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Ferroelectric domain switching kinetics of a lead-free $AgNbO_3$ thin film on glass substrate

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ABSTRACT

Lead-free silver niobate ($AgNbO_3$) thin film was deposited on glass substrate by pulsed laser deposition. Ferroelectric properties of the $AgNbO_3$ thin film were investigated. The $AgNbO_3$ thin film capacitor exhibited good ferroelectricity with a remnant polarization of about 15.7 $\mu C/cm^2$ ($2P_r \sim 31.4\ \mu C/cm^2$) at room temperature and fast switching behavior within about 130 ns. Triangular grains on the ANO thin film surface were observed by atomic force microscopy (AFM). By using piezoelectric force microscopy (PFM), we investigated ferroelectric domain switching and domain wall motion of the $AgNbO_3$ thin film. From the domain wall speed as a function of applied electric field in the $AgNbO_3$ thin film, activation energy of domain wall motion was derived. Compared to the $PbTiO_3$ thin film reported previously, the $AgNbO_3$ thin film showed faster switching behavior which could be attributed to its lower activation energy for domain wall motion.

1. Introduction

Ferroelectric materials have been widely applied to electronic devices such as non-volatile random access memory (NVRAM), actuator, senor, and energy harvesting devices due to their spontaneous polarization and piezoelectricity [1–5]. However, because lead-based materials, particularly Pb(Zr, Ti)O_3 system, are very toxic in their process and applications, environment-friendly (lead-free) ferroelectric materials are receiving great attention as potential substitutes [6–8]. Among these lead-free materials, $AgNbO_3$ is one of the promising candidates due to its remarkable ferroelectric characteristics and relatively simple perovskite structure [9–11]. In spite of extensive study on structure and ferroelectric polarization of bulk $AgNbO_3$, there has been little report of ferroelectric domain and polarization switching behavior on $AgNbO_3$ thin films.

To satisfy a demand for the fast-operating devices at the present day, investigating of polarization switching behavior is indispensable for ferroelectric application. Generally, polarization switching phenomenon in ferroelectrics has been understood by nucleation of the switched domain and lateral domain wall motion [12–14]. With the advance of piezoresponse force microscopy (PFM), it is possible for us to explore domain structure and domain wall motion at the nanoscale in ferroelectric materials. The ferroelectric domain wall motion has been investigated experimentally and theoretically, especially predicted by Merz's law [12]. By applying an external electric filed, the ferroelectric domain wall can be moved and the speed of a domain wall also can be changed. These significant behaviors are thought to have originated from the electrical activation energy for the domain wall motion [15]. In this sense, understanding the kinetics and domain wall motion in the $AgNbO_3$ thin film are important aspect for the application of information storage like NVRAM.

In this study, we fabricated lead-free $AgNbO_3$ thin film on glass substrate by pulsed laser deposition (PLD). Here, a Ta film as adhesion layer was deposited on glass substrate to enhance crystalline orientation and electrical properties for Pt bottom electrode. Ferroelectric polarization and switching current of the $AgNbO_3$ thin film are demonstrated. By using PFM, we observe ferroelectric domain structure and a switching behavior. Finally, the domain wall speeds are measured and activation energy for domain wall motion is derived from a relation between the domain wall speed and applied electric field.

2. Experimental

$AgNbO_3$ thin films were deposited on glass substrates by PLD. The fused silica glass as substrate was used for the deposition of the ANO thin films. As an adhesion layer on glass, Ta film was prepared using radiofrequency (RF) magnetron sputtering, which

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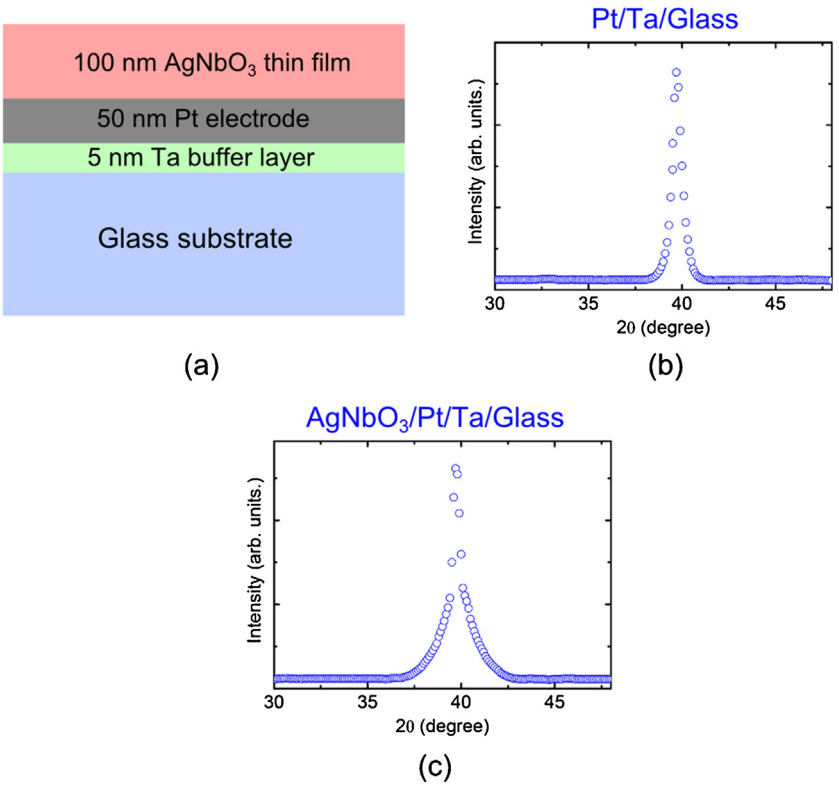

Fig. 1. (a) A schematic diagram of $AgNbO_3$ thin film on Pt/Ta/glass substrate. (b) The XRD pattern of Pt/Ta/glass substrate (c). The XRD pattern after deposition of the $AgNbO_3$ thin film on Pt/Ta/glass substrate.

deposited at 500 °C under 10 mTorr Ar gas and 100 W of RF power. Then, Pt bottom electrode film was deposited on Ta/glass substrate by RF sputtering of 50 W RF power at 600 °C and 10 mTorr Ar gas. The $AgNbO_3$ target for the PLD was prepared by using a conventional solid-state reaction according to its stoichiometry. A KrF excimer laser with a wavelength of 248 nm and an energy density of 0.5 J/cm^2 was focused onto the $AgNbO_3$ target. The distance between the target and substrate was −4 cm. After the base pressure reached -5×10^{-7} Torr, the substrate temperature was set at 750 °C with an oxygen partial pressure of 100 mTorr. After deposition, the $AgNbO_3$ thin film sample was immediately cooled to room temperature in oxygen ambient at 300 Torr. For the fabrication of Pt/$AgNbO_3$/Pt capacitor, Pt top electrode (circular-shape with diameter of 100 μm and thickness of 100 nm) was layered on $AgNbO_3$/Pt/Ta/glass substrate by RF sputtering. After deposition of Pt top electrode, the capacitor sample was annealed at 400 °C for 5 min.

The structure of the $AgNbO_3$ thin film was investigated by X-ray diffraction (XRD, CuKα radiation 1.542 Å). The thickness of the $AgNbO_3$ thin films was measured in a cross-sectional scanning electron microscopy (SEM). The surface morphology and roughness of the $AgNbO_3$ thin film were evaluated by AFM. The ferroelectric domain was also observed by PFM. The ferroelectric hysteresis loop was measured by an RT66A (Radiant Technologies, Inc.) test system

3. Results and discussion

Fig. 1(a) shows a schematic diagram of our sample, where each layer was sequentially deposited on glass substrate. The thicknesses of Ta buffer layer, Pt bottom electrode, and $AgNbO_3$ thin film were 5, 50, and 100 nm, respectively. We characterized the crystal structures of multilayer films on glass substrate by XRD measurement. In the absence of the $AgNbO_3$ thin film on Pt/Ta/glass substrate, the XRD θ–2θ pattern scanned over a range of angles (30–48°) showed a peak which reflect (1 1 1) orientation of crystalline Pt on Ta adhesion layer as shown in Fig. 1(b). For comparison, XRD measurement was also performed after deposition of the $AgNbO_3$ thin film on Pt/Ta/glass substrate. In Fig. 1(c) one dominant peak was also observed at the same position, but it was broader than single peak of Pt electrode. This is ascribed to that a (1 1 1) peak of the $AgNbO_3$ thin film was overlapped with the peak of the Pt electrode. The deposited the $AgNbO_3$ thin film shows highly (1 1 1)-oriented single-phase perovskite structure. Generally, the crystallinity of a bottom electrode plays an important role in the texture of sequentially deposited layers in thin films process.

To investigate ferroelectric polarization and properties of the $AgNbO_3$ thin film, we made a Pt/$AgNbO_3$/Pt capacitor by depositing a Pt top electrode on $AgNbO_3$/Pt/Ta/glass sample. Fig. 2(a) demonstrates a $P-E$ hysteresis loop of the Pt/$AgNbO_3$/Pt capacitor measured at a frequency of 10 kHz. A remnant polarization (P_r), saturation polarization (P_s), and coercive electric field (E_c) of the $AgNbO_3$ capacitor exhibited 15.7 μC/cm^2, 26.1 μC/cm^2, and 152 kV/cm, respectively. Our remnant polarization value is comparable to that (11.5 μC/cm^2) of (1 1 1) $AgNbO_3$ thin film on (1 1 1) Nb-doped $SrTiO_3$ substrate [16,17]. To check polarization switching behavior of $AgNbO_3$ ferroelectric thin film capacitor, we measured the switching current curve. As mentioned earlier, the ferroelectric polarization switching process is a key indicator for device applications such as non-volatile random access memory. Here, switching current was actually recorded by the response difference of two consecutive (switching and non-switching) pulses train which consisted of square pulses with an input width of 1 μs and a rising time of about 1 ns [18]. For the positive and negative pulses, we applied +5 V and −5 V as switching bias. The pure (net) switching current curve was obtained as shown in Fig. 2(b). The $AgNbO_3$ thin film exhibited fast switching behavior within a switching time of about 130 ns. This switching behavior is comparable to that of the $PbTiO_3$ thin film previously reported [19].