Kinetics of ferroelectric domain structure during switching: Theory and experiment

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KINETICS OF FERROELECTRIC DOMAIN STRUCTURE DURING SWITCHING: THEORY AND EXPERIMENT

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Abstract Some trends in modern research of domain kinetics during switching are reviewed. The importance of the complex investigations of model crystals by local and integral experimental methods simultaneously with computer simulation of domains evolution is stressed. Some new experimental results concerning superfast switching are presented. It is shown that the momentary domain patterns are defined by kinetics of switching process. The general theoretical approach based on nucleation theory in analogy with any phase transformations is used for explanation of all observed effects. The influence of geometrical transformations occurring during domain evolution on transient current shape is confirmed by computer simulation.

INTRODUCTION

The switching under the action of electric field is an attribute of ferroelectrics. So the research of domain structure evolution is always in fashion. Recently the interest in exploration of fast switching process has grown rapidly in view of some promising applications of thin films mainly in memory devices. Moreover this phenomenon is of fundamental interest as an example of the phase transformation which can be in situ studied under precisely controlled external conditions.

It must be mentioned that there exists local and integral levels of polarization reversal research. The first is connected with the direct observations of the domain structure evolution during switching and the registration of the definite domains behaviour. Thus velocities of domain arising per area and its sideways/forward growth are measured. The nucleation is the only mechanism for describing these phenomena. The next level is the measurement of integral characteristics of switching process. The most informative and popular experimental method is the registration of transient current. So field and temperature dependencies of switching time and shape of current pulse were measured in different ferroelectrics. Really the polarization reversal is the result of arising and growing of great number of interacting domains accompanied by their coalescence. Thus statistical properties of domain evolution during switching must be taken into account for explanation of the behaviour of any integral transient characteristic.

The classic reviews dealing with these problems were published mainly more then twenty years ago. Since that time a lot of new experimental and theoretical
results were obtained. The aim of our paper is to point out some interesting experiments and consider them from the unified point of view.

THE DYNAMICS OF THE DOMAIN IN THE ELECTRIC FIELD: LOCAL CHARACTERISTICS

In the classic direct experiments Stadler and Zachmanidis, Miller and Savage measured the field dependences of sideways and forward domain walls motion velocity. In all investigated ferroelectrics the exponential and power laws were observed. These results were successfully explained through the nucleation theory by Merz, Miller and Weinreich, Fatuzzo, Wieder and Hayashi. Nevertheless the detail investigations of domain local behaviour carried out in real ferroelectrics by various authors differed sufficiently from sample to sample and did not present the unified picture of the phenomena.

For clarifying the situation one have to use a model object for the careful investigations. It must be a proper chosen ferroelectric single crystal with following characteristics: 1) simple domain structure (uniaxial ferroelectric); 2) optically distinguished domains; 3) high quality; 4) easily reversible in electric field domain structure (without damage of crystals); 5) experimentally convenient temperature region of ferroelectric phase existence. The choice of such crystals permits to use optical methods of real-time registration.

A few experimental methods: electron microscopy (SEM and TEM), nematic liquid crystals (NLQ) and pyroelectric probe scanning were used successfully for in situ research of domain kinetics but the optical ones if possible are the most informative and convenient and moreover the only one which allow to register superfast kinetics.

According to formulated requirements we have chosen model materials clearly demonstrating the limiting geometric types of domain shape existing in uniaxial crystals. The first is the collinear ferroelectric - semiconductor lead germanate Pb5Ge3O11 (PGO). Its domains are optically distinguishable due to existence of the optical activity which sign is reversed under switching. The other is improper ferroelectric - ferroelastic gadolinium molybdate Gd2(MoO4)3 (GMO).

Optical unit for real-time domains registration with the pulse dye laser as a light source was used to explore the domain kinetics with high spacial (about micron) and time (about 10 ns) resolution in wide range of switching times (up to microseconds). The peculiarities of domain shape and mechanisms of the domain walls sideways motion in weak, strong and superstrong fields were investigated.

The domain structure evolution on switching is none but an example of phase transformation. Its kinetics have much in common with such well-known phenomena as switching in ferromagnetics and liquid crystals or crystallization. As any phase transformation it must be and can be described in terms of elementary nucleation
processes of various dimensionalities. During ferroelectric switching the local electric field in the region of nucleation site is the driving force of transformation. This field is a superposition of the fields produced by applied voltage, bound charges (depolarization field), external and bulk screening charges of various nature. Moreover one must take into account the deformation in the vicinity of the domain wall which defined its orientation especially in ferroelectrics - ferroelastics.

**Sideways Domain Wall Motion**
The sideways motion of plane domain walls in GMO and PGO with the model domain structure was investigated in details. The wall position during switching was directly registered with high precision. After applying of field pulse the non-uniform movement was observed. The walls velocity reached quickly the maximum, then decreased slowly and at last became constant. In weak fields the wall stopped at some distance from initial position and when the field was switched off it spontaneously returns to start place.

To explain these results one must take into account the values and relaxation times of electric fields of different nature. It's sufficient to consider the simple model of ferroelectric capacitor with effective dielectric surface layers/gaps. In this system the screening of depolarization field occurs as at electrodes so in bulk near the layers. In PGO the time constant of external screening is about microseconds and bulk one - about hours. So the field produced by bulk charges is nearly constant during these experiments. The evaluation of the spacial distribution of total field in the vicinity of initial position of the wall allowed to explain quantitatively all obtained results. The same approach can be used for explanation of a lot of memory and retardation effects which were observed in real ferroelectrics.

**Domains Shape**
It is well-known that domains shape depends upon the value of switching field. In the weak fields the quasi-equilibrium domain shape is observed during all switching period. It is determined by crystal symmetry and elastic properties. In PGO (trigonal symmetry) it leads to the existence of regular shaped hexagonal domains. In GMO plane walls of two orthogonal orientations were observed. This situation is similar to the regular crystal growth under small oversaturations. In analogy the domain kinetics can be explained by lateral (layer by layer) growth through one-dimensional (1-D) nucleation.

In strong fields the irregular-shaped domains grow due to the 2-D-nucleation at the walls as usual. It's interesting that application of a train of short strong field pulses again leads to the formation of regular domains but of triangular shape. It's due to arising (during the pulse) of 2-D-nuclei as a steps on the walls and lateral wall smoothing in pause by 1-D-nucleation. Such competition of different
FIGURE 1a. The hexagonal domains, arising in PGO in weak fields: A - domain pattern; B - schematic drawing of domain growth. Thin arrows - the directions of real steps motion, thick arrows - observed walls motion. The dashed line - the facets of single crystal.

FIGURE 1b. The switching in GMO in weak field: A - the domain pattern; B - the domain walls and the facets of single crystal; C - the domain walls forming during switching from different single domain states.

FIGURE 1c. The switching in PGO in the train of strong field pulses: A - the domain pattern, B - schematic drawing of domain growth. The dashed line - the single crystal facets.
nucleation processes as a mechanism of common nature can be used for explanation of domain shape changes observed in real ferroelectrics.

Superstrong Fields
The field increasing leads to the qualitative changes of the character of domain structure evolution. In weak fields only already existing domains are growing. In strong fields a great number of small domains are arising on the entire sample surface. Under «superstrong field» (more then 12 kV/cm for PGO) the switching time is about microseconds and the lateral motion of domain boundary is observed again. However the mechanism of the motion is qualitatively different. A great number of small domains arise near the domain wall (correlated nucleation) forming a chain-like structure (Fig. 2a). Then they expand and merged with initial domain and at the same time a new chain appears and so on \(^{22,23}\).

![FIGURE 2a. Correlated nucleation in PGO in superstrong fields.](image)

This behaviour can be explained by local field spacial distribution anomalies near the moving domain wall. The depolarization field is not completely screened and the total field has a maximum on some distance from the wall. As a result the correlated nucleation occurs. This electrical remote action of domain walls results in inhomogeneous domain motion observed in different crystals.

The observed dynamical structure can be «freeze in» by application of the train of single polar pulses of superstrong field. Their duration must be many times shorter then switching time. As a result the special type of stable domain structure («wide domain wall») appears. It's a fractal-type structure consisted of the nonthrough domains. Its wideness can exceed 0.1mm (Fig. 2b). Important to stress that all observed peculiarities are common in nature with the kinetics of any highly nonequilibrium system and rather typical for them.

Incoherent Domain Walls
The dynamics of incoherent domain walls in ferroelectrics-ferroelastics was intensively studied both theoretically and experimentally \(^{24,25}\). It was shown that elastic
properties of crystals determines domains shape and kinetics. Nevertheless as in all considered situations the screening effects play the important role too. The dependence of the local field spacial distribution upon the domain shape determines the mobility and the start fields value of spike-like domains and zig-zag domain walls in GMO

In conclusion it is necessary to point out that all considered mechanisms can be used for explanation of observations reported by many authors in various crystals. The different individual behaviour can be attributed to the variety of material parameters: elastic constants, conductivity, symmetry, dielectric permittivity and so on.

TRANSIENT CURRENT INVESTIGATIONS: INTEGRAL CHARACTERISTICS

The most popular experimental method of studying the polarization reversal (the only one possible in thin films) is the registration of transient current proposed by Merz. In this case the total response of the system is formed by great number of domains arising and growing during switching. Thus the statistical properties of «many-domain» system plays the principle role. In analogy with many-body systems its behaviour can be described only within some statistical theory. Such approach was developed by Kolmogorov and Avrami for explanation of crystallization kinetics. Their ideas were used in phenomenological descriptions of transient current by Wider and Fatuzzo.

Ishibashi and Takagi were the first who consistently applied Kolmogorov-Avrami (K-A) theory for the analysis of ferroelectric domain switching. Two limiting situations were considered: alpha-model (I category) - the nucleation probability remains constant during switching and beta-model (II category) - all the nuclei are formed at the very beginning. Two-dimensional (2-D) approach can be applied because for the main part of experimental situations (especially for thin films) one can neglect transit time of domains through the thickness of the crystal.

The K-A theory was formulated for infinite media only. But in real systems (especially in polycrystal thin films) the edge effects are very important. Recently

FIGURE 2b. Fractal-type domain structure formation in the train of superstrong field pulses.
Duiker and Beale\textsuperscript{31}, Ishibashi and Orihara\textsuperscript{32} made an attempt to apply the K-A approach to real finite systems. It was shown also that computer simulation is useful for testing the theory.

The attempt to approximate experimental data by K-A formula with one set of parameters only leads to fractal dimensionalities of nuclei ($n$)\textsuperscript{33,34}. This indicates that K-A theory can't be used in such direct simple form. To solve this problem we suggest to divide all the time of switching process into some intervals according to some geometrical transformations of the domains. In each interval pure K-A law with definite set of parameters can be used for fitting the switching current curve with high accuracy (Fig. 3a). It is important to point out that in this case $n$ always (in all intervals) remain integer. We shall name the boundaries between intervals «the time of the catastrophe».

\begin{center}
\begin{figure}
\centering
\includegraphics[width=\textwidth]{fig3a.png}
\caption{Transient current for thin film of PLT approximated by K-A formula with integer $n$ taken into account catastrophical transformation of domain geometry.}
\end{figure}
\end{center}

The computer simulation was carried out for alpha- and beta-models with isotropic and anisotropic domain walls sideways motion\textsuperscript{35}. The 2-D-model of 400*400 unit cells represents a thin sample of uniaxial ferroelectric. It was shown that in all cases the initial interval is described by usual K-A formula with appropriate integer value of $n$. It means that during this interval domains does not «feel» the edges. The first catastrophe occurs at the moment equal to the average time interval which is needed for the domains to reach the nearest edge. At the moment of touching the domains are «magically transformed» into two parts with different behaviour.
Let us consider the simplest situation: beta-model with anisotropic growth (Fig.3b). In this case at the first catastrophe the domains touch the nearest edge and at second - the opposite one. During the first time interval there is no edge effects and derivative of switched area is described by K-A formula with $n = 2$. The domain growth is decomposed in two parts during the second interval: one half of the domain continue 2-D-growth ($n = 2$) and another one is growing as a ribbon along the edge ($n = 1$). At last interval the strip domain arise and only 1-D-growth remains ($n = 1$). It is possible to calculate the average time of any catastrophe. Computer simulation proves that this simple model can give an extremely good approximation for the data averaged upon the large number of realizations (computer experiments) (Fig.3b). It's important to stress that experimental data for real polycrystal thin film are averaged for a great number of grains. The similar approach was successfully applied to all considered situations.

Such statistical description of real domain kinetics during switching allow to use new physically clear parameters characterizing the ensemble behaviour in terms of geometric phase transitions. Moreover the mathematical treatment of transient current curves allow to solve the reverse problem and obtain the main averaged parameters of domains kinetic in real ferroelectric crystals and thin films. We confirmed the validity of this approach for the model crystals.
CONCLUSION

From our point of view the most interesting trends in modern research of ferroelectric domain switching are connected with: 1) superfast switching phenomena; 2) using of model crystals for careful in situ investigations of domain kinetics; 3) computer simulations of domain processes. It was shown that «fast evolution» of the domain structure is a kinetic process during which non-equilibrium momentary domain patterns arise. The competition between the nuclei of different dimensionalities under the action of time-dependent local electric field determine the behaviour of system. This total field is influenced by various screening processes with different time constants. Thus numerous retardation and memory effects take place.

The model crystals are very useful not only for investigating and understanding of domain dynamics by direct methods but also for carrying out correlation between integral and local characteristics. Moreover the results obtain during the investigations of model ferroelectric crystals are of fundamental interest and can be used for explanation of superfast crystallization of amorphous films, ultrafast switching in liquid crystals and ferromagnetics etc.

The computer simulations in couple with complex of traditional experimental methods reveal itself as an extremely powerful tool for the detail investigations of the switching phenomena even in heterogeneous systems (e.g. thin films).

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