Non-Kolmogorov-Avrami switching kinetics in ferroelectric thin films

Yu Yun

10/04/2019

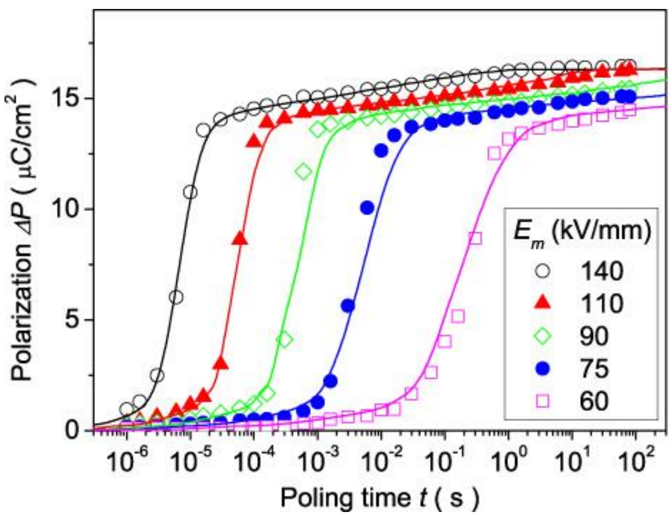
UNIVERSITY OF Nebraska Lincoln

UNIVERSITY OF Nebraska Lincoln

Motivation & Background

Kolmogorov-Avrami-Ishibashi (KAI) model

- focused on the statistics of domain coalescence
  - the classical theory of nucleation and growth of reversed domains.



$$\Delta P(t) = 2P_s c(t) = 2P_s(1 - exp(-\frac{t}{t_0})^d)$$

J. Phys. D: Appl. Phys. 45 (2012) 165301

Non-KAI model
Nucleation-limited-switching (NLS) model

- focused on the statistics of nucleation

PHYSICAL REVIEW B **66**, 214109 (2002)

Non-Kolmogorov-Avrami switching kinetics in ferroelectric thin films

Alexander K. Tagantsev, Igor Stolichnov, and Nava Setter
Ceramics Laboratory, Swiss Federal Institute of Technology (EPFL), CH-1015 Lausanne, Switzerland

Jeffrey S. Cross and Mineharu Tsukada
Fujitsu Laboratories, Ltd., 10-1 Morinosato-wakamiya, Atsugi, 243-0197 Japan
(Received 27 March 2002; published 23 December 2002)

The switching kinetics in ferroelectric thin films has been intensively studied during the past decade. It is widely accepted that this kinetics is basically governed by the dynamics of domain coalescence (the Kolmogorov-Avrami-Ishibashi model). This conclusion is mainly supported by fitting the time dependence of the switching currents to that predicted by this model, the fit being typically performed in a 1–2 decade interval of time. The present paper reports on a study of the switching kinetics in modified Pt/Pb(Zr,Ti)O_3/Pt thin films as a function of time and applied voltage, performed in time intervals from 10 ns to 1s. Our experimental data show that both the time and applied field dependences of the switching polarization (when monitored over a wide enough time interval) are in a strong qualitative disagreement with the predictions of the Kolmogorov-Avrami-Ishibashi approach. For the interpretation of our result, an alternative approach is forwarded. In contrast to Kolmogorov-Avrami-Ishibashi approach, we assume that the film consists of many areas, which have independent switching dynamics. The switching in an area is considered to be triggered by an act of the reverse domain nucleation. The switching kinetics is described in terms of the distribution function of the nucleation probabilities in these areas. The developed approach enables a good description of the polarization dynamics in typical ferroelectric thin films for memory applications.

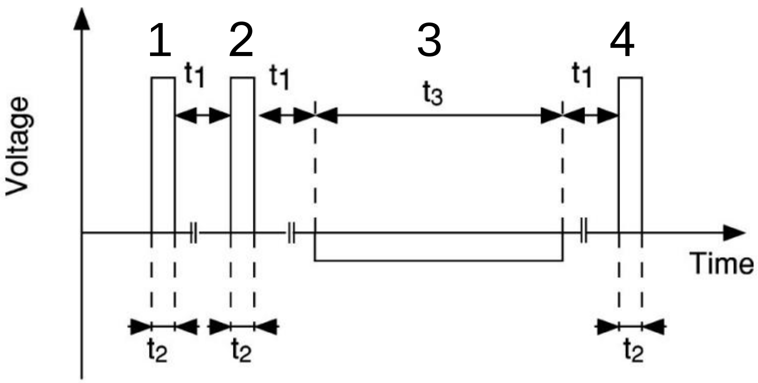
1) They assume that the film consists of many areas, which have independent switching dynamics.
2) The switching in an area is considered to be triggered by an act of the reverse domain nucleation.
3) The switching kinetics is described in terms of the distribution function of the nucleation probabilities in these areas.

Experimental

Sample information:

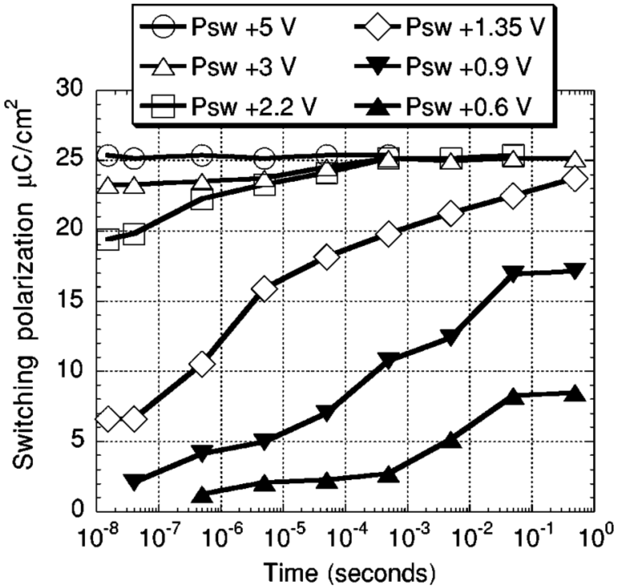
- **PZT** films of (111)-orientation grown on Pt (111) bottom electrode by chemical solution deposition.
- The films with Zr/Ti ratio of 40/60 contained dopants including Ca, Sr, and La.
- Thickness ~ **135 nm**, 15 by 15 μm IrO_x top electrodes.

❖ **Sequence of voltage pulses used for measurements of the switching polarization**



- Intervals t_1 = 1s,
- t_2 = 50ns, amplitude for pulse 1,2,4 is 5V
- t_3, ranged from 10 ns to 0.5 s, for voltage amplitudes of 0.6, 0.9, 1.35, 2.2, 3, and 5 V

❖ **Switching polarization as a function of switching time, for different voltages**



I. The role of pulse 1 was to define the polarization state of the film.

II. Since pulse 2 had the same polarity as pulse 1, the current response corresponding to pulse 2 determined the non-switching polarization.

III. Pulse 3 switched polarization partially or completely depending on its **amplitude and width**.

IV. Pulse 4 measured the polarization switched by pulse 3.

Model for Switching Kinetics – review KAI model

The basic idea of KAI model

- FE domains have been initiated from **independent nucleation centers**, **unrestrictedly grow** under the action of the applied electric field.
- At early stages of switching, **the appeared domains grow without overlapping**, so that **the volume where the polarization is reversed can be found as the sum of those of the individual domains.**

- ❑ In terms of the KAI model one nucleus, in principle, **can provide switching of the whole sample**.
- ❑ The **switching occurs region-by-region**, and **the switching in a region does not necessarily lead to the subsequent switching of the neighboring** ones as is expected according to the KAI model.

KAI model fitting

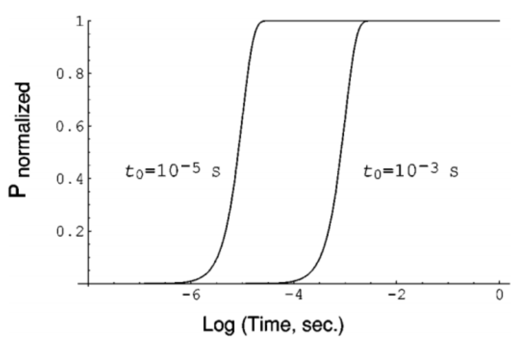
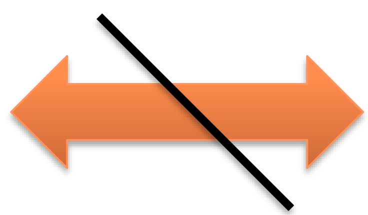
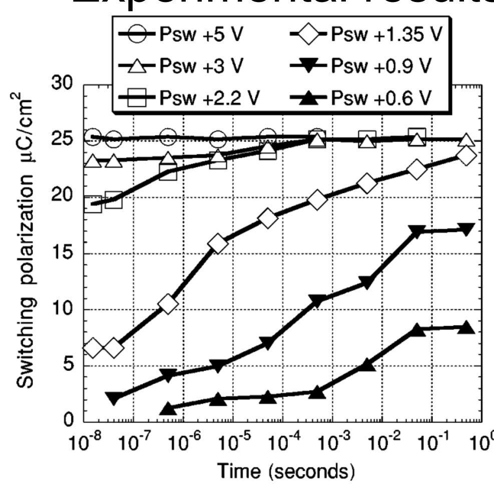


FIG. 3. Prediction by the Komlogorov-Avraami-Ishbashi model: fraction of switched polarization normalized to its maximal value as a function of time, for different values of parameter t_0 according to Eq. (1) with $n=2$.

contradict



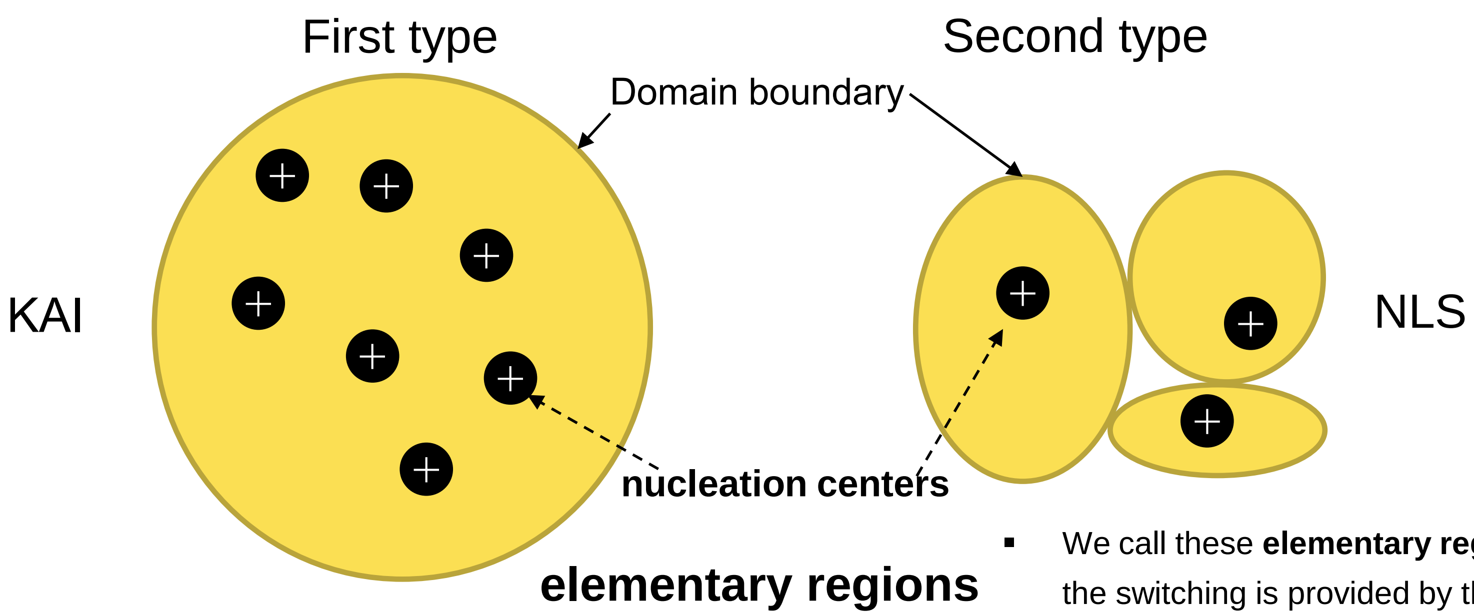
Experimental results



Model for Switching Kinetics

Two types of switching kinetics

- **First type**, in large enough elementary regions, which contain **a large number of nucleation centers**, the switching kinetics should **obey the KAI model.**
- **Second type**, in regions containing **only a few nucleation centers**, a very different type of the switching kinetics takes place. **The total contribution of the second type of regions is controlled by the statistics of virtually independent switching of these regions**



- We call these **elementary regions**, where the switching is provided by the domains nucleated at centers belonging to this region.

Model for Switching Kinetics

Assumptions of NLS model

i. **The film is presented as an ensemble of elementary region.**

ii. **The switching of an elementary region occurs once a domain of reversed polarization is nucleated in the region.**

iii. **Time needed for switching of an elementary region is equal to the waiting time for the first nucleation, i.e., the time needed for filling the region with the expanding domain is neglected compared to the waiting time.**

iv. **The distribution of the waiting times for the ensemble of the elementary regions is smooth and exponentially broad, i.e., covering many decades.**

- **T_i as the typical waiting time** of the elementary region (i),
- $1/T_i$ being the nucleation rate in the region
- and $\gamma_i = S_i / \langle S_i \rangle$ for the ratio of the area of region (i) S_i to the average area $\langle S_i \rangle$ of elementary regions in the film.
- The fraction of the volume of the ferroelectric switched by time t, p(t) is given by the obvious relation

$$p(t) = 1 - \frac{\sum_{NS} S_i}{\sum_{all} S_i}, \qquad (1)$$

The special case

- **The special case of this problem where the elementary regions are identical**, **i.e., all T_i and all S_i are equal.**
- the classical decay equation for the number of non-switched areas N_{NS}

$$dN_{NS} = -N_{NS}\frac{dt}{\tau} \qquad (2)$$

Boundary condition $N_{NS}=N_0$ at $t=0$

- N_0 is the total number of the elementary regions in the film

Leads to $N_{NS} = N_0 e^{-t/\tau}$ (3)

identical to the prediction of **KAI model** for the case of **one-dimensional domain growth**

$$p(t) = 1 - e^{-t/\tau} \qquad (4)$$

the NLS model comprises a distribution of T_i and γ_i

approximation

$$p(t) = 1 - \langle \gamma_i e^{-t/\tau_i} \rangle \simeq 1 - \langle e^{-t/\tau_i} \rangle \qquad (5)$$

Since the waiting time enters the exponent of this expression, averaging over its broad distribution is decisive for our problem. For this reason, addressing in this paper the main trends of the phenomenon, **we neglect with the logarithmic accuracy the correlation between the distributions of T_i and γ_i**

Replace $\langle \gamma_i e^{-t/\tau_i} \rangle$ by $\langle \gamma_i \rangle \langle e^{-t/\tau_i} \rangle = \langle e^{-t/\tau_i} \rangle$

Model for Switching Kinetics

- Approximation widely used in the theory of disordered systems

$$\langle e^{-t/\tau_i}\rangle = \int_{-\infty}^{\infty} e^{-t/\tau} g(\ln\tau)\, d(\ln\tau)$$

the distribution function g(z) meets the normalizing condition

$$\int_{-\infty}^{\infty} g(z)\, dz = 1$$

$$z = \ln\tau \quad z_0 = \ln t,$$

$$\langle e^{-t/\tau_i}\rangle = \int_{-\infty}^{\infty} \exp(10^{z_0 - z}) g(z)\, dz$$

the step function centered at $z=z_0$

$$\langle e^{-t/\tau_i}\rangle = \int_{z_0}^{\infty} g(z)\, dz$$

$$p(t) = 1 - \langle \gamma_i e^{-t/\tau_i}\rangle \simeq 1 - \langle e^{-t/\tau_i}\rangle$$

$$p(t) = \int_{-\infty}^{\ln t} g(z)\, dz$$

Application of the NLS Model

$$p(t)=\int_{-\infty}^{\ln t} g(z)\,dz$$

a distribution function of the waiting times, which is **flat for z lying between z_1 and z_2**

$$\tau_{\min}=10^{z_1} \qquad \tau_{\max}=10^{z_2}$$

out of this region, decays as $1/z^2$, the rate of the decay being controlled by parameter Γ

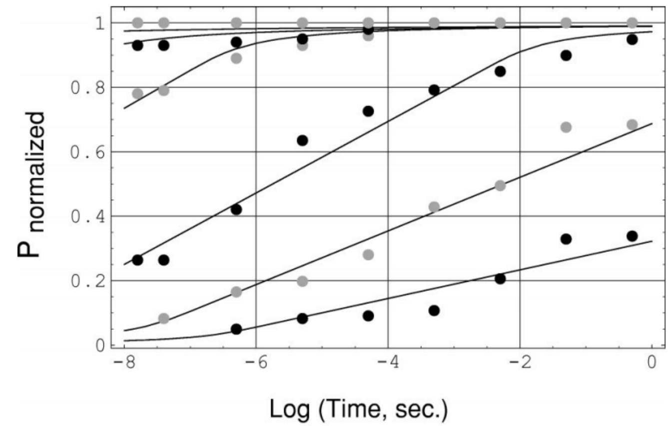


FIG. 4. Fit of the switching curves from Fig. 2 using the prediction of the NLS model, Eq. (10). Experimental data from Fig. 2 for voltages of 0.6, 1.35, and 3 V are marked with black dots, the data for 0.9, 2.2, and 5 V are marked with gray dots.

$$g(z)=\frac{\Gamma^2 h}{(z-z_1)^2+\Gamma^2} \quad \text{for } z_0<z_1,$$

$$g(z)=h \quad \text{for } z_1<z_0<z_2,$$

$$g(z)=\frac{\Gamma^2 h}{(z-z_2)^2+\Gamma^2} \quad \text{for } z_2<z_0,$$

$$h=(z_2-z_1+\Gamma\pi)^{-1}.$$

$$p(t)=\Gamma h\left(\frac{\pi}{2}-\arctan\frac{z_1-z_0}{\Gamma}\right) \quad \text{for } z_0<z_1$$

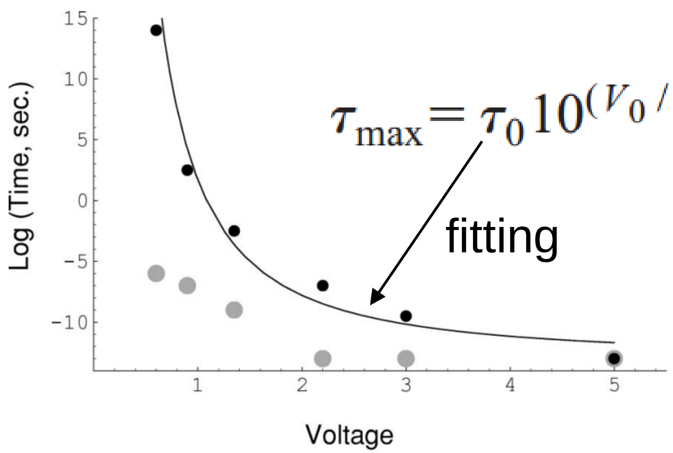
$$p(t)=\Gamma h\left(\frac{\pi}{2}+\frac{z_0-z_1}{\Gamma}\right) \quad \text{for } z_1<z_0<z_2,$$

$$p(t)=\Gamma h\left(\frac{\pi}{2}+\frac{z_2-z_1}{\Gamma}+\arctan\frac{z_0-z_2}{\Gamma}\right) \quad \text{for } z_2<z_0,$$

$$z_0=\log t. \tag{10}$$

Application of the NLS Model

- The observed switching kinetics can be described in terms of the NLS model where the spectrum of the **waiting times of the elementary regions rapidly shrinks with increasing applied voltage**



$$\tau_{\max} = \tau_0 10^{(V_0/V)^n} \quad (11)$$

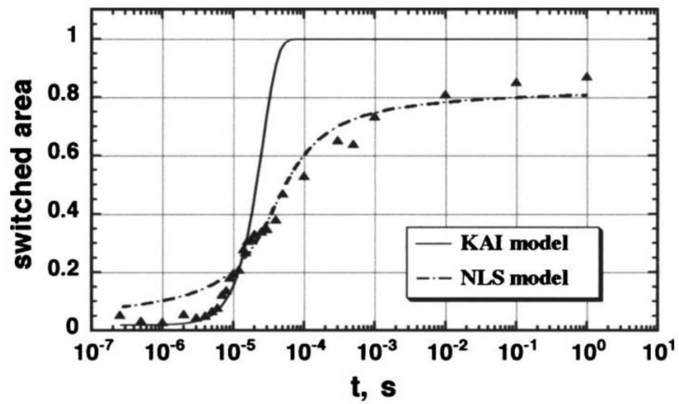
FIG. 5. Upper (black dots) and lower (gray dots) limits for the spectrum of waiting times for domain nucleation used for fitting curves in Fig. 4 as functions of the applied voltage. The solid line shows a fit of the experimental data for voltage dependence of the maximal waiting time to the function given by Eq. (11).

$$\tau_{\max} = \tau_0 e^{(E_0/E)^{1.5}}$$

$\tau_0 = 10^{-13}$ s, $n = 1.5$, and $V_0 = 6$ V.

Summary

1. The NLS model suggests that the switching in thin films is limited by the nucleation of reversed domains rather than by the sideway motion of domain walls.
2. As a result, in the NLS model the polarization response is controlled by a distribution of relaxation (waiting) time whereas a unique-relaxation-time kinetics is inherent to the KAI model.
3. An increase of the field results in a shrinkage of the spectrum.



Gruverman et al. Appl. Phys. Lett. **87**, 082902 (2005)

Thanks for your attention