1. INTRODUCTION

A decrease in the switching charge as a result of long-time cyclic switching of polarization in thin ferroelectric films, which is known as the fatigue effect, remains a key problem, whose solution is essential for wide practical application of nonvolatile storage devices based on such films [1–16]. Recently, several alternative mechanisms have been proposed for explaining the fatigue effect in films. In accordance with the bulk-locking mechanism, charged domain walls in a film subjected to cyclic switching are fixed (pinning effect) by charge carriers localized at traps [9, 10]. It should be noted that this mechanism was proposed for the first time by Kudzin et al. [16] for explaining fatigue effects in barium titanate single crystals. According to another mechanism (interface scenario), fatigue is associated with the slowing down of domain growth, which is due to suppression of nucleation (seed termination) at the film–electrode interface [5, 6]. The role of oxygen vacancy redistribution in an electric field has also been discussed extensively [14]. In [12], it is proposed that fatigue can be explained by the growth of a nonferroelectric passive surface layer upon cyclic switching. It should be noted that, in all contemporary models, the relation between the fatigue effect and the domain-structure kinetics upon cyclic switching is disregarded while considering fatigue in thin films. At the same time, recent direct observations of the domain structure of thin films with the help of scanning-probe microscopy proved that the fatigue effect is accompanied by the formation of a complex-shaped region being switched and by the appearance and growth of regions with a frozen orientation of polarization (“frozen domains”) [8, 17].

In this study, we propose an approach to describing the evolution of switched regions during cycling switching, which is based on the decisive role of retardation in the bulk screening of the depolarization field. For cyclic switching, this retardation leads to the self-organized formation of a spatially inhomogeneous internal bias field due to retardation of bulk screening of the depolarization field. Variations in the switching charge and in the amplitude of switching current, which are calculated with the help of computer simulation of domain kinetics upon cyclic switching, are in good agreement with experimental data obtained for thin lead zirconate–titanate (PZT) thin films. © 2002 MAIK “Nauka/Interperiodica”.

2. MODEL

It is well known that, after switching of polarization in a ferroelectric capacitor, external screening (charge redistribution on the electrodes accompanied by switching current) rapidly compensates the depolarization field $E_{\text{dep}}$. However, there is no complete field compensation in the bulk, since a nonpolar surface layer (dielectric gap) exists in ferroelectrics [18, 19]. After termination of external screening, the field $E_{\text{dep}}$ is compensated only partly and a residual depolarization field $E_{\text{rd}} = E_{\text{dep}} - E_{\text{ex.scr}}$ is preserved in the bulk [19, 20]. The compensation of $E_{\text{rd}}$ due to slow processes of bulk screening leads to the formation of an internal bias field $E_{\nu}$. This field is responsible for the unipolarity of the switching process, which is manifested in a shift of a hysteresis loop upon a quite rapid (as compared to the
bulk screening) cyclic switching in an alternating field [20, 21]. It has been shown, using lead germanite single crystals as an example, that the field \( E_b \) is spatially nonuniform [22] and varies upon a long-time cyclic switching [19–23]. Bulk screening can be regarded as a result of competition between three groups of mechanisms: (1) orientation of defect dipoles [24–26], (2) redistribution of bulk charge carriers [18, 19], and (3) injection of charge carriers from electrodes through a dielectric gap.

We take into account the fact that the domain kinetics during cyclic switching are a self-organized process, since the spatial distribution of \( E_b \) is determined by the preceding evolution of domains and determines, in turn, their subsequent kinetics. Earlier, we demonstrated experimentally that the field \( E_b \) is virtually uniform over the entire switched region in the case of long-time cyclic switching with asymmetric (\( T_{\text{pos}} \neq T_{\text{neg}} \)) rectangular pulses if the switching time is much shorter than the period of the switching field (\( t_s \ll T \)). In this case, the mean value of \( E_b \) is determined by the asymmetry of pulses \( c \sim \frac{T_{\text{pos}} - T_{\text{neg}}}{T_{\text{pos}} + T_{\text{neg}}} \) [28]. It follows that in the case of slow switching (\( t_s \leq T \)), the internal bias field \( E_b \) becomes spatially nonuniform even when symmetric pulses are used, since different regions in the sample are in states with opposite directions of polarization for different periods of time. During long-time switching, the local value of \( E_b \) relaxes to a value determined by the relative difference in the residence times of a given part of the sample in the states with opposite directions of polarization, \( (T^+ - T^-)/T \) (Fig. 1b). In this case, the magnitude and sign of the internal bias field \( E_b(x, y) \) are spatially inhomogeneous and the distribution function \( f(E_b) \) varies upon cyclic switching. The computer simulation of the domain kinetics carried out by us revealed that the variation of \( f(E_b) \) upon cyclic switching leads to the appearance and growth of unswitched regions (kinetically frozen domains) [29, 30].

3. COMPUTER SIMULATION

The domain kinetics for cyclic polarization switching in a thin plate (film) of a uniaxial ferroelectric under the action of rectangular field pulses were simulated on a two-dimensional (2D) matrix. We used the classical model of polarization switching, in which the domain structure kinetics involve the formation of new domains and their subsequent growth (nucleation and growth model) [31, 32]. We assumed that both processes are controlled by nucleation. The formation of nuclei of three different types was considered (Fig. 1a).

(i) Formation of nuclei at the ends of the existing steps leads to step growth.

(ii) Nucleation at a domain wall leads to the formation of new steps.

(iii) The appearance of isolated nuclei leads to the formation of new domains.

The probability \( p_k \) of formation of nuclei of a specific type in a given element of the matrix (i, j) during the Nth switching cycle is determined by the local field \( E_{\text{loc}}(i, j, N) \):

\[
p_k(i, j, N) = \exp\{-E_{\text{ac}, k}/E_{\text{loc}}(i, j, N) - E_{\text{th}, k}\},
\]

where \( E_{\text{ac}, k} \) and \( E_{\text{th}, k} \) are the activation field and the threshold field, respectively [19].

It is well known that the nucleation probability at a domain wall is much higher than that at a distance from the wall, since \( E_{\text{ac}, 1} < E_{\text{ac}, 2} < E_{\text{ac}, 3} \) [19]. Consequently, an increase in the length of a domain wall facilitates polarization switching.

The local field \( E_{\text{loc}}(i, j, N) \) is the sum of the uniform external field \( E_{\text{ex}} \), the residual depolarization field \( E_{\text{rd}} \), and the spatial nonuniform internal bias field \( E_b(i, j, N) \) formed by the end of the preceding switching cycle:

\[
E_{\text{loc}}(i, j, N) = E_{\text{ex}} + E_{\text{rd}} + E_b(i, j, N).
\]

In experiments on fatigue, bulk screening proceeds, as a rule, quite slowly, and \( \tau \gg T \). In this case, \( E_b(i, j, N) \) tends to compensate the sum of the external and the residual depolarization field averaged over a switching cycle, \( \langle E_{\text{ex}} + E_{\text{rd}} \rangle \). For symmetric pulses of rectangular shape, we have \( \langle E_{\text{ex}} \rangle = 0 \) at any point, while \( \langle E_{\text{rd}} \rangle \) is spatially nonuniform: \( \langle E_{\text{rd}} \rangle = E_{\text{rd}}(T^+ - T^-)/T \), where \( T^+ \) and \( T^- \) are the times for which the spontaneous polarization at a given point is directed along one of the two opposite directions, respectively, during a switching cycle (Fig. 1b). It should be noted that the local values of \( T^+ \) and \( T^- \) for each cycle depend on the domain kinetics in the entire sample (since the probability of switching at a given point is determined by the state of its surroundings) and, hence, can be determined only as a result of

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**Fig. 1.** (a) Three types of nuclei considered in simulation: (1) a nucleus at the end of a step, (2) a nucleus on a domain wall, and (3) an isolated nucleus. (b) Diagram showing variations in the relative local value of spontaneous polarization and in the relative value of the external field during a simulated switching cycle.
simulation of the domain kinetics in the entire matrix. In our analysis, we did not aim at choosing a preferred screening mechanism from those described above and characterized the kinetics of screening only by a time constant \( \tau \).

The domain structure kinetics in each switching cycle were simulated by taking into account the field \( E_b(i, j, N) \) formed by the end of the preceding cycle. The calculated local values of \( T^+ \) and \( T^- \) were used for recalculating the spatial distribution of \( E_b \) for the next switching cycle in accordance with the relation

\[
E_b(i, j, N) = E_b(i, j, N - 1) \exp(-T/\tau) + E_{rd}[1 - \exp(-T/\tau)]\Delta T(i, j, N)/T. \tag{3}
\]

Assuming the condition \( T \ll \tau \) to be satisfied, we used the simplified relation

\[
E_b(i, j, N) = E_b(i, j, N - 1)(1 - T/\tau) - \langle E_{rd}(i, j, N - 1) \rangle T/\tau. \tag{4}
\]

Obviously, the results of simulation must depend to a considerable extent on the spatial distribution of \( E_b \) prior to the first switching cycle. In the course of simulation, we considered two versions of the initial state: an idealized version with a uniform zero internal bias field (without bulk screening) \( E_b(i, j, 1) = 0 \) and a realistic version with a completely screened polydomain initial state \( E_b(i, j, 1) = -E_{rd}(i, j) \). The latter version corresponds to the spatial distribution observed after prolonged holding of the domain structure in a static state. In this case, the fields \( E_b \) in domains with opposite orientations are equal in magnitude but opposite in sign.

3.1. Initial State with Zero Internal Bias Field

The evolution of the domain structure (a set of consecutive domain configurations each of which corresponds to a single switching cycle) at various stages of fatigue process, which is obtained for the initial state with zero internal bias field, is presented in Fig. 2. It can be seen that complete switching occurs during the first cycle (Fig. 2a), while after a long-time cyclic switching, polarization reversal occurs predominantly in narrow regions separating kinetically frozen domains of different polarities (Fig. 2b). Such a change in the geometry of the switching region leads to a qualitative change in the type of domain kinetics. In the initial state, switching is mainly due to a 2D growth of individual domains (Fig. 2a), while the appearance of frozen domains leads to an increase in the contribution from 1D motion of domain walls. After a long-time cyclic switching, the domain kinetics are mainly determined by a reversible parallel displacement of domain walls (Fig. 2b). It is important to note that a similar change in the geometry of the switching region was also detected by directly observing domain kinetics in PZT films using scanning-probe microscopy [8].

![Fig. 2](image)

**Fig. 2.** Sequence of instantaneous domain configurations forming during a switching cycle, found with the help of computer simulation (a) during the first switching and (b) a long-time cyclic switching. Switched domains of opposite polarities are shown by black and white, while frozen domains of various polarity are shown by light-gray or dark-gray color. The initial state corresponds to zero internal bias field.

![Fig. 3](image)

**Fig. 3.** Computer simulation (initial state with zero internal bias field): (a) distribution functions of the internal bias field for various values of \( N \) approximated by a Gaussian; and (b) switching current caused by a triangular pulse for \( N = 50: \Delta E/w = 0 \) corresponds to the Preisach theory, and \( \Delta E/w \neq 0 \), to a modified approach [25], where \( \Delta E \) is the difference between the threshold fields required for the formation of an isolated seed and for domain growth, respectively. The inset shows the dependence of \( j_{max} \) on \( 1/w \) fitted by formula (5).

The spatially nonuniform field \( E_b \) formed after the completion of the \( N \)th cycle was characterized by the instantaneous value of the distribution function \( f(E_b, N) \) of the internal bias field:

\[
f(E_b, N) = L^2 \sum \delta[E_b - E_b(i, j, N)]. \tag{5}
\]

The distribution functions \( f(E_b, N) \) obtained as a result of simulation are closely fitted by a Gaussian
The initially narrow distribution function spreads in the course of cyclic switching (variance $w$ increases significantly). The formation and growth of two peaks of the distribution function at $E_b = \pm E_{rd}$ correspond to the formation and increase in the area of kinetically frozen domains (Fig. 3a).

According to the Preisach theory, the distribution function for the internal bias field determines the dependence of switching charge and current on the applied voltage during testing with triangular pulses [33, 34]. The spread of $f(E_b, N)$ upon cyclic switching decreases the switching charge, since polarization switching is terminated in the regions in which the local field $E_{loc}$ becomes smaller than the threshold field $E_{th}$. In our modification [35] of the Preisach approach, we took into account the above-mentioned fact that the threshold fields required for the formation of a single nucleus and for domain growth (nucleation at a domain wall) differ significantly. It is shown that the dependence of the maximum value of switching current $j_{max}$ on the variance $w$ of the distribution function, which is found in our model with the help of computer simulation, can be fitted by the formula (Fig. 3) [35]

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

where $J$ and $a$ are constants.

It is important to note that the maximum value of the switching current, $j_{max}$, decreases in the fatigue process to a considerably greater extent than the switched charge does (Fig. 4).

3.2. Completely Screened Polydomain Initial State

A simulation of cyclic switching from a polydomain completely screened state revealed an initial increase in the switching charge and current on the applied voltage during testing with triangular pulses. The spread of $f(E_b, N)$ upon cyclic switching decreases the switching charge, since polarization switching is terminated in the regions in which the local field $E_{loc}$ becomes smaller than the threshold field $E_{th}$. In our modification [35] of the Preisach approach, we took into account the above-mentioned fact that the threshold fields required for the formation of a single nucleus and for domain growth (nucleation at a domain wall) differ significantly. It is shown that the dependence of the maximum value of switching current $j_{max}$ on the variance $w$ of the distribution function, which is found in our model with the help of computer simulation, can be fitted by the formula (Fig. 3) [35]

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

where $J$ and $a$ are constants.

It is important to note that the maximum value of the switching current, $j_{max}$, decreases in the fatigue process to a considerably greater extent than the switched charge does (Fig. 4).

3.2. Completely Screened Polydomain Initial State

A simulation of cyclic switching from a polydomain completely screened state revealed an initial increase in the switching charge. This feature enabled us to single out an additional rejuvenation stage preceding the fatigue stage (Fig. 5a). It can be seen that the geometries of the switching region in the rejuvenation and fatigue stages are qualitatively different (Fig. 6). In the course of rejuvenation, the width of the switching region increases considerably and its connectivity changes. After the completion of rejuvenation, this region has the form of a connected labyrinth structure. At the fatigue stage, a self-consistent smoothing and simplification of the labyrinth structure is observed. The stimulation revealed that frozen domains of the same polarity are predominantly formed and grow (unipolar fatigue). The sign of unipolarity is determined by the geometry of the initial domain structure. This ten-
dency is in good agreement with the reported experimental results [13].

The evolution of the internal-bias-field distribution function in this case differs qualitatively from that in the case of a zero initial internal bias field considered above. Two peaks corresponding to initial domains of opposite signs spread and merge into a single broad peak during rejuvenation (Fig. 7a). The subsequent behavior of this peak repeats the variation of \( f(E_{bi}, N) \) described earlier (Fig. 7b). At the rejuvenation stage, the switching current is the sum of two contributions corresponding to switching in the regions differing in the sign of the internal bias field (Fig. 8a).

4. EXPERIMENT

The rejuvenation and fatigue were studied in lead zirconate–titanate \( \text{Pb(Zr}_{0.2}\text{Ti}_{0.8})\text{O}_3 \) films 100–200 nm thick. The films were deposited by sol–gel method on Pt/Ti/SiO\(_2\)/Si substrates [11]. The upper platinum electrode was used for applying rectangular bipolar pulses with 100% filling (without a pause between the pulses). The amplitude of pulses \( U_{cyc} \) was 5–8 V, and the frequency \( f_{cyc} \) was chosen in the interval from 10 Hz to 1 MHz. Hysteresis loops and switching currents were measured by applying triangular pulses \( (f_m = 10–100 \text{ Hz}, U_m = 5–7 \text{ V}) \) following a certain number of switching cycles with rectangular pulses. Cyclic switching and measurements were carried out at room temperature using an automated measuring complex [29].

The measured variations in switching charge (Fig. 5b) and in current (Fig. 8b) upon cyclic switching in PZT films confirm the existence of the rejuvenation stage, which is manifested most clearly in the increase in \( j_{\text{max}} \) (Fig. 9). The experimental data on the evolution of the shape of the current during cyclic switching is in qualitative agreement with the predictions of computer simulation (Figs. 5a, 8a). It should be noted that, in order to reduce the computer time required for simulation, the value of the time constant \( \tau \) characterizing the kinetics of screening was chosen to be much smaller than its experimental value, which leads to the observed discrepancy between the theoretical and experimental values of \( N \), for which the magnitude of the switched charge decreases to half the initial value (endurance).

5. CONCLUSION

Thus, the proposed model of self-consistent evolution of the local internal bias field enabled us to describe the variations in the geometry of the switching region during cyclic switching and to predict the existence of the rejuvenation stage. With the help of computer simulation, we observed a correlation between the variations in the internal-bias-field distribution function and in the shape of switching current upon cyclic switching. The established correlation makes it possible to extract important information concerning the fatigue kinetics from measurements of the switching current. The good agreement between the results of simulation and the experimental data obtained for thin PZT films confirm the applicability of the proposed approach.

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