PFM Studies of Polarization Dynamics in Ferroelectric Capacitors

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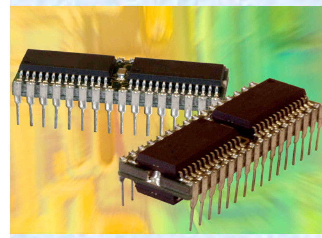
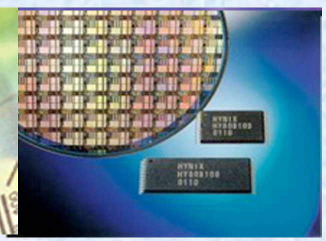
Outline

- **Introduction**
  Ferroelectric switching: theory and experiment
- **Bias Dependent Measurements in PFM**
  Variability of switching parameters at the nanoscale
- **Time Dependent Measurements in PFM**
  Applicability to fast switching processes
- **Domain Switching Kinetics in Thin Film Capacitors**
  Polycrystalline vs epitaxial
- **Capacitor Scaling Effect**
  Nucleation vs wall motion
- **Conclusion**

Ferroelectric Memories

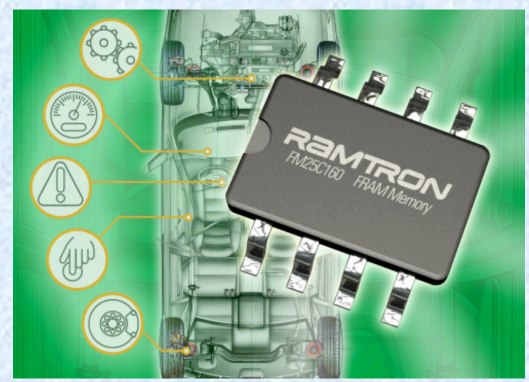




FeRAM - a dream semiconductor memory:

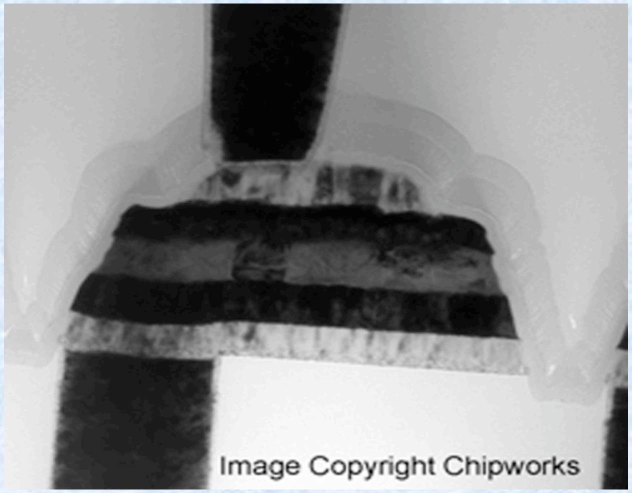
- **High-density (as DRAM)**
- **High-speed (as SRAM)**
- **Nonvolatile (as Flash)**

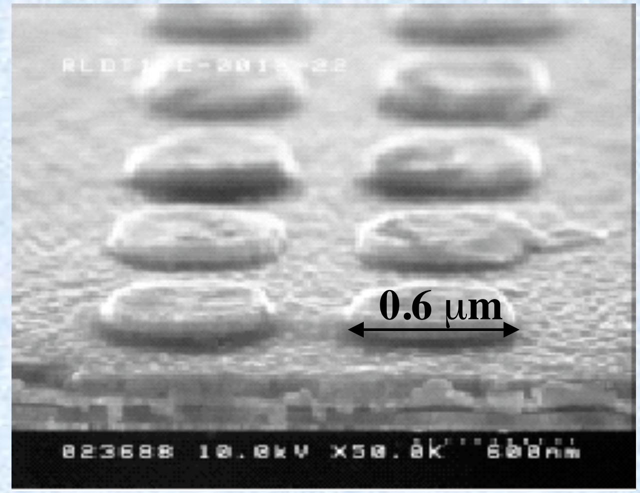
Over 30 million products using FRAM have already been shipped, including metering, RFID and smart-card devices (source: Ramtron, 2007).



Scaling of Ferroelectric Memories

PZT FeRAM cells



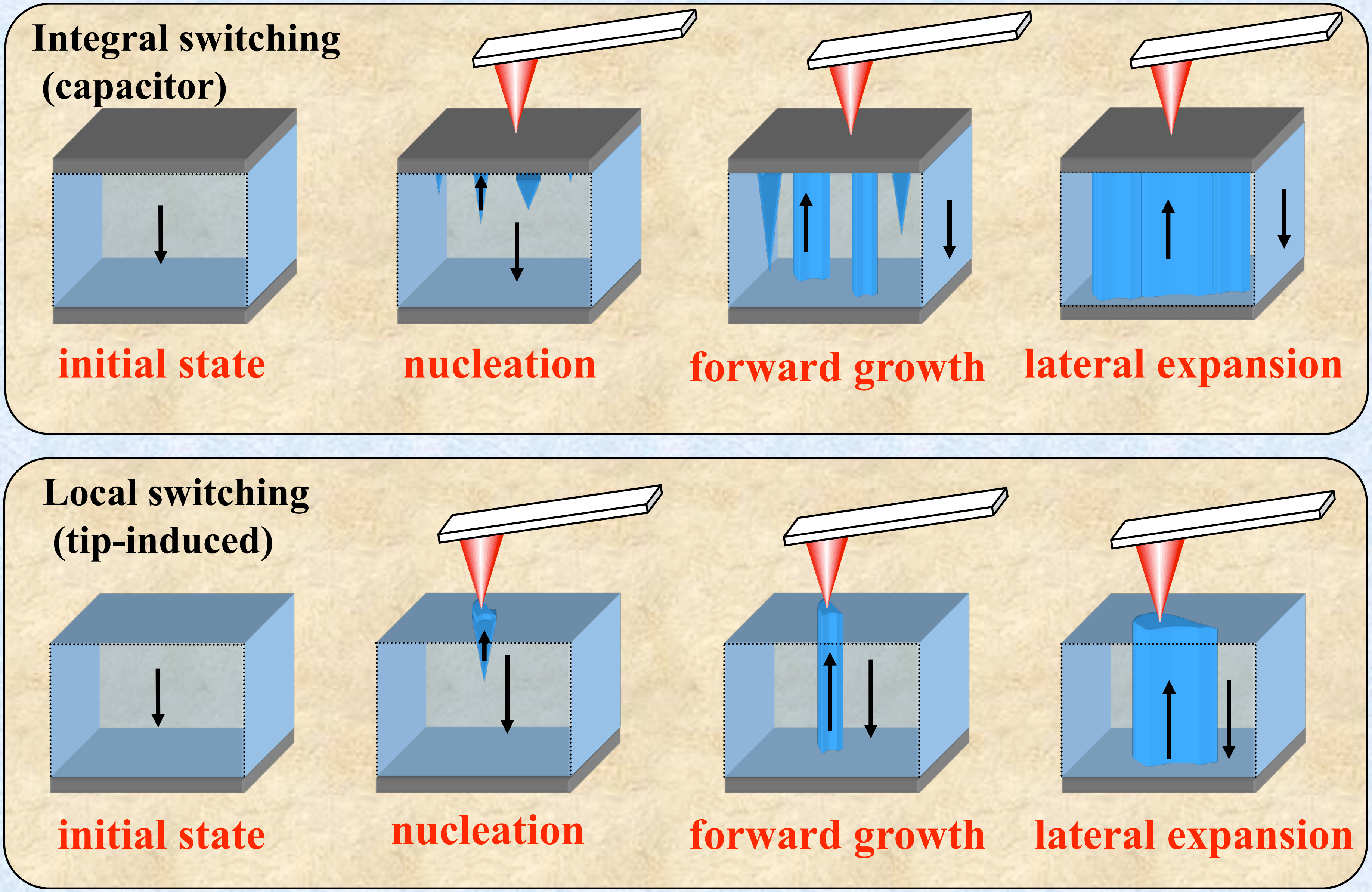


Size effects become critical:

- **Intrinsic limit for stable polarization**
- **Extrinsic effect on a stable domain and mechanism of switching**
- **Size limit for an operational device**

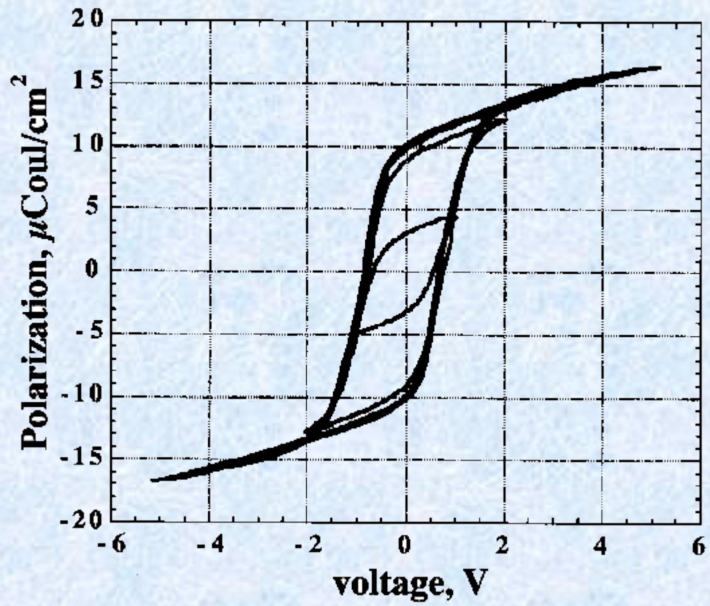
Macroscopic measurements → **Local probe testing**

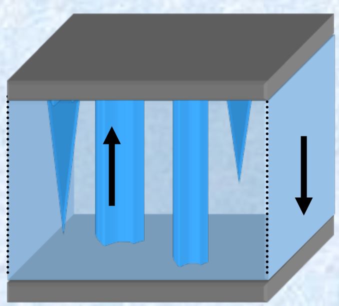
Mechanism of Ferroelectric Switching in PFM



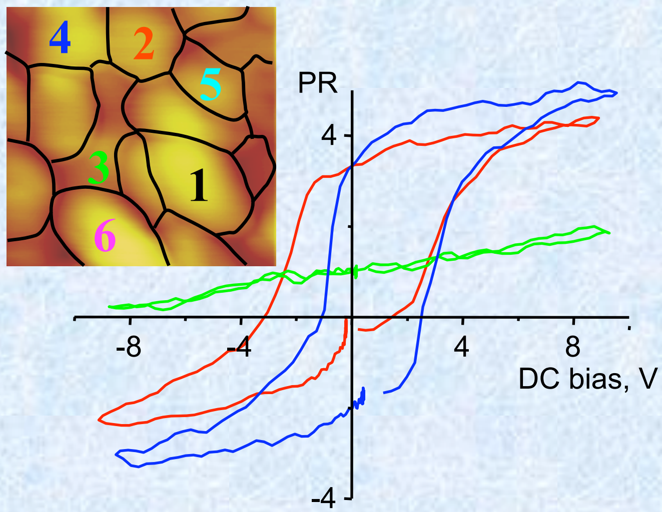
Hysteresis Loop: Global Switching vs Local

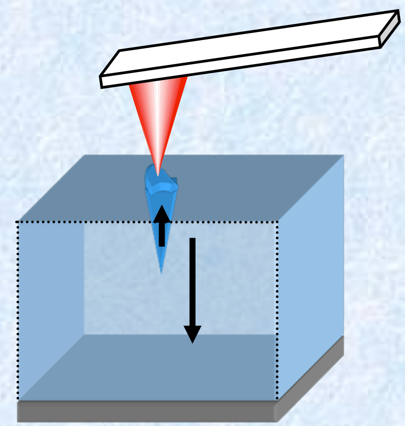
Polarization Hysteresis Loops





PFM Hysteresis Loops



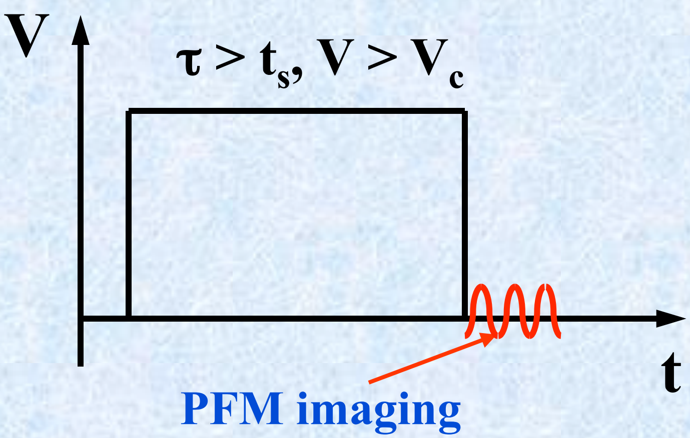


$$PR = d_{\mathrm{eff}}\{V_0 - 2V_{\mathrm{w}}\}$$

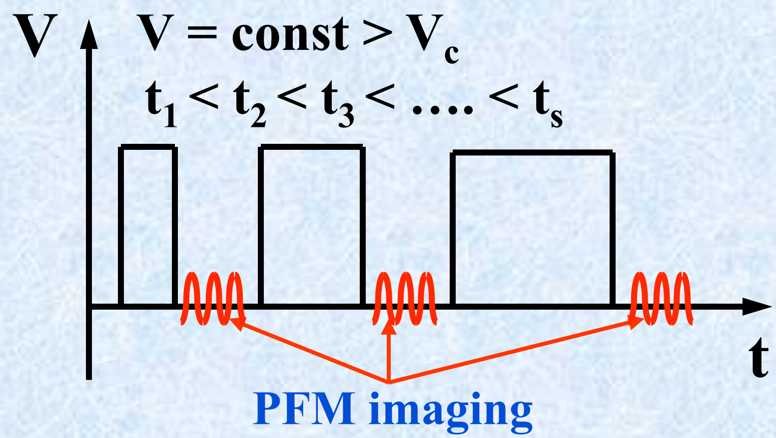
Kalinin et al, APL (2004)

Pulse Trains for PFM Switching Studies

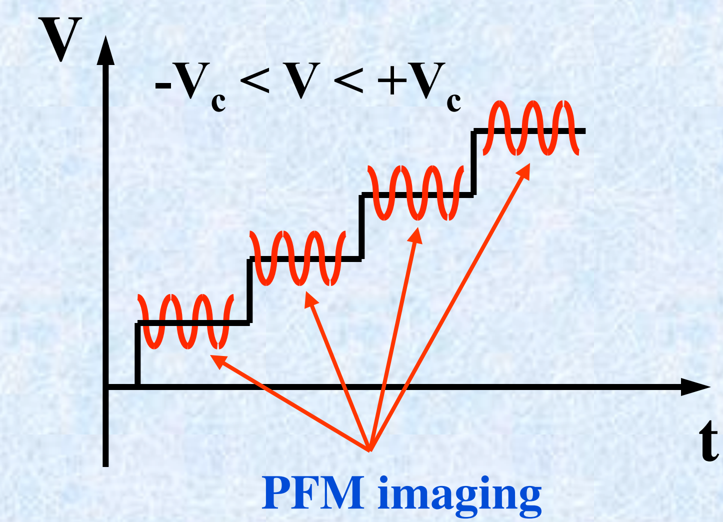
1. Conventional (switch-and-image) testing



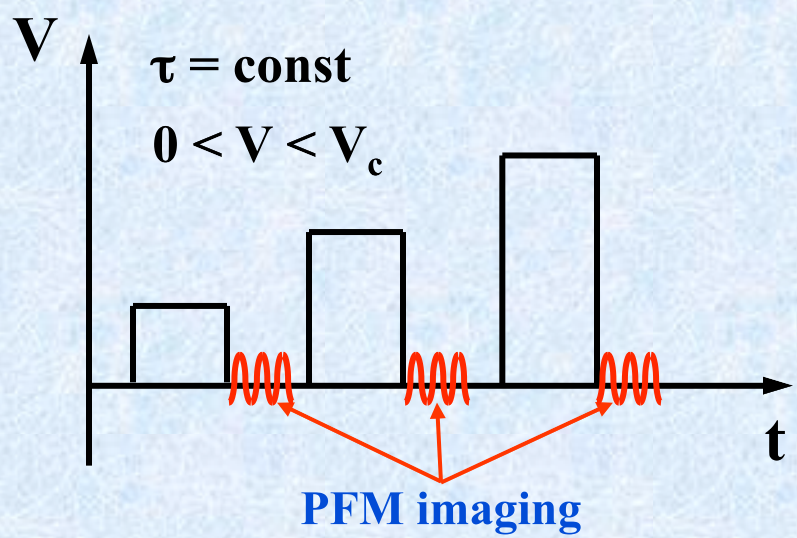
2. Stroboscopic (time dependent) testing



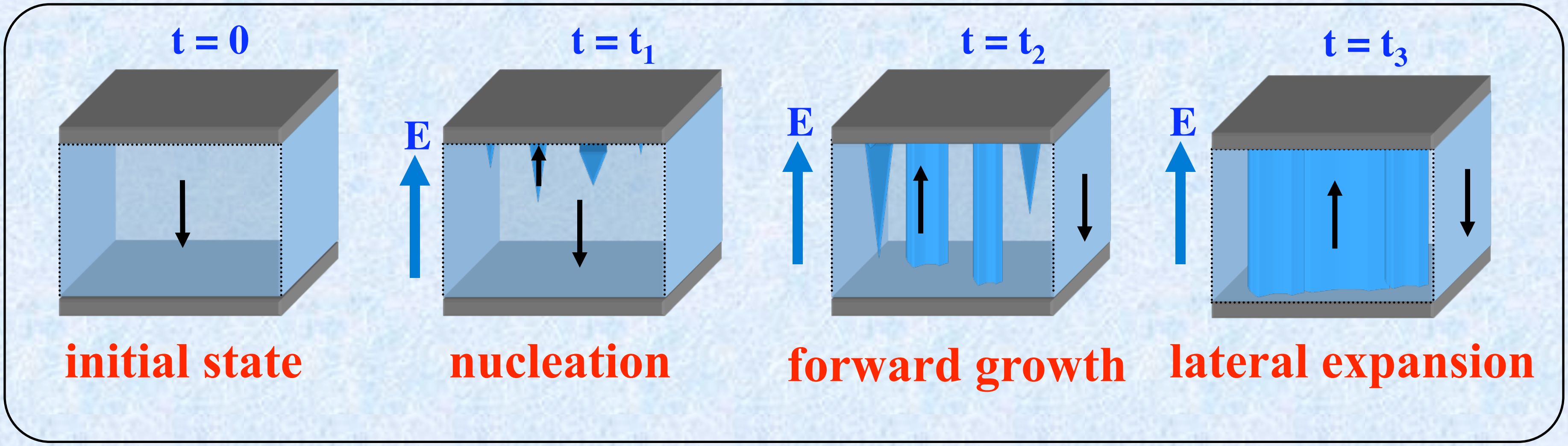
3. Spectroscopic (step bias) testing



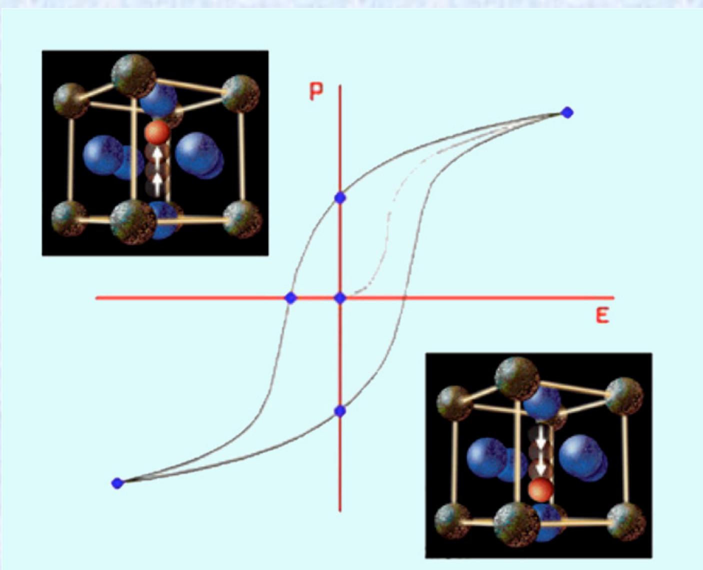
4. Spectroscopic (pulsed bias) testing



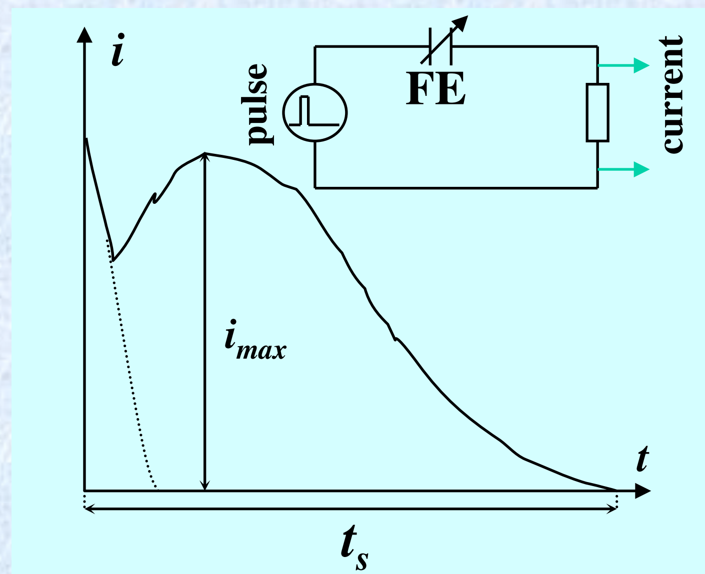
Mechanism of Switching in Ferroelectrics



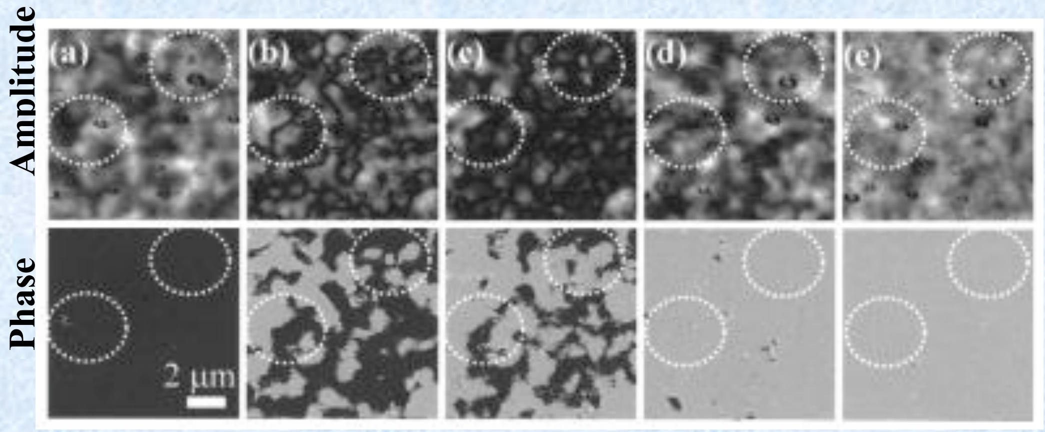
Bias dependence



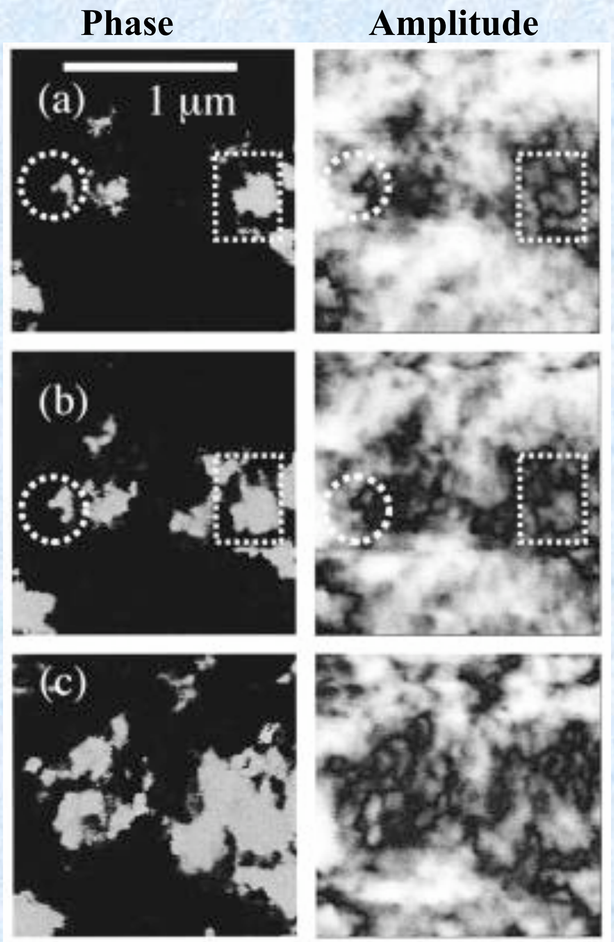
Time dependence



Domain Evolution as a Function of Applied Bias



S. Hong and N. Setter, APL **81**, 3437 (2002)

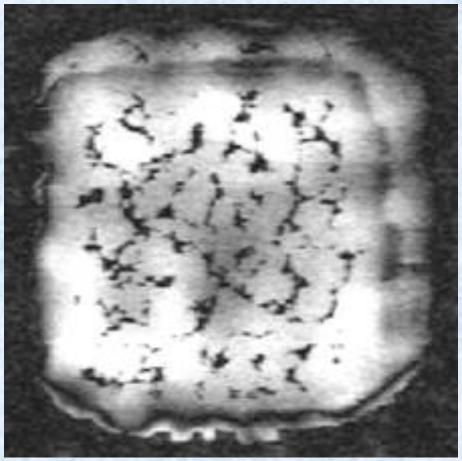


I.Stolichnov et al, APL **86**, 012902 (2005)

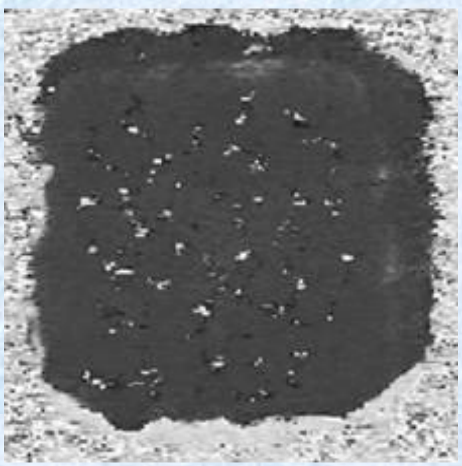
Polarization as a function of applied DC bias

1.1 x1.1 μm^2 capacitor

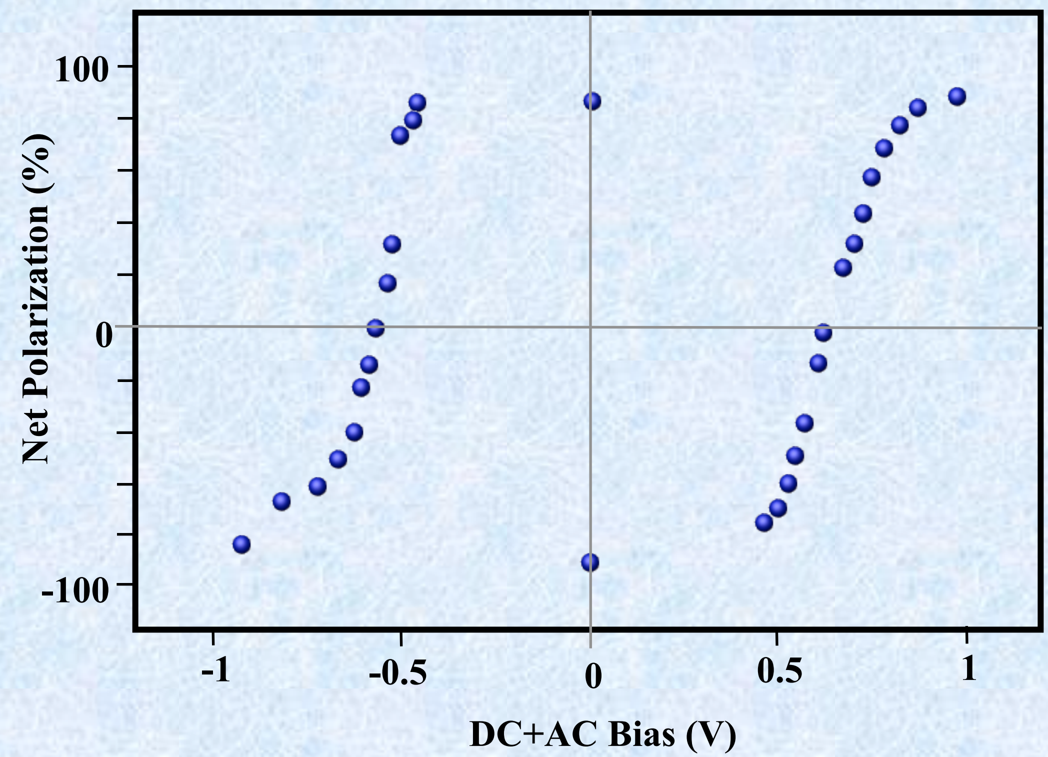
VPFM Amplitude



VPFM Phase



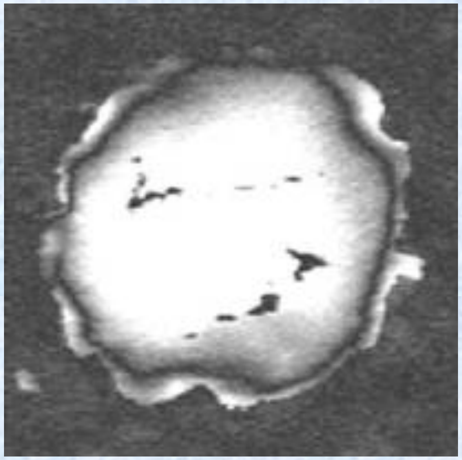
PFM imaging of ferroelectric capacitors provide direct microscopic evidence of the microscopic switching behavior.



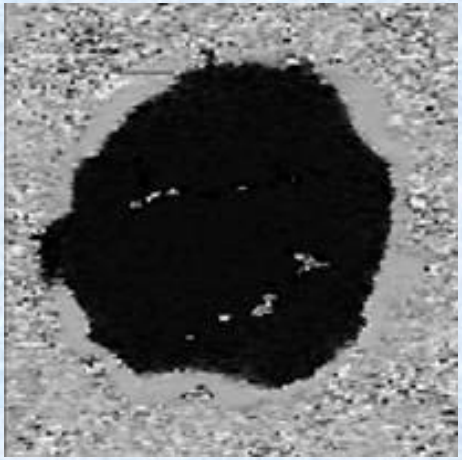
Polarization as a function of applied DC bias

$0.5 \times 0.5\ \mu m^2$ capacitor

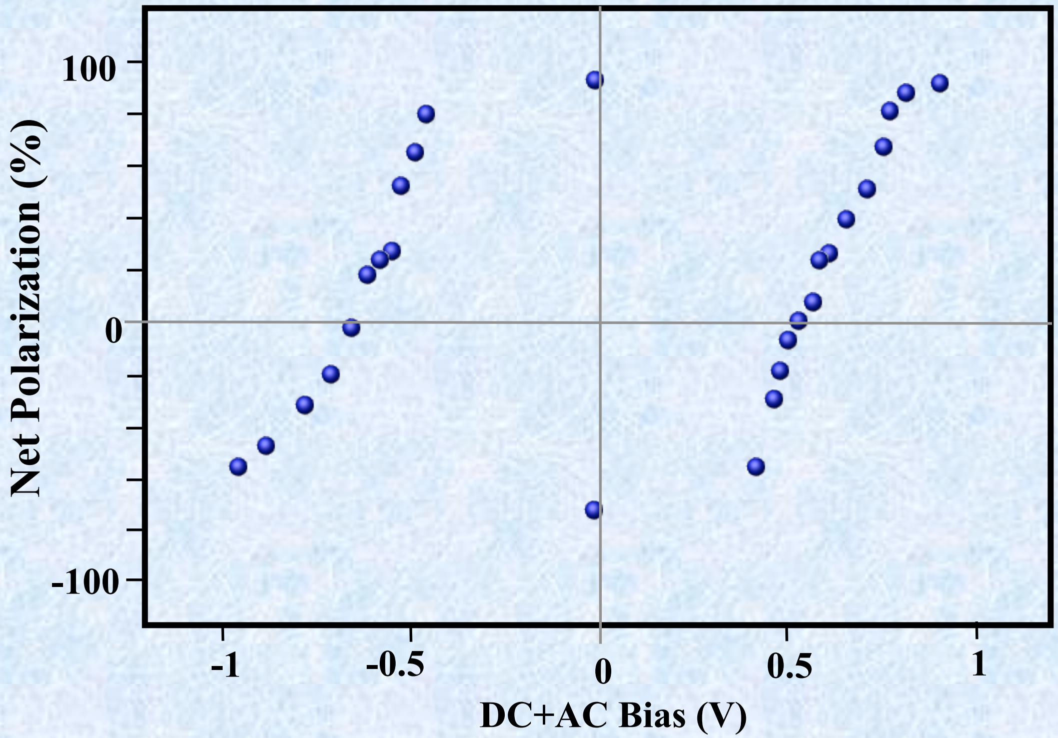
VPFM Amplitude



VPFM Phase



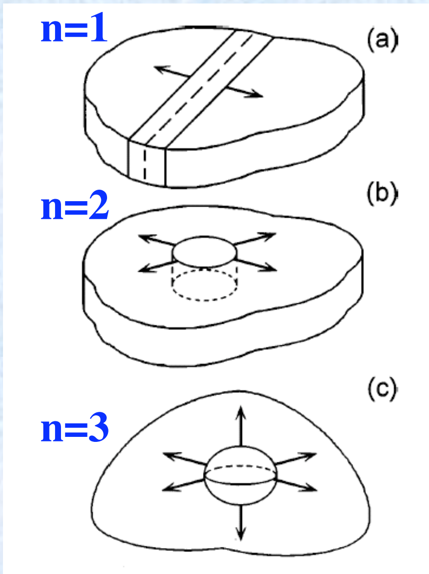
Electrode edge effect is observed: non-switched domains along the edge serve as seeds for switching. Reverse switching is almost exact re-trace of the forward switching.



Switching Kinetics in Ferroelectrics

Kolmogorov-Avrami-Ishibashi model

(nucleation in infinite medium)



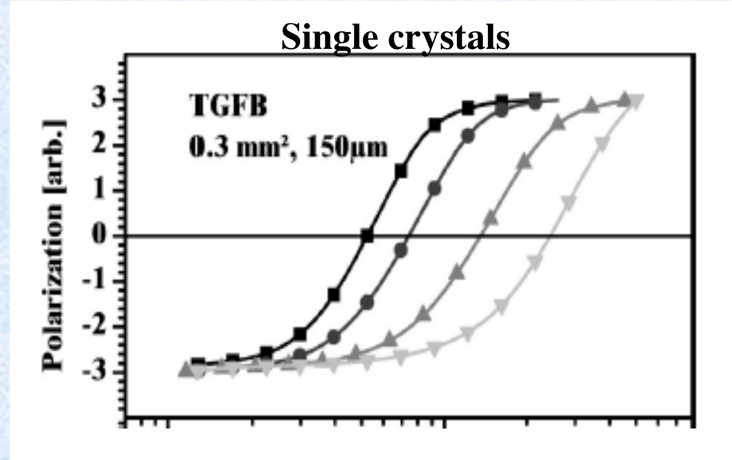
Fraction of switched polarization:

$$f(t) = P(t)/P_{total}$$

$$f(t) = 1 - \exp[-(t/t_0)^n]$$

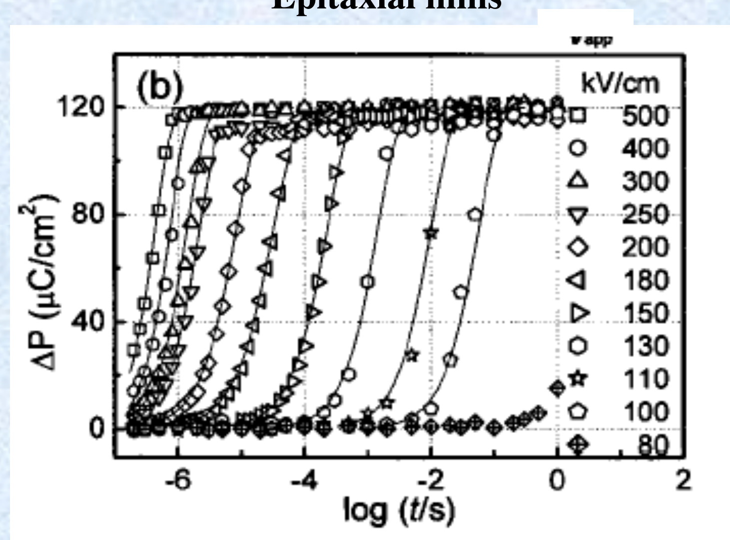
t_0 - characteristic switching time
n - dimensionality of domain growth

A. N. Kolmogorov, Izv. Akad. Nauk **3**, 355 (1937)
M. Avrami, J. Chem. Phys. **8**, 212 (1940)
H. Orihara *et al*., J. Phys. Soc. Jpn. **63** 1031 (1994)



C. Pulvari and W. Kuebler, JAP **29**, 1742 (1958)

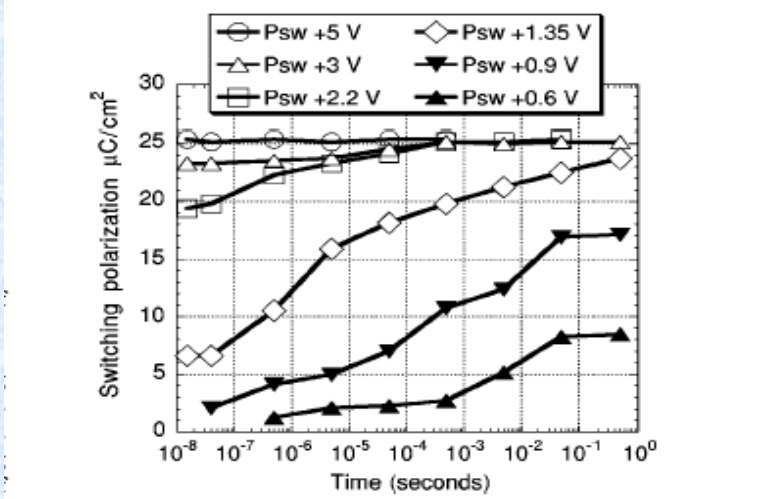
Epitaxial films



Y. W. So *et al*, APL **86**, 92905 (2005)

Switching in Polycrystalline Films

• **Nucleation limited switching (NLS model)**

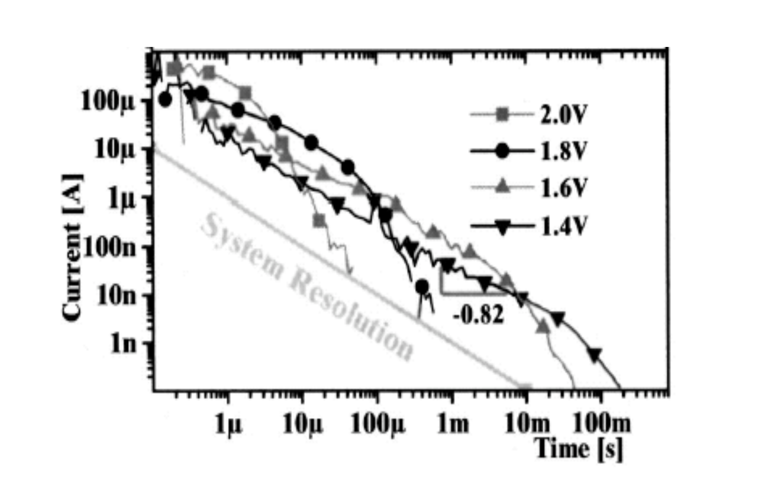


Nucleation limited switching mechanism (no effect of domain coalescence)

$$\frac{P(t)}{P_m} = 1 - \left\langle e^{-t/\tau} \right\rangle$$

Jung et al, Integrated Ferroelectrics 48, 59 (2002)
Du and Chen, MRS Proc. 493, 311 (1998)
Tagantsev et al, PR B 66, 214109 (2002)

• **Curie–von Schweidler behavior**



Ferroelectric switching is a result of polarization processes with a broad distribution of relaxation times:

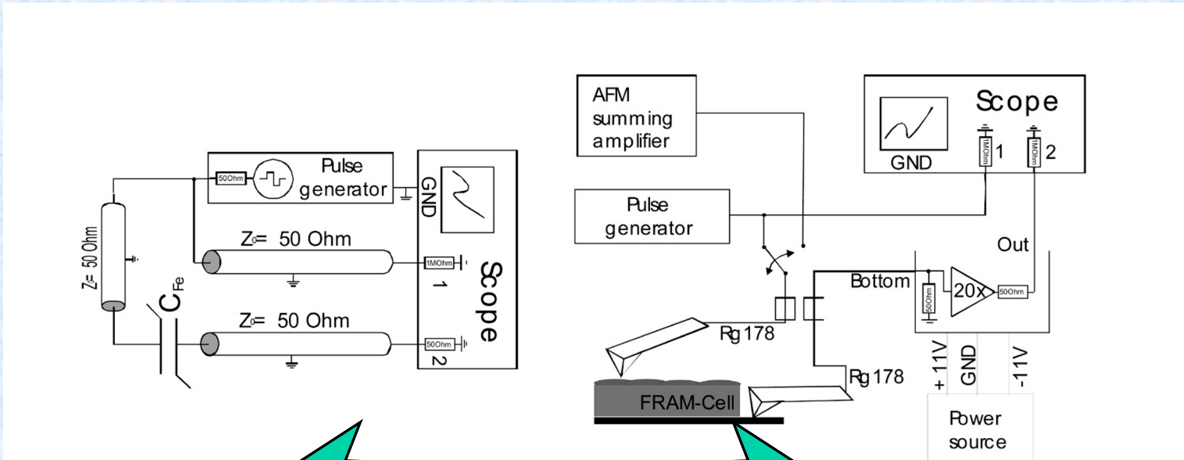
$$i \sim t^{-\alpha}$$

Lohse et al, JAP 98, 2332 (2001)

PFM Setup for Time-Dependent Switching Studies

Rise time: ~ 5-20 ns
Pulse width: > 40 ns

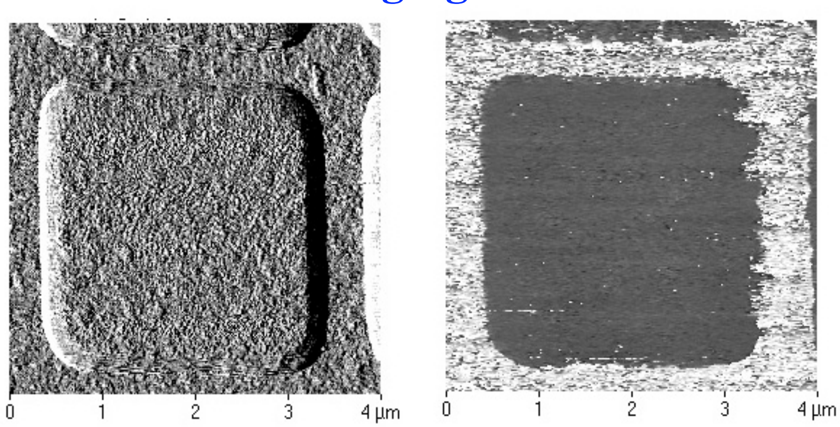
Transient current 10 μA
Min cap size: 1 μm^2



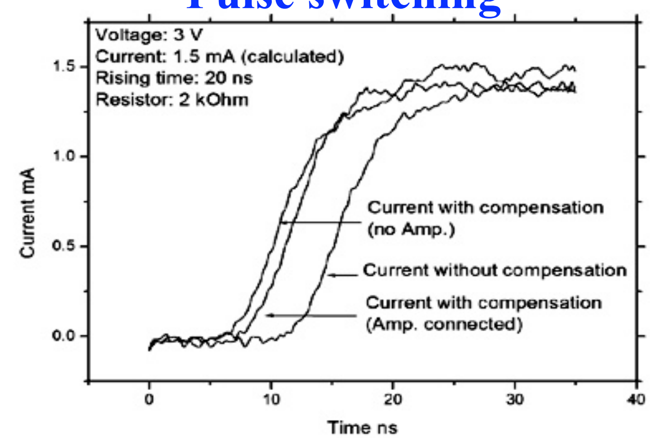
Key parameters:

- Pulse rise time
- RC-constant
- Impedance matching

PFM imaging



Pulse switching

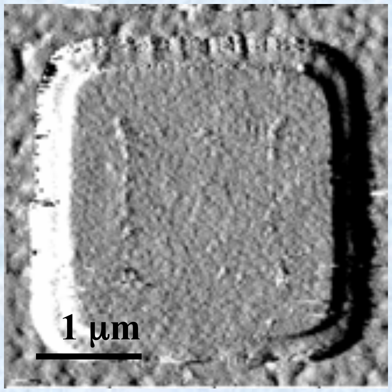


PZT Capacitors for Switching Studies

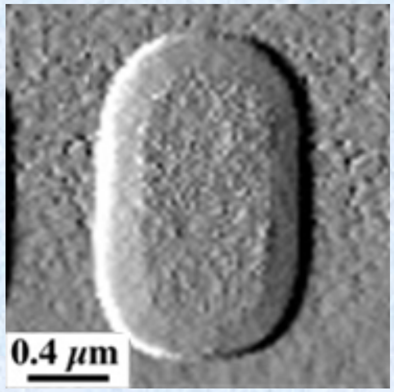
Polycrystalline (111) $Pb(Zr_{0.4}Ti_{0.6})O_3$

Epitaxial (001) $Pb(Zr_{0.2}Ti_{0.8})O_3$

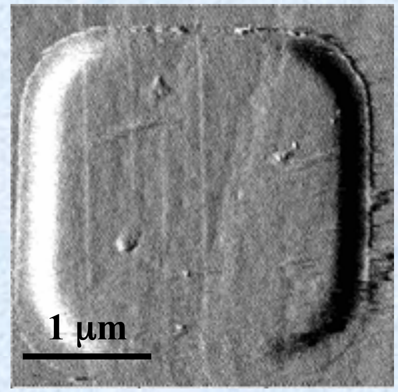
$3x3\ \mu m^2$



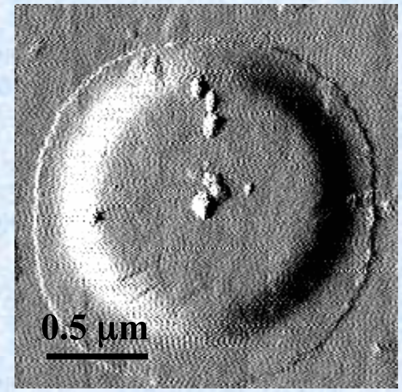
$1x1.5\ \mu m^2$



$2.5x2.5\ \mu m^2$

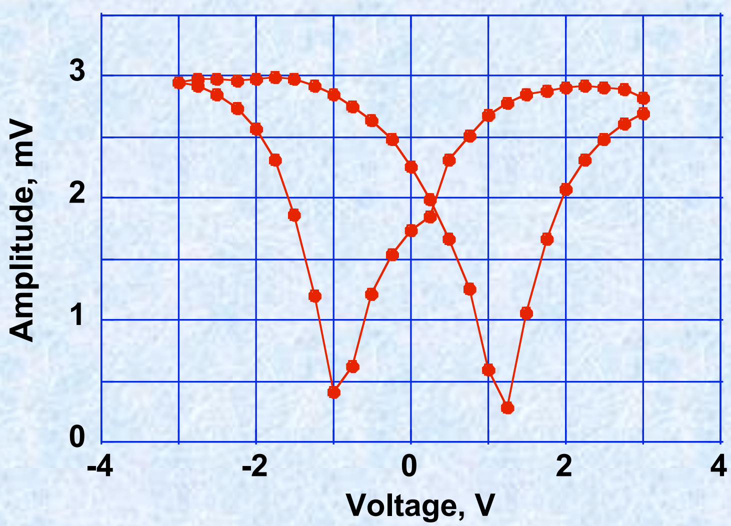


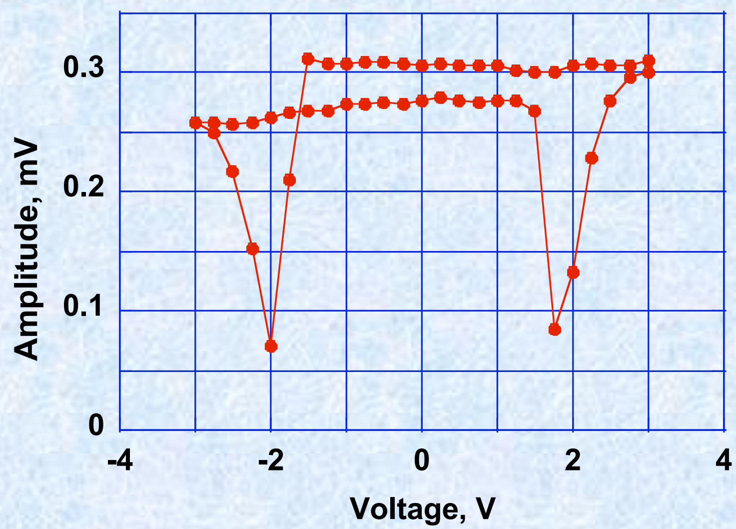
1.1 μm diameter



IrO_2(50nm)/PZT(180nm)/Pt(175nm)/Ti/SiO2/Si

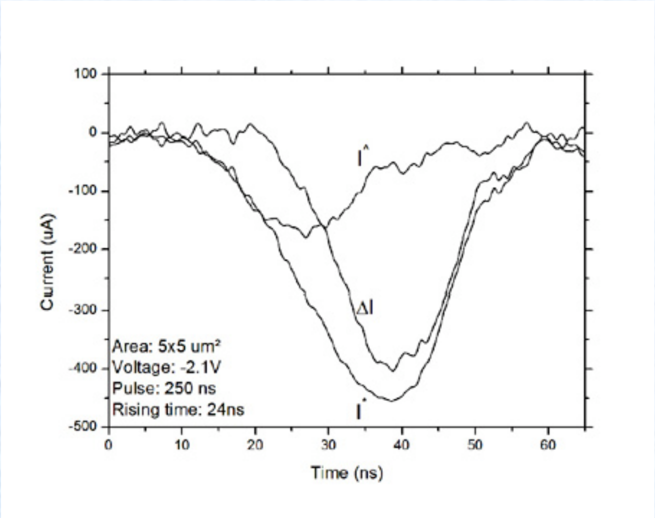
IrO_2(50 nm)/PZT(50 nm)/SRO(20 nm)/STO



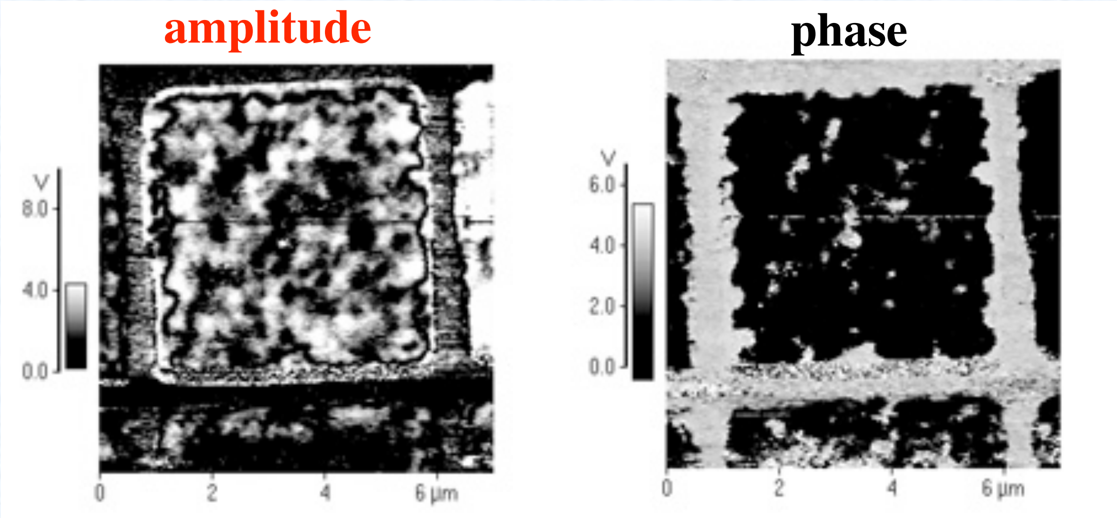


Comparison of PFM and Switching Current Data

Pulse: 2.1 V, 250 ns



5x5 μm² capacitor



I(t) data suggest *complete* switching of the capacitor

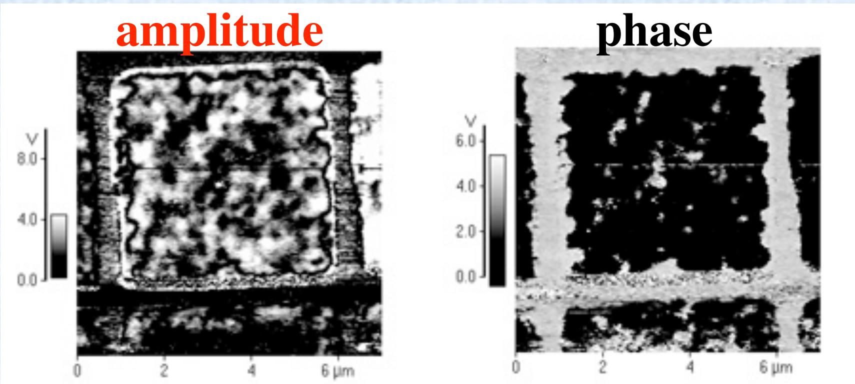
But...

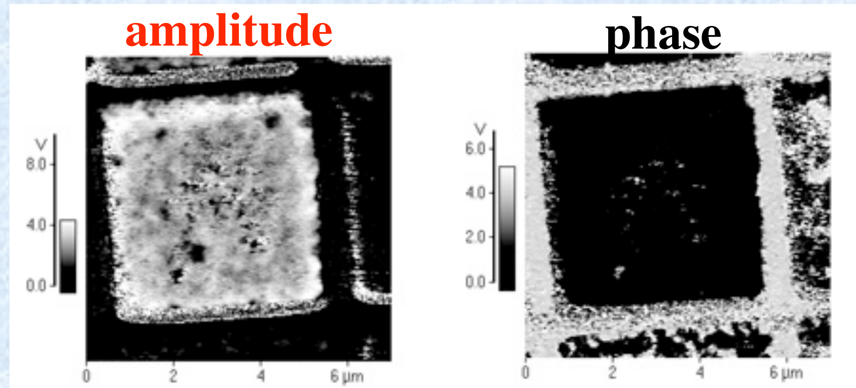
PFM images suggest *incomplete* switching

Pulse Parameters Effect on PFM Image

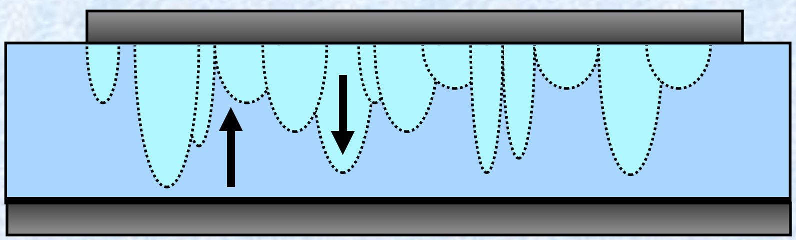
Pulse: 2.1 V, 250 ns **Pulse: 5 V, 1 s**

Piezoresponse images after pulse application

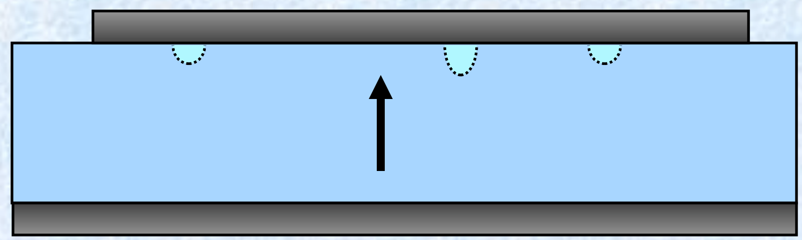




Schematics of domain pattern (side view)



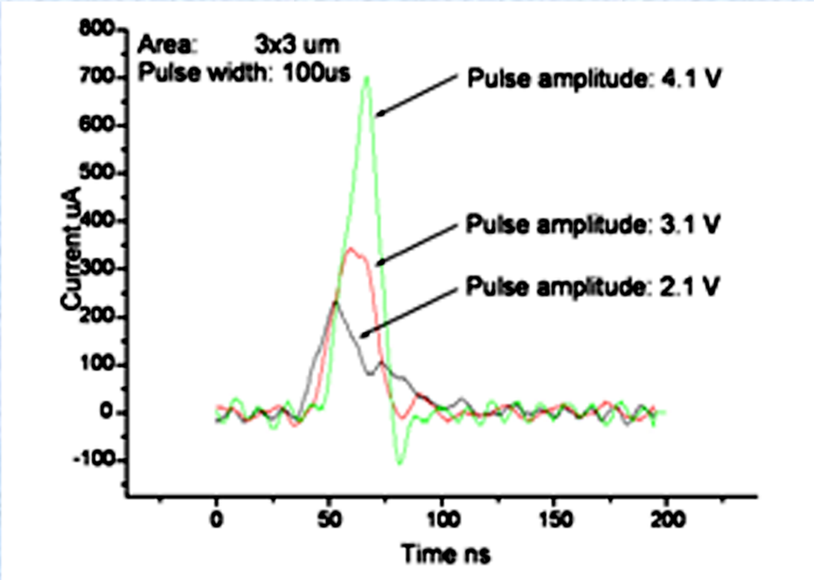
Partial switching



Complete switching

Note: Intermediate domain pattern corresponding to partial switching exists in a metastable state for a time period long enough for PFM imaging

PFM for Fast Domain Switching Dynamics



Switching times in FE caps: from 50 ns to 1 ms

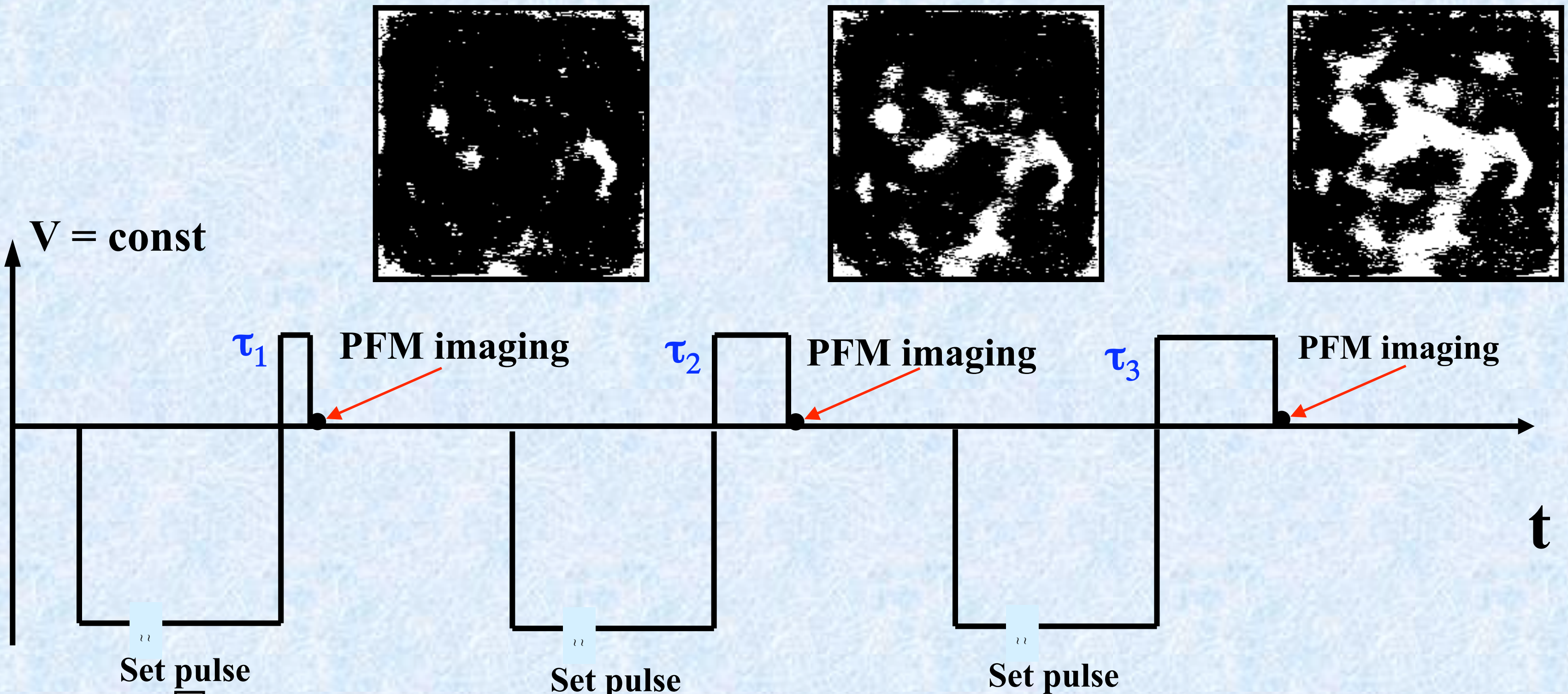
Acquisition of the PFM image: several minutes

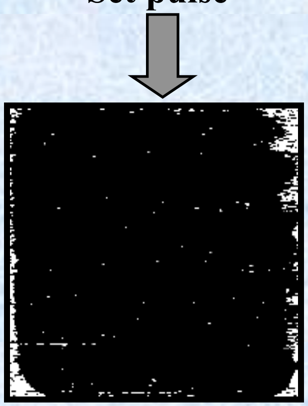
How can we use PFM to study fast switching processes?

STROBOSCOPIC PFM

PFM for Fast Domain Switching

Pulse train for stroboscopic PFM studies of domain kinetics:





Condition to be fulfilled:

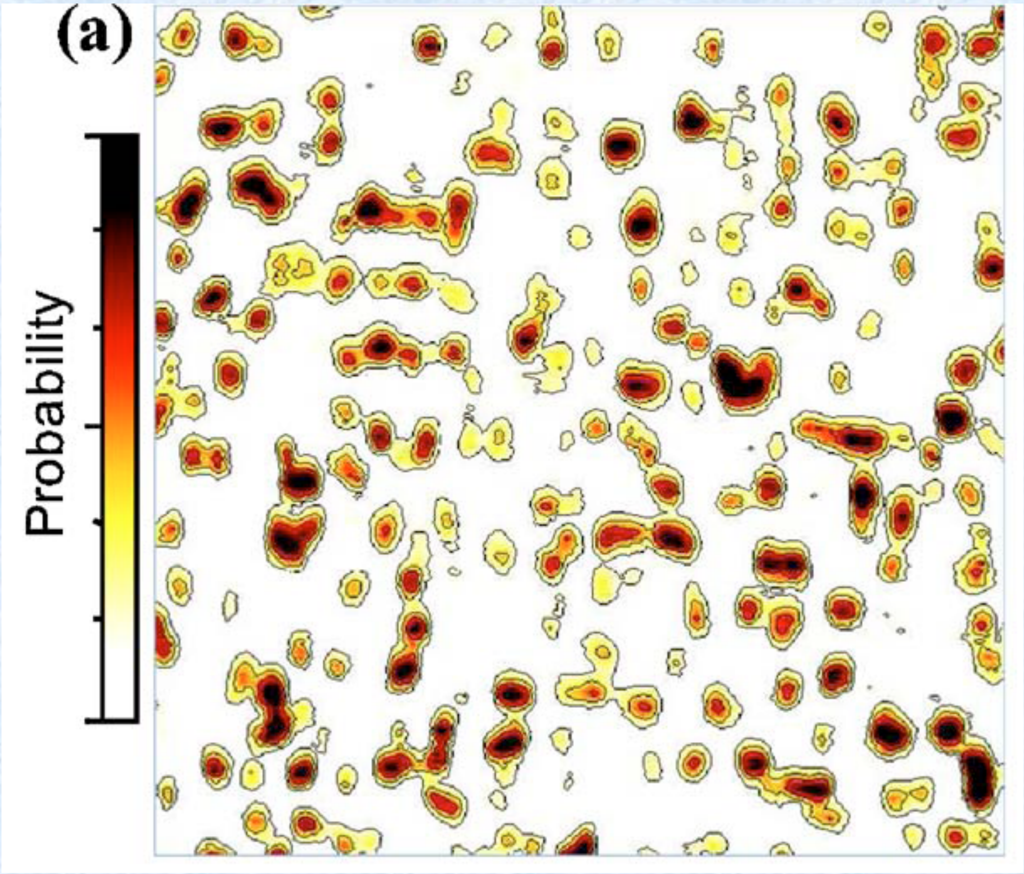
$$\tau_1 < \tau_2 < \tau_3 < \ldots . < t_s$$

Time resolution depends on:

- **Pulse rise time**
- **First pulse duration**
- **Pulse length increment**

C.Dehoff et al, Rev. Sci. Instr. (2005).

Heterogeneous Nucleation in Capacitors



The nucleation sites in capacitors can be visualized directly.

D. J. Kim et al, Appl. Phys. Lett. (2007)

Domain Switching in FeRAM Capacitors

Shortest pulse: 10 ns

Voltage range: <2.0 V

Coercive voltage: 1.1 V

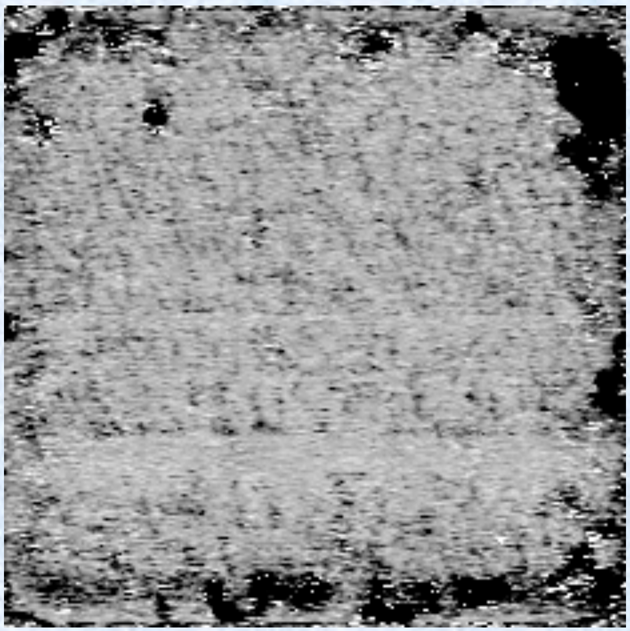
Direct measurements of:

Nucleation rate

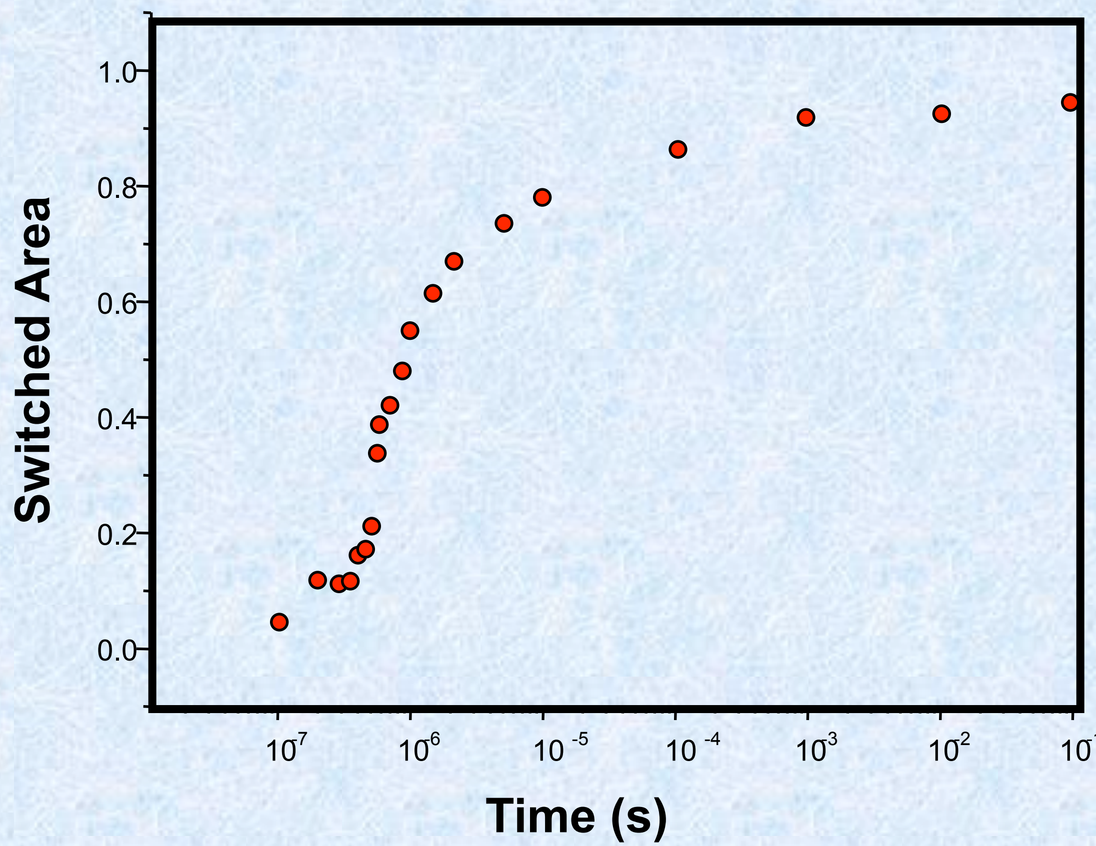
Forward growth velocity

Lateral growth velocity

5x5 μm²

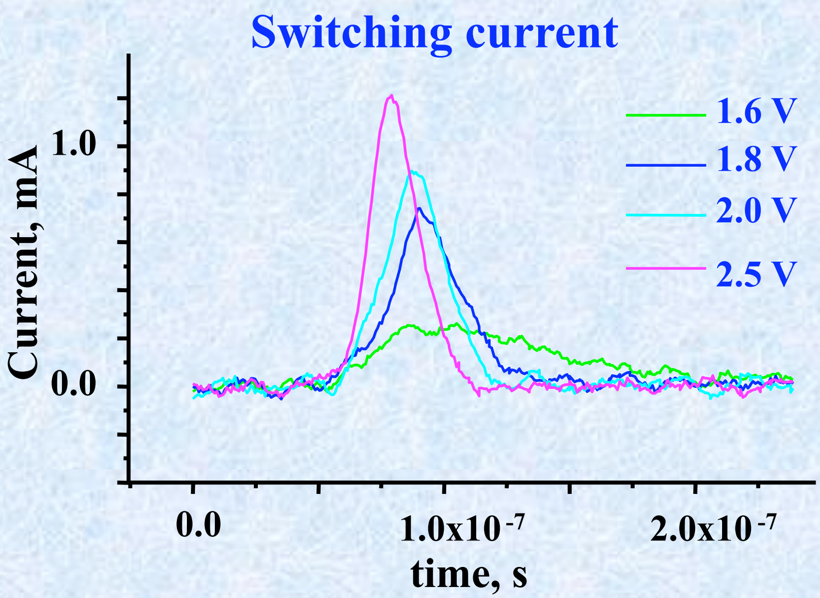


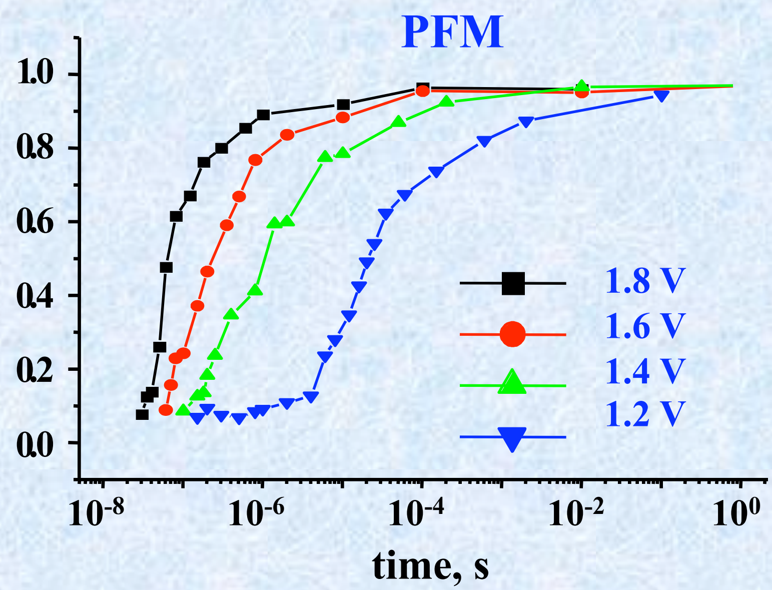
1.6V (~89 kV/cm)

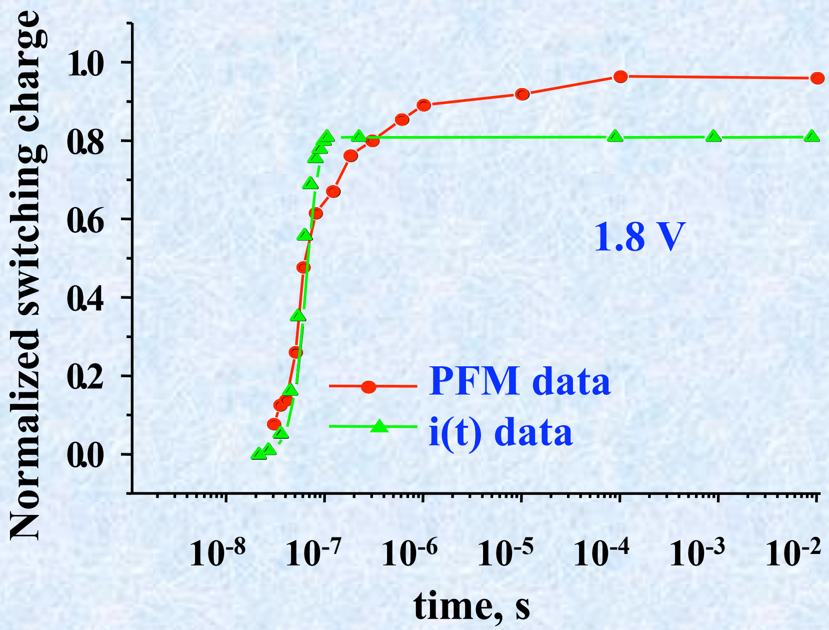


Note switching time variations within the capacitor.

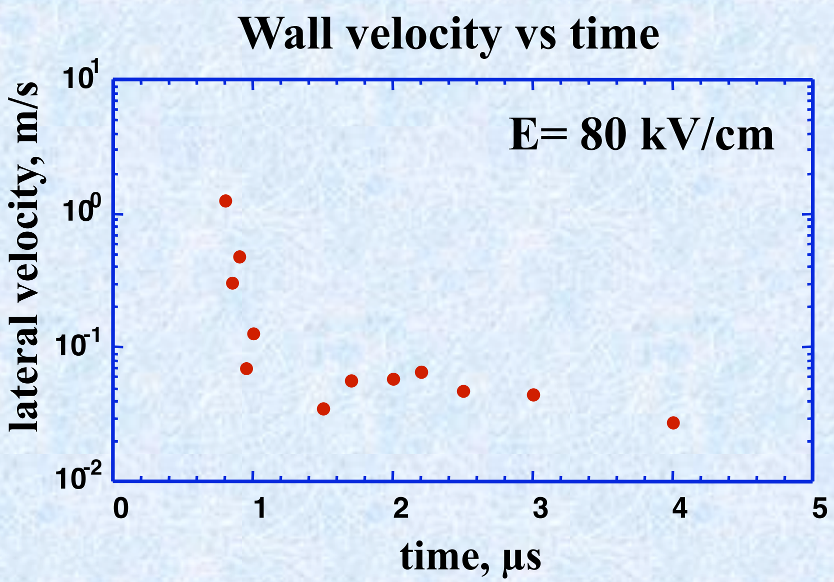
PFM vs Switching Current



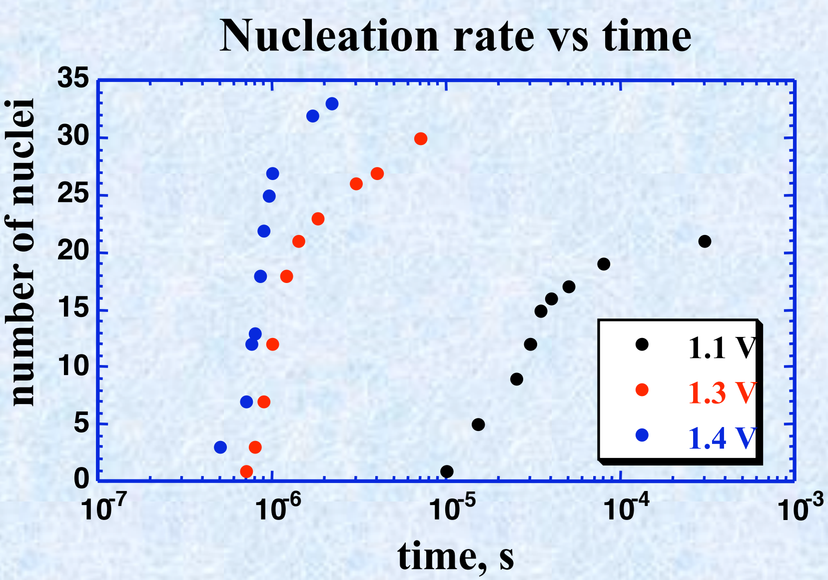


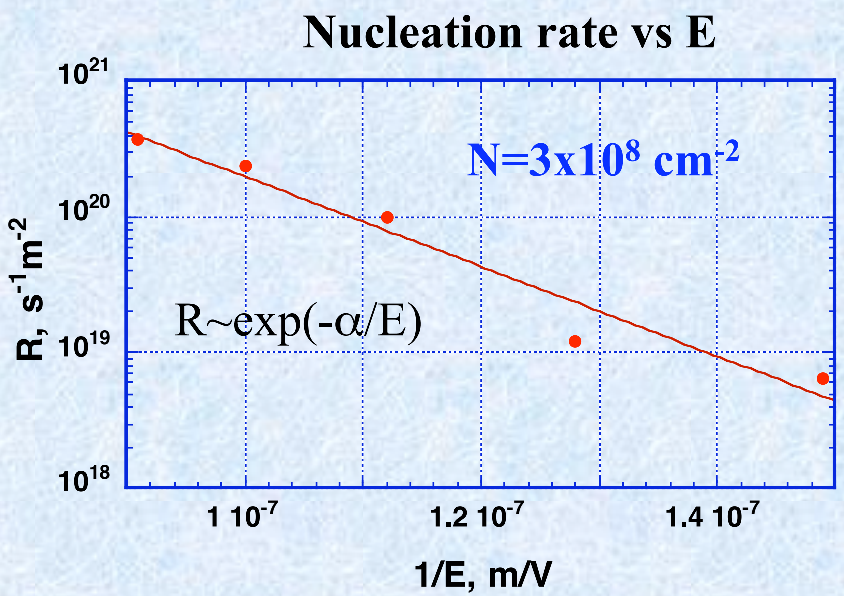


Parameters of Domain Switching

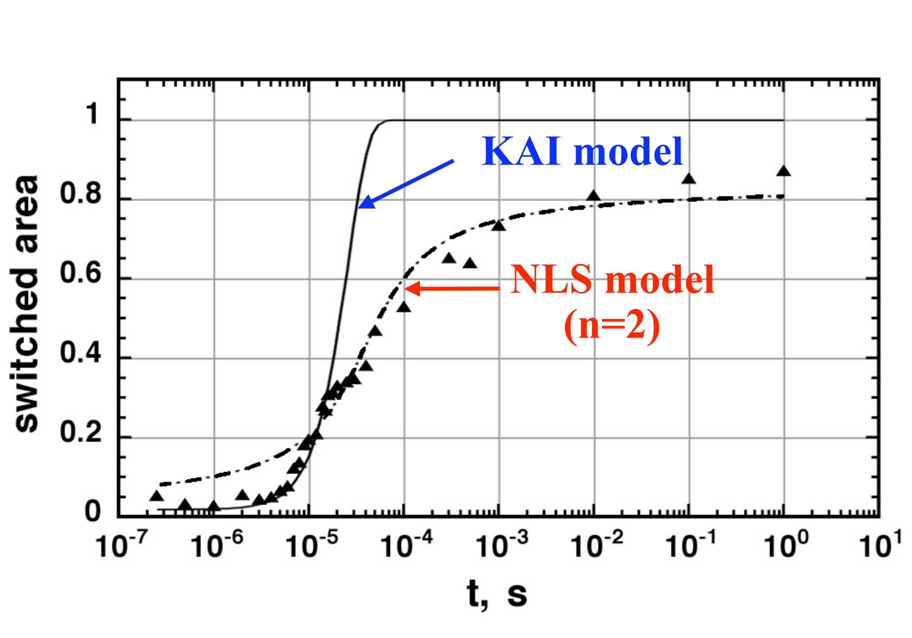


- Wall velocity varies by two orders of magnitude during switching
- Velocity decreases before domain merging with nearby domains
- Area fraction switched via nucleation increases with voltage





Fitting Domain Switching Dynamics



NLS model: broad distribution of local switching times

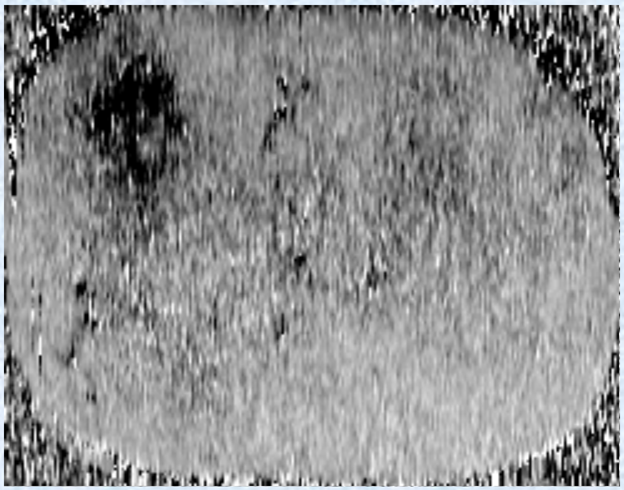
$$\Delta p(t) = 1 - \sum F(\log t_0) e^{-(t/t_0)^n}$$

Experimentally observed domain dynamics differs from the basic assumptions of the KAI model

Domain Switching in Small Capacitors

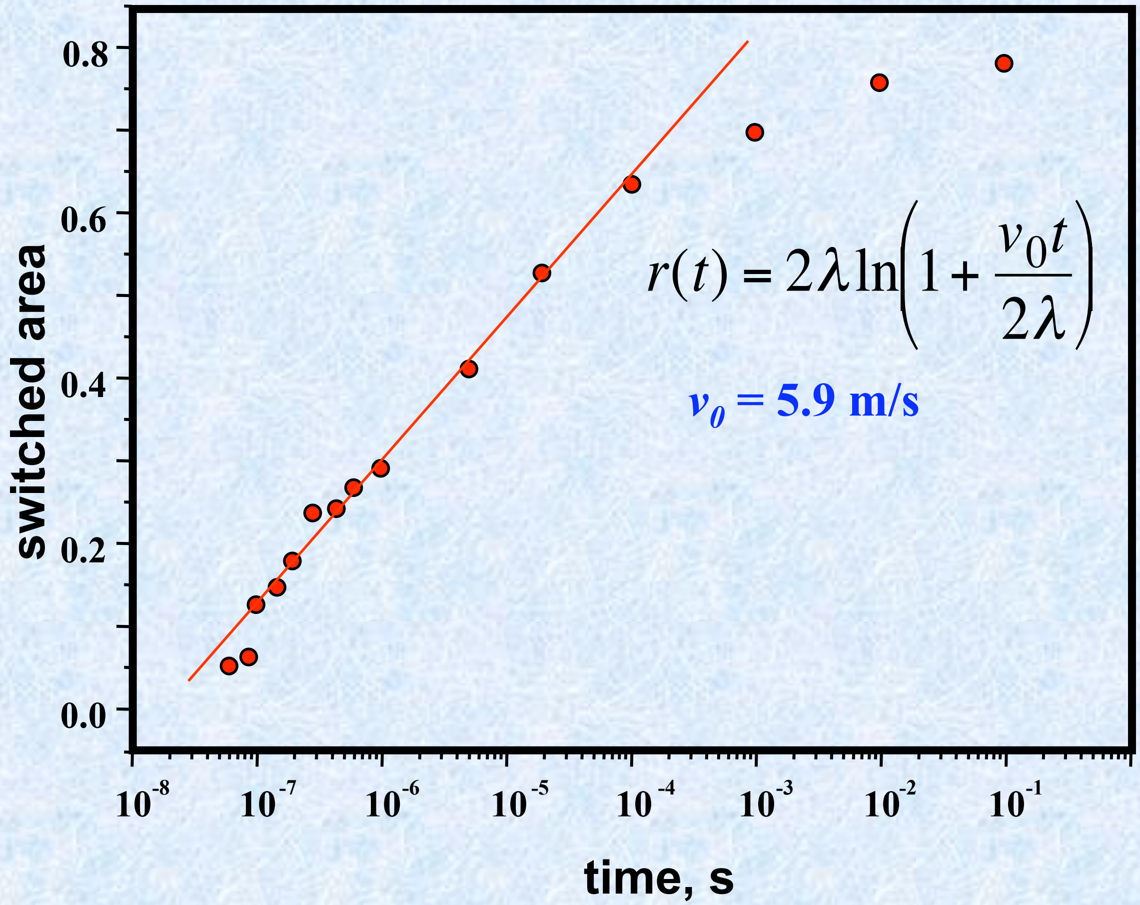
Capacitor scaling: change in the switching mechanism

$1\times1.5\ \mu m^2$



1.6V (1.45 V_c)

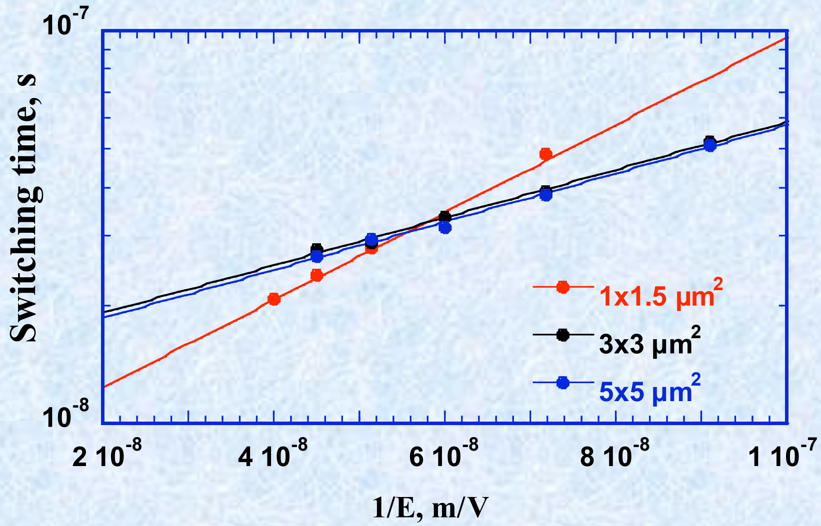
- **Nucleation is reduced**
- **Switching is via domain wall**



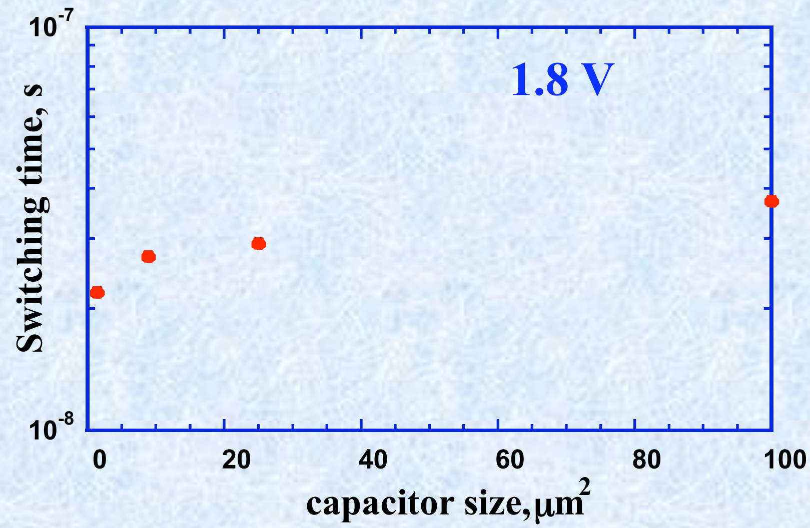
M. Dawber et al, APL 82, 436 (2003)

Capacitor Size Effect

Switching time vs E



Switching time vs capacitor size



5x5 μm^2 capacitor: **α=1.4 · 10^5 V/cm**

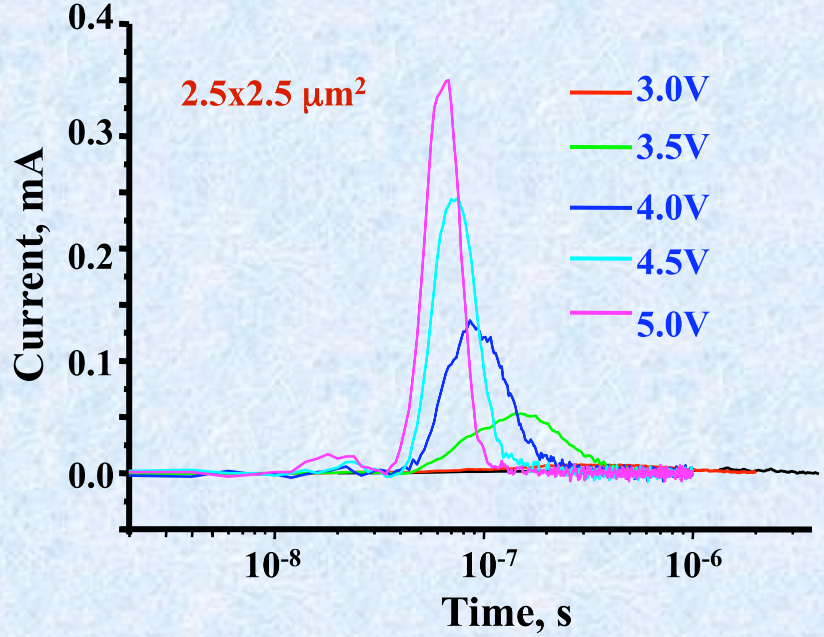
3x3 μm^2 capacitor: **α=1.4 · 10^5 V/cm**

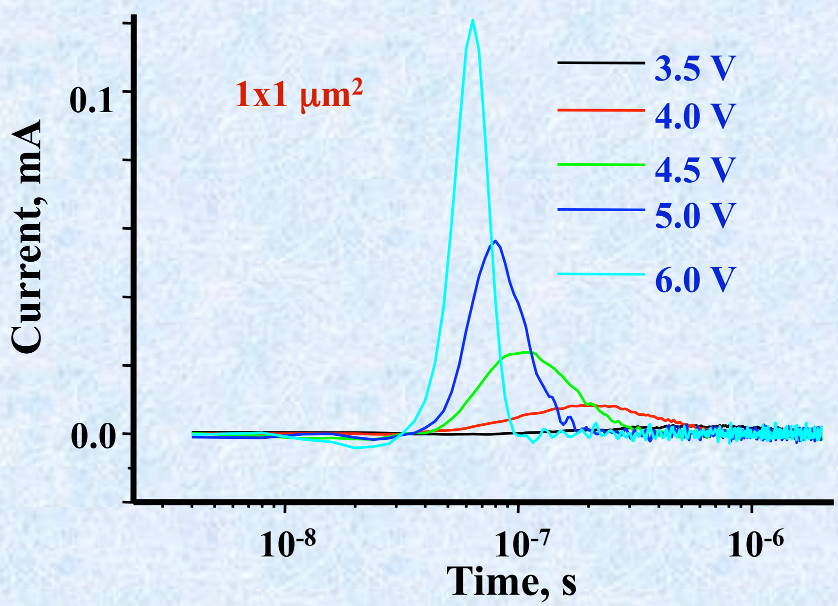
1 x1.5 μm^2 capacitor: **α=2.6 · 10^5 V/cm**

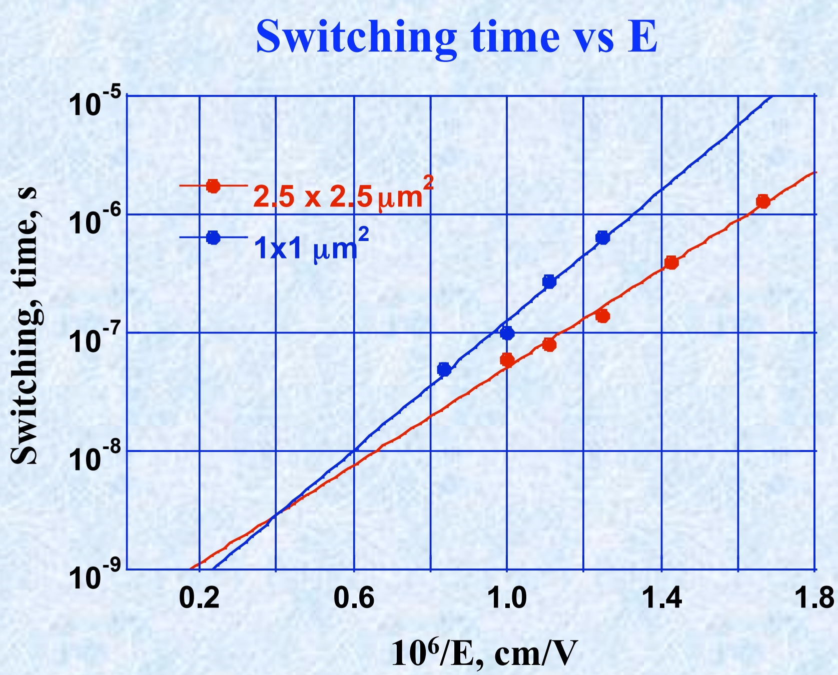
Capacitor scaling:

- Decrease in a number of nuclei
- Larger contribution of wall motion to polarization reversal with decrease in capacitor size
- Both NLS and KAI model fail to describe the switching kinetics in small capacitors

Switching Behavior of Epitaxial Capacitors







$$t_{sw} = t_0 \exp(\alpha / E)$$

2.5x2.5 μm² capacitor: **α=4.8 · 10⁶ V/cm**

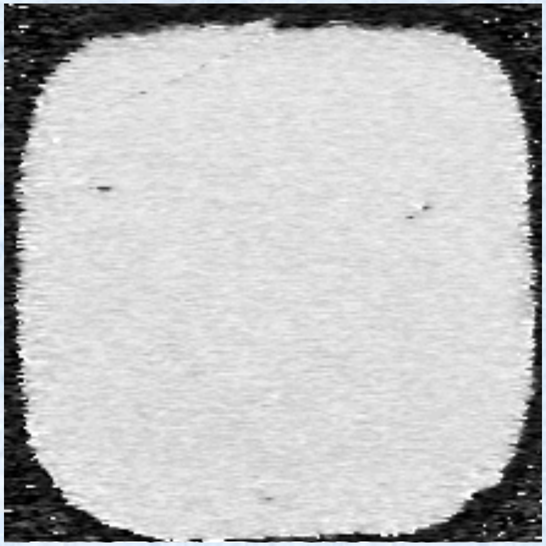
1x1 μm² capacitor: **α=6.3 · 10⁶ V/cm**

Domain Switching in Epitaxial Capacitors

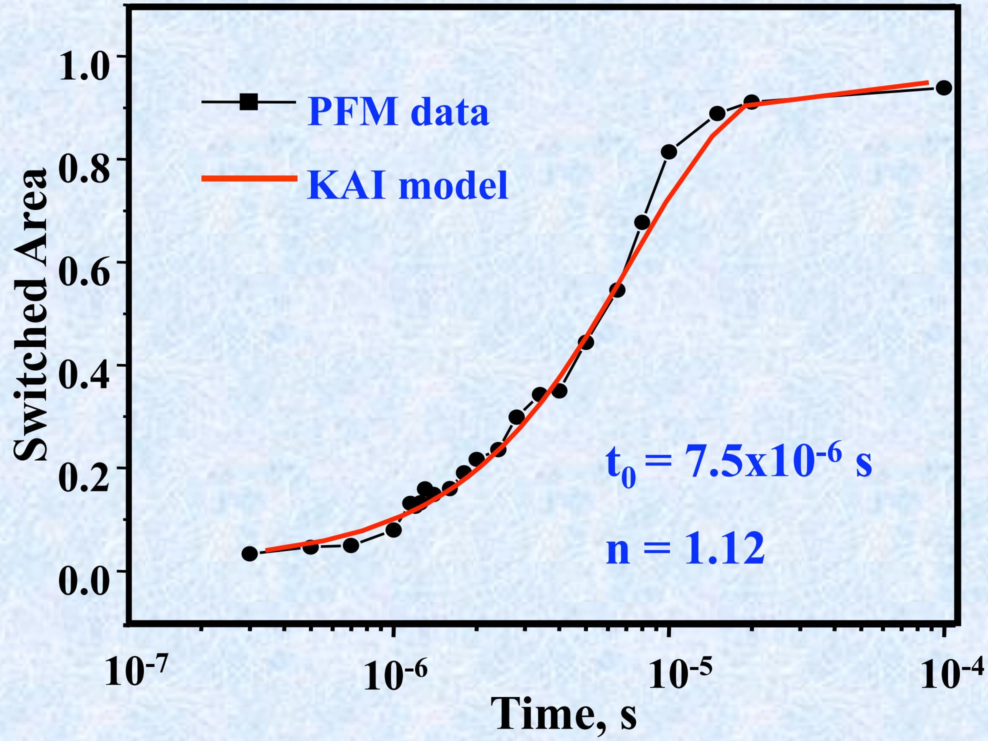
Coercive voltage: 2.0 V

P_s= 90 μC/cm²

2.5x2.5 μm²



2.5V (1.38 V_c)



- Smaller number of nucleation sites due to the improved film/electrode interface ($N=8\times10^7$ cm^{-2})
- Strong anisotropy of wall velocity

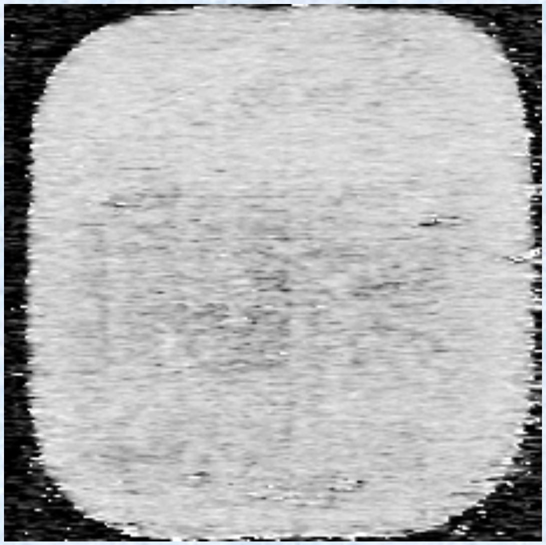
Sample courtesy M. Alexe

Domain Switching in Epitaxial Capacitors

Coercive voltage: 2.0 V

P_s= 90 μC/cm^2

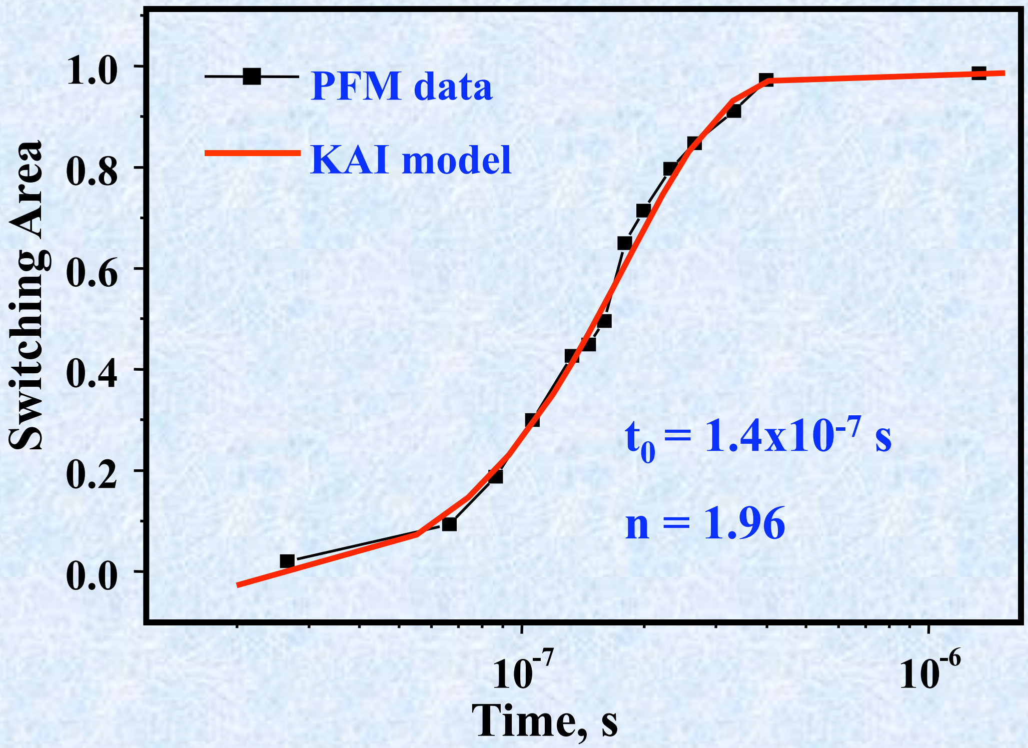
2.5x2.5 μm^2



3.5V (1.9 V_c)

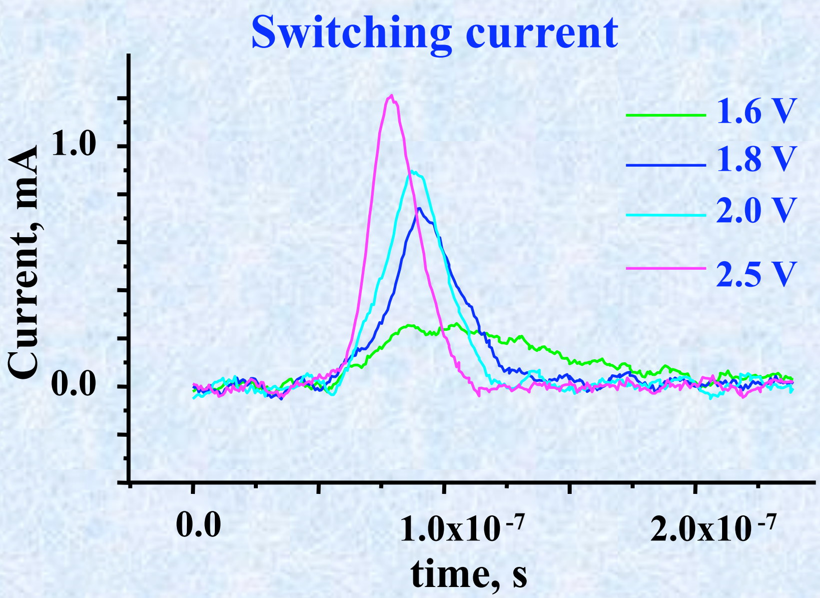
Nucleation density

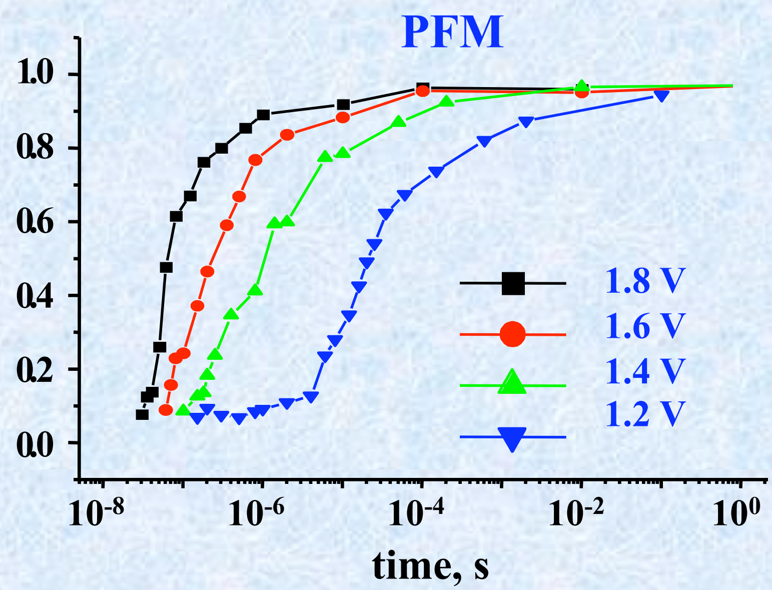
N=7x10^8 cm^{-2}

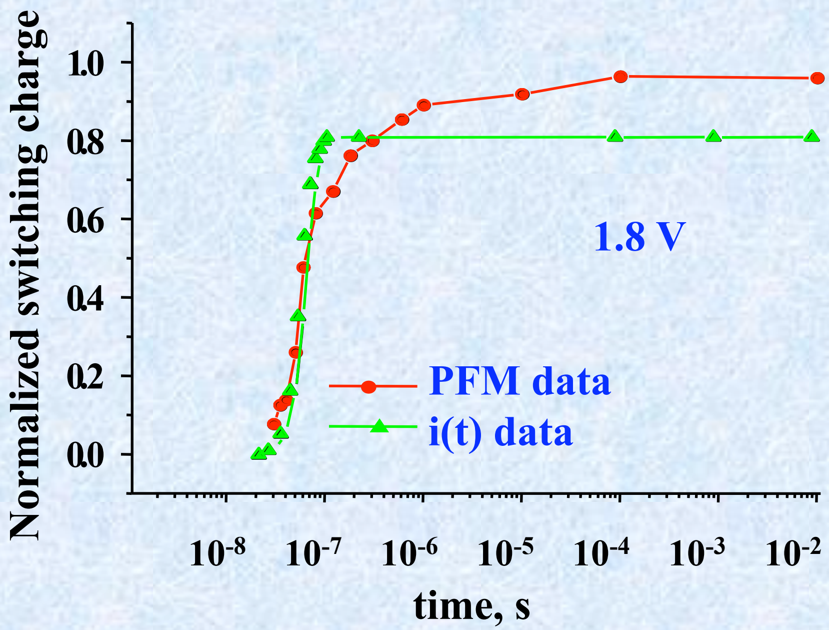


Sample courtesy M. Alexe

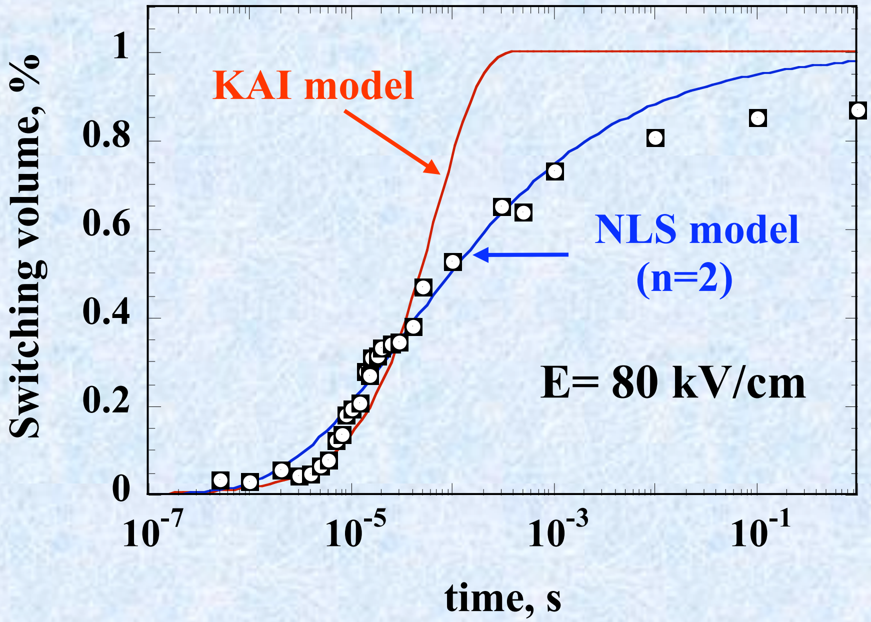
PFM vs Switching Current







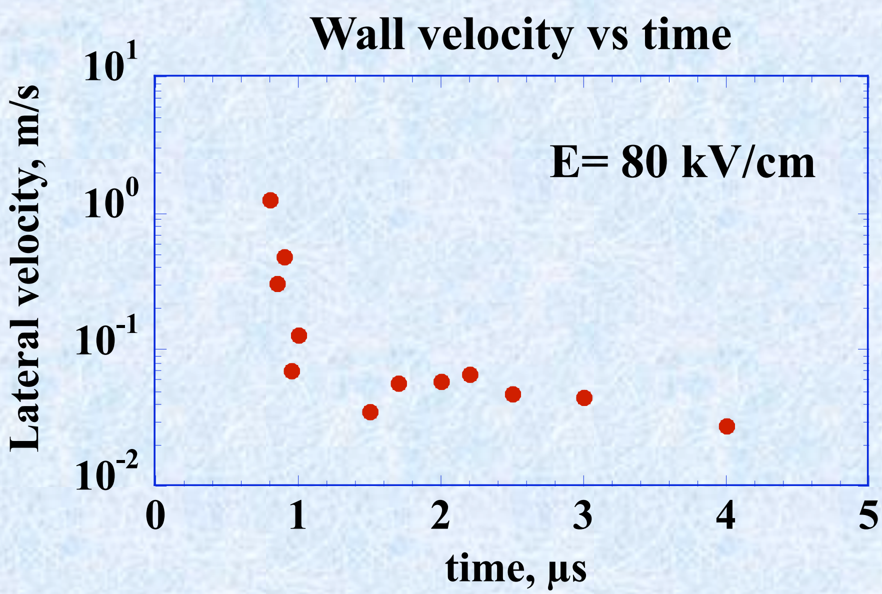
Fitting Domain Switching Dynamics



- Polycrystalline PZT films: broad distribution of local switching times

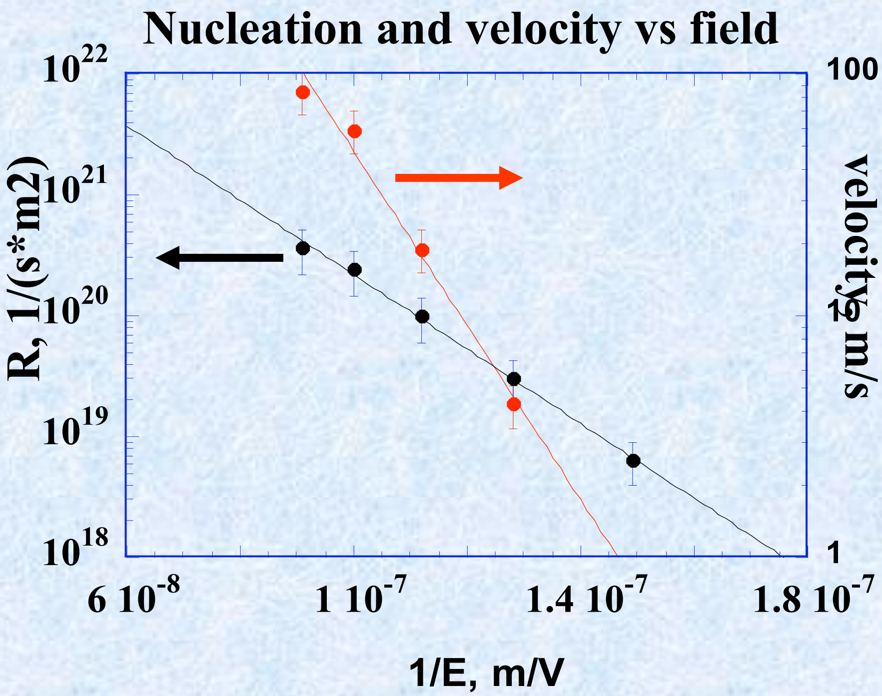
$$\Delta p(t) = 1 - \sum F(\log t_0) e^{-(t/t_0)^n}$$

Experimentally observed domain dynamics differs from the basic assumptions of the KAI model



- Wall velocity varies significantly during switching
- Area fraction switched via nucleation increases with voltage

Parameters of Domain Switching Kinetics



Two main mechanisms of switching in large capacitors:

- **nucleation** $\alpha_n = 7.1 \cdot 10^7$ **V/cm**
- **wall motion** $\alpha_w = 8.3 \cdot 10^7$ **V/cm**

- Nucleation rate

$$R = R_\infty \exp(-\alpha_n / E)$$

$\alpha_n = 7.1 \cdot 10^7$ **V/cm**

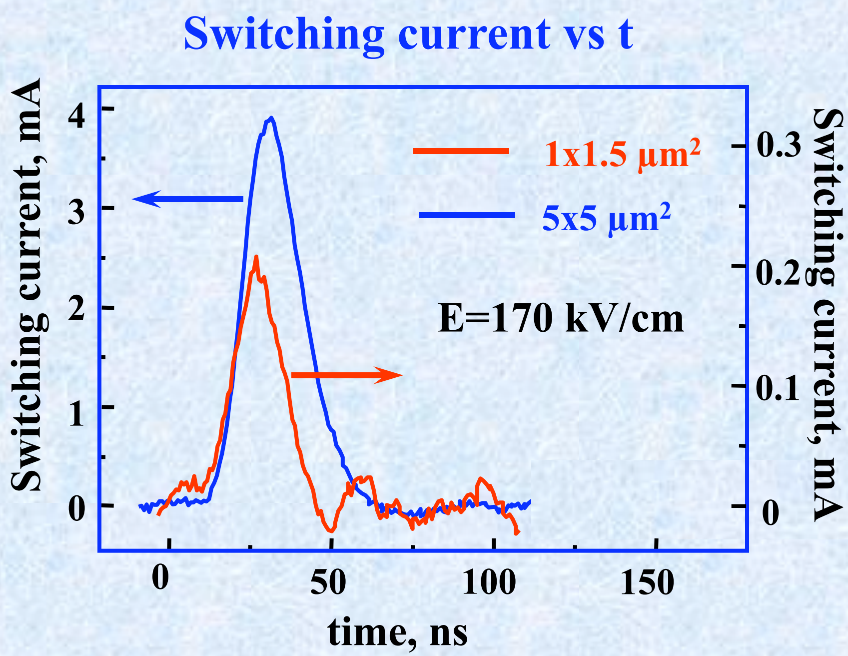
- Wall velocity

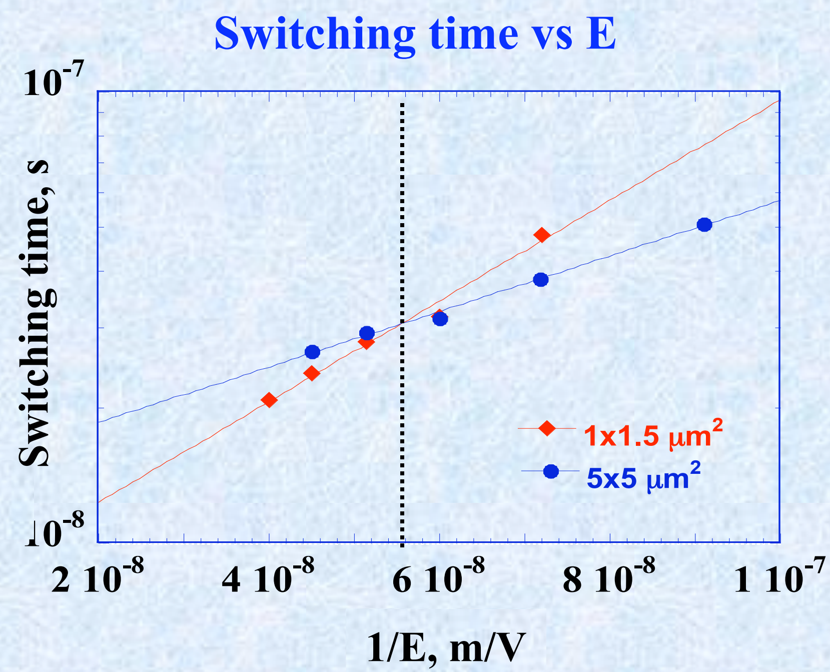
$$v = v_\infty \exp(-\alpha_w / E)$$

$\alpha_w = 8.3 \cdot 10^7$ **V/cm**

A difference between α_n and α_w activation fields implies transition from wall-limited to nucleation limited switching: in high fields, nucleation becomes a rate-limiting parameter.

Capacitor Scaling Effect on Switching





Small capacitors switch faster than large capacitors at high fields (> 150 kV/cm) but more slowly at low fields.

5x5 μm² capacitor: **α=1.4 · 10⁵ V/cm**

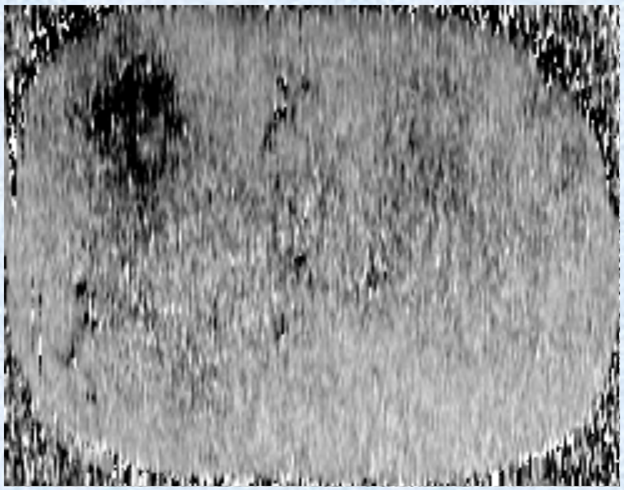
3x3 μm² capacitor: **α=1.4 · 10⁵ V/cm**

1 x1.5 μm² capacitor: **α=2.6 · 10⁵ V/cm**

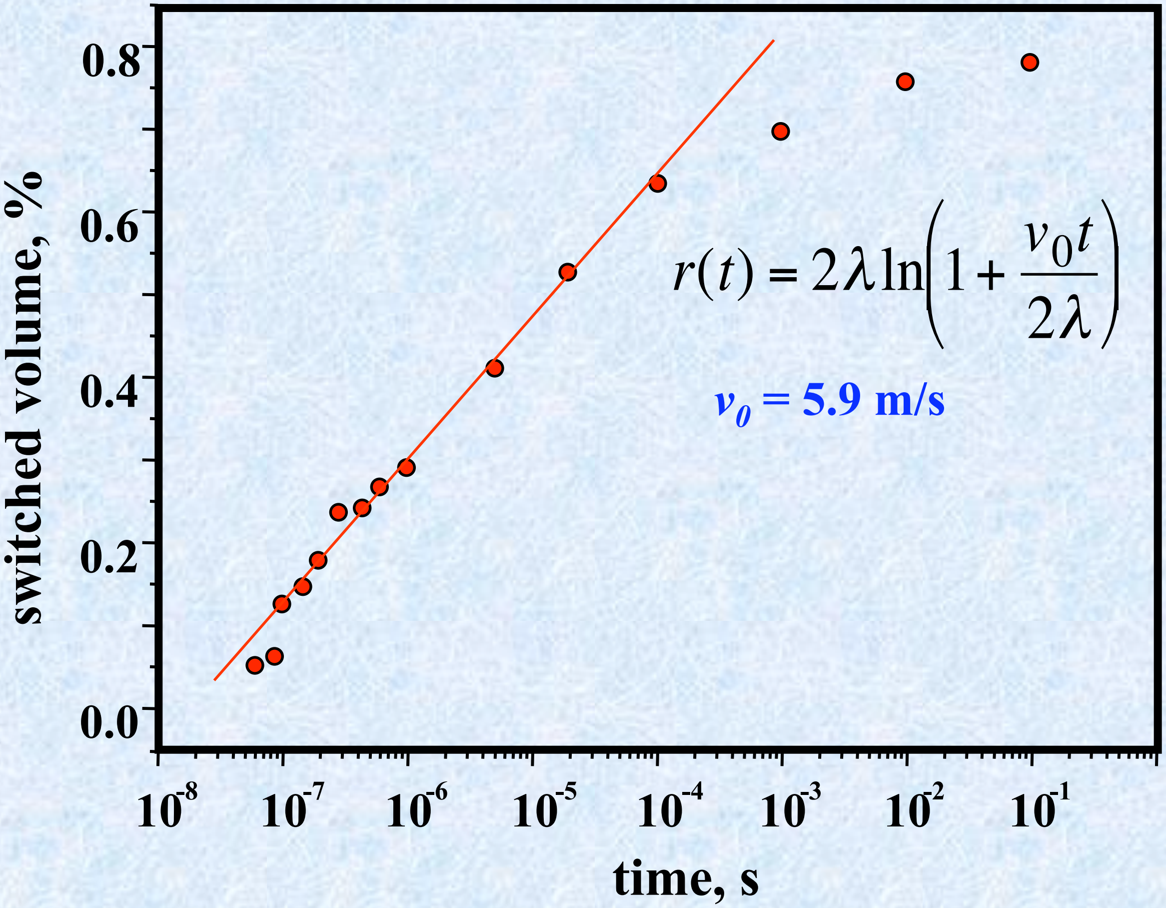
Domain Switching in Small Capacitors

Capacitor scaling: change in the switching mechanism

$1x1.5\ \mu m^2$



1.6V (~89 kV/cm)

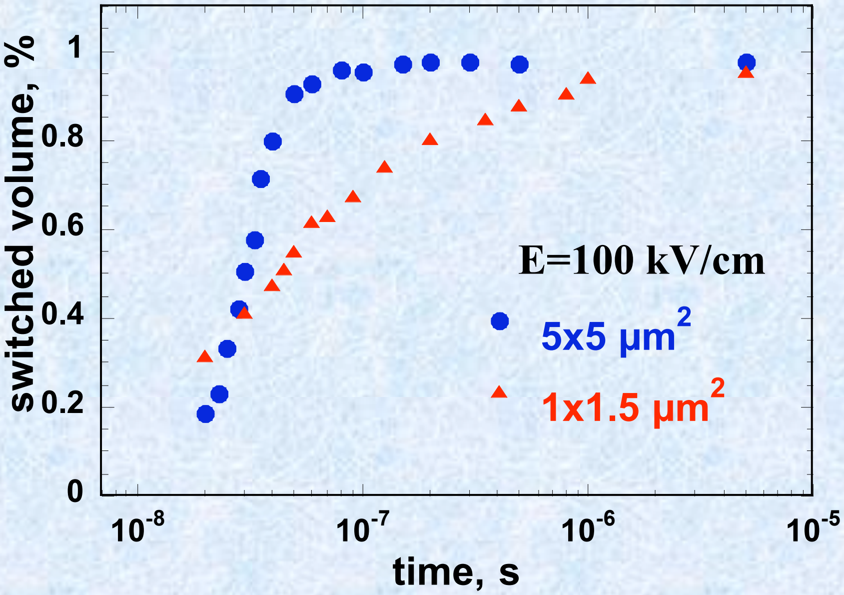


Capacitor scaling:

- **Nucleation is reduced**
- **Switching is via domain wall**
- **Both NLS and KAI are not applicable**

M. Dawber et al, APL 82, 436 (2003)

Capacitor Scaling Effect on Switching Mechanism

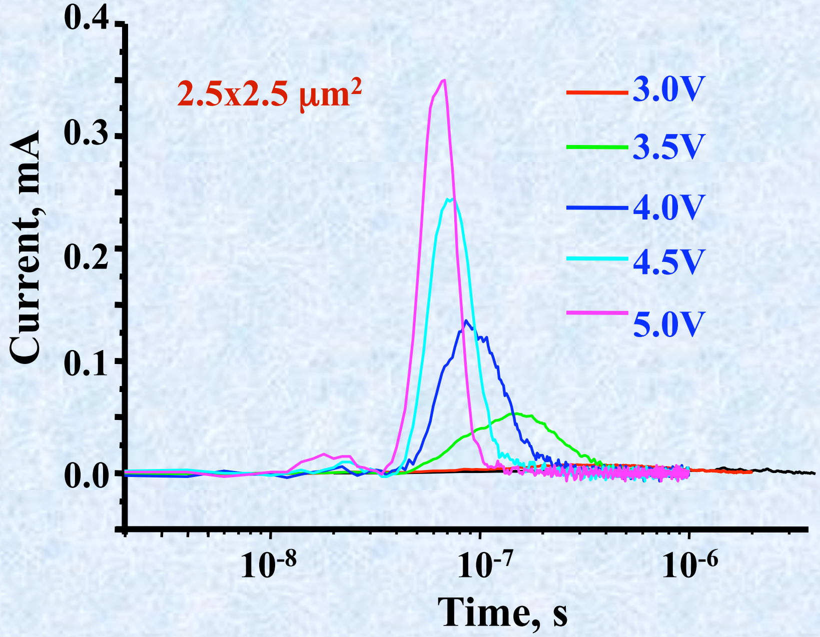


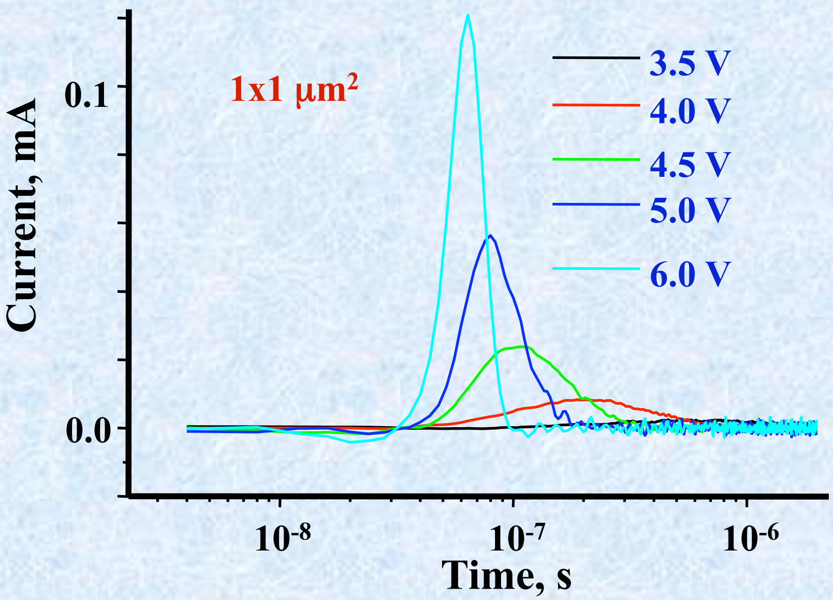
- Low fields (<150 kV/cm): wall motion is a rate-limiting mechanism (small capacitors switch more slowly).
- High fields (>150 kV/cm): nucleation is a rate-limiting mechanism (large capacitors switch more slowly)

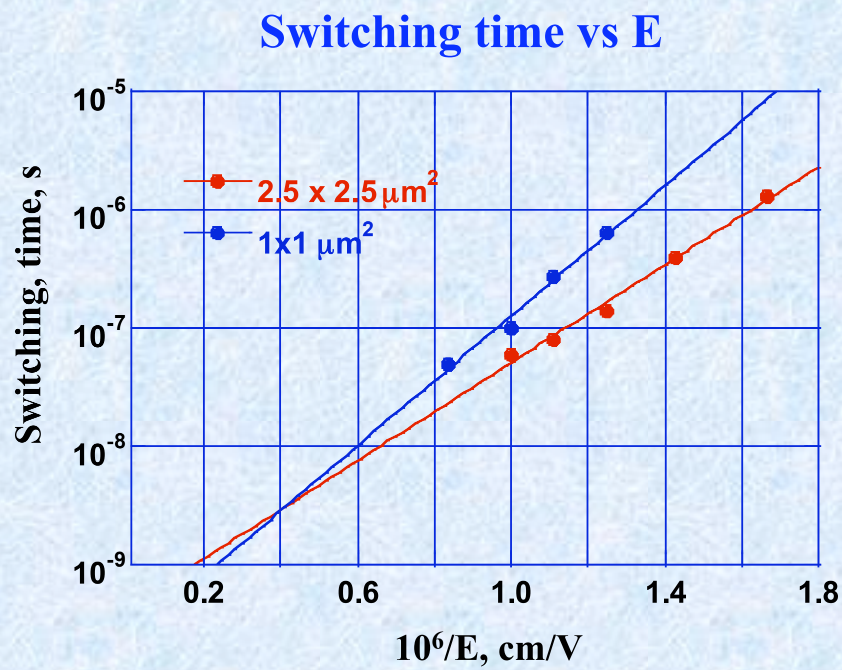
Criterion for capacitor size effect on rate-limiting mechanism:

$$\frac{v_\infty}{NR_\infty}\exp\left(\frac{\alpha_n - \alpha_v}{E}\right) << A^{3/2}$$

Switching Behavior of Epitaxial Capacitors







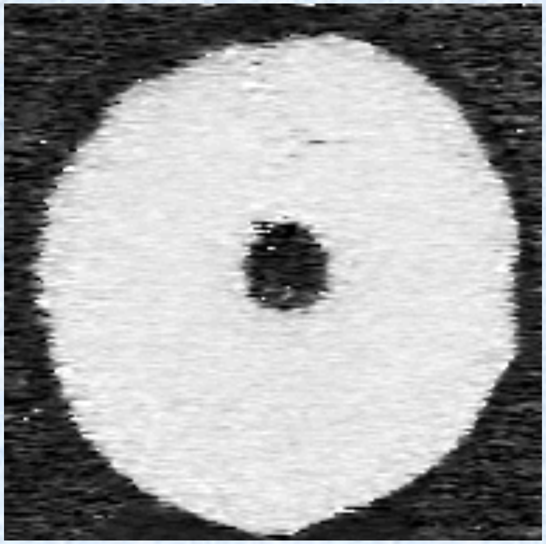
$$t_{sw} = t_0 \exp(\alpha / E)$$

2.5x2.5 μm² capacitor: **$\alpha = 4.8 \cdot 10^6$ V/cm**

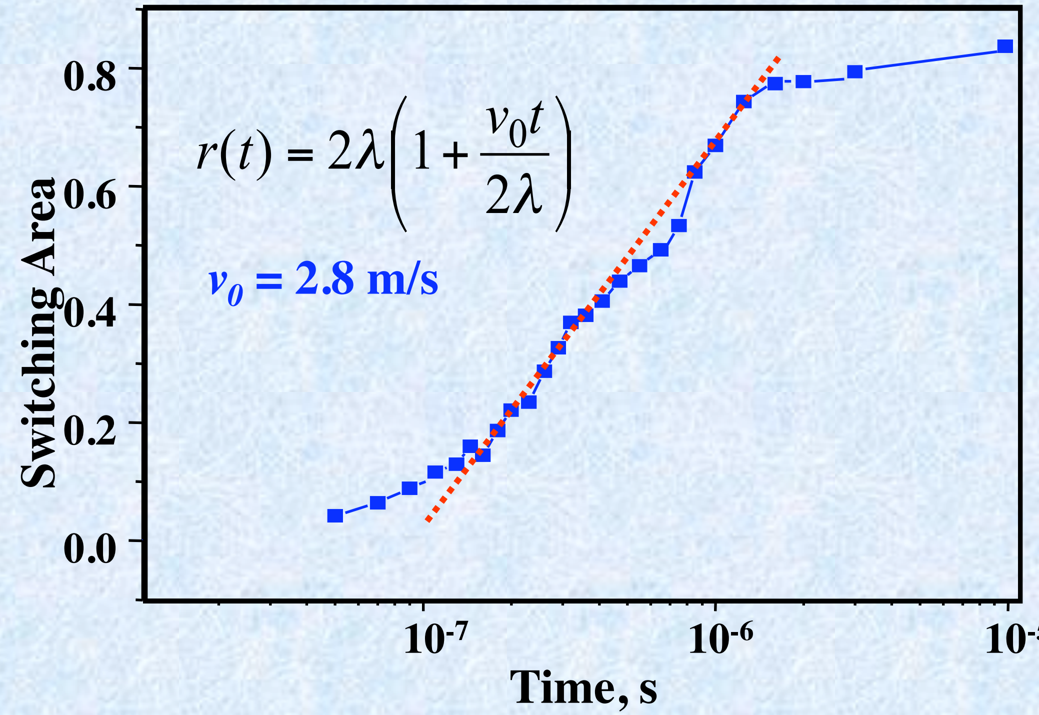
1x1 μm² capacitor: **$\alpha = 6.3 \cdot 10^6$ V/cm**

Domain Kinetics in Small Epitaxial Capacitors

1.0 μm



3.5V (1.9 V_c)



- **Strong perimeter effect**
- **Reduced nucleation**
- **Pinned region in the center**
- **Dynamic vortex domain?**

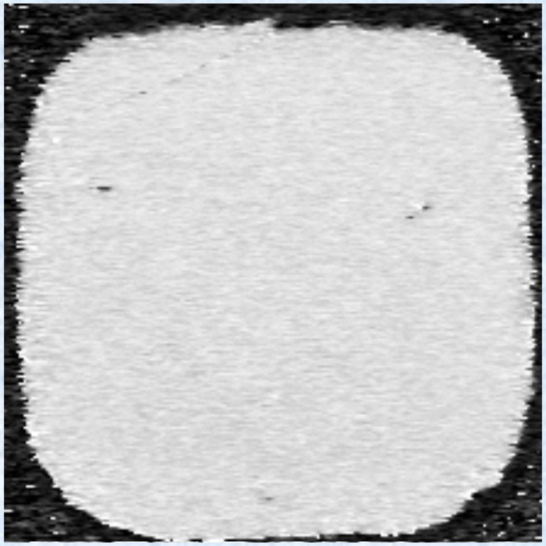
M. Dawber et al, APL **82**, 436 (2003)

Domain Kinetics in Epitaxial Capacitors

Coercive voltage: 2.0 V

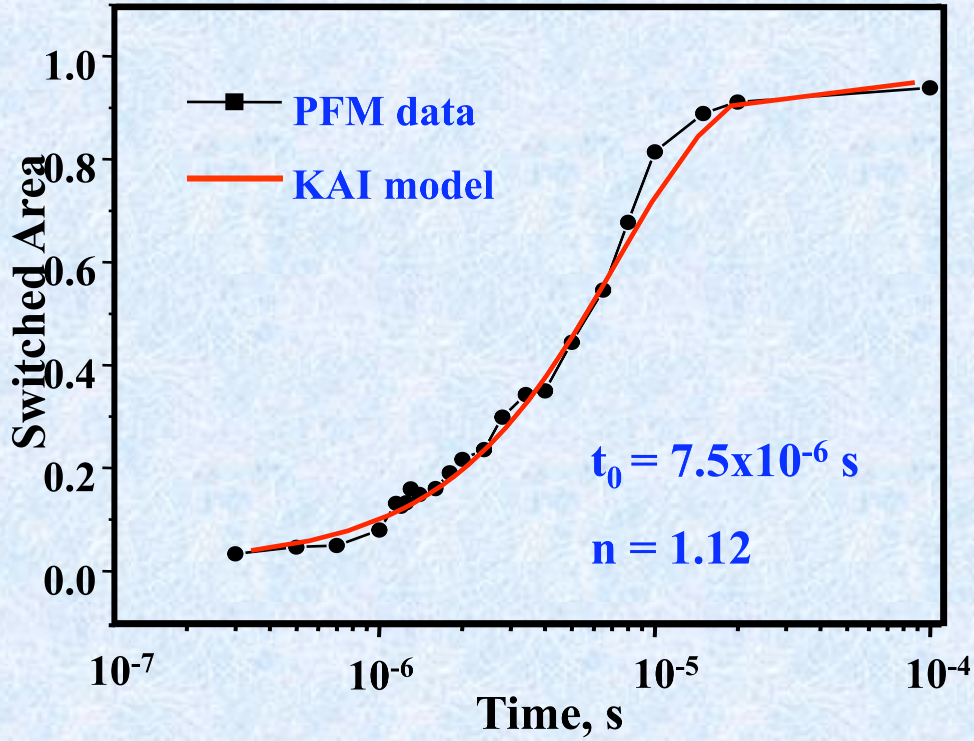
P_s= 90 μC/cm^2

2.5x2.5 μm^2



2.5V (1.38 V_c)

Kinetics of switching in low fields



- Smaller number of nucleation sites due to the improved film/electrode interface (N=8^{x}10^7 cm^{-2})
- Strong anisotropy of wall velocity

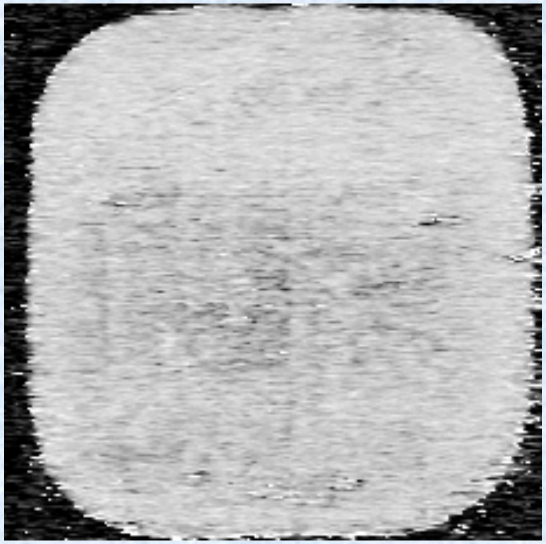
Sample courtesy M. Alexe

Domain Kinetics in Epitaxial Capacitors

Coercive voltage: 2.0 V

P_s= 90 μC/cm^2

2.5x2.5 μm^2

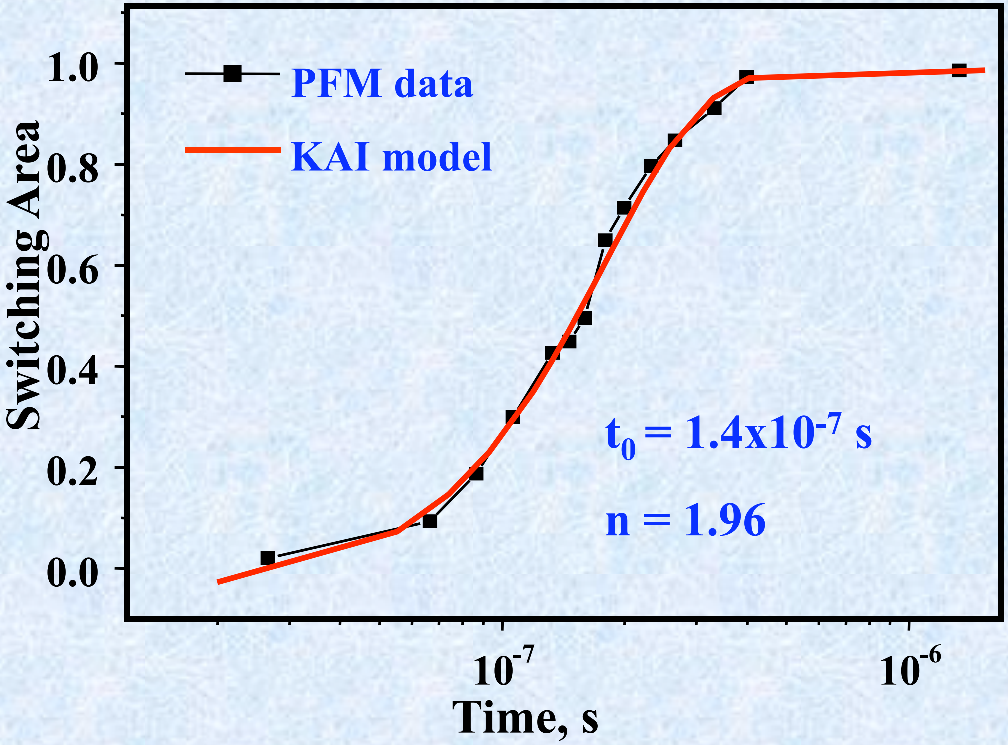


3.5V (1.9 V_c)

Nucleation density

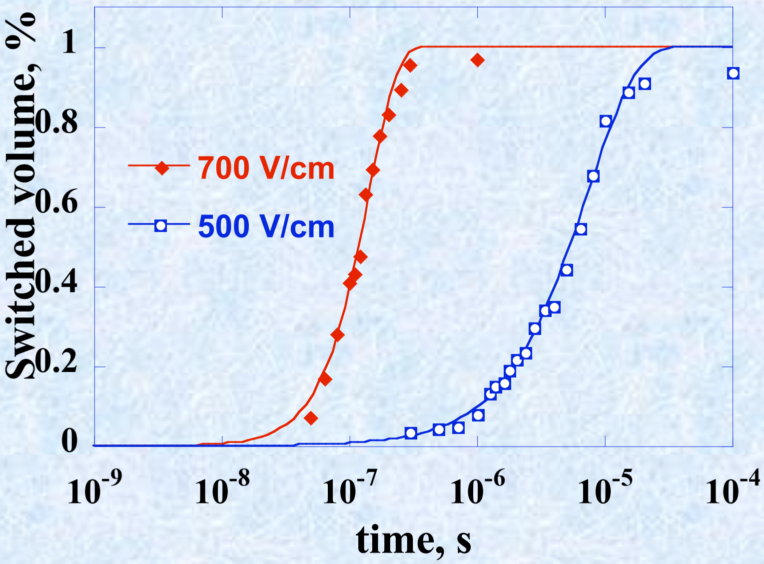
N=7^{x}10^8 cm^{-2}

Kinetics of switching in high fields

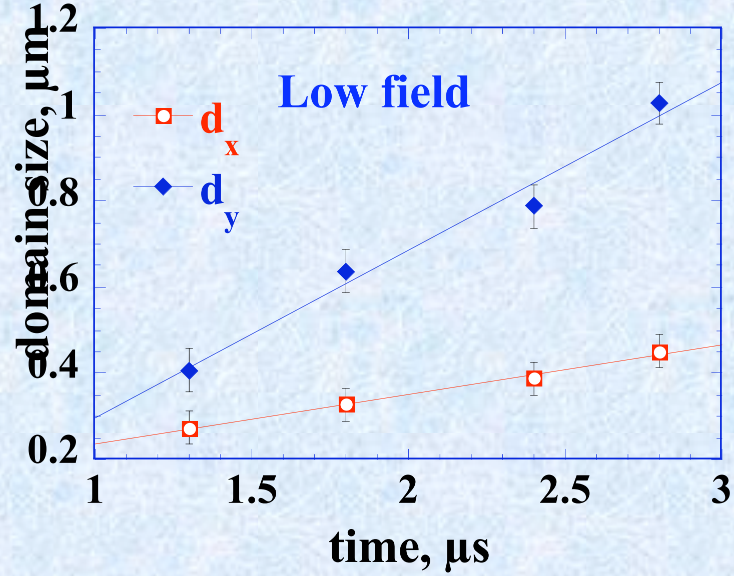


Sample courtesy M. Alexe

Fitting Domain Switching Dynamics



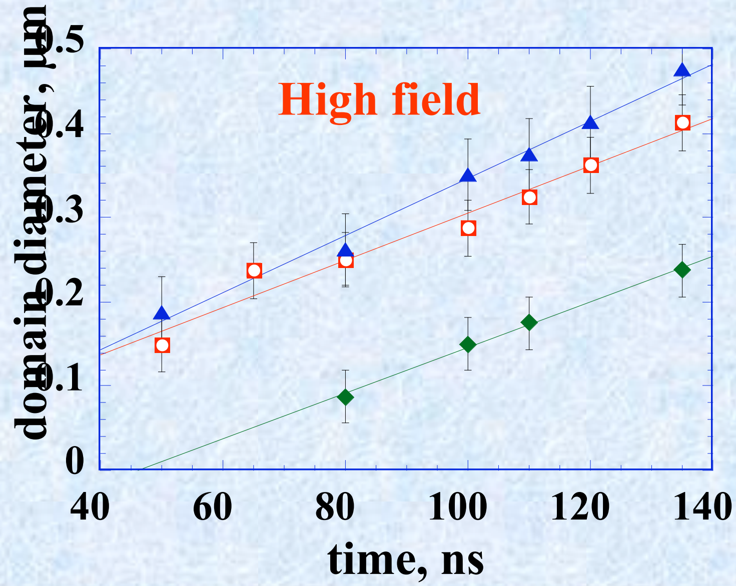
Domain size vs time



- Epitaxial PZT films: KAI model provides adequate fitting

$$f(t) = 1 - \exp[-(t/t_0)^n]$$

- Field-induced change in the switching mechanism is reflected in domain dimensionality change:

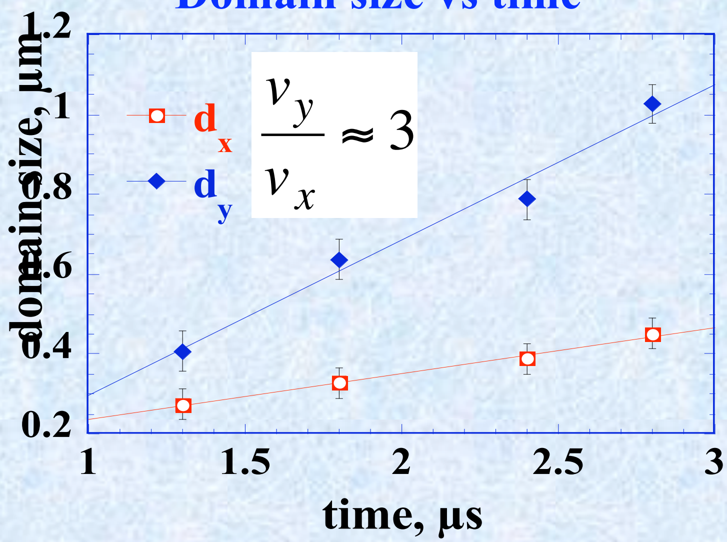


High fields: n = 1.96

Low fields: n = 1.12

Anisotropy of Domain Wall Motion

Domain size vs time



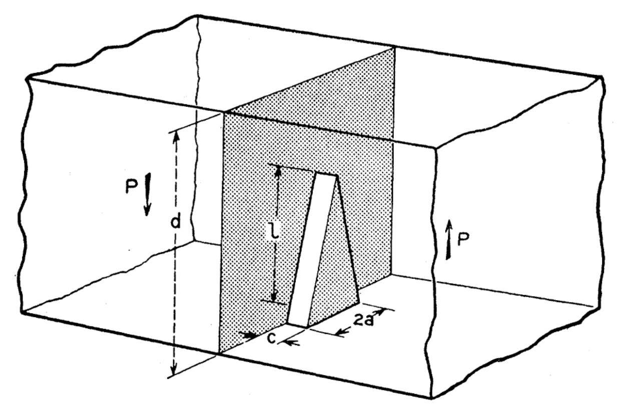


FIG. 1. Schematic drawing of a triangular step on a 180° domain wall. The applied electric field is parallel to the spontaneous polarization on the left side of the figure.

Miller & Weinreich, Phys. Rev. (1960)

Activation type of wall motion yields

$$v = v_\infty \exp\left(-\frac{\Delta U}{kT}\right)$$

Then $$\frac{v_y}{v_x} = \exp\left(-\frac{\Delta U_y - \Delta U_x}{kT}\right)$$

where $\Delta U_x, \Delta U_y$ **- activation energies in x, y directions**

Since $$\Delta U = \frac{8b}{3\sqrt{3}} \frac{\sigma_p^{1/2}\sigma_w^{3/2}}{P_s E}$$

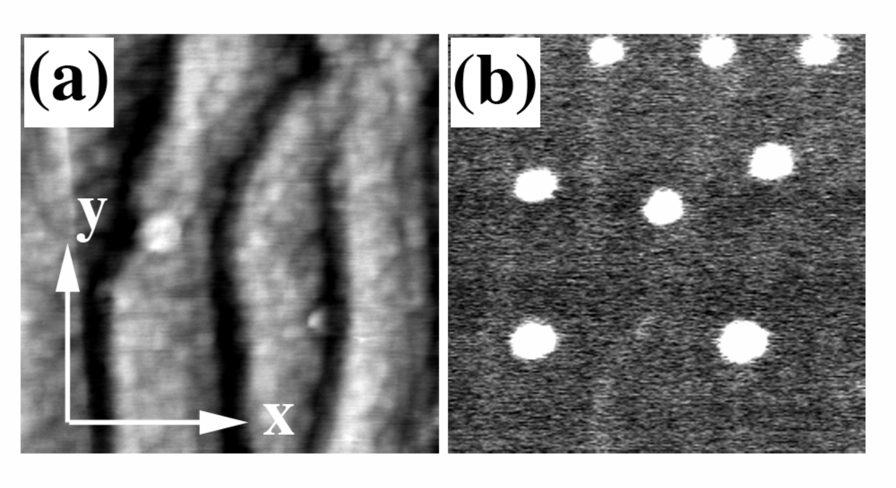
We have $$\frac{\Delta U_x}{\Delta U_y} = \left(\frac{\sigma_{wx}}{\sigma_{wy}}\right)^{3/2}$$

$$\sigma_{wx} / \sigma_{wy} = 1.07$$

Orientational variation of wall energy density by 7% will produce wall velocity anisotropy of factor 3

Effect of Microstructure on Domain Wall Motion

Topography and PFM of PZT film surface



Direction of fast wall motion coincides with the orientation of morphological ridges which may be a reason for anisotropy of wall energy density and anisotropic domain growth in low fields.

No anisotropy of domain growth has been observed in PFM tip-induced switching: in high fields any directional variations of ΔU become negligibly small:

$$\Delta U = \frac{8b}{3\sqrt{3}} \frac{\sigma_p^{1/2} \sigma_w^{3/2}}{P_s E}$$

Conclusion

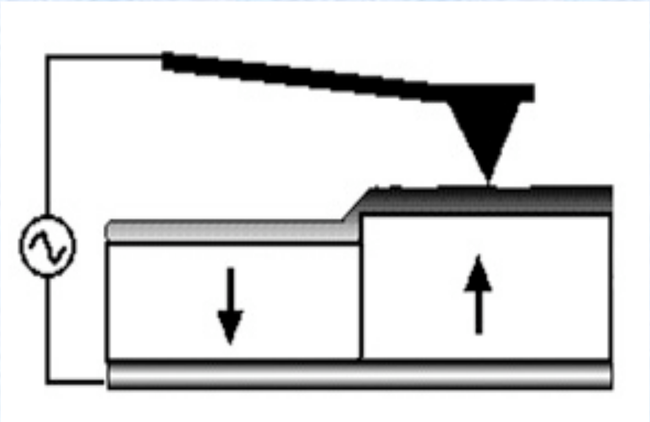
- Inhomogeneous nucleation results in cycle-to-cycle reproducibility of switching kinetics which allows application of stroboscopic PFM and direct measurements of domain nucleation rate and wall velocity.

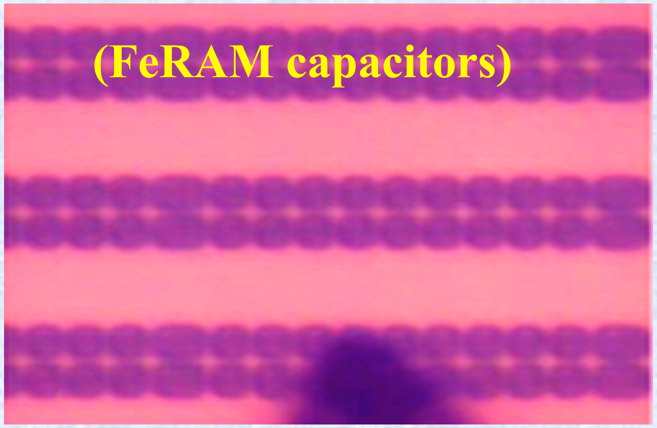
- It has been shown directly that the rate-limiting mechanism depends upon both the capacitor size and field range.

- In small capacitors (~1μm^2) switching is dominated by the wall motion (nucleation limited). As a result, in high fields, small capacitors switch faster than large capacitors (>> 1μm^2) where nucleation, rate-limiting mechanism, plays significant role. In low fields, wall velocity as a rate-limiting mechanism causes slower switching of small capacitors.

- Capacitor microstructure plays a major role in switching kinetics. In polycrystalline capacitors, broad variation of local switching times leads to NLS-type switching kinetics. In epitaxial capacitors (of even microscale dimensions), KAI model provides adequate description of switching.

PFM for Testing Ferroelectric Capacitors





PFM allows delineation of domain patterns in FE capacitors as a function of time and voltage and correlate this information with integral switching parameters (switching charge, coercive voltage, time of switching)