New Approach to Analysis of the Switching Current Data, Recorded During Conventional Hysteresis Measurements

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(Received May 19, 2003)

The explanation of fatigue as a spatially non-uniform kinetic imprint due to evolution of the local threshold field distribution function has been proposed by us recently. In this paper we propose a new switching current analysis which allows to extract the distribution function from the data of conventional hysteresis loop measurements. Domain kinetics in slow-increasing field in spatially inhomogeneous ferroelectric was investigated by computer simulation. Domain arising and growth were considered under assumption that the threshold field for appearance of isolated nuclei is essentially higher than for nuclei at the domain wall. The obtained approach essentially differs from Preisach one. The proposed method has been applied for analysis of the switching current data measured during fatigue cycling in PZT and PLZT thin films. The fitting formula for dependence of switching charge and current maximum on the cycle number has been proposed and successively used for experimental data. The proposed method can accelerate the fatigue testing.

Keywords: Ferroelectric thin films; PZT; PLZT; fatigue; switching current analysis

INTRODUCTION

Polarization reversal is used in ferroelectric memory devices for information writing and reading [1]. It has been shown in classical works [2, 3] that all 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, which are applied in ferroelectric memory devices [4–20]. Knowledge of the fatigue origin is necessary to improve the film endurance essential for ferroelectric non-volatile memories (FeRAM).
Several alternative scenarios for the suppression of the switching polarization have been proposed [11]. Bulk-locking mechanism is based on the assumption that the charged domain walls are pinning with the help of electric charge trapping centers [13]. The interface scenario supposes the inhibition of domain growth due to inhibition of nucleation at the electrode interfaces [8, 10]. The crucial role of electromigration of oxygen vacancies is discussed [17, 18]. The growth of the passive surface layer with damaged ferroelectric properties during cycling is proposed recently [15]. It must be stressed that the fatigue is considered without relation with domain evolution during cycling, though it was shown by direct observation by scanning probe microscope that fatigue in thin films manifests itself through growth of “frozen” domains [12, 21]. Therefore the knowledge of the domain kinetics in electric field is very important.

Domain visualization during switching in thin films is very difficult so the charge and current recording during polarization reversal (“switching”) is the most popular experimental technique. Two types of field pulses have been used usually for such investigations: (a) rectangular pulses (Merz technique [22]) and (b) triangular ones (observation of hysteresis and current loops) [23]. In turn two approaches have been used for data analysis: (a) the kinetic approach [24–27] and (b) the quasi-static one [28–31]. Application of both approaches is restricted. The first cannot be applied for real spatially nonuniform films, while the second completely neglects the domain growth.

In this paper we propose the new approach for analysis of the current data in spatially nonuniform ferroelectrics obtained during quasi-static switching. It allows to extract the evolution of the bias field distribution function during fatigue in thin films. Moreover the fitting formula for dependence of switching charge and current maximum on the cycle number based on kinetic approach to fatigue effect has been proposed and applied for PZT and PLZT films. The method can be used for acceleration of the fatigue testing in thin film memory devices.

**FATIGUE DUE TO SPATIALLY NON-UNIFORM IMPRINT**

In our fatigue model we account for the fact that the domain kinetics during cycling is a self-organized process, because the spatial distribution of internal bias field $E_b$ depends on the previous domain evolution and in turn defines subsequent domain kinetics [32]. $E_b$ for the given region tends to the averaged on switching cycle value of depolarization field. This effect leads to kinetic spatially non-uniform imprint while the sign and value of $E_b$ after cycling become spatially nonuniform.
Simulation of domain kinetics during ac cycling shows that $E_b$ distribution function varies with the number of cycles (Fig. 1(a)). The simulation procedure described earlier [32–35]. The switching charge decreases as a result of the distribution function widening when the bias field in wings exceeds the amplitude of applied field (Fig. 1(b)). This effect manifests itself as a growth of regions with frozen domains.

Slow compensation of depolarization field by bulk screening [36, 37] leads to formation of $E_b$. Three bulk screening mechanisms are revealed and discussed: (1) reorientation of the defect dipoles [38, 39], (2) redistribution of bulk charges [36, 40], and (3) injection from the electrodes [8, 10]. Formation of $E_b$ in static state represents a well-known imprint effect [41, 42].

Simulated dependences of the maximum polarization and switching current on the switching cycle number are displayed in Fig. 2. It is seen that the current maximum is essentially more sensitive to change of the distribution function during fatigue. The parameters of the model have been chosen in such a manner which allows to minimize the number of switching cycles.

![Figure 1](image1.png)

**FIGURE 1** (a) Simulated internal bias field distribution functions for different switching cycle number. (b) Schematic representation of the decreasing of switchable area due to the widening of distribution function.

![Figure 2](image2.png)

**FIGURE 2** Simulated dependences of (a) maximum polarization and (b) maximum value of switching current on the number of switching cycles.
during simulation. It is clear that this approach can be verified only after extracting the information about evolution of the distribution function by proper analysis of the switching current data.

**KINETIC APPROACH TO CURRENT ANALYSIS**

The most popular experimental method for switching measurements is the recording of the switching current under the action of rectangular electric field pulses proposed by Merz [22]. The kinetic approach is applied for analysis of the data as it is supposed that the switching current is determined by domain nucleation and growth [23]. This approach is based on Kolmogorov-Avrami (K-A) formula written for crystallization of metals [24, 25]. This formula has been applied for switching current in ferroelectrics in [26]. It is known that K-A formula can be used only for complete switching by rectangular pulse from single-domain state in uniform infinite ferroelectrics. Modification of K-A formula for finite size media has been proposed in [27, 43].

It is necessary to point out that Merz method requires the constant value of the switching field during whole switching. The field rise time $t_r$ must be negligible as compare with the switching one $t_s$. This condition is never realized during fast switching in thin ferroelectric films as far as $t_r$ is about $t_s$. Moreover the thin films are essentially spatially nonuniform. Therefore the kinetic approach is inapplicable for quantitative analysis of the current data measured in thin films by Merz method.

**QUASI-STATIC APPROACH TO CURRENT ANALYSIS**

Quasi-static approach was suggested and used for analysis of the current data during slow switching when the voltage rise time is much longer than the local switching time. For thin films this relation is typical for conventional hysteresis measurements with frequency range from 1 Hz to 10 kHz. The quasi-static approach has been proposed by Preisach for explanation of the hysteresis loop shape in ferromagnets [28] and applied for ferroelectrics by Turik [29]. Recent years applications of Preisach approach in ferroelectric thin films were made [30, 31]. It must be pointed out that the current loops are essentially more sensitive to switching conditions and therefore more useful for quantitative analysis than polarization loops [32, 44].

According to Preisach approach each small part of nonuniform ferroelectric is characterized by the local value of threshold field and is switched when
the switching field has reached this value. Thus the switching current shape correlates with threshold field distribution function. While the Preisach approach neglect the domain growth it represents a **nucleation only model**. At the same time it is known that the input of domain growth in switching current is significant.

For quasi-static switching conditions the whole area of uniform ferroelectric should be switched at the same value of applied field. However such case is never obtained in real thin films due to spatially nonuniform switching characteristics.

There are several mechanisms of thin film nonuniformity: (1) random orientation of crystallographic axes in grains leading to nonuniform distribution of the polar field components in polycrystalline films, (2) variation of the thickness and surface shape peculiarities in thin films leading to spatially nonuniform field, (3) structural defects leading to nonuniform threshold field, (4) spatially nonuniform internal bias field formed by bulk screening during cyclic switching [32–35].

For analysis of the switching current in nonuniform ferroelectrics we have proposed **nucleation and growth model**—taking into consideration both domain nucleation (arising) and growth. The domain kinetics in ferroelectric with spatially inhomogeneous threshold field under the action of slow increasing field has been studied by computer simulation. The polarization reversal has been considered as a switching in step-by-step increasing external field (Fig. 3(a), in the inset).

\[ E_{ex}(n) = n \delta E \]  

where \( n \) is the step number.

**FIGURE 3**  (a) Simulated time dependence of the switching current for different \( \Delta E_{th}/w \), in the inset—scheme of step-by-step increase of applied field. (b) Simulated current maximum versus \( \Delta E_{th}/w \) value fitted by Eq. (5).
It was assumed that switching at each field-step occurs through formation of isolated nuclei (arising of new domain) and subsequent nucleation at the domain walls (domain growth) (Fig. 4). Similarly to the Preisach model we suppose that duration of each step is long enough for switching in all regions, where

\[ E_{ex}(n) > E_{th} - E_b \]  

(2)

The threshold value for the nucleation at the wall \( E_{th, gr} \) (domain growth) is essentially lower than far from the wall (formation of isolated domain) \( E_{th,n} \),

\[ E_{th,n} = E_{th, gr} + \Delta E \]  

(3)

where \( \Delta E \geq 0 \).

The total area of the regions \( \Delta A(n) \) switched at each step (duration \( \delta t \)) determines the switching current at field \( E_{ex}(n) \)

\[ j(n) = 2P_r \Delta A(n)/\delta t \]  

(4)

The Gaussian distribution function of threshold field with dispersion \( w \) has been chosen. The simulated currents for different \( \Delta E/w \) are shown at Fig. 3(a). The field dependence of current is proportional to the distribution function of threshold and internal bias field for \( \Delta E = 0 \) only corresponding to Preisach model. Otherwise the relation between current shape and distribution function is more complicated. The simulation results for nucleation and growth differ significantly from the Preisach approach and correlate with experiment (Fig. 3). The input of the domain growth in the current leads to

![FIGURE 4 Nucleation and growth during single field step. Domain configurations: (a) initial, (b) after nucleation, (c) after growth. Areas switched: black—previously, hatched gray—by nucleation, gray—by growth.](image)
following dependence of the current maximum on $\Delta E/w$ (Fig. 3(b))

$$j_{\text{max}}(\Delta E/w) = j_{\text{max}}(0) + A \left[ \exp \left( \frac{a}{\Delta E/w} \right) - 1 \right]$$  \hspace{1cm} (5)

where $A$ and $a$ are constants.

The current pulse can be divided in stages. At the first it is determined by domain growth, while at the second it is proportional to the threshold field distribution function shifted on $\Delta E$. Transition to the second stage is obtained when the whole nonswitched area is bordered with the switched one. The switching process at the second stage is determined by nucleation only with modified condition: $E_{\text{th,n2}} = E_{\text{th,n1}} - \Delta E = E_{\text{th,gr1}}$. Such behavior allows to reveal the threshold field distribution function by fitting the second stage by Gaussian with integral equal to the switched charge (Fig. 5).

**EXPERIMENT**

The proposed approach has been applied for analysis of the experimental current data recorded in sol-gel PLZT 5/40/60 and PZT 20/80 thin films (100–200-nm-thick) grown on Pt/Ti/SiO$_2$/Si substrate with Pt top electrodes. The fatigue cycling was carried out by applying a bipolar rectangular pulse trains (100% duty cycle, the amplitude $V_{\text{cyc}} = 5–8$ V, the cycling frequency $f_{\text{cyc}}$ ranged from 10 Hz to 100 kHz). The hysteresis loops and switching currents were measured in triangular waveform ($f_m = 10–100$ Hz, $V_m = 5–7$ V) after a certain number of switching cycles. Both cycling and measurements have been done at room temperature. The experimental procedure has been described in details elsewhere [32, 34].
It was shown that the shape of the currents measured during switching is very similar to simulated one (Fig. 5). The fitting of the experimental data by proposed method allows to extract the main parameters of the bias field distribution function.

It was shown recently that this model is applicable for describing the fatigue effect [32–35]. The evolution of the parameters of the distribution function extracted by fitting of experimental data measured during fatigue for PLZT and PZT thin films is shown at Figs. 6 and 7. It is seen that increase of the switching charge, which corresponds to the “wake-up” stage, is obtained at the beginning of cycling switching (up to $10^5$ cycles for PLZT and $10^3$ cycles for PZT 20/80) and correlates with slow decrease of the dispersion. While decreasing of the switching charge at the fatigue stage is followed by essential increasing of the Gaussian dispersion (Fig. 6(a)). Increase of the dispersion leads to incomplete switching due to arising of the frozen domains in regions with bias field corresponding to wings of the distribution function (Fig. 4(b)). It has been observed that dispersion of the internal bias field

FIGURE 6 Evolution of the dispersion of the bias field distribution function extracted by treatment of the experimental data fitted by Eq. (6) and residual polarization and current maximum in PLZT 5/40/60 thin films.

FIGURE 7 (a) Current maximum fitted by Eqs. (5) and (7), (b) maximum and remnant polarization fitted by Eqs. (6) and (7) in PZT 20/80 films.
has the following dependence on the number of cycles during fatigue stage (Fig. 4a):

\[ w = B(N - N_0)\gamma + w_0 \]  

(6)

where \(N_0\)-switching cycle number corresponding to the beginning of fatigue stage, \(w_0\)-dispersion of bias field distribution function after \(N_0\) cycles, \(B\) and \(\gamma\)-constants.

The value of the power \(\gamma\) is equal to 0.5. The obtained dependence can be explained due to the similarity between the distribution function widening and the process of “Brownian motion” of bias field value. Such tendency one-to-one correlates with predictions of our model of fatigue effect [32].

The obtained Gaussian distribution function of bias field and its evolution during fatigue cycling allows us to propose the fitting formula for the fatigue dependence of the switching charge and switching current maximum. The switching charge for quasi-static switching can be calculated as an integral of the internal bias field distribution function over the range of applied triangular electric field:

\[ P(w) = \frac{1}{\sqrt{2\pi}w} \int_{-E_{m}}^{E_{m}} e^{-\frac{(x-E_{b0})^2}{2w^2}} dx \]  

(7)

where \(E_{b0}\)-mean value of the internal bias field (macroscopic bias of the hysteresis loop), \(E_{m}\)-amplitude of the applied linear increasing field.

Substitution of the dispersion \(w\) dependence on the number of cycles during fatigue stage (7) in Eq. (7) gives the fitting formula for fatigue dependence of the switching charge. Evolution of the switching current maximum during fatigue has been obtained by substitution of (7) in Eq. (5). The result of application of the proposed fitting formulas for the experimental data in PLZT thin films are presented in Fig. 5(b),(c) and for PZT 20/80–in Fig. 7.

On the other hand, excluding the dispersion from the Eqs. (5) and (6) we can obtain the relationship between switching charge and maximum value of the switching current. The experimental dependence of \(P_{\text{max}}\) on \(J_{\text{max}}\) fitted by proposed formula is presented in Fig. 8.

The proposed method of analysis of the switching current and switching charge data during fatigue can be used for acceleration of the fatigue testing procedure. It is possible by analysis of the current data due to higher sensitivity of its maximum value to cyclic switching as compare to switching charge. The proposed fitting formulas allow to estimate the endurance by extrapolation.
CONCLUSION

We have shown how the information about the distribution function of the threshold fields and the spatial correlation function can be extracted from the switching current data measured in triangular pulses at low frequencies. The proposed approach has been applied for analysis of the experimental currents in PZT and PLZT thin films measured during fatigue cycling. Our investigations revealed that the proposed method opens the possibilities for quantitative characterization of the ferroelectric films produced by different technologies and can be used for detail investigation of the fatigue effect kinetics.

ACKNOWLEDGMENTS

The authors are grateful to T. Schneller for providing thin films. The research was made possible in part by RFBR (Grant 01-02-17443), by RFBR-DFG (Grant 02-02-04006), by Education Ministry RF (Grant E02-3.4-395), by program “Basic Research in Russian Universities” (Grant ¥P.06.01.031), and by Award No.REC-005 of CRDF.

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