How to extract information about domain kinetics in thin ferroelectric films from switching transient current data

V. Ya. Shur \textsuperscript{a}, E. L. Rumyantsev \textsuperscript{a}, S. D. Makarov \textsuperscript{a}, V. V. Volegov \textsuperscript{a}

\textsuperscript{a} Institute of Physics and Applied Mathematics Ural State University. Lenin ave.51, Ekaterinburg, 620083, Russia

To cite this Article: V. Ya. Shur, E. L. Rumyantsev, S. D. Makarov and V. V. Volegov, 'How to extract information about domain kinetics in thin ferroelectric films from switching transient current data', Integrated Ferroelectrics, 5:4, 293 - 301

To link to this article: DOI: 10.1080/10584589408223886

URL: http://dx.doi.org/10.1080/10584589408223886
HOW TO EXTRACT INFORMATION ABOUT DOMAIN KINETICS IN THIN FERROELECTRIC FILMS FROM SWITCHING TRANSIENT CURRENT DATA

V. Ya. SHUR, E. L. RUMYANTSEV, S. D. MAKAROV and V. V. VOLEGOV

Institute of Physics and Applied Mathematics, Ural State University, Lenin ave.51 620083 Ekaterinburg, Russia

(Received May 16, 1994)

The paper presents the new method of mathematical treatment which allows to extract essential information about domain kinetics in polycrystal films from the traditional measurements of switching current. This information can not be obtained within the commonly used treatment. The method was verified by computer simulation and direct experiments in model single crystals.

Keywords: domain structure, ferroelectric thin films, switching current, phase transformations, computer simulation

INTRODUCTION

It is well-known that the measurement of transient current after application of opposite polarity square voltage pulses is the most popular and practically the only one method of experimental investigation of fast switching in thin ferroelectric films.\(^1\) Up to now the extracted information is limited because the traditional mathematical treatment of experimental data commonly yield only field and temperature dependencies of switching time,\(^2\) whereas it is clear, that the switching current data contains undoubtedly the information about domain structure kinetics: arising, growth and coalescence of domains.\(^3,4\) It is reasonable to propose that with adequate theoretical description the kinetic parameters of the main stages of domain evolution can be extract even from these integral measurements.

PROBLEMS OF THE CLASSICAL APPROACH

The theoretical description of the switching process\(^5,6\) is usually based on Kolmogorov-Avrami (K-A) statistical theory\(^7,8\) developed initially for the description of metal crystallization in infinite media (e.g. in the approximation that the average distance between the centers of crystallization is much less than the crystallizing media sizes). The most consistent application of K-A theory for the description of switching kinetics and transient current shape in ferroelectrics was given by Ishibashi and Takagi.\(^9\)

Recently there were made attempts to modify K-A approach for the description of the behavior of real finite objects.\(^10,11\) It was pointed out that the effects due to
the finite sizes are very important in polycrystal films and ceramics. In such objects the reconstruction of domain structure goes simultaneously and independently in individual grains of extremely small sizes. The averaging over the great number of independent experimental realizations favours the application of statistical description in this case.

Two limiting situations are considered usually by this statistical model. Following classification of Kolmogorov they are: firstly, $\alpha$-model—in which the number of domains is progressively increasing (the nucleation probability per volume $\alpha$ usually remains constant during switching) and, secondly, $\beta$-model—when all domains involved in switching are arising instantaneously at the very beginning of the process with the density $\beta$ per volume. In both cases domain wall velocity $v$ is taken to be constant. Within these approximations the main formula describing the variation of the fraction of the area occupied by unswitched domains runs as follows:

$$q(t) = \exp\left(-c\alpha v^n t^{n+1}/(n + 1)\right)$$ for $\alpha$-model \hfill (1)

$$q(t) = \exp(-c\beta v^n t^n)$$ for $\beta$-model \hfill (2)

where $n$—the dimensionality of the kinetic process, depending upon the type of domain structure ($n = 1$ or 1-D process for lamellar domains, $n = 2$ or 2-D process for isolated cylindrical domains and $n = 3$ or 3-D process for bulk ones), $c$—the shape constant.\(^7\)

While describing the experimental transient current pulses in ferroelectric films by these expressions the fractal values of the kinetic process dimensionalities $n$ are usually obtained.\(^{12,13}\) Moreover it appears impossible to describe the decaying part of the current pulse by these formulas. It is apparent because the proposed theory did not account for the fact that at the instant the growing domain touches the sample boundaries there occurs the geometrical catastrophe. The domain ceases to grow in one of the directions—in other words the abrupt change of the shape constant occurs. This process drastically changes the shape of transient current. Such effect is the most pronounced while observing the growth of wedge-like or lens-like domains (at the high anisotropy of domain wall velocity) which is rather typical for thin ferroelectric films.\(^{14,15}\)

SWITCHING IN FINITE MEDIA

Let us consider the polarization reversal in thin square plate (with area $S = l^2$) oriented perpendicular to polar axis. We can consider this kinetic process as two-dimensional (2-D) because the domain growth velocity in polar direction is always considerably greater than in others. Let us take into account that during any experiment there exists limited time interval between consecutive measurements $\Delta t$. Thus for any value of the anisotropy of the velocity of sideways domain growth $A = v_x/v_y$ we can consider the process as the isotropic movement of the domain walls occurring on the discrete matrix $L_x \times L_y$, where $L_x = l/(v_x \Delta t)$ and $L_y = l/(v_y \Delta t)$.

Let us consider at first the $\beta$-model with the appropriate number of the simultaneously arising domains $N_0 = \beta_0 l^2$, which are growing in the shape of rhombus with high anisotropy $A >> 1$ ($L_x << L_y$). One can choose the 1-D line with the
length $L_i$ (i-th row of the matrix) situated at the distance $\Delta_i = |L - i|$ from the middle of the matrix (Figure 1). The switching process in such a line satisfies all the requirements needed for the application of K-A theory. As it is seen on Figure 1 the number of the domains in the considered line is increasing when the domains from another regions reach it. So the switching process in the line occurs according to $\alpha$-model.

The limitations on the matrix sizes in $x$ direction lead to the abrupt changes of the intensity of domains arising in the line per time during switching. At the beginning $\alpha_i(t) = \alpha_0 = N_0/(2AL^2) = \beta/L$, where $2L = L_x$, but then it undergoes two catastrophic abrupt changes:

$$
\alpha_i(t) = \begin{cases} 
\alpha_0, & \text{for } 0 < t < L - \Delta_i \\
\alpha_0/2, & \text{for } L - \Delta_i < t < L + \Delta_i \\
0, & \text{for } L + \Delta_i < t 
\end{cases}
$$  \hspace{1cm} (3)

The initial value of $\alpha$ undergoes twofold decrease as a result of the first catastrophe and after the second—even the type of the model is changed. In other words the $\alpha(1-D) \rightarrow \beta(1-D)$ geometrical phase transition occurs.

In order to verify the validity of proposed description the computer simulation of the switching in 2-D matrix for $\beta$-model was carried out with the random distribution of growing rhombic domains. For every step the variation in the fraction of the area occupied by arising domains $\Delta S/\Delta t$ (the analog of transient current) was averaged over great number of independent realizations (computer experiments). It is seen on Figure 1 that the switching in definite line (1-D part of the sample) is approximated extremely good by Equation (1) with $n = 2$ and two catastrophic changes of parameters in accordance with Equation (3).

For the time dependence of the fraction of the area occupied by unswitched domains in the whole matrix we have:
It is seen that the $\beta(2-D)$ kinetic process is the sum of local $\alpha(1-D)$ processes. As a result we obtain the formula similar to Equation (2) in which the density of the domains per square $\beta(t)$ effectively diminishes during switching (in other words the gradually decreasing of shape constant obtain):

$$\beta(t) = 2 \sum_{i=1}^{L} \alpha_i / L = \beta_0 (1 - t/t_c)$$  (5)

where $t_c = 2L$.

It is seen that during switching in restricted area with high anisotropy of sideways motion of domain walls there are two important peculiarities: at first, the number of the domains involved in the $\beta(2-D)$ process effectively diminishes and, at second, the dimensionality of the kinetic process catastrophically changes at the appropriate moment $t_c$. It means that $\beta(2-D) \rightarrow \beta(1-D)$ transition occurs. The contribution of these peculiarities diminishes with the increasing of effective sizes and in the limit ($L \rightarrow \infty$) the well-known K-A formula for $\beta$-model is obtained.

It is possible to show that the expression analogous to Equation (5) can be used for the description of the transient current shape for any model situations: arbitrary value of anisotropy (account for the limitations in both directions) and $\alpha$-model as well.

For $\alpha$-model:

$$i(t) = (2PI^2n^{n+1)/t_{0\alpha}}[1 - (1 + 1/(n + 1))t/t_m\exp[-(t/t_{0\alpha})^{n+1}(1 - t/t_m)],$$  (6)

for $\beta$-model:

$$i(t) = (2P, l^2n^{-1/t_{0\beta}}[1 - (1 + 1/n)t/t_m\exp[-(t/t_{0\beta})^{n}(1 - t/t_m)],$$  (7)

where $t_{0\alpha} = [c\alpha v^{n+1}/(n + 1)]^{-1/(n+1)}$, $t_{0\beta} = (c\beta v)^{-1/n}$, $P_s$—spontaneous polarization.

In this case the decay time $t_m$ (the characteristic time of the process of the effective decreasing of domain number) is the function of anisotropy, shape constant and nucleation intensity. In the above considered case for $\beta$-model with high anisotropy $t_m$ is equal to $t_c$.

**COMPUTER EXPERIMENTS**

The computer simulations of the switching process was carried out on the 2-D matrix of 500 $\times$ 500 unit cells. The averaging of current pulses over great number of realizations (more than 1000) was held for obtaining statistically reliable results corresponding to the experimental conditions in polycrystal thin films. The computer experiments were done for $\alpha$- and $\beta$-models in the wide range of the model parameters. The various shapes of growing domains with different orientation relative to the (samples) boundaries were examined as well.

The results of computer experiments for $\alpha$- and $\beta$-models are approximated
extremely good by the Equations (6) and (7) with integer values of kinetic process dimensionalities. It is important that these formulas succeeded in describing the decaying part of the pulses which is impossible within the framework of the classical approach. The typical results for β-model with isotropic domain growth and α-model with high anisotropy are shown on the Figure 2. The existence of the geometrical catastrophes is clearly seen in both cases.

EXPERIMENTAL RESULTS

The investigations of the switching were done in thin plates of model single crystal lead germanate PGO (with optically distinguished domains) for the experimental testing of the method. Two different types of initial domain structure were prepared:
the single domain state and the controlled maze structure with high density of domain walls (Figure 3).\textsuperscript{16} The main stages of domains' evolution obtained as a result of mathematical treatment of transient current data are in qualitative agreement with results of direct observations of domain kinetics.

The mathematical treatment allows us to show the qualitative and quantitative difference in the domains kinetics in these two cases. Switching from single domain state demonstrate the $\beta(2-D) \rightarrow \beta(1-D)$ geometrical transition in the decaying part of the current pulse. It is necessary to mention that the first catastrophe on Figure 3a is due to the relatively large dimension of the sample in polar direction ($d = 130 \, \mu m$) and never can be realized in thin films. During switching from the maze domain structure we reveal other type of geometrical transition $\beta(1-D) \rightarrow \alpha(2-D)$. This fact enables us to state that the change from the maze to isolated cylindrical domains occurs. All predicted transformations of the types of domain structures are confirmed by direct real-time visualization of the domain kinetics.
The width of maze elements in our experiments was about few microns and it was difficult to observe the momentary domain patterns by optical methods. The proposed method was very effective even in this case for extracting the information about the ((geometry)) (type) of the domain structure and its changing during switching. It must be stressed that in contrast to any method of visualization of momentary domain patterns the proposed method has practically no resolution limits.

The method has been applied with much success for the studying of thin polycrystal films of PLT and PZT (Figure 4). The $\beta(2-D) \rightarrow \beta(1-D)$ and $\alpha(2-D) \rightarrow \alpha(1-D)$ transitions during switching are rather typical in thin films. This kind of transitions were discussed above in details and confirmed by computer simulation.
The considered transformations can be interpreted as the result of anisotropic growth of residual domains in finite media. The predicted domain evolution is in accordance with the results of the TEM observation of static domain patterns in thin films.\textsuperscript{14,15}

With proposed mathematical treatment we can obtain such important parameters of domain structure evolution as: kinetic process dimensionality $n$, anisotropy of domain growth $A$ and characteristics time constants $t_0$, $t_m$ and $t_c$. The knowledge of such parameters in turn makes it possible to describe the reconstruction of the domain structure in terms of the number of arising domains per volume ($\alpha$ and $\beta$) and their sideways velocities ($v_\alpha$ and $v_\beta$). The obtained parameters are depended upon the voltage and temperature. They are changing considerably during aging and degradation.

CONCLUSIONS

It is seen that proposed statistical description makes it possible to extract qualitatively new information from integral experimental characteristics. This information is of principal interest for the understanding of domain structure kinetics. The proposed method allows to investigate the actual processes such as degradation, aging and so on in real integrated devices in terms of physically clear parameters describing the domain structure evolution. This is very important for the application purposes.

ACKNOWLEDGEMENTS

This work was supported in part by Russian Foundation of Fundamental Research under Grant No. 93-02-2451. It is a pleasure to acknowledge the many helpful stimulating discussions with P. K. Larsen and J. F. Scott. We are very grateful to V. I. Sarapulov for technical assistance and to L. A. Suslov, V. V. Starostin and L. V. Gorodnic for providing some samples.

REFERENCES