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Electrical properties of $SrBi_2Ta_2O_9$ ferroelectric thin films at low temperature

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The temperature dependence of electrical properties for $SrBi_2Ta_2O_9$ thin film capacitors with platinum electrodes (Pt/SBT/Pt) on silicon wafers was studied from 10 to 300 K. With a decrease in temperature from 300 to 200 K, the remanent polarization of the thin films shows about an 11% reduction from its 300 K value; however, it is reduced by about 87% reduction from its 200 K value when the temperature drops from 200 to 100 K. With a decrease to 200 K, the polarization fatigue was significant, and the capacitor shows an approximate 29% reduction in polarization from its initial value following 10^{10} cycles. The dielectric response and leakage current of the thin films were also studied over the same lower temperature region. These results are helpful in the understanding of the fatigue-free behavior observed in $SrBi_2Ta_2O_9$ thin films at room temperature and provide additional insight into their use for ferroelectric memory applications. [DOI: 10.1063/1.1527700]

Recently, the use of the ferroelectric materials for nonvolatile memory devices, in particular, ferroelectric random access memories (FRAM), has generated much interest.[1–11] In FRAM, information can be stored by using the two polarization states of the ferroelectric thin film capacitor. FRAM has a lower power requirement, a faster access time, and potentially, a lower cost than many other nonvolatile memory devices. To meet commercial requirements for device lifetime, the ferroelectric thin films in the FRAM must have reliable polarization cycling characteristics. One of the most popular materials for this application was initially lead zirconate titanate (PZT). However, PZT thin films exhibit severe polarization fatigue during electric field cycling when standard Pt metal electrodes are employed. In recent years, it has been discovered that ferroelectric materials belonging to the layered perovskite family, such as $SrBi_2Ta_2O_9$ (SBT), show essentially no polarization fatigue with electric field cycling.[4] Several models have been proposed to explain the fatigue-free response of SBT thin films, such as the contribution of the $(Bi_2O_2)^{2+}$ layer,[3,4] which prohibits carriers from coming into perovskite layers due to charge compensation by self-regulating their position in the lattice;[3,4] and easy unpinning of domain walls from space charge,[5,6] which can be viewed as a self-recovery mechanism.

Most of the data in previous reports can be explained by one of these mechanisms. However, this does not mean that SBT thin films have no fatigue under all conditions. In fact, SBT films are known to exhibit fatigue at temperatures greater than room temperature. When SBT films were heated up to 125 °C, for example,[11] fatigue began to appear and polarization decreased by more than 10% of its initial value following 10^{12} cycles. Data for FRAM performance over a wide range of temperature are needed to meet some automotive applications (i.e., −50 °C), as well as military applications (called Mil Spec). In spite of intensive investigations, the ferroelectric and dielectric properties of films at low temperature have not yet been reported, although other aspects of these materials have been investigated. For example, Yan *et al.* reported the internal friction of SBT ceramics in the low temperature region (~100 K),[12,13] and from these results, polarization fatigue is predicted at lower temperatures. They strongly recommend that the electrical properties of SBT at lower temperatures should be further studied. In this letter, we will report the electrical properties of SBT thin films in the temperature regime of 10 to 300 K.

The SBT thin films were synthesized by pulsed laser deposition (PLD) combined with a thermal annealing technique, followed by completion of the sandwich structure Pt/SBT/Pt film capacitor devices, as described elsewhere.[14] Compositional analysis was performed using an inductively coupled plasma quantemeter, yielding a ratio of Sr:Bi:Ta to be 1.0:1.98:2.0 for the SBT film. The crystallographic structure of the films was characterized by x-ray diffraction. The dominant orientations of the films on the platinized silicon substrates were (008) and (115). The microstructure of the films was observed by transmission electron microscopy (plan-view and cross-section), and showed a mean grain size of about 200 nm, thickness of about 200 nm, and a dense film morphology with abrupt interfaces. This microstructure is typical of PLD films.[14–16]

Ferroelectric properties of the SBT thin films in the temperature regime of 10 to 300 K were measured using an RT66A ferroelectric tester from Radiant Technologies. Dielectric properties, such as capacitance–voltage ($C-V$) characterization, dissipation factor ($\tan\delta$) and dielectric constant (ε_r) of the films, were also measured by an HP4194A

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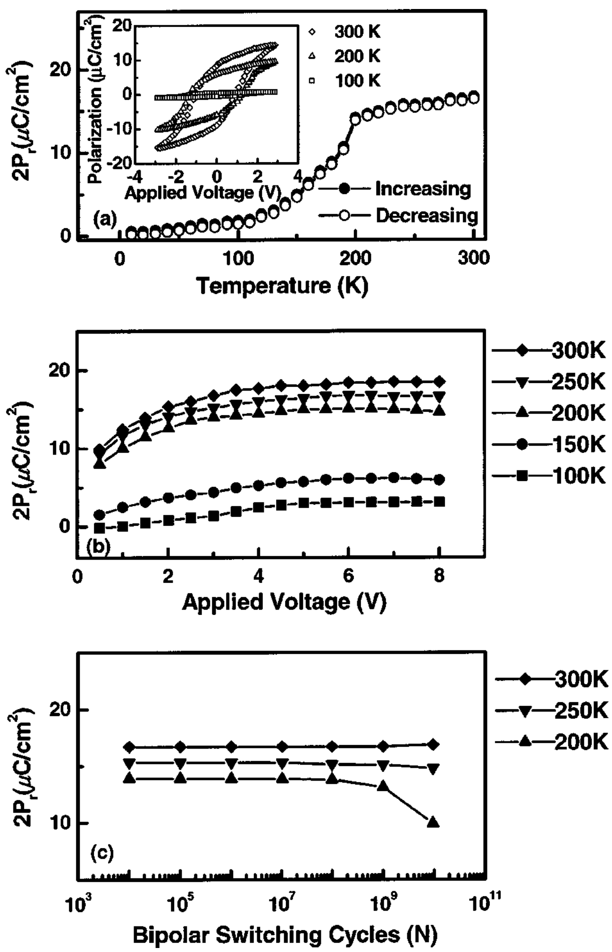

FIG. 1. The temperature dependence of twice the remanent polarization ($2P_r$) of SBT thin films: (a) $2P_r$ at 3 V for both decreasing temperature (open), increasing temperature (solid), and the hysteresis loop at variable temperature (inset) and (b) $2P_r$ vs voltage at various temperature; and (c) the fatigue behavior of $2P_r$ as a function of temperature at 3 V bipolar pulse with 1 kHz.

impedance/gain-phase analyzer in the same temperature regime.

Figure 1 shows the temperature dependence of twice the remanent polarization ($2P_r$) of the SBT thin films. In Fig. 1(a), with a decrease in temperature from 300 to 200 K, the remanent polarization of the thin films shows about an 11% reduction from its 300 K value; an 87% reduction is observed when the temperature is decreased from 200 to 100 K. The curves for both decreasing temperature (open) and increasing temperature (solid) in Fig. 1(a) are completely symmetric, which indicates that the sharp decrease in $2P_r$ below 200 K is not caused by an extraneous effect such as electrode delamination at lower temperature, but is an intrinsic property of the SBT film. As shown in Fig. 1(b), $2P_r$ increases with increasing voltage and then saturates at higher voltages. In addition with increasing temperature, these polarization values saturate at lower voltage. Fig. 1(c) shows the fatigue behavior of $2P_r$ as a function of temperature. Near room temperature (300 to 250 K), SBT has excellent fatigue resistance. However, for lower temperature (i.e., at 200 K), the onset of fatigue occurs at about 10^{10} cycles and the magnitude of $2P_r$ drops significantly. At 200 K, the capacitor shows an approximate 29% reduction in polarization from its initial value following 10^{10} cycles in Fig. 1(c).

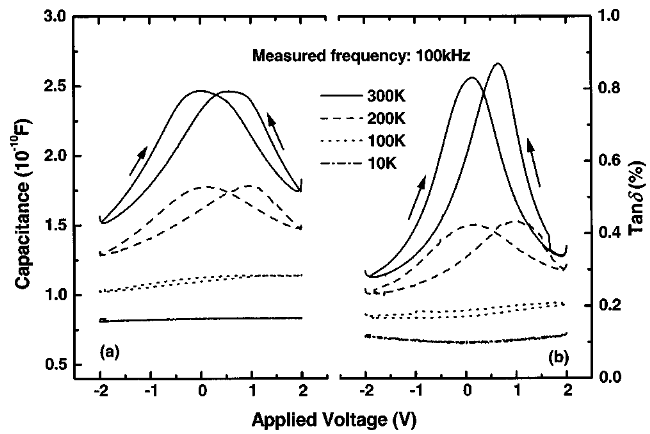

FIG. 2. Measured capacitance (a) and dissipation factor (tan δ) (b) vs voltage relations in SBT films as a function of temperature.

Figure 2 shows the capacitance and dissipation factor (tan δ) versus voltage relations in SBT films as a function of temperature. The curves in Fig. 2(a) show maxima in the capacitance values at approximately the coercive field ($E_c = V_c/d$). The magnitudes of the maxima weaken with decreasing temperature, and essentially disappear at 10 and 100 K. From Fig. 2(b), the tan δ value at zero field is about 0.0077 at room temperature, which is typical of the value reported for SBT films prepared by the metalorganic decomposition.[10] As shown in Fig. 2, the peaks with the direction of applied field scanning are asymmetric, which originates from the asymmetric conduction of the capacitor.[17]

Information on predicted phase transitions at lower temperature has not been reported for SBT films or ceramics. Our capabilities for studying this issue are limited. Furthermore, the results of internal friction[12,13] from 100 to 270 K and far-infrared spectra[18] down to 30 K studies for SBT did not give any indication of phase transitions at lower temperature. Herein, we believe that origin for the temperature effect of $2P_r$ may arise from switchable polarization that is frozen at a lower temperature. In our SBT sample, 180° domain walls dominate. The wall potential below a certain temperature has a "boxlike" shape,[19] which induces a defect to strongly pin it, and the potential well is deepened with decreasing temperature. These factors make depinning difficult and reduce switchable polarization. This results in a decrease in $2P_r$ with decreasing temperature as well as fatigue at lower temperatures, both of which are observable in Fig. 1(a) (open) and (c). Conversely, when the temperature is increased, the switchable polarization again increases due to shallowing of the potential well and the corresponding reduction in the tendency of a defect to act as a pinning site. The result of this is that the original $2P_r$ values are recovered, which can be seen in Fig. 1(a) (solid).

We assume that the capacitance consists of linear and nonlinear parts:[20] $C = C_f + (dP(E)/dE)S/L$, where C_f is the linear capacitance, $P(E)$ is the polarization, E is the applied field, L is the thickness, and S is the area of the capacitor. From this equation, the capacitance peaks in Fig. 2(a) correspond to polarization reversal, and the peak intensity depends on the amount of switchable polarization. The dissipation in the films is mainly from the field-induced motion of domain walls. Thus, the peaks in Fig. 2(b) also result from

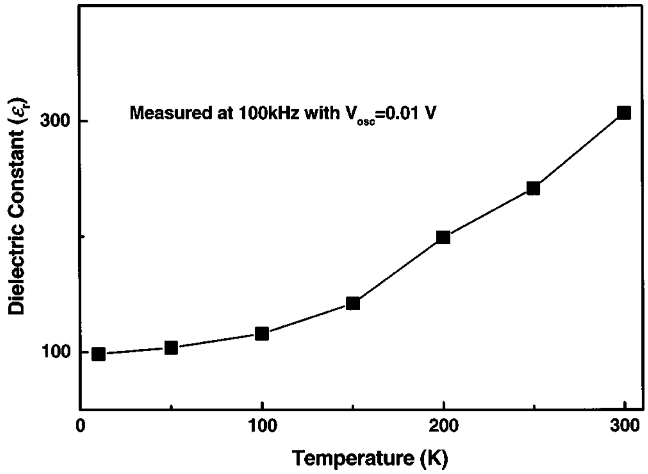

FIG. 3. Temperature dependence of the dielectric constant of SBT films.

polarization reversal. As shown in Fig. 2, with decreasing temperature, the amount of switchable polarization lessens due to domain pinning, or the freezing of domains, which suppresses the magnitude of peaks. Because of this, polarization reversal in the films is diminished resulting in the disappearance of the peaks in both the capacitance and the dissipation curves, as illustrated in Fig. 2. The results of dielectric constant measurements are in good agreement with the ferroelectric measurements.

The temperature dependence of dielectric constant for SBT below room temperature was calculated from capacitance measurement and is presented in Fig. 3. In this figure, we see that ε_r increases slowly below 200 K, then rises quickly. The shape of curve is typical of the temperature dependence for dielectric materials, but no anomaly, as might be associated with a phase transition, was found in the temperature region investigated.[12] The dielectric constant at room temperature is about 305, which is similar to the value reported by Taylor *et al.*,[11] but higher than the value (~228) reported by Takemura *et al.*[10]

Finally, the temperature dependence of the leakage current of the films from 10 to 300 K is presented in Fig. 4. The leakage current density lessens by about three orders when the temperature drops from 300 to 100 K. The differences in leakage current for the two bias directions are likely due to the different thermal treatments experienced by the two electrodes during capacitor fabrication. This effect has been widely observed previously and likely results from interfaces with different characteristics. Thus, while the shapes of the curves for different polarities are similar, the absolute magnitudes of the leakage currents for the two bias directions are different.[17]

In summary, the temperature dependence for SBT thin film capacitors with platinum electrodes (Pt/SBT/Pt) on silicon wafers was investigated from 10 to 300 K. With decreasing temperature, the unpinning process becomes difficult due to the deepening of the potential well–defect interaction and *kT*, and polarization fatigue becomes significant. The origin

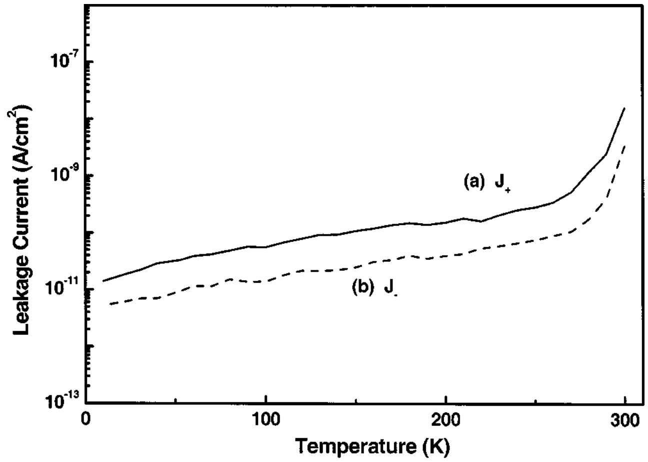

FIG. 4. Temperature dependence of the leakage current of a Pt/SBT/Pt sample at 3 V: (a) J_+ and (b) J_- are leakage currents with the top electrode positively and negatively biased, respectively.

for the temperature effect of $2P_r$ at lower temperature, i.e., the freezing of the switchable polarization, is discussed. These results are helpful in understanding the fatigue-free response observed at room temperature, and are also necessary to fully characterize SBT for memory applications in which a broader temperature range is anticipated.

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