Kinetic approach to fatigue phenomenon in ferroelectrics

Vladimir Ya. Shur,a) Evgenii L. Rumyantsev, Ekaterina V. Nikolaeva, Eugene I. Shishkin, and Ivan S. Baturin
Institute of Physics and Applied Mathematics, Ural State University, Ekaterinburg 620083, Russia
(Received 14 June 2001; accepted for publication 11 September 2001)

We propose an approach to the explanation of the fatigue effect as an evolution of the switching area during cyclic switching as a result of self-organized domain kinetics due to retardation of bulk screening of the depolarization field. The formation of spatially nonuniform internal bias field during cycling (kinetic imprint effect) slows the domain kinetics in some regions leading to formation of the kinetically frozen domains. Presented fatigue and rejuvenation experimental data measured in sol-gel PbZr1−xTixO3 thin films are in accordance with the results of computer simulation. © 2001 American Institute of Physics. [DOI: 10.1063/1.1418008]

I. INTRODUCTION

It has been shown in classical works1,2 that ferroelectric materials demonstrate essential decreasing of the switching charge during ac voltage cycling known as fatigue phenomenon. In recent years this effect has been intensively studied in thin films (mostly in PbZr1−xTixO3), which are widely discussed for application in ferroelectric memory devices.3–18 The knowledge of the fatigue origin is of principal importance for improving the film endurance, which is essential for ferroelectric nonvolatile memories (FeRAM).

Several alternative scenarios for the suppression of the switching polarization have been proposed. Bulk-locking mechanism is based on the assumption that the charged domain walls are pinning with the help of electric charge trapping centers.12 The interface scenario supposes the inhibition of domain growth due to inhibition of nucleation at the electrode interfaces.7–9 The crucial role of electromigration of oxygen vacancies is discussed.16,17 The growth of the passive surface layer with damaged ferroelectric properties during cycling is proposed recently14 and must be stressed that the fatigue process is always considered without relation with domain structure evolution during cycling, though it was shown by direct observations using scanning probe microscope that fatigue in thin films manifests itself through arising and growth of complicated structure of nonswitched regions (“frozen” domains).11,19

In this work we propose an approach, which allows us to describe an evolution of switching area during cycling. We point out the crucial role of retardation effect during bulk screening of the depolarization field. During cycling this effect leads to self-organized formation of spatially nonuniform internal bias field (kinetic imprint), which suppresses arising/nucleating of new domains and slows the polarization reversal. As a result the switching becomes incomplete due to inhibition of nucleation in the regions with maximum value of internal bias field. The main model predictions such as the existence of rejuvenation stage and the evolution of the geometry of switching area are in accord with the experiment.

II. MODEL

It is known that in the ferroelectric capacitor after polarization reversal the external screening through charge redistribution at the electrodes (switching current) rapidly compensates the depolarization field $E_{\text{dep}}$. Nevertheless the residual depolarization field $E_{\text{rd}} = E_{\text{dep}} - E_{\text{est},\text{cr}}$ remains due to the existing intrinsic surface dielectric gap.20,21 Slow compensation of $E_{\text{rd}}$ by bulk screening20–24 leads to the arising of the internal bias field $E_b$ observed as a loop shift (imprint effect25,26) for ac cyclic switching with period $T$ much shorter than the bulk screening time constant $\tau$.21,22 It was shown that $E_b$ can be spatially nonuniform and can change during long-enough cycling.21–23 Three groups of bulk screening mechanisms are revealed and widely discussed: (1) reorientation of the defect dipoles,27,28 (2) redistribution of bulk charges/carriers,20–23 and (3) injection from the electrodes.7–9

In our model we account for the fact that the domain kinetics during cycling is a self-organized process, because the spatial distribution of $E_b$ depends on the previous domain evolution and in turn defines subsequent domain kinetics. The local value of $E_b$ tends to the field value defined by the relative difference of stay duration of the given sample region in the states with opposite polarization directions [Fig. 1(b)]. Thus both sign and value of $E_b$ become spatially nonuniform and $E_b$ distribution function varies with the number of cycles. Our simulation of domain kinetics shows that this kinetic imprint effect leads to arising and growth of kinetically frozen regions.

The domain kinetics during periodical switching in rectangular pulses in uniaxial thin ferroelectric was simulated on 2D matrix $L \times L$. We use the classical model of polarization reversal,29,30 where the domain kinetics is achieved through domain nucleation and domain wall motion by generation and growth of steps. Thus we have distinguished three nucleation processes: (1) step growth by nucleation at the step edge, (2) step generation by nucleation at the wall, (3) new domain formation by appearance of isolated nucleus [Fig. 1(a)]. The nucleation probability at a given point $(i,j)$ during considered switching cycle $N$ is defined by the local field $E_{\text{loc}}(i,j,N)$:
$p_i(i,j,N) \sim \exp[-E_{ac_1i}/(E_{loc}(i,j,N)-E_{th_1i})],$  
where $E_{ac_1i}$ and $E_{th_1i}$ are the activation and threshold fields for given nucleation process $k$.

It is known that the probability of nucleation at the wall exceeds essentially the bulk one as $E_{ac_1i} < E_{ac_2i} < E_{ac_3i}$. Thus the increase of the domain boundary length facilitates the switching.

The local field is a sum of uniform external field $E_{ex}$, residual depolarization field $E_{rd}$, and local value of internal bias field $E_{bi}(i,j,N)$ formed by the end of previous cycle:

$$E_{loc}(i,j,N) = E_{ex} + E_{rd} + E_{bi}(i,j,N).$$

For slow bulk screening, when $\tau \approx T$, which is typical for fatigue experiments, $E_{bi}(i,j,N)$ relaxes compensating the sum of external and residual depolarization fields averaged over the switching cycle ($E_{ex} + E_{rd}$). For symmetric rectangular pulses $\langle E_{rd} \rangle = 0$. The local value of $\langle E_{rd} \rangle = E_{rd}(T^+ - T^-)/T$, where $T^+$ and $T^-$-time of given point residence in the states with opposite sign of spontaneous polarization during the switching cycle [Fig. 1(b)]. It is clear that the values of $T^+$ and $T^-$ being local nevertheless can be obtained only by simulation of the domain kinetics in the whole matrix. We understand that in real ferroelectrics the bulk screening is a complicated process due to competition of several above-mentioned mechanisms. Within our approach it is difficult to favor any of them, because in our model $\tau$ is the only parameter characterizing the screening kinetics.

After each simulation of domain kinetics during the cycle we recalculate the spatial distribution of the internal bias field:

$$E_{bi}(i,j,N) = E_{bi}(i,j,N-1)(1 - T/\tau) - \langle E_{rd}(i,j,N-1) \rangle T/\tau.$$  

We have considered two variants of the initial spatial distribution of the internal bias field $E_{bi}(i,j,1)$: (1) uniform zero internal bias and (2) completely screened multidomain initial state. The first corresponds to idealized initial state without bulk screening. In the second one $E_{bi}$ takes the equal positive value at domains of one sign and the negative one elsewhere.

III. RESULTS AND DISCUSSION

A. Initial state with zero internal bias field

The set of patterns corresponding to one switching cycle at the different fatigue stages simulated for the initial state with zero internal bias $\langle E_{bi}(i,j,1) = 0 \rangle$ are presented in Fig. 2. It is seen that initially the switching occurs in the whole area [Fig. 2(a)]. After fatigue cycling the switching occurs mainly in narrow regions surrounding the frozen domains.

![Fig. 1.](image1.png)
![Fig. 2.](image2.png)
![Fig. 3.](image3.png)
![Fig. 4.](image4.png)
Such evolution of the switching area geometry demonstrates the qualitative change of domain kinetics. In the initial state the process is governed by 2D growth of isolated domains [Fig. 2(a)]. Increasing of the input of 1D wall motion follows arising and growth of the frozen regions. For strong fatigue, the domain kinetics is defined mostly by reversible parallel translation of the walls [Fig. 2(b)]. It is important to point out that such geometry change has been observed directly in fatigued PZT films.\textsuperscript{11}

We characterize the nonuniform $E_b$ at a given cycle by internal bias field distribution function

$$f(E_b,N) = L^{-2} \sum \delta(E_b - E_b(i,j,N)).$$

The simulated $f(E_b,N)$ is well fitted by the Gaussian (Fig. 3) and fatigue induced evolution shows the smearing of initial narrow distribution with dispersion $w$ increasing during cycling. The formation of two peaks at $E_b = \pm E_{rd}$ corresponds to the arising of frozen domains (Fig. 3).

According to the Preisach theory this field distribution function defines the voltage dependence of switching charge and current\textsuperscript{31,32} for testing in triangular pulses. The broadening of $f(E_b,N)$ during cycling leads to decrease of the switching charge due to termination of polarization reversal in the regions where $E_{loc}$ is lower than threshold field $E_{th}$. Our modification of this approach\textsuperscript{33} accounting for the difference between the threshold fields for nucleation and growth allows us to simulate the dependence of the switching current maximum $j_{max}$ on $w$, which is well-fitted by the following formula (Fig. 4):

$$j_{max}(1/w) = j_{max}(0) + J[\exp(a/w) - 1],$$

where $J$ and $a$ are constants.

It is important that the fatigue diminishing of current maximum is more pronounced than that for switching charge (Fig. 5).

B. Completely screened multidomain initial state

This demonstrates the existence of additional rejuvenation stage (increasing of the switching charge) preceding the fatigue process [Fig. 6(a)]. The rejuvenation after the fatigue stage has been experimentally discovered in bulk ferroelectrics and thin films.\textsuperscript{3,34} We found that rejuvenation and fatigue stages differ by geometry of switching area (Fig. 7). The formation of complicated maze structure during rejuvenation is accompanied by the increase of switching area width. During fatigue stage the self-consistent flattering and simplification of maze pattern is observed. Simulation shows that contrary to fatigue from the initial state with zero internal bias, the unipolar growth of the frozen domains of only one sign is usually obtained. The sign of unipolarity is defined by geometry of the initial domain structure. Such tendency is in accord with published experimental results.\textsuperscript{15}

Evolution of internal bias field distribution function differs qualitatively from the one for zero internal bias initial state. Two peaks corresponding to initial domain areas with opposite sign of polarization smear and merge during rejuvenation thus forming one wide peak [Fig. 8(a)]. Its subsequent behavior follows the above discussed evolution of $f(E_b,N)$ [Fig. 8(b)]. At the rejuvenation stage the switching current is a sum of two inputs corresponding to the switching in the regions with different sign of internal bias field [Fig. 9(a)].

C. Experiment

The fatigue/rejuvenation cycling was carried out in Pb(Zr\textsubscript{0.2}Ti\textsubscript{0.8})O\textsubscript{3} films (100–200 nm thick) grown by chemical solution deposition on Pt/Ti/SiO\textsubscript{2}/Si substrates.\textsuperscript{13} Pt top electrodes have been used for applying a bipolar rectangular
pulse trains (100% duty cycle, the amplitude \( U_{\text{cyc}} = 5-8 \) V, the cycling frequency \( f_{\text{cyc}} = 10 \text{ Hz} - 100 \text{ kHz} \)). The hysteresis loops and switching currents were measured in triangular waveform (\( f_{\text{cyc}} = 10-100 \text{ Hz}, \ U_{\text{cyc}} = 5-7 \) V) after a certain number of switching cycles. Cycling and measurements have been done at room temperature.

The fatigue-induced evolution of the switching charge [Fig. 6(b)] and current [Fig. 9(b)] pulses for PZT films confirms the existence of the predicted rejuvenation stage, which is most pronounced for \( f_{\text{cyc}} \) (Fig. 10). The evolution of the shape of switching current pulse during cycling is in accord with simulated behavior [Figs. 6(a) and 9(a)].

IV. CONCLUSIONS

The proposed model of self-organized evolution of local internal bias field (kinetic imprint effect) allows us to describe the change of the geometry of switching area during cycling and predicts the existence of rejuvenation stage. The simulation shows that the evolution of the internal bias field distribution function and switching current are correlated. Thus the switching current data provide the essential information on fatigue kinetics. The comparison with experimental data for PZT thin films confirms the validity of our model. Within our model the nonswitched regions, formed during self-organized domain evolution, qualitatively differs from the seeds discussed by Tagantsev et al.\textsuperscript{7–9} The regions are kinetically frozen because the ac field amplitude and period are insufficient for complete switching. As a result the considered fatigue effect is reversible and partial rejuvenation occurs after increasing the field amplitude in accordance with experiment\textsuperscript{\textsuperscript{34}} and in contrast with Ref. 14. The recently observed fatigue anisotropy\textsuperscript{35} can be attributed to dependence of endurance on the geometry of initial domain structure. The discussion of the dependence of fatigue behavior on the cycling frequency and geometry of the initial domain structure will be presented in a subsequent article.

ACKNOWLEDGMENTS

The authors are grateful to T. Schneller and R. Gerhardt, who fabricated the investigated PZT films and to O. Lohse for technical assistance. The research was made possible in part by a Grant of the Ministry of Education of the Russian Federation, by the Program “Basic Research in Russian Universities” and by Award No. REC-005 of the CRDF.