## Retention of Thin Ferroelectric VDF–TrFE Copolymer Films Evaluated from Dielectric Non–Linearities

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*Abstract*—Usual retention tests are based on the application of read pulses to a ferroelectric sample while the charge response is recorded. These tests, however, do not allow the continuous recording of retention as after the application of the read pulse the ferroelectric is in a new state defined by the direction of the read pulse. We propose a novel approach which is based on the nondestructive readout of the remanent polarisation by measurement of small signal dielectric nonlinearities. The temporal development of the remanent polarisation is directly accessible from the measured first and second harmonics in the current response to a small sinusoidal voltage signal. The novel technique has been used to investigate the retention of thin VDF-TrFE copolymer films of molar ratio 70/30 with thickness below 200 nm. This technique may also be useful for the nondestructive readout of ferroelectric memory cells.

## I. INTRODUCTION

Polarisation retention of a ferroelectric is an important feature for most of its applications, in particular for memories [1] but also for pyroelectrics or piezoelectrics. On the other hand, a reduction or loss of remanent polarisation might be favorable when the ferroelectric is used as the dielectric in a capacitor to store energy.

In the common procedure to investigate retention the ferroelectric sample is polarised first and after a specified time a further voltage pulse is applied while the charge response is recorded. Various kinds of pulse sequences are applicable for same state, opposite state, switching, and non-switching tests [2]. The limitation of this technique is that the sample is polarised again by the second read pulse (either in the same or in opposite direction) and a continuous recording of polarisation loss is not possible. Furthermore, already relative small numbers of switching pulses can cause some fatigue [3]. Therefore, we propose an alternative procedure which is based on the continuous measurement of the small signal linear and second order non–linear dielectric permittivity as a function of time.

## II. THEORY

A sinusoidal electric field  $E(t) = E_{\sim} \cos(\omega_0 t)$  applied to a non-linear dielectric material causes a dielectric displacement

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Fig. 1. Working principle of a ferroelectric memory.

D(t) with components at higher harmonics [4],[5]:

$$D(t) = D_0 + D_1 \cos(\omega_0 t) + D_2 \cos(\omega_0 t) + \dots$$
(1)

Using the Fourier decomposition theorem and:

$$D = \varepsilon_1 E + \varepsilon_2 E^2 + \varepsilon_3 E^3 \tag{2}$$

one can link the displacements components to the non-linear permittivities  $\varepsilon_n$  [6],[7]:

$$\varepsilon_0 \varepsilon_n = \lim_{E \to 0} 2^{n-1} \frac{D_n}{E_{\sim}^n}.$$
(3)

Furthermore, one can use the Landau expansion of the free energy F as a function of the dielectric displacement:

$$F = F_0 + \frac{\alpha}{2}D^2 + \frac{\gamma}{4}D^4 + \frac{\delta}{6}D^6 - ED$$
 (4)

and the knowledge, that at electric field E = 0 the dielectric displacement is equal to the spontaneous polarisation  $P_s$  or, in the case of a multi-domain ferroelectric material, the remanent polarisation  $P_r$ , at the minimum of the free energy  $\delta F/\delta D = 0$ . That leads us to:

$$E = \alpha D + \gamma D^3 + \delta D^5.$$
<sup>(5)</sup>