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A MODEL FOR OPTICAL AND ELECTRICAL POLARIZATION FATIGUE IN $SrBi_2Ta_2O_9$ AND $Pb(Zr,Ti)O_3$

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Abstract We find that significant polarization fatigue (> 90%) can be induced in $SrBi_2Ta_2O_9$ (SBT) thin films using (a) broad-band optical illumination combined with a bias near the switching threshold and (b) electric field cycling under broad-band optical illumination. In the latter case, the extent of polarization fatigue increases with decreasing cycling voltage. In either case, the optically fatigued SBT capacitors can be fully rejuvenated by applying a saturating dc bias with light or by electric field cycling without light, which suggests a field-assisted recovery mechanism. A similar behavior was observed in $Pb(Zr,Ti)O_3$ (PZT) films with LSCO electrodes. Based on these results, we suggest that *polarization fatigue in ferroelectrics is essentially a dynamic competition between domain wall pinning due to electronic charge trapping, and field-assisted unpinning of the domain walls.* Thus, domain wall pinning is not necessarily absent in nominally fatigue-free systems. Instead, these systems are ones in which domain wall unpinning occurs at least as rapidly as any pinning. Factors which may affect the pinning and unpinning rates will be discussed.

INTRODUCTION

Polarization fatigue, the decrease in switchable polarization with electric field cycling, has received considerable attention lately because ferroelectric thin films are being evaluated for use in nonvolatile memory applications.[1-6] $Pb(Zr,Ti)O_3$ (PZT) and $SrBi_2Ta_2O_9$ (SBT) thin films are currently the two most promising ferroelectric materials for nonvolatile memory applications. PZT is attractive because it is a well-studied system which has excellent ferroelectric properties and can be processed at relatively low temperatures (550-650°C). However, PZT thin films with Pt electrodes exhibit significant polarization fatigue, although this problem has been mostly alleviated by using oxide electrodes such as $La_{1-x}Sr_xCoO_3$[7], RuO_2[8], $SrRuO_3$[9], and IrO_2[10]. In comparison, SBT is attractive because it exhibits negligible polarization fatigue even with simple metal electrodes, such

as Pt.[11,12] However, SBT films require rather high processing temperatures (750-800°C), which poses serious challenges for their integration with Si-based microelectronic devices.

Several models have been proposed to explain polarization fatigue in the PZT system.[1-6] In one model[1], it is suggested that oxygen vacancies accumulate at the film/electrode interface during cycling and subsequently form oxygen-deficient dendrites which grow into the bulk of the film. The regions of the film in between the dendrites are screened from the applied field and can no longer switch, leading to polarization loss.[1] Another model[2] is based on the effective one directional motion of oxygen vacancies and their entrapment at the film/electrode interface, resulting in polarization loss due to structural damage at the interface. These two models implied that oxygen vacancies are the dominant point defect which controls fatigue in Pt/PZT/Pt capacitors.

Recently, additional insight has been provided by studies which demonstrated that polarization suppression in PLZT films and ceramics can be induced by optically illuminating the films with band-gap light while applying a dc bias just below the switching threshold.[13] Furthermore, it was demonstrated that the switchable polarization of optically-suppressed and electrically-fatigued Pt/PZT/Pt capacitors could be fully rejuvenated by the simultaneous application of a saturating bias and band-gap light.[3] These results indicated that fatigue is basically due to pinning of domains by charge trapping at domain boundaries and that the polarization discontinuity at domain boundaries acts as the driving force for charge trapping.

The dependence of fatigue on the electrode material in PZT films provided compelling evidence that fatigue is controlled, at least in part, by the interface properties. It was also shown that both the top and bottom electrode interfaces play a nearly equivalent role in the fatigue process.[14] To account for fatigue dependence on the electrode material, it has been suggested that oxide electrodes act as sinks for oxygen vacancies, thereby preventing their accumulation at the film/electrode interface.[4] Thus, the currently available data suggest that both oxygen vacancies and electronic charge carriers play a role in the fatigue process. Warren at al. suggested the oxygen vacancies ($V_O^{\cdot\cdot}$) may actually stabilize the trapped electronic charge and consequently stabilize domain wall pinning.[15]

In the case of SBT, we have recently discovered that optical illumination can result in significant polarization fatigue in Pt/SBT/Pt capacitors.[5-6] Similar effects were also observed in LSCO/PZT/LSCO capacitors.[6] These results[5-6] indicated that the Pt/SBT/Pt and LSCO/PZT/LSCO systems are in fact susceptible to fatigue effects even though they exhibit negligible loss in the switchable polarization using standard fatigue testing conditions (cycling to saturation/no light). In this paper, we propose a qualitative model

which explains the lack of electrical fatigue in Pt/SBT/Pt and LSCO/PZT/LSCO capacitors, the photo-induced changes in their fatigue behavior, and the electrode dependence of electrical fatigue in PZT films.

EXPERIMENT

SBT and PZT films were synthesized using solution chemistry techniques described elsewhere.[16,17] The SBT films were deposited on Pt coated SiO_2/Si substrates, while the PZT films where deposited on Pt or LSCO coated SiO_2/Si substrates. The films were deposited using a multilayer spin-coating approach. Each layer was spin-coated at 3000 rpm for 30 s and then heat treated at 300°C for 5 min. The films were 0.4-0.5 μm thick after crystallization. The SBT films were crystallized in an oxygen ambient at 750°C for 30 min, while the PZT films were crystallized at 650°C for 30 min in air.

Top semi-transparent electrodes (300 μm diameter) of Pt (SBT and PZT films) and LSCO (PZT films) were sputter-deposited to permit the study of photo-induced effects. The ferroelectric hysteresis loops were measured using an RT66A ferroelectric tester from Radiant Technologies. The electrical fatigue tests were performed using a 25 kHz, sinusoidal drive voltage. A 200 W Oriel Hg arc lamp was used to optically illuminate the SBT and PZT capacitors. Although, strong photo-induced changes in the hysteresis and fatigue behavior were observed using band-pass (λ <550 nm) and narrow-band interference (λ = 365 nm) filters, broad-band unfiltered light was used in these experiments to maximize the photo-induced changes.

RESULTS AND DISCUSSION

Normal Fatigue Tests

Fig. 1 shows the fatigue behavior of Pt/SBT/Pt, LSCO/PZT/LSCO, and Pt/PZT/Pt capacitors performed under normal testing conditions (cycling to saturation/ no light). As expected, both Pt/SBT/Pt and LSCO /PZT/LSCO capacitors show negligible polarization fatigue, while the Pt/PZT/Pt capacitor undergoes substantial polarization fatigue during electric field cycling. It is important to note here that subjecting the fatigued Pt/PZT/Pt capacitor to a combination of light and saturating dc bias restores more than 90% of the switchable polarization.[3] This result clearly demonstrates that electronic charge carriers are involved in the fatigue process. In the remainder of this paper, we present results of optical and electrical experiments aimed at understanding the differences in fatigue behavior shown in Fig. 1.

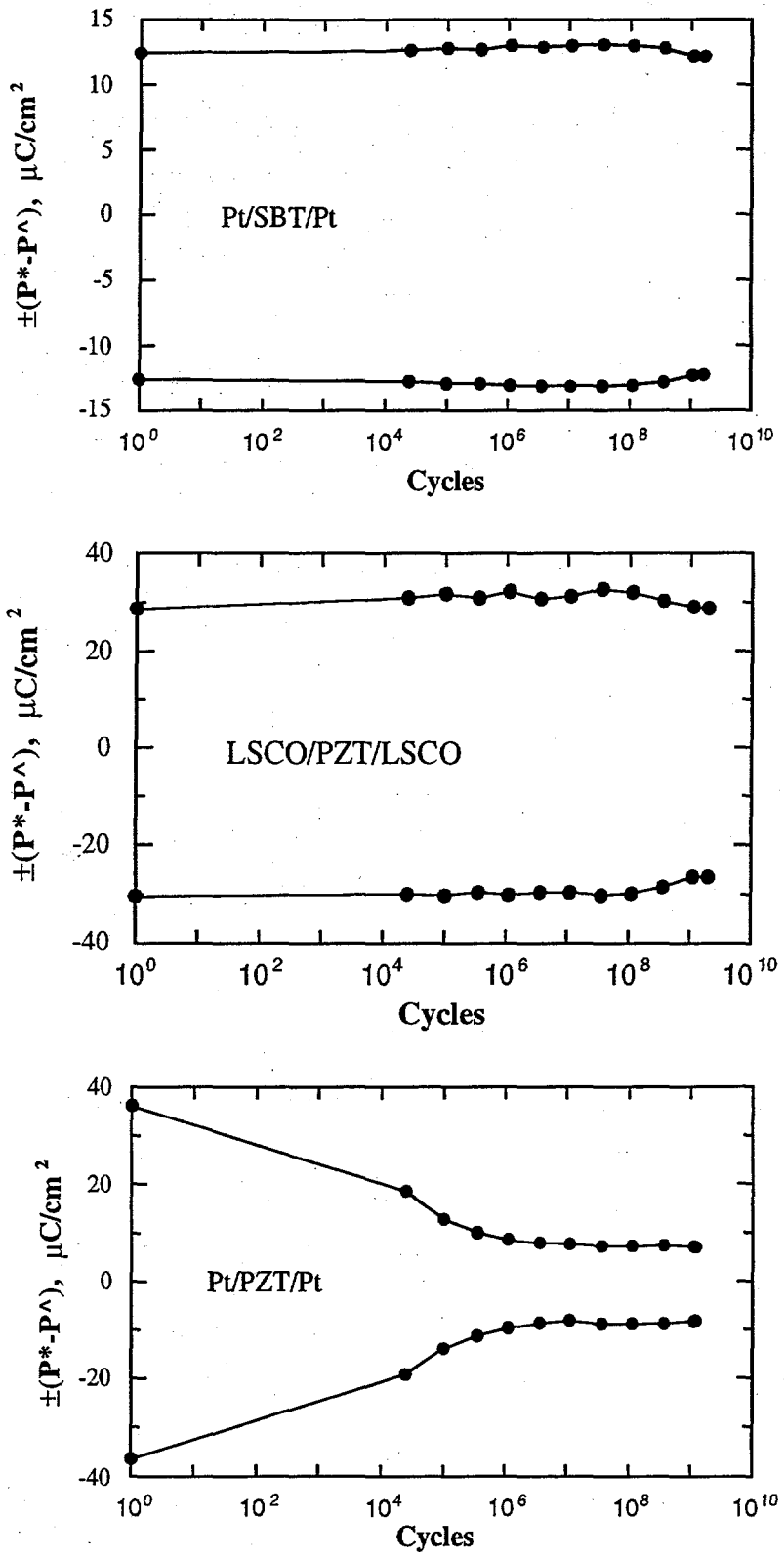


FIGURE 1 Electrical fatigue behavior of Pt/SBT/Pt, LSCO/PZT/LSCO, and Pt/PZT/Pt capacitors.

Optically-Induced Polarization Suppression

Fig. 2 shows the effect of simultaneously applying a dc bias near the switching threshold (~Vc) and broad-band light to the SBT and PZT capacitors. Notice that under these conditions (+0.8V, light), significant polarization suppression is induced in all three capacitors, regardless of the electrode material. The use of a bias near the switching threshold (~Vc) of these films, was based on previous results which demonstrated that the optically-induced polarization suppression in Pt/PZT/Pt films is maximum when a dc bias near the switching threshold of the illuminated film is used.[13]

The polarization suppression in Fig. 2 shows, for the first time, that the Pt/SBT/Pt and LSCO/PZT/LSCO capacitors are susceptible to fatigue-like effects. It is worth noting that the optically suppressed state shown in Fig. 2 is a very stable one. In fact, very little polarization could be restored by applying up to 20 Volts (without light) to the optically suppressed capacitors. In addition, the amount of polarization suppression depends on the illumination time used. For instance, Fig. 2 shows that 30% of the switchable polarization of the Pt/SBT/Pt capacitor is suppressed using a 30 sec illumination time. However, when the exposure time was increased to 10 min, about 90% of the switchable polarization was suppressed.

The ability to suppress the switchable polarization using the light/+0.8V combination can be explained based on a model previously proposed for Pt/PZT/Pt thin films.[13] Basically, we suggest that the polarization suppression in all three capacitors shown in Fig. 2 results from trapping of the photogenerated carriers (electrons and holes) at domain boundaries which have significant polarization discontinuity. These boundaries constitute an electrostatic potential well which attracts the photogenerated carriers. Once the photogenerated carriers are trapped at such a boundary, they tend to pin it, thereby reducing the switchable polarization. [13] The population of domain boundaries with a polarization discontinuity is expected to be maximized during switching, which explains why maximum polarization suppression is achieved by biasing the capacitors near the switching threshold (~ 0.8V for these films). Additional support for the electronic nature of the polarization suppression shown in Fig. 2 is that the simultaneous application of a saturating bias (±8V for these films) and light results in essentially complete capacitor rejuvenation. In this case, the photogenerated carriers recombine with the trapped charge and the saturating bias serves to realign the domains.

Voltage Cycling of Optically Suppressed Capacitors

Next we investigated the effect of voltage cycling, without light, on polarization recovery in optically-suppressed capacitors. Fig. 3 shows the effect of electric field cycling (±8V, without light) on polarization recovery in the optically-fatigued capacitors shown in

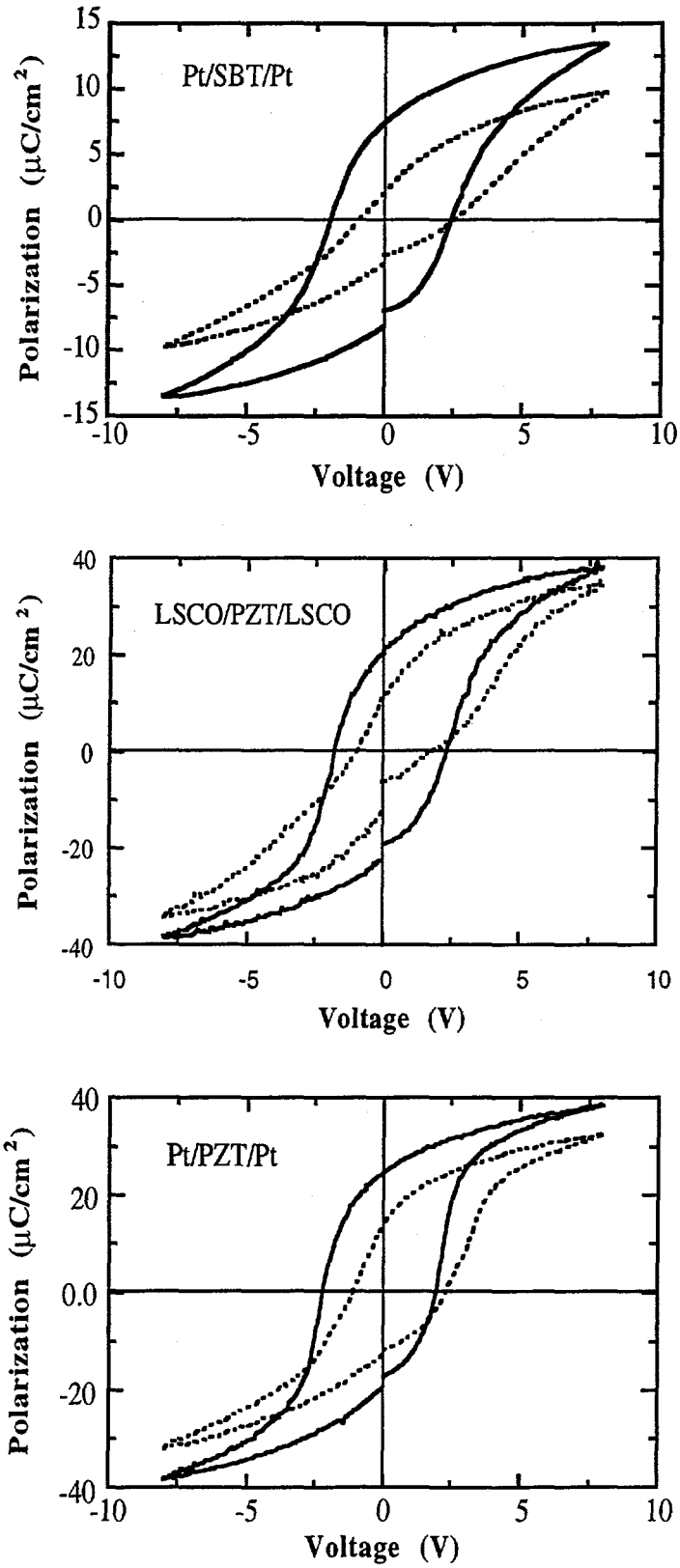


FIGURE 2 Hysteresis loops of three capacitors before (solid line) and after (broken line) illuminating the capacitors with broad-band light while applying a +0.8V dc bias.

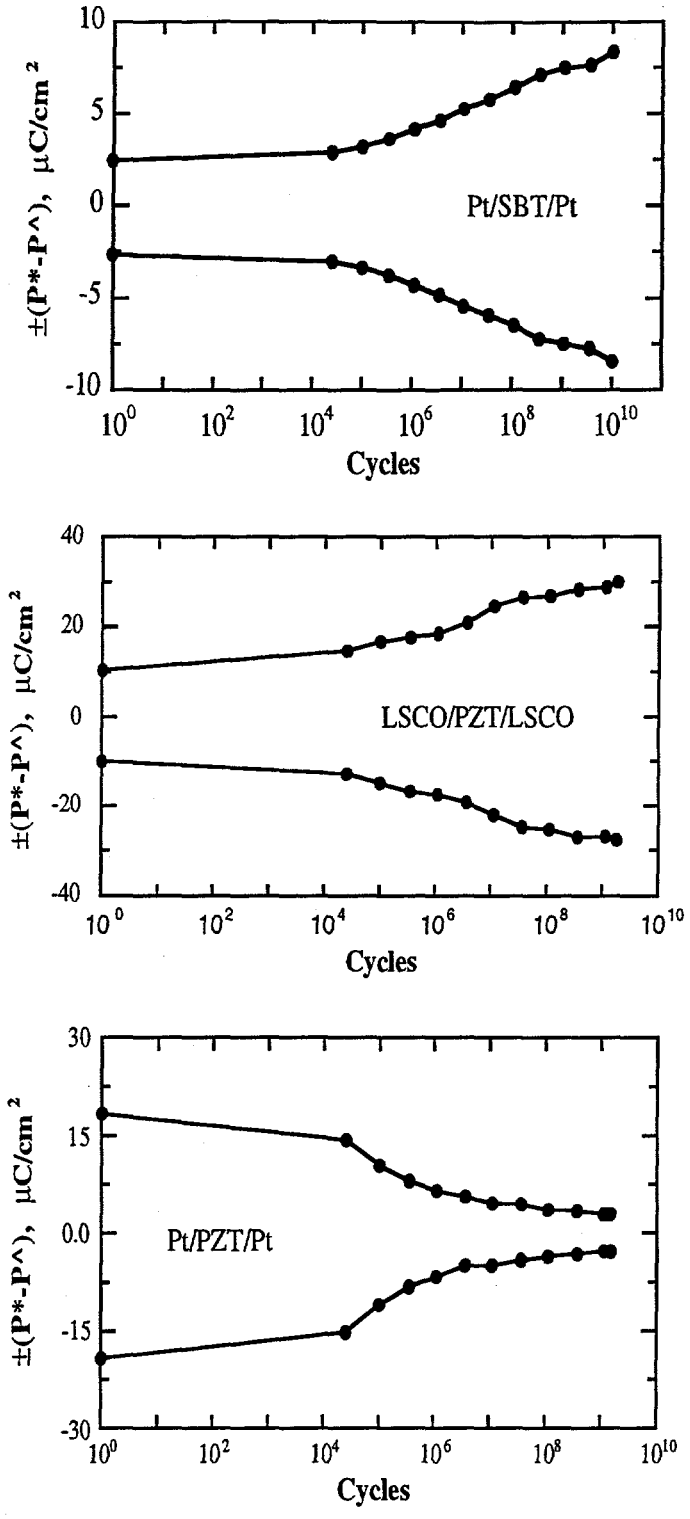


FIGURE 3 Effect of voltage cycling (±8V) on polarization recovery in the same three capacitors of Fig. 2. The voltage cycling was performed in the dark after the polarization has already been optically-suppressed as shown in Fig. 2.

Fig. 2. Notice that the Pt/SBT/Pt and the LSCO/PZT/LSCO capacitors tend to recover to their initial polarization state with electric field cycling. In contrast, electric field cycling of the Pt/PZT/Pt capacitor leads to additional suppression in the switchable polarization. The behavior shown in Fig. 3 was also reflected in the hysteresis loops (not shown) taken before and after subjecting the optically-suppressed capacitors to voltage cycling.

The recovery behavior shown in Fig. 3 indicates that domain wall unpinning by the cycling field is the dominant process which occurs during cycling of Pt/SBT/Pt and LSCO/PZT/LSCO capacitors. In contrast, optically-suppressed Pt/PZT/Pt capacitors continue to lose switchable polarization when subjected to electric field cycling, which suggests that domain wall pinning (due to electronic charge trapping) is the dominant process during cycling. In the absence of light, the trapped electronic charge carriers are most likely injected from the electrode into the film since the cycling electric fields are quite high (~200 kV/cm).

We therefore suggest that the difference in the normal fatigue (Fig. 1) and polarization recovery (Fig. 3) behavior between Pt/SBT/Pt and LSCO/PZT/LSCO on the one hand, and Pt/PZT/Pt on the other, implies the following: the extent of polarization fatigue in a given material system is determined by a dynamic competition between domain wall pinning due to trapping of electronic charge carriers, and field-assisted domain wall unpinning. This conclusion is further substantiated by additional experimental data presented next.

Fatigue Testing Under Light

In order to verify the above proposed view of polarization fatigue in ferroelectrics, we performed electrical fatigue tests at different cycling voltages and under optical illumination (Fig. 4). The fatigue tests were performed on fresh capacitors using 25 kHz, ±8V and ±4 V cycling voltages combined with broad-band light. Fig. 4 reveals several interesting points. First, the extent of polarization fatigue in all three capacitors increases with decreasing cycling voltage. This voltage dependence supports the idea that field-assisted unpinning of the domain walls occurs during voltage cycling.

Secondly, since illuminating the samples during a fatigue test increases the concentration of electronic charge carriers, the more pronounced fatigue of the Pt/SBT/Pt and LSCO/PZT/LSCO capacitors under light (Fig. 4) compared to cycling in the dark (Fig. 1) is consistent with the notion that electronic charge trapping is an important component of the fatigue process. The higher concentration of electronic charge carriers in the presence of light can result in enhanced charge trapping and consequently enhanced domain wall pinning. One exception to this behavior is the Pt/PZT/Pt capacitors when the fatigue tests are performed using a saturating bias (±10 Volts). In this case, reduced

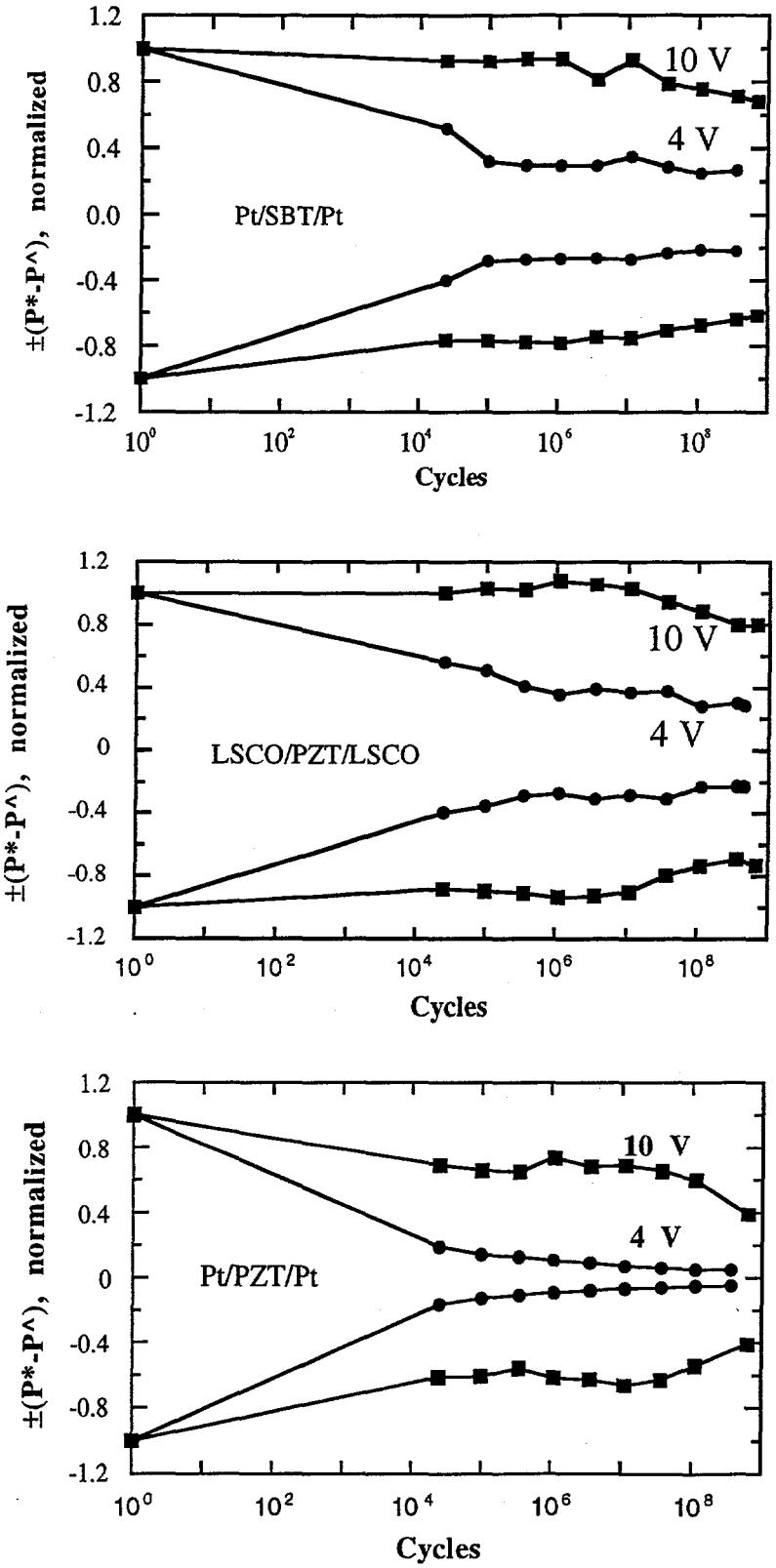


FIGURE 4 Electrical Fatigue tests performed under broad-band optical illumination at two voltages (±4 and ±10 V).

polarization fatigue is observed when cycling is performed under light. This effect has also been previously reported for Pt/PZT/Pt capacitors.[18] One possible explanation for this behavior might be that significant charge injection from the Pt electrode into the PZT film occurs when cycling with a saturating bias (±10V); this results in recombination with the photogenerated carriers. As a result, the electronic carrier concentration in the film is reduced, leading to less charge trapping and therefore less domain wall pinning (i.e., reduced fatigue).

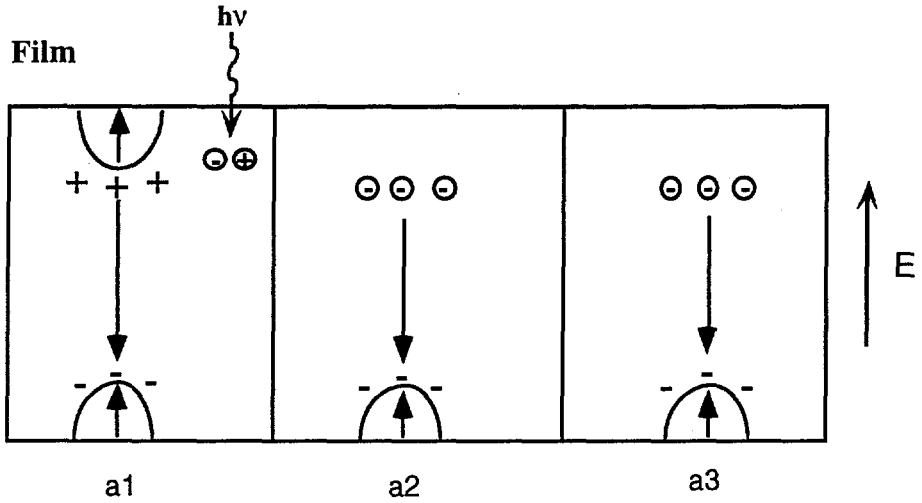
MODEL SUMMARY

Based on the above results we suggest that electrical fatigue in ferroelectric materials is a dynamic *competition between domain wall pinning due to electronic charge trapping, and field-assisted unpinning of the domain walls.* In accordance with this argument, we suggest that during the normal fatigue tests (cycling to saturation/ no light), the trapping electronic charge carriers at defect sites near the domain boundaries leads to domain wall pinning. In the absence of light, the electronic charge carriers are likely to be injected from the electrode or created within the ferroelectric material during electrical fatigue testing. The idea of charge being injected from the electrode or created within the ferroelectric during fatigue testing is plausible given the magnitude of the cycling fields (~200 kV/cm). Additional evidence for the electronic nature of the fatigue process was evident from the experiment in which electrically fatigued Pt/PZT/Pt capacitors could be rejuvenated by simultaneously applying light and a saturating bias.[3]

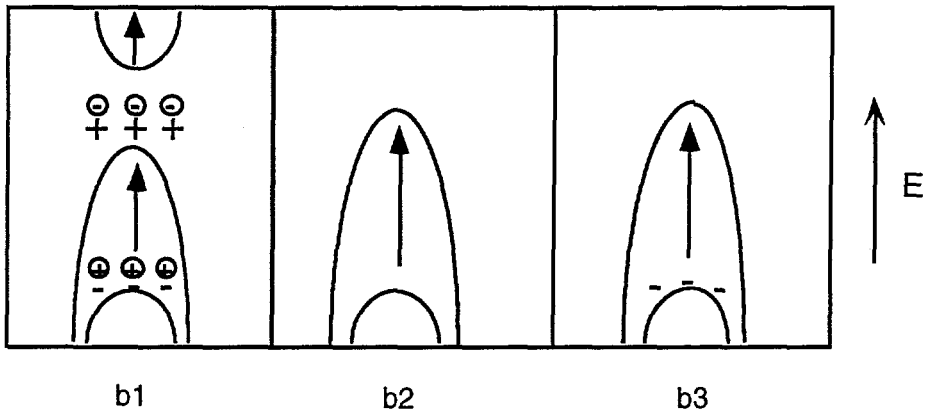
In addition to domain wall pinning due to charge trapping, field-assisted domain wall unpinning also occurs during electrical fatigue testing. This fact is substantiated by the voltage dependence of fatigue shown in Fig. 4. Thus, during voltage cycling of a ferroelectric material both domain wall pinning and unpinning are expected to occur. As Fig. 5 illustrates, the pinning process results from electronic charge trapping at domain boundaries with a polarization discontinuity. The trapped electronic charge carriers may be injected from the electrode material or photogenerated by illuminating the sample. The unpinning processes might include (a) overcoming the pinning forces by the cycling field, (b) field-assisted detrapping of trapped carriers, which results in domain wall unpinning, and (c) recombination of photogenerated or injected carriers with the trapped charge, which can also lead to domain wall unpinning.

Thus, domain wall pinning is not necessarily absent in nominally fatigue-free systems. Instead, these systems are ones in which domain wall unpinning exceeds any pinning. Therefore, the fatigue performance of a given material system will be determined by its relative pinning and unpinning rates. This implies that domain wall

(a) Domain Wall Pinning :



(b) Domain Wall Unpinning:



⊖ and ⊕ : injected or photogenerated electronic charge carriers

- and + : electronic charge carriers trapped at a domain boundary

⊖/+ and ⊖/- : recombination of charge carriers

FIGURE 5 A schematic illustrating (a) domain wall pinning due to charge trapping, and (b) domain wall unpinning. The schematic shows a hypothetical thin film in which three domains are initially poled down. The switching process proceeds by nucleating domains of the opposite direction to the intial state. During switching, the injected (a2 and a3) or photogenertaed (a1, if sample is illuminated) charge carriers are attracted by the polarization discontinuity near the domain boundaries, where they become trapped at defect states near the domain walls, leading to domain wall pinning. Domain wall unpinning can now occur by recombination of injected or photogenerated charge carriers with those already trapped(b1), by field-assisted detrapping of trapped charge (b2) or by simply overcoming the pinning forces by the applied field (b3).

unpinning in Pt/SBT/Pt and LSCO/PZT/LSCO systems must exceed the pinning rate, leading to no net polarization loss during normal fatigue tests (cycling to saturation/no light), while the opposite is true for the Pt/PZT/Pt capacitors.

In the remainder of this paper, we present plausible reasons as to why the unpinning rates apparently exceed domain wall pinning in the Pt/SBT/Pt and LSCO/PZT/LSCO systems, but not in the Pt/PZT/Pt system.

FACTORS WHICH MAY AFFECT THE PINNING AND UNPINNING RATES

Pt/SBT/Pt Capacitors

We suggest that domain wall pinning in SBT is weak as compared to PZT, leading to the dominance of the unpinning process during electric field cycling. The weak domain wall pinning in SBT is proposed to result from (a) a smaller magnitude of ferroelectric polarization, (b) shallow electronic charge trapping centers, and (c) lower $V_O^{\cdot\cdot}$ concentration in the SBT perovskite sublattice as compared to PZT.

The relatively small magnitude of ferroelectric polarization in SBT can lead to weaker domain wall pinning by either (1) weak trapping of individual charges, or (2) a lower concentration of trapped charge at the domain boundary. In the case of weak trapping, rejuvenation by field-assisted detrapping requires that the applied field (~200 kV/cm) is sufficient to detrap the charge carriers. The trap depth for the charge is expected to depend on both the type of defects present and on the depth of the electrostatic potential well at the domain boundary, which is determined by the magnitude of the ferroelectric polarization. Since SBT has lower polarization than typical PZT compositions, the contribution of ferroelectric polarization to the trap depth should be smaller in SBT.

In the other case, the density of trapped charge at a domain boundary is determined by the availability of trapping sites (i.e., defects) and by the value of the remanent polarization, Pr. The remanent polarization sets the upper limit on the trapped charge density, because the charge is trapped to compensate the polarization discontinuity at a domain boundary. The smaller remanent polarization of SBT may, therefore, lead to lower trapped charge densities and, thus, to weaker domain wall pinning than exhibited by PZT thin films.

The second factor which may contribute to the proposed weak domain wall pinning in SBT is the recent calculations which show that defect states in SBT are shallow.[18] Specifically, it was found that a hole trapped at the Bi^{3+} ion, Bi^{4+} center, and an electron trapped at a Ta^{5+} ion, Ta^{4+} center, are both shallow.[19] These results indicate that charge trapping in SBT may be relatively weak and therefore field-assisted detrapping (which

leads to domain wall unpinning) can be readily induced by the cycling voltage during a fatigue test.

A third possibility which may also contribute to weaker domain wall pinning in SBT is a lower oxygen vacancy concentration in the perovskite sublattice of SBT as compared to PZT. While the volatility of Bi_2O_3 in SBT may lead to $V_O^{\bullet\bullet}$ formation (associated with Bi vacancies), it is plausible to expect these oxygen vacancies to be mostly located in the Bi_2O_3 layer. Compared to PZT, the A-site cation in the perovskite sublattice of SBT (Sr) is less volatile than Pb, and therefore it is plausible to expect a lower $V_O^{\bullet\bullet}$ concentration in the perovskite sublattice of SBT compared to PZT. This conclusion is important as most of the ionic displacements responsible for the appearance of the ferroelectric polarization in SBT occur in the perovskite sublattice.[20] Warren et al. have recently shown that oxygen vacancy-induced distortions in the oxygen octahedron of PZT tend to stabilize the trapped charge during electrical fatigue, thereby leading to enhanced domain wall pinning.[15] This result implies that it is oxygen vacancy formation in the perovskite sublattice that is important for the stabilization of domain wall pinning. The possibility of lower $V_O^{\bullet\bullet}$ concentration in the perovskite sublattice of SBT may lead to less stable domain wall pinning, which can also contribute to the lack of electrical fatigue in SBT.

LSCO/PZT/LSCO Capacitors

In contrast, the lack of electrical fatigue in the LSCO/PZT/LSCO capacitors cannot be attributed to weaker domain wall pinning. This conclusion is supported by the fact that replacing the top LSCO electrode by Pt leads to severe polarization fatigue in the same film. Since replacing the top electrode does not affect the bulk of the film, it can be concluded that the strong dependence of polarization fatigue on electrode material for PZT must be related to the effect of the film/electrode interface on the relative pinning and unpinning rates during electric field cycling. We suggest that the electrode-dependent polarization fatigue in PZT films can be explained by (a) differences in the charge injection from the electrode into the film during cycling, and (b) the ability of the oxide electrode (LSCO) to act a sink for oxygen vacancies.[4]

In the first case, it is possible that the metallic electrode (Pt) can inject more charge into the film during cycling than the oxide electrode (LSCO); this implies that charge trapping and therefore domain wall pinning is smaller in LSCO/PZT/LSCO capacitors. In the other case, the ability of LSCO to act as a sink for $V_O^{\bullet\bullet}$ can prevent their accumulation near the film/electrode interface during electric field cycling. The reduced accumulation of oxygen vacancies in PZT films with LSCO electrodes can influence their fatigue behavior in two ways. First, oxygen vacancies have been shown to stabilize

charge trapping and consequently domain wall pinning.[15] Thus, a reduced oxygen vacancy concentration near the interface implies less stable charge trapping and domain wall pinning near the interface of the LSCO/PZT/LSCO capacitors. Secondly, the accumulation of $V_O^{\bullet\bullet}$ can potentially lead to the formation of an n-type layer near the interface;[1,21] such an n-type layer would result in an increase in the electron injection rate, leading to an increase in charge trapping and, thus, pinning rates. In fact, an increase in the injection rate was indicated by the dramatic increase in the leakage current exhibited by electrically fatigued Pt/PZT/Pt capacitors.[1] Therefore, a reduced $V_O^{\bullet\bullet}$ accumulation near the interface of LSCO/PZT/LSCO suggests reduced electronic charge injection rates; this implies reduced charge trapping and consequently less domain wall pinning.

Pt/PZT/Pt Capacitors

The significant polarization fatigue of Pt/PZT/Pt capacitors implies a dominance of domain wall pinning over unpinning during electric-field cycling. This can be attributed to (a) larger ferroelectric polarization as compared to SBT, leading to enhanced electronic charge trapping and/or deeper trap depth (both effects lead to stronger domain wall pinning in PZT) and (b) oxygen vacancy accumulation near the film/electrode interface, since Pt cannot act as a sink for $V_O^{\bullet\bullet}$.

CONCLUSIONS

A qualitative fatigue model based on the premise that *fatigue is essentially a dynamic competition between domain wall pinning due to electronic charge trapping, and field-assisted domain wall unpinning* is proposed. This result implies that domain wall pinning is not necessarily absent in nominally fatigue-free systems, but rather that these systems are ones in which unpinning occurs at least as rapidly as pinning. The pinning and unpinning rates can be influenced by at least four factors: (a) availability of electronic charge carriers (b) the magnitude of ferroelectric polarization (b) concentration and mobility of oxygen vacancies and (c) the nature and concentration of the trapping centers present in the material. These arguments can explain the lack of electrical fatigue in SBT and the electrode dependence of fatigue in PZT thin films. They can also explain the photo-induced changes in the fatigue behavior of both materials.

ACKNOWLEDGMENTS

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