Solid State Communications 192 (2014) 89–92

Contents lists available at ScienceDirect

Solid State Communications

journal homepage: www.elsevier.com/locate/ssc

Temperature dependence of dynamic hysteresis behavior in $Pb_{0.4}Sr_{0.6}TiO_3$ ferroelectric films

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ARTICLE INFO

Article history:
Received 22 December 2013
Accepted 13 May 2014
by T. Kimura
Available online 20 May 2014

Keywords:
A. Thin films
B. Sputtering
C. Ferroelectric property
D. Dynamic hysteresis.

ABSTRACT

The temperature (T) dependence of dynamic hysteresis behavior is investigated in low temperature crystallized $Pb_{0.4}Sr_{0.6}TiO_3$ thin films between 85 K and 340 K. It was found that the temperature scaling relations for hysteresis area $\langle A\rangle$, remnant polarization P_r and coercive field E_c take the different forms in two different temperature regions divided by a transition region ranging from 190 K to 225 K and the $\langle A\rangle$, P_r and E_c decreased in the first region and increased in the second region with increasing T. The different scaling relations can be predicted by the effect of thermal activated de-freezing of domain wall and dielectric response of defects with applied electric field. Moreover, the effect of substrate temperature on the dynamic hysteresis behavior versus T is also investigated.

1. Introduction

Considering its superior ferroelectric as well as dielectric performances [1,2], low crystallized temperature compatible with the current silicon-based microelectronic technology [3] and weak dependence of electrical properties on grain size [4], the lead strontium titanate ((Pb_xSr_{1-x})TiO_3, (PST)) thin film has been extensively investigated as one of the most promising candidate materials for applications in capacitors for ferroelectric random access memories (FRAM) and frequency agile devices [1,2,4,5]. It is well known that the dynamic hysteresis behavior deemed to a non-equilibrium behavior associated with first-order phase transitions is of special theoretical and technological importance for the applications such as FRAM [6], because it can provide a lot of domain-reversal information [7–11]. The power law of $\langle A\rangle \propto E^{\alpha} f^{\beta}$ (where $\langle A\rangle$ is hysteresis area, E is the amplitude of electric field, f is frequency, and α as well as β are scaling exponents) is the center for the investigation of the dynamic hysteresis behavior and have been conducted theoretically for ferromagnetic by Rao et al. with the scaling exponents α and β of 2 and -1 respectively in terms of the three-dimensional $(\Phi^2)^2$ and $(\Phi^2)^3$ models [12]. Moreover some experimental investigations were carried out in both bulk ceramics and thin films [12–17]. In addition to the E and f, the temperature dependence of dynamic hysteresis behavior is also indispensable for the investigation of the ferroelectric property in ferroelectric films and Rao et al. theoretically predicated that the exponents in the power law scaling of $\langle A\rangle$ against T were -0.7 and 1.0 for $(\Phi^2)^2$ and $(\Phi^2)^3$ models respectively [12,18].

Unfortunately, there is a lack of sufficient investigation on the temperature dependence of dynamic hysteresis behavior of PST thin films and the effect of the defects on the temperature dynamic scaling behavior in PST thin films. It is thus the aim of this investigation to experimentally study the temperature dependence of the dielectric response and dynamic hysteresis behavior in PST thin films.

2. Experimental section

About 300 nm thick $Pb_{0.4}Sr_{0.6}TiO_3$ thin films were deposited by in-situ r.f.-magnetron sputtering method on $LaNiO_3$ coated Si substrate [3]. The deposition was carried out under a substrate temperature compatible with the silicon based microelectronic technology ranging from 450 to 500 °C (denoted as PST-450 and PST-500) with pure Ar atmosphere. The detailed parameters for the preparation of PST thin films were described elsewhere [3].

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http://dx.doi.org/10.1016/j.ssc.2014.05.007
0038-1098/

The Pt top electrodes with a diameter of 150 μm were fabricated onto the top of PST thin films by sputtering through the traditional photolithography and lift-off methods to form the capacitor structure. The X-ray diffraction results showed that the PST-450 and PST-500 are perfectly (100)-orientated while the PST-500 possessed better crystalline quality. HP4192A (Agilent Technologies, Santa Clara, CA) impedance analyzer was adopted to investigate the capacitance and loss tangent as a function of T at various f. The polarization hysteresis versus electric field (P–E) loops were measured by an aixACCT TF Analyzer 2000 system with sine wave at a frequency of 1 kHz. The temperature was maintained using a computer controlled cryostat system (ST-500, JANIS Research Company, Inc.).

3. Results and discussion

The hysteresis loops of PST-500 measured at several typical temperatures with various voltage amplitudes are demonstrated in Figs. 1(a–c). It can be observed that the loops do not saturate at low voltage amplitude while gradually become saturated with further increasing electric field due to the E improved ferroelectric domain reversal. The remnant polarization ($P_r=(P_r^+-P_r^-)/2$) and coercive electric field ($E_c=(E_c^+-E_c^-)/2$) increase monotonically with the temperature increasing from 85 K to 185 K, which can be explained by the decrease in the domain wall mobility at low temperature [1,19,20]. A larger applied field is required to overcome the energy barrier and drive the motion of domain wall at lower temperature because the switching of some domains is restrained, consequently leading to the increases in V_c, P_r and thus the hysteresis area [19,20]. However, the remnant polarization, coercive electric field and hysteresis area become larger with the temperature further increasing to 340 K, which may be the result of the dielectric response of the defects with applied electric field. To further confirm the effect of the temperature on the dielectric and ferroelectric properties, the dielectric constant and loss tangent as a function of T at various f were tested and displayed in Fig. 1(d). An abrupt change in the dielectric constant and loss can be observed. Considering the composition of $Pb_{0.4}Sr_{0.6}TiO_3$ and its single phase transition, this abrupt change in dielectric response can be explained by the increasing activity of defects such as oxygen vacancies formed during the sputtering under an atmosphere of pure Argon. It is well known that the restraining of the defects minished at temperature around 200 K [19,20], which will bring out the increasing of dielectric response, loss tangent. Considering the relation between the dielectric response and polarization described as the following equation [21]:

$$\varepsilon_r = 1+\varepsilon_0{}^{-1}\partial P/\partial E \quad (1)$$

the ferroelectric property is consequently improved because of the increasing of dielectric response of the defects with the applied electric field. Moreover, the change is more obviously at lower frequency and similar behavior of the effect of defects on the dependence of dielectric response on frequency was also observed in previous study in $BiFeO_3$ films [22].

Fig. 2 shows the dependence of the hystersis area ⟨A⟩ on the electric field tested at various temperatures. It can be observed that the hysteresis area first decreases with temperature increasing from 85 K to 185 K, while increases with temperature further increasing to 340 K, which is consistent with the aforementioned analysis. Moreover, the ⟨A⟩ first grows up rapidly with an electric field less than 300 kV/cm, and then increases slowly under higher E, implying that the motion ferroelectric domains is suppressed under higher electric field [23] and thus larger drive force is required to reverse the domains. The dependence of the electric field scaling exponent on the temperature thus can be divided into two sections, separated by $E\sim$300 kV/cm, and displayed in the inset of Fig. 2. One can see that the variation of temperature affects the scaling exponent dramatically in the first region, while shows weak effect in the second region. It can be found that the α first

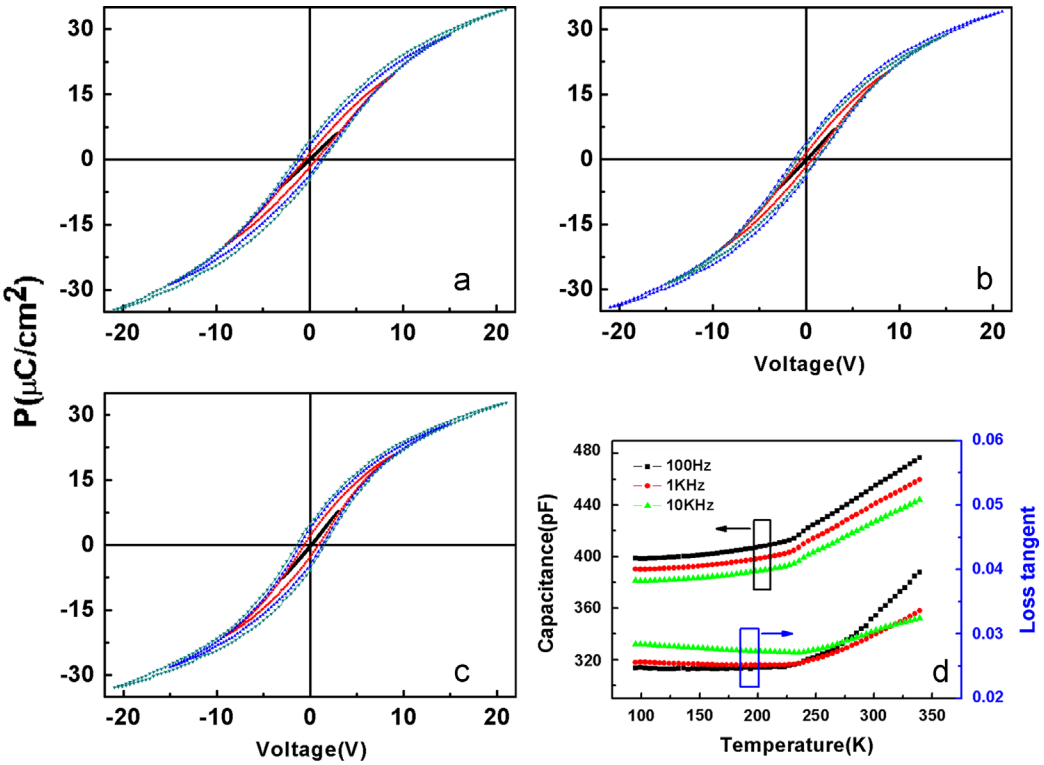

Fig. 1. (Color online) The P–E hysteresis loops of PST-500 under various E with at fixed temperature (a) 85 K, (b) 185, (c) 340 K and (d) its dielectric constant and loss as a function of temperature ranging from 95 K to 340 K.