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Effects of poling on the switching properties of $\text{SrBi}_2\text{Ta}_2\text{O}_9$ films

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The effect of poling on the switching properties of $\text{SrBi}_2\text{Ta}_2\text{O}_9$ films was investigated via the technique of switching current testing. The samples with 660 nm thickness were poled under dc voltage ranging from 1 to 8.25 V, and for different duration from 1 to 80 min. After poling, both the net-switched charge and switching time first jumped to higher values, then decreased with the elapse of time. The decreases were separated into two regimes, a fast and slow regime. The change of net-switched charge and switching time with poling voltage and poling time showed nonmonotonic behavior. These results were explained by the change of domain kinetics and the redistribution of charge carriers under both the applied and depolarization field. © 2002 American Institute of Physics. [DOI: 10.1063/1.1471935]

Recently, much attention has been paid to the ferroelectric materials due to their potential applications in piezoelectric, dielectric, optical, pyroelectric, and nonvolatile random access memory (NVRAM) devices.¹ To obtain the optimal pyro- and piezoelectric effect, a poling treatment has to be performed during packaging of these devices.² In the study of NVRAM materials, it was also found that fatigued³ and irradiation damaged⁴ films were rejuvenated through a larger voltage poling. Both of the above could be caused by the effect of poling electric field on the conformation of domains and the mobility of domain walls. However the underlying science of poling remains unsolved.^{5–9} In recent years, $\text{SrBi}_2\text{Ta}_2\text{O}_9$ (SBT) has attracted great interest because of its superior fatigue behavior over $\text{PbZr}_x\text{Ti}_{1-x}\text{O}_3$, while the report of poling effects on SBT materials was seldom found.⁸

Recording of the switching current is the most popular method used to investigate domain kinetics.^{10–13} In this letter, the switching current of SBT films before and after being poled under dc voltage were recorded and compared to investigate the effect of poling on the configuration and mobility of domain walls. This work may provide a different insight into the effect of electric field on ferroelectric films, and help to understand comprehensively the movement of domain walls under electric field.

The SBT thin films were coated on Pt/Ti/SiO₂/Si substrates using conventional metalorganic decomposition (MOD) method, and then dried at 400 °C for 10 min. After this process was performed several times to achieve the desired film thickness, the films were annealed at 750 °C in oxygen ambient for 30 min. A Pt top electrode, with a size of 0.0314 mm², was deposited by sputtering, then a postannealing procedure was used to insure good contact between the Pt electrode and the SBT film. The thickness of the samples was 660 nm.

The samples were first poled for different periods under dc voltage generated by an arbitrary generator (HP33120A).

The measurement of the switching process began right after the dc signal was turned off. Continuous series of bipolar-double-pulse signal (frequency 100 KHz, pulse with 1 μs, amplitude 2 V) were applied from the above generator to the samples, and the switching current was recorded at different moments on a multichannel oscilloscope (HP54825A). The schematics of the testing signal and the typical switching current of our samples were described elsewhere.¹² The switching current was analyzed according to the literature by Larsen *et al.*¹³ The difference between the transient current corresponding to the first positive pulse and that corresponding to the second is called the net-switching current. The integration of this current is called net-switched charge ΔQ , which is equal to the electrode area times ($P_{r+} - P_{r-}$). Here P_{r+} and P_{r-} represent positive and negative remnant polarization, respectively. Switching time t_s is defined as the time from the onset to a point 90% down from the maximum value of net-switching current, and the switching time t_s of our samples varies with the pulse amplitude in a range from 100 to 600 ns.¹² We report here the typical behavior of our samples.

Figure 1 depicts the variation of ΔQ and t_s with the elapse of time after the sample being poled at 8.15 V for 10 min. The y axis is normalized by the corresponding values ΔQ_0 and t_{s0} before poling. It shows that after poling, both ΔQ and t_s increase compared with original values. The films deposited by the MOD method have a multi-crystal structure, hence the domains can orient in any direction. Figure 2 illustrates a simple example about what may happen during poling. Figure 2(a) is the original domain structure of a grain having a 90° domain wall. The domain with the self-polarization originally perpendicular to the external field (E_p), which is usually unswitchable under 2 V testing signal due to the larger energy needed for the growth of 90° domains,^{14,15} can be reoriented to the parallel direction [Fig. 2(b)] during poling because of the much longer poling time compared with that of testing pulse. The whole grain can then be easily switched during testing through the reverse of

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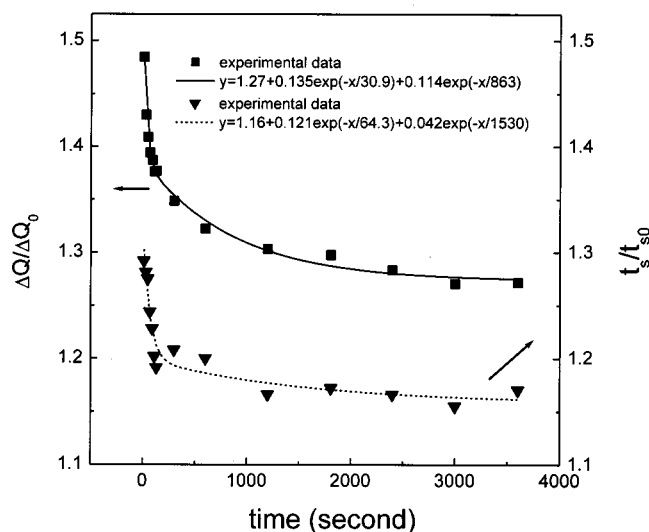


FIG. 1. Normalized net-switched charge and switching time with the lapse of time after being poled at 8.15 V for 10 min.

180° domains, which causes the increase of switchable polarization and in turn the net-switched charge. The switching process includes the following three steps: the nucleation of reverse domains, the forward growth of these domains, and the sideways spreading out of the domains to fill the entire film.¹⁶ After poling, the domain walls of reverse domains should move a longer distance before achieving the reverse of the whole samples due to the larger switchable region. Thus, the switching time must also increase in the case that the nucleation rate and the moving velocity of domain walls remain under the same testing voltage.

The fitting curves in Fig. 1 indicate that the decay behavior afterwards is governed by two mechanisms correlated with the short and the long time regimes as described by

$$\Delta Q/\Delta Q_0 = 1.27 + 0.135 \exp(-t/30.9)$$

$$+ 0.114 \exp(-t/863),$$

$$t_s/t_{s0} = 1.16 + 0.121 \exp(-t/64.3)$$

$$+ 0.042 \exp(-t/1530).$$

After poling, several factors may affect the switching properties. First, depolarization field may cause serious back switching of domains in thin films.¹⁶ Although the depolarization field caused by the discontinuity of polarization on the surface can be quickly compensated by external screening effects¹⁷ through redistribution of charges on the electrodes by the current in the external circuit, the spatial inhomogeneity of the polarization near the interface can still be a source of the decrease of switchable polarization. Second,

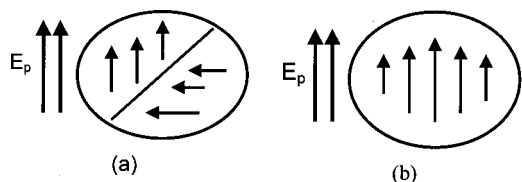


FIG. 2. Schematic diagrams of what may happen during poling. (a) Original domain structure in a grain, (b) domain structure after poling. The polarization direction is marked with arrow, E_p represents the poling field.

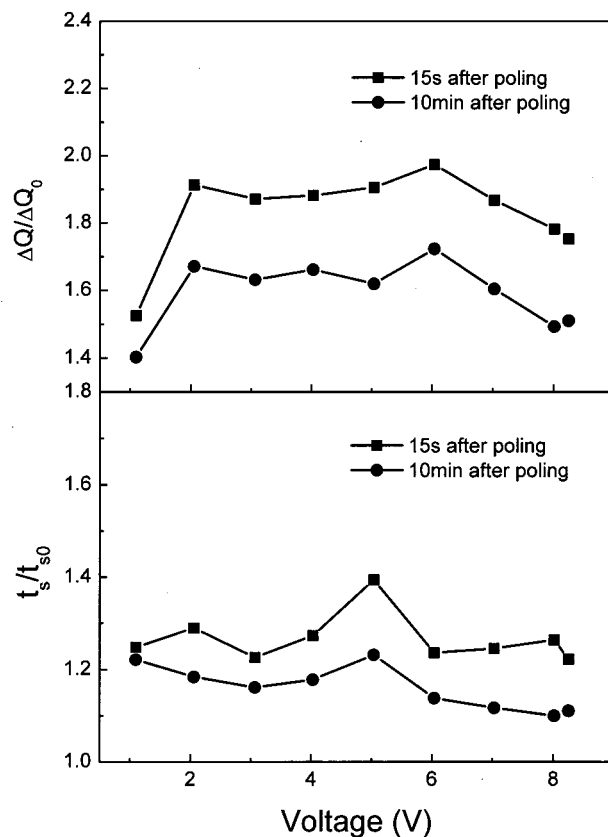


FIG. 3. Normalized net-switched charge and switching time with poling voltage tested at 15 s and 10 min after poling.

internal stresses may affect the switching properties. During poling, the 90° domain switching should cause additional internal stress,^{18,19} which may also induce the domains switching back to their original directions in ferroelectric materials with perfect structures. However, the stress in our sample can be well relaxed by the large amount of imperfect structures in grain boundaries. Third, the redistribution of space charge, dipolar defects and charged defects under both depolarization and external field may affect the switching properties. The redistribution of charge carries in the bulk to compensate the depolarization field is called bulk screening as described by Shur.¹⁷

So, the fast decay of ΔQ in Fig. 1 must be due to the depolarization field and the somewhat retarding bulk screening effect. The long time regime might come from the pinning effect of domain walls by charge carriers, which was usually mentioned as the main cause of fatigue.¹⁶ Contrary to poling effects, the decrease of switchable polarization is sure to be related to the smaller distance the domain walls should move during switching, and in turn the decrease of switching time.

The samples discussed below were poled for 5 min at different voltages from 1 to 8.25 V (Fig. 3). Poling effects are so obvious even at 1 V, a voltage smaller than the testing voltage 2 V. The net-switched charge rises obviously with the voltage changing from 1 to 2 V, which indicates that more domains might turn to the direction parallel to the applied field during poling at 2 V through 90° domain switching. The condition of 2 V dc voltage and 5 min duration seems sufficient to approach the optimal poling effect, so the net-switched charge keeps little change in the voltage range from

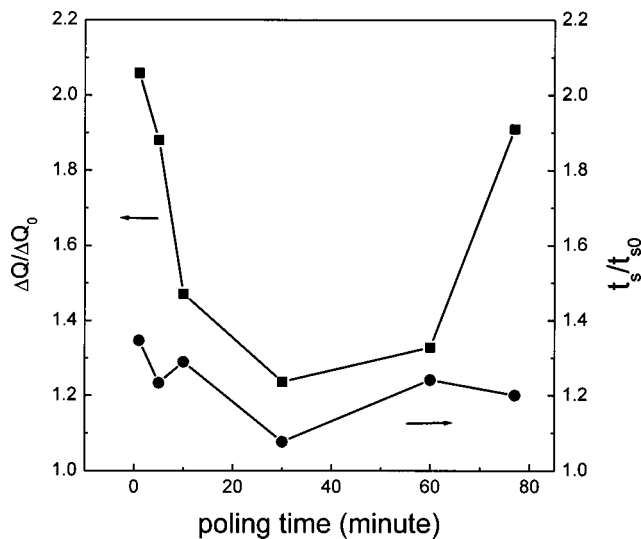


FIG. 4. Normalized net-switched charge and switching time with poling time tested at 15 s after poling.

2 to 6 V. With the further increase of voltage, large amounts of space charge and charged defects most probably accumulate near the electrodes, such as, positively charged oxygen vacancies Vo^{oo} in front of the cathode, with negative charge carriers in the anode.^{6,9} During poling, these charge carriers could shield the applied field and lead to the decrease of effective field. After poling, although these charge carriers will diffuse away thermally or under the promotion of alternating testing signal, the less movable charge carriers such as oxygen vacancies and dipolar defects can still act to some extent as a prevention of domain switching, which leads to the decrease of ΔQ .

The variation of switching time with voltage can be explained as follows: Switching time is mostly determined by the switching time needed for big domains. Whereas the domain size is restricted by the grain size in films, so the switching time changes only a little with voltage. There seems to be abnormal around 5 V, which might be the extreme when approaching the optimal poling effect.

Figure 4 is the variation of ΔQ and t_s with poling time tested at 15 s after poling at 8.2 V. With the increase of time from 1 to 80 min, ΔQ decreases first, then increases. One minute seems to be sufficient for the optimal poling effect. Then with the time increasing to 30 min, more charge carriers could accumulate near the electrodes, which would decrease ΔQ as described above. With the further increase of time, the rapid increase of the leakage current was observed during poling, and most samples would break down if being poled for a period longer than 80 min. This demonstrates that some of the charge carriers are likely to accumulate on grain boundaries to form current access, thus the charge carriers near the electrodes might be partially neutralized, so as to decrease the screening effect on both the poling and testing

field. That is why ΔQ increase with poling time in the period from 30 to 80 min.

In Fig. 4, t_s also almost remains except for an abnormal minimum at 30 min, which can be explained similar to Fig. 3. The biggest size of switchable domains keeps to be almost the same except that the screening effect of charge carriers reaches maximum around 30 min.

It is valuable to note that the switching properties of our samples without breakdown can be restored after two days without any additional treatment. It seems that with the re-conversion of the domain structure, the charge carriers that redistributed during poling might also be restored through thermal diffusion and trapping of space charge.

In summary, the effects of poling on the switching properties of SBT films were studied by the technique of switching current testing. After poling, both the net-switched charge and switching time first jumped to high values, then decayed. The relation between the change of the above values and the poling conditions (voltage and time) was non-monotonic based on our typical results. Our results could be explained by the domain kinetics and redistribution of charge carriers under both the external and depolarization field. Although our experiments shine some lights on poling effects, further experiments are still needed to verify the real mechanism.

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