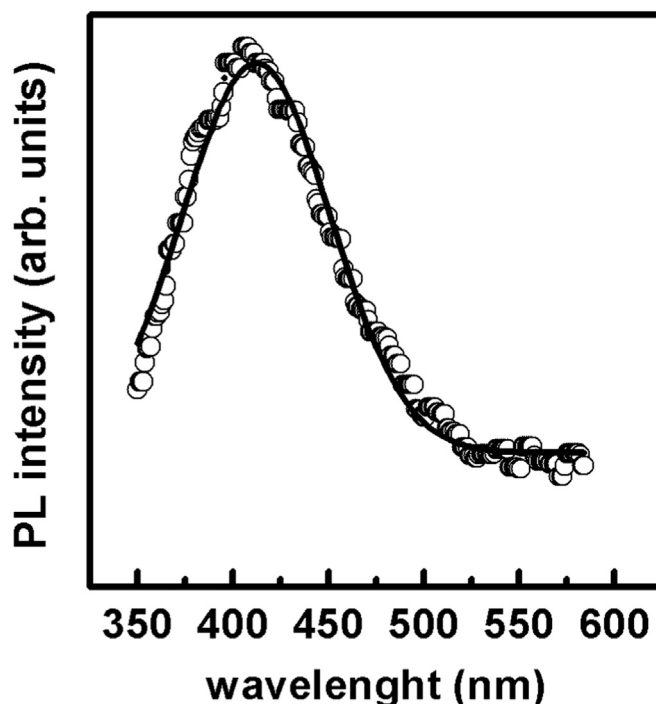


Fig. 2. PL spectrum of a polycrystalline BTO thin film deposited on Pt/TiO₂/SiO₂/Si substrate.

perovskite phase of BTO without any secondary phase. The peaks marked with (*) are related to the TiO₂ layer of the substrate [10,13,14]. It is noticed that diffracted peak positions of BTO film shift towards lower 2θ values compared to that of JCPDS data. This reveals the presence of tensile strain in the film, which is generally related with the presence of defects [15]. The presence of defects in the films was further confirmed from the PL study. Fig. 2 depicts room temperature PL spectrum of the BTO thin film. The film exhibited a strong violet emission centered at 412 nm, which is usually related to defect levels associated with oxygen vacancies [16]. This is in good agreement with the fact that the formation of oxygen vacancies is more probable as the present films were annealed in oxygen deficient environment. Further, the tensile strain might be responsible for the oxygen vacancies in BTO films [15,17].

Fig. 3(a) depicts the I–V characteristic curves of Au/BTO/Pt device without an initial electroforming process. The dc voltage was swept from $-V_{\max}$ to $+V_{\max}$ in forward direction and $+V_{\max}$ to $-V_{\max}$ in vice-versa. Various bias ranges (different $V_{\max}$) have been applied to the device in order to investigate the dependence of the resistive hysteresis on the amplitude of $V_{\max}$. We have fixed $V_{\max} \leq 7$ V only to prevent the device from unrecoverable breakdown due to local Joule heating at high dc bias voltage [18]. As illustrated in Fig. 3(a), I–V curves display the hysteresis behavior strongly dependent on the applied $V_{\max}$. The resistance of two states, such as low resistance state (LRS) and high resistance state (HRS), was read-out at -1 V and the resistance ratio $[R_{\text{HRS}}/R_{\text{LRS}}]$ versus $V_{\max}$ is shown in inset of Fig. 3(a). The resistance ratio increases with the applied $V_{\max}$ and is found to be ≈ 25 when $V_{\max} = 7$ V. This resistance ratio is higher than the value found in BTO epitaxial films prepared by PLD technique [1,19]. The decreases in magnitude of resistance of HRS state in the I–V curve obtained for $V_{\max} = 7$ V compared to that of $V_{\max} = 4$ and 5 V can be attributed to joule heating as it can rupture conductive filaments [20].

In order to get deep information regarding the mechanism underlying the resistance switching behavior in BTO films, several conduction models, such as space-charge limited current (SCL)

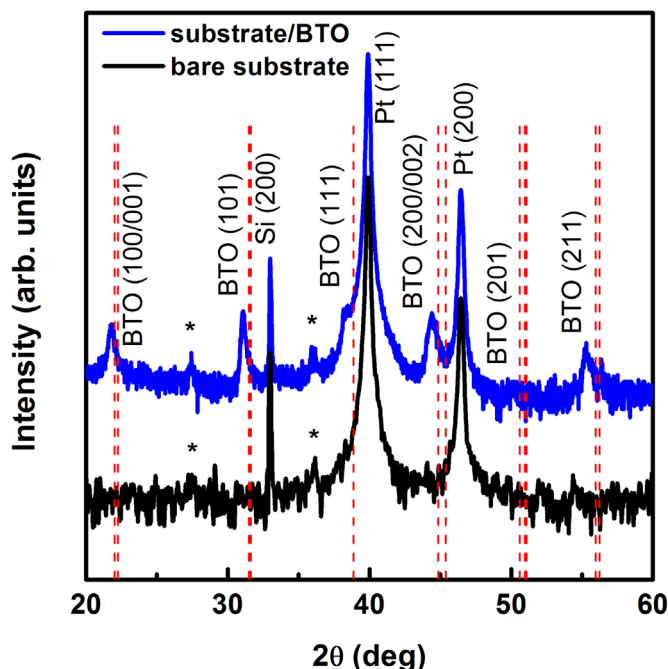


Fig. 1. XRD pattern of bare substrate and substrate/BTO thin film.

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