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Switching kinetics in nanoferroelectrics

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Abstract

We have measured the switching in ferroelectric capacitors of lead zirconate titanate (PZT) over three orders of magnitude in lateral area, from $A = 166$ to $0.19\ \mu m^2$ (the latter being the size of the smallest ferroelectric random access memory (FRAM) cells in production), and over three orders of magnitude in ramp rate of applied voltage ($dE(t)/dt = 10^7$–10^{10} V cm^{-1} s^{-1}). In accord with the model of Scott (1998 *Ferroelectr. Rev.* **1** 1), the submicron cells follow a different dependence to the larger cells: for $A \gg 1\ \mu m^2$, the data fit a theory due to Landauer *et al* (the LYD model), which neglects nucleation; whereas the nanoscale devices satisfy the functional dependence predicted by Pulvari and Kuebler (the PK model), albeit with a modified coefficient. This crossover behaviour has implications for Gbit FRAM device performance at high speed. Fringing field effects measured agree with a simple model from Feynman.

1. Introduction

Recently there has been strong interest from both the fundamental physics and the need for nanoscale engineering of capacitors for RAMs (random access memories) points of view in understanding the switching kinetics in submicron capacitors. Two early theories [1, 2] of the coercive field as a function of the ramp rate of applied voltage give rather different predictions, but neither theory has ever been tested over a range of lateral sizes sufficient for determining their respective regimes of applicability. Over most of the past two decades the theory generally used to fit data was that of Ishibashi [3], which in turn was based upon the crystal growth model of Avrami. This model has as its rate-limiting parameter the domain wall speed, but in the past few years it has been shown that nucleation is more often the rate-limiting parameter. Following the development of a nucleation-limited model by Du and Chen [4, 5], Jung *et al* [6] showed that such nucleation models gave better fits to experimental data in various materials, and Tagantsev *et al* [7] later independently arrived at the same conclusions. What has not been established, however, is the size range over which these theories are applicable. In the

present paper we examine ferroelectric capacitors from Samsung Co. down to about 450 nm on a side (at 0.19 μm^2, the smallest commercial ferroelectric memory cells), and show that their switching kinetics are described more accurately by the Pulvari–Kuebler model than that of Landauer *et al*.

Several early theories were developed in the 1950s for switching in ferroelectrics and in particular for the dependence of coercive field on voltage ramp rate. The Landauer, Young and Drougard theory (LYD) developed relationships [1] for barium titanate based upon empirical data reported by Epstein [8] and Merz [9]. Wieder [10] provided a rather different model which had good accord with observed frequency dependences.

Both the breakdown field E_B and the coercive field E_c in ferroelectrics vary significantly with the rise time (ramp rate) of the applied voltage. Breakdown field typically varies as a power law of switching time, $t_S^{-1/2}$ [11], whereas there are several different theories for $E_c(t_S)$ [1–7]. In the latter case, the dependence arises physically from the behaviour of domain walls in a viscous medium [12]. In high fields such domain walls can often be described as ballistic, with damping similar to that of mechanical projectiles [13].

Ramp rates can vary considerably in real devices. The coercive field of a ferroelectric capacitor in a random access memory (FRAM) is typically of order 50–100 kV cm^{-1} (10 MV m^{-1}), and present FRAMs have 60 ns access times, so that rise times <10 ns are required. This is a ramp rate $dE(t)/dt = 10^{13}$ V cm^{-1} s^{-1}. There are no published reports in the literature on the size dependence of the behaviour of coercive fields in this ramp rate regime. Some years ago one of us [14, 15] derived a criterion for switching to be limited by domain wall speed v, rather than nucleation:

$$\frac{v}{Nr} \ll A^{3/2}, \tag{1}$$

where A is the electrode area, r the nucleation rate at field E and N the number of nucleation sites per unit area. Both v and r vary as $E^{3/2}$ [16], so the field dependence drops out of this equation. Using independently determined values of N [17], we find that nucleation will dominate in PZT for $A < 1$ μm^2; this is in the range of cell areas studied in the present work. (We note parenthetically that the critical size is much larger in some other ferroelectrics, e.g., $A < 100$ μm^2 for KNO_3.) Hence, even before doing any experiments, we had reason to suppose that the switching kinetics in PZT cells of submicron lateral width would differ from those of larger cells. Peripherally, we note that the theory of Duiker *et al* [17] is valid for finite size cells, whereas as pointed out by Dalton *et al* [18], the Ishibashi solutions [3], based upon the Avrami model, are not. To a good approximation, we found (equation (47*c*) of [15]) that the high-voltage switching time t_{HV} is given by

$$t_{HV} = \frac{1}{v}\left(\frac{9v}{4\pi r}\right)^{1/4} = 0.6 \pm 0.1 \text{ ns}, \tag{2}$$

using $v = 440$ m s^{-1} and $r = 7 \times 10^{28}$ s^{-1} m^{-3} [17], in reasonable accord with the most recent experimental value of 0.3 ns [19].

In the present work we examine the behaviour of E_c as a function of $dE(t)/dt$ from 10^7 to 10^{10} V cm^{-1} s^{-1} and for ferroelectric PZT capacitors from 0.19 to 166 μm^2. The large-area capacitors satisfy the coercive field theory of Landauer *et al* (LYD), whereas the submicron devices satisfy the theory of Pulvari and Kuebler (PK model). This PK theory explicitly includes both a nucleation-limited regime and a domain wall velocity-limited regime in its mathematics, by choosing an exponential form for the time dependence of the response of displacement current $I(t)$ to the field applied; this gave a fast linear dependence at high fields E, due to domain wall speeds, and a slow, nonlinear dependence at low voltages, due to nucleation. Thus, in the PK theory, nucleation and domain wall speeds are each the

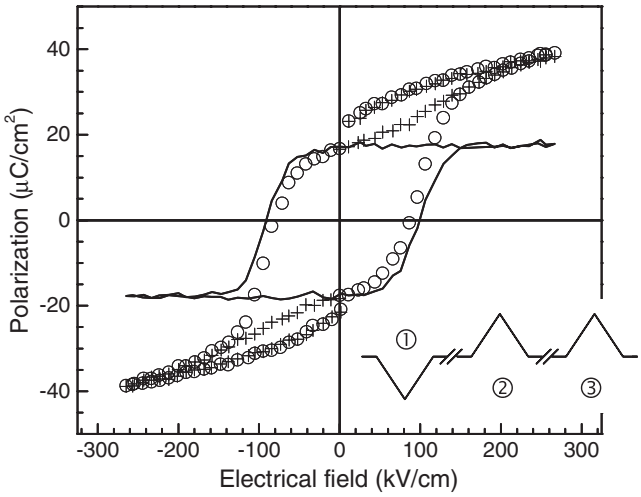

Figure 1. Hysteresis loops showing how the coercive field is different when the non-remanent polarization component is incorporated (○) or not (solid line). The non-remanent polarization component (+) could be separated by applying the three-pulse chain as shown in the inset. The coercive field deduced from the remanent hysteresis loop corresponds to the inflection point at which $\delta P/\delta E$ is maximal. These loops are obtained from PZT memory cell capacitors of size of 0.19 μm^2.

rate-limiting parameter in different time or voltage regimes. The LYD theory, on the other hand, while not ignoring nucleation, concluded that it was extremely unimportant, because it would produce an unobserved inductive response in the displacement current transients (since domains cannot contribute to the displacement current until a finite time *after* they are created). Thus, qualitatively speaking, the PK theory is expected to be more applicable for submicron ferroelectric kinetics, which Scott's model [14, 15] shows to be nucleation limited, and the LYD theory more applicable to large cells; this is in accord with our experiments. In general, nucleation-limited kinetics describes ferroelectric switching in several different materials over a wider range of voltage and frequency than do domain wall speed-limited kinetics [6, 4, 5, 7].

2. Experimental details

P–E hysteresis is a very useful tool for researching ferroelectric thin films. It delivers information on the nonlinear polarization response of the ferroelectric capacitors, typically stimulated by a bipolar triangular voltage in our measurements. It also provides us with not only the nonlinear response including non-remanent polarization but also the switching resistance, such as the coercive field—the field at which polarization equals zero. By 'non-remanent' polarization, we mean the polarization that would be induced by an electric field in a paraelectric substance; this polarization can be linear or nonlinear in applied field E.

Figure 1 shows that hysteresis loops may cause an underestimation of the coercive field due to incorporation of the non-switching polarization into the resultant nonlinear response. Therefore, it is vital to minimize the non-switching polarization, in pursuit of which three consecutive triangular pulses are applied to the samples with a 1 s quiescent period in between. If a sample is pre-poled into a polarization state (e.g., $-P_r$ in this case), that ensures that the first pulse (①) in the inset of figure 1 reinforces the preset state; and then both non-remanent polarization and remanent polarization will be switched and measured (○) when the second

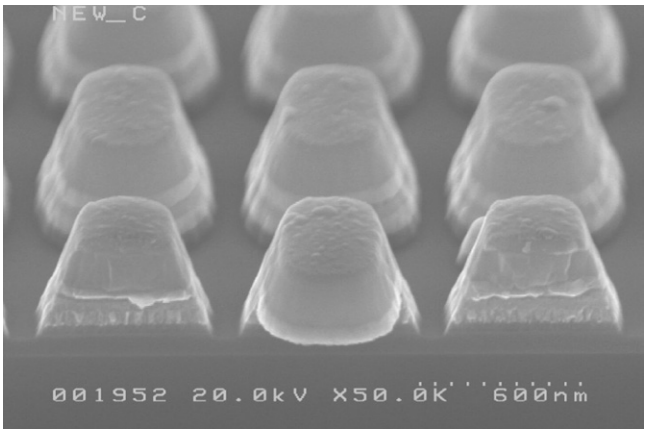

Figure 2. Plan view of PZT capacitors with an Ir/IrO_2 top electrode of area 0.32 μm^2 with a bottom electrode of $Pt/IrO_2/Ir$.

pulse (②) is applied. But for the last pulse (③), only the non-remanent polarization will be encountered (+), since the remanent polarization is already switched into the particular state during the pulse ②. Subtracting the non-remanent polarization from the resultant polarization gathered in the stage ② allows the remanent polarization to be derived.

Repeating the experiment for the opposite polarization state permits the two halves of the bipolar non-remanent response to be independently measured. Recombining the two halves results in a full non-remanent hysteresis loop being constructed, as indicated in the solid line of figure 1. From the remanent hysteresis loop, both positive and negative coercive fields are deduced and averaged. Averaging here is used to eliminate the effect of internal bias fields ('imprint').

The PZT capacitors studied vary from 166 and 0.19 μm^2 in size. The P–E hysteresis loops in figure 1 are measured from a group of capacitors, each of which is as large as 0.19 μm^2. All the applied pulses are at a ramp rate of 1.6×10^5 V s^{-1}. All the capacitors investigated here have the same vertical structure: an Ir/IrO_2 stack as the top electrode (TE); PZT as the ferroelectric film (FE); and a $Pt/IrO_2/Ir$ stack as the bottom electrode (BE). Figure 2 exhibits an SEM plan view of the PZT capacitors used.

3. Results and discussion

P–E hysteresis loops describe how the polarization P is related to the switching field E and the time during switching in an expression of the form $P = f(E, t)$ with which various physical parameters such as the activation field α and domain wall displacement velocity per volt γ can be derived. Moreover, the activation field α obtained is a material property quantity similar to the coercive field E_c. Furthermore, the parameter γ is closely related to the displacement mobility of domains, μ_D.

The coercive field E_c depends explicitly on not only a field rise rate $dE(t)/dt$ that can be achieved in different ways by adjusting the amplitude or frequency of the triangular driving voltage, but also the past history of the material. To make sure that each hysteresis measurement has the same history, before each remanent hysteresis measurement, a bipolar triangular pulse with 100 ms period and 3.0 V amplitude was applied to the samples. Figure 3 shows that the remanent hysteresis curve depends strongly on the field ramp rate varying from 3.3×10^7 to

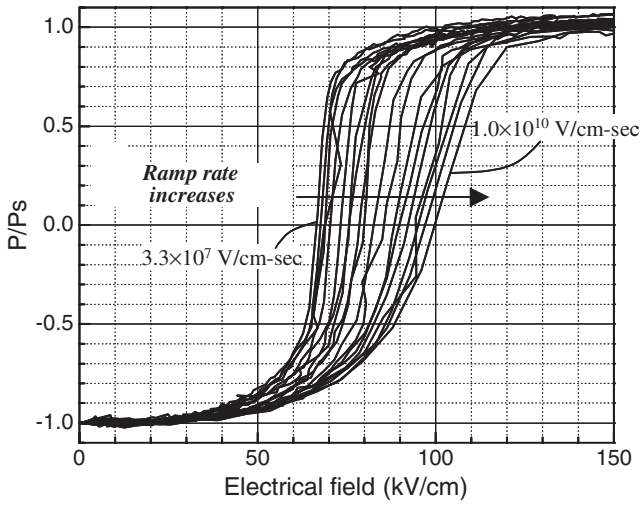

Figure 3. Normalized remanent P–E hysteresis loops as a function of the frequency of the applied triangular pulse with various ramp rates that range from 3.3×10^7 to 1.0×10^{10} V cm^{-1} s^{-1}: each P–E curve is normalized to its saturation polarization P_S in 166 μm^2 PZT capacitors.

1.0×10^{10} V cm^{-1} s^{-1} to reach its saturation value and thus its coercive field. In the LYD model, the polarization $P(t)$ depends on the electric field $E(t)$ and its ramp rate $\mathrm{d}E(t)/\mathrm{d}t$:

$$\frac{P(t)}{P_S} = 1 - 2\exp\left[\frac{\nu E(t)}{\frac{\mathrm{d}E(t)}{\mathrm{d}t}\mathrm{e}^{\alpha/E(t)}}\sum_{k=1}^{n}(-1)^{k+1}\frac{k!E(t)^k}{\alpha^k}\right], \tag{3}$$

where P_S is the saturation polarization in a P–E hysteresis loop and ν is a constant which is independent of the field and polarization. By definition, the polarization $P(t)$ equals zero when the field $E(t)$ reaches the coercive field E_c. Then equation (3) is expressed as follows:

$$\ln\left(\frac{\mathrm{d}E(t)}{\mathrm{d}t}\right) = \ln\left[\frac{\nu E_c}{b}\sum_{k=1}^{n}(-1)^{k+1}\frac{k!E_c^k}{\alpha^k}\right] - \frac{\alpha}{E_c}, \tag{4}$$

where $b = \ln(1/2)$ and α is an activation field of order 10 times E_c. This equation (4) is very important because it provides a simple analytical form for relating a field rise rate $\mathrm{d}E/\mathrm{d}t$ to the coercive field, particularly in an applied field of triangular form, due to the fact that the field rise rate is a constant value.

Furthermore, as pointed out in earlier works studying the dependence of the activation field on the thickness of ferroelectric materials [20–22], the coercive field is strongly dependent on the sample thickness. Unlike for bulk ferroelectrics, surface effects in the case of thin films should be considered, because a certain amount of voltage drop is expected across the surface layers in the presence of an electric field. For simplicity, if we assume that the coercive field depends inversely on the thickness of the sample as Merz found [21], the coercive field E_c can be written as

$$E_c = E_{\mathrm{bulk}} + E_{\mathrm{surface}}, \tag{5}$$

where E_{bulk} is the true coercive field strength, which can be obtained on very thick samples, and E_{surface} is the field due to surface layers. From equations (4) and (5), the modified LYD model is now compared to data obtained for PZT thin film capacitors as thin as 144 nm and as large in area as 166 μm^2. Figure 4 shows the correspondence of the model (solid line) to

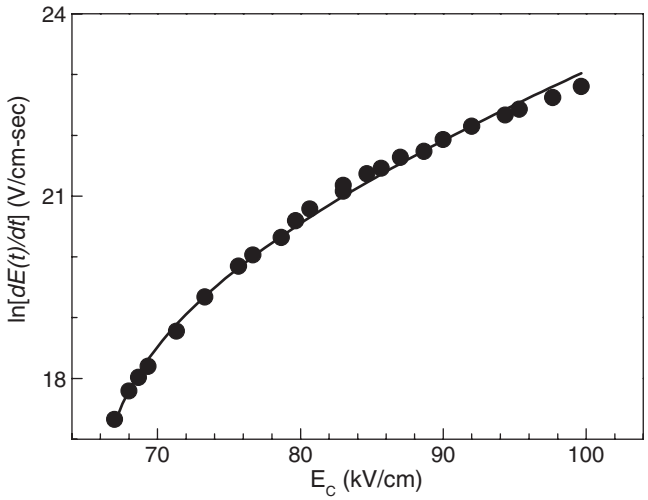

Figure 4. Log of ramp rate $dE(t)/dt$ versus coercive field E_c for 166 μm^2 PZT capacitors, compared with LYD theory [1].

the data (filled circle). This modified LYD model predicts the switching behaviour of thin film PZT ferroelectrics, as shown in figure 4. The activation field α is 6.2×10^5 V cm^{-1} and $E_{surface}$ is 5.3×10^4 V cm^{-1} from the fit to the data. The value of the activation field here is in good agreement with those obtained by Song *et al* [23] which range from 600 to 800 kV cm^{-1}. They explored the activation field of various PZT families with a different approach, in which the maximum switching current i_{max} was investigated as a function of the electric field E, rather than by studying the coercive field versus ramp rate.

The primary concern here is to understand how switching properties of a submicron capacitor are different from those of a larger one. To determine the dependence of the coercive field on the ramp rate of the electric field, remanent hysteresis loops were measured for nanocapacitors of size 0.32 and 0.19 μm^2, as displayed in figure 1. Similarly to the case for large capacitors, the remanent hysteresis curves were also taken at various ramp rates, varying from 5.3×10^6 to 1.0×10^{10} V cm^{-1} s^{-1}. From the PK empirical model, we can relate the field ramp rate to a function of the coercive field when a triangular field is applied across the films:

$$\ln\left(\frac{dE(t)}{dt}\right) = \ln\left(\gamma E_c^2\right) - \frac{\alpha}{E_c} + \delta, \tag{6}$$

where γ is proportional to the domain wall mobility μ_D and δ is a constant. Figure 5 illustrates the correspondence of the modified LYD model and the PK model to the data from 0.32 (figure 5(a)) and 0.19 μm^2 (figure 5(b)) capacitors. The modified LYD model is unable to fit the data from these capacitors. However, the PK model is well fitted to the data measured at area $A = 0.32$ μm^2, and even those for the 0.19 μm^2 capacitors.

Pulvari and Kuebler (PK) found that $\mu_D = 10$ cm^2 V^{-1} s^{-1} for barium titanate [7], whereas in the present work we find $\mu_D = 14$ and 7 cm^2 V^{-1} s^{-1} for 0.32 and 0.19 μm^2, respectively. These are several orders of magnitude larger than the experimental domain mobility values measured directly by Gruverman [24], and this may be due to the assumptions of PK's model: Pulvari and Kuebler assume that all domains nucleate on the cathode or anode and propagate a distance d all the way through the sample. In order to fit their apparent velocities, they therefore derive high mobilities. However, in reality in a PZT ceramic domain walls will

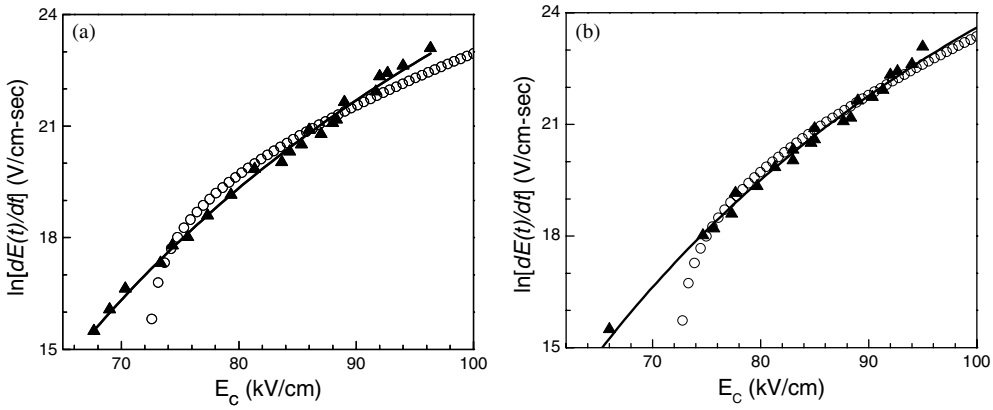

Figure 5. Log of ramp rate $\mathrm{d}E(t)/\mathrm{d}t$ versus coercive field E_c for (a) 0.32 and (b) 0.19 $\mu\mathrm{m}^2$ PZT capacitors, compared with LYD theory [1] and Pulvari–Kuebler theory [7].

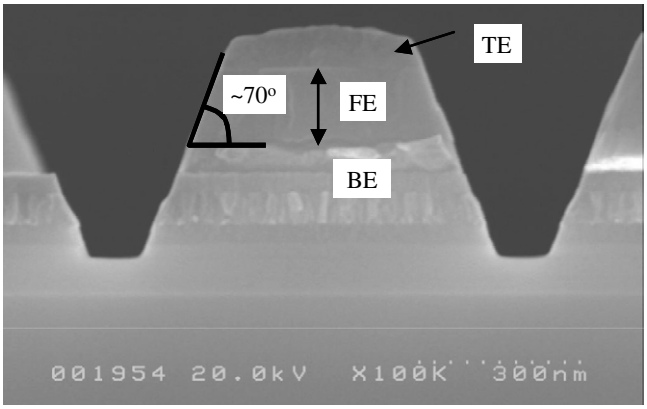

Figure 6. A cross-sectional SEM picture of 0.32 $\mu\mathrm{m}^2$ PZT capacitors, showing the aspect ratio and the sidewall angle of 70°.

nucleate throughout the interior of the film; and in order to form a single columnar domain from cathode to anode, each domain need only grow a few lattice constants a. Therefore the fitting parameter μ_D in PK's model may relate to Gruverman's actual measured domain wall mobilities μ_exp as $\mu_\mathrm{D} \simeq (d/a)\,\mu_\mathrm{exp} \simeq 360\,\mu_\mathrm{exp}$ for a 144 nm thick PZT film and lattice constant $a = 0.40$ nm. Note that the smallest cells tested here already meet the area requirements for 2007 for FRAMs as detailed in the International FRAM 'Roadmap' [25].

As shown in the SEM picture in figure 6, the step coverage of 0.32 $\mu\mathrm{m}^2$ PZT FRAM capacitors is not perfectly vertical; an angle of 70° is obtained. This results in the need for correction of the measured capacitance for fringing field effects [26]. Figure 7 shows that the fringing field correction (equation (7)) to second order (approximating the capacitor geometries as truncated cones and ignoring the corners):

$$\frac{C_\mathrm{corrected}}{C_\mathrm{elementary}} = 1 + \left(\frac{d}{\pi R}\right)\ln\left(\frac{16\pi R}{\mathrm{e}d}\right) + \left[\frac{d}{2\pi R}\ln\left(\frac{d}{16\pi R}\right)\right]^2, \tag{7}$$

(where R is the cell radius and e the base of natural logarithms) gives only about 50% of the observed capacitor correction that would occur for perfect parallel plates. We have found rather

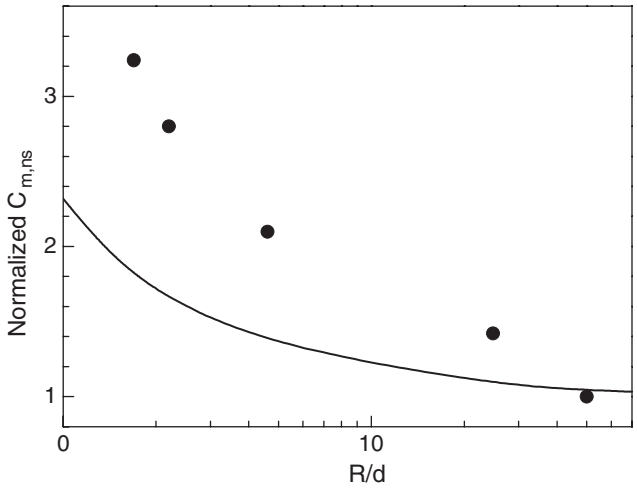

Figure 7. Fringing field correction of measured non-switching capacitance $C_{\mathrm{m,ns}}$ versus aspect ratio (radius R divided by thickness d) for PZT capacitors, showing the failure of equation (7). The capacitors are treated as approximating truncated cones.

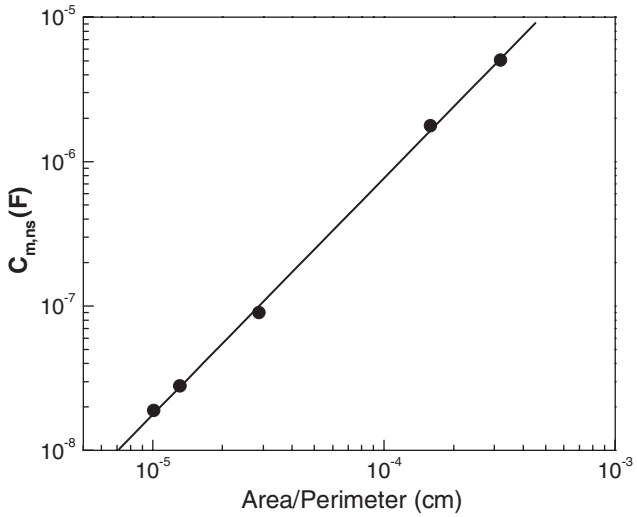

Figure 8. Measured non-switching capacitance $C_{\mathrm{m,ns}}$ of PZT capacitors versus area/perimeter ratio A/P for areas from 0.19 to 166 μm^2; the straight line is an empirical fit to $(A/P)^{1.64}$.

surprisingly (figure 8) that the excess capacitance satisfies an empirical correction formula $\Delta C = L^{1.64}$, where L is the cell lateral width; or, since for nearly square cells, C is proportional to L^2, $\Delta C/C = L^{-0.36}$. This formula is reminiscent of the work of Maier *et al* [27, 28], and of Shchukin and Bimberg [29] and Williams *et al* [30], who showed that the energetics of nanoparticles is related to the number of edges and corners that they have [31]. One possible explanation of the formula above is that there is a perimeter damaged by OH in the devices [32] which reduces the effective area A by an amount that is proportional to the perimeter; in this case a correction term of the functional dependence $\Delta C/C \cong gP/A \cong L/L^2$, and hence $\Delta C/C \cong L^{-1.0}$ might be expected, where g approximates the width of the strip

along the perimeter damaged by hydrogen ions in the plasma etching process. The fact that experimentally the exponent is about 0.4 rather than 1.0 might arise from effects at the corners of the cells or from a nonlinear effect of OH hydroxyl ion concentration on the local dielectric constant. However, a simpler argument follows Feynman [33] who pointed out that including fringing fields gives a correction to the capacitance $C = \varepsilon A/d = \varepsilon L^2/d$ for a square of side L and thickness d less complex than that in equation (7), such that one can approximate the effective area A as increased on each side by $3/8$ of the thickness d:

$$C \cong \frac{\varepsilon\,(L + 3d/8)^2}{d} \cong \varepsilon\frac{L^2}{d} + \frac{3}{4}\varepsilon L. \tag{8}$$

In the present case L is of about the same magnitude as d (150–450 nm), so an average of the two terms (quadratic and linear) on the right-hand side of equation (8) gives an effective dependence on length L midway between linear and quadratic:

$$C \cong L^{1.5} \qquad \text{or} \qquad \Delta C \cong \left(\frac{A}{P}\right)^{1.5}, \tag{9}$$

as observed.

4. Summary

We explored switching properties of PZT capacitors ranging from $>100\ \mu\text{m}^2$ down to $0.19\ \mu\text{m}^2$ by examining a field ramp rate versus the coercive field in P–E hysteresis loops stimulated by a triangular pulse. From the fit of the modified LYD model to the data, we found that the LYD model fits PZT capacitors of size $166\ \mu\text{m}^2$, and implies an activation field α of $622 \pm 20\ \text{kV cm}^{-1}$. However, nanocell capacitors of areas 0.32 and $0.19\ \mu\text{m}^2$ conform with the PK theory, in which nucleation and domain movement are both important.

Fringing field effects due to mesa-structured capacitors give insufficient non-remanent capacitance to explain the measurements, particularly as the aspect ratio increases. However, the excess capacitance satisfies an empirical correction formula of $\Delta C/C = L^{-0.36}$. This formula agrees reasonably well with a prediction from Feynman for thickness d comparable to lateral dimension L.

As a peripheral comment, we note that for a triangular wave form, the ramp rate can be changed by varying the frequency at constant voltage or by varying the voltage at constant frequency f. Thus, the dependence of the coercive field E_c upon the ramp rate is not necessarily the same as $E_c(f)$, which was first characterized by Scott up to 2 MHz [34, 35] and later by Waser *et al* [36].

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