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THE INFLUENCE OF LIGHT ON FATIGUE IN PZT FILMS WITH PLANAR ELECTRODES

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Degradation properties, such as a low voltage breakdown, fatigue, and ageing of ferroelectrics are believed to be the major problems of ferroelectric films affecting their lifetime. Although these properties have been studied for a long time, the information is still insufficient for both a quality improvement of ferroelectric devices and their lifetime predictions. Degradation mechanisms should be studied for the lifetime prediction, as well as for material development.

Fatigue is the decrease in switchable polarization with increasing number of polarization reversals. In this paper, general features and the mechanisms of fatigue are briefly referred to and the light influence on fatigue in PZT films is discussed.

A set of fatigue measurements has been performed on RF-sputtered PZT films of about 2 µm in thickness. Polarization switching characteristics by applying sinusoidal a.c. electric fields were studied both with sandwich-type and planar electrodes. The planar structure has allowed for the study of the influence of the free charge carries induced by UV – illumination in the planar capacitor gap. The fatigue became noticeable after 10^6 switching cycles for a sandwich structure and after 10^9 those cycles for films with planar electrodes. The film illumination during the polarization switching accelerates significantly the fatigue process. The additional fatigue induced by the photoactive light was completely reversible.

Of special importance is the fact that the films on metal substrates (sandwich-capacitor) were fatigued more rapidly than those on dielectric substrates (planar capacitor), though they were prepared under the same conditions. This evidences the major contribution of transition layers to the development of the fatigue process that agrees well with the model of fatigue proposed in the literature.

1. Introduction

Due to the unique properties, such as a high value of dielectric permittivity ε, spontaneous polarization P_s, piezoelectric moduli d_{ij} and electromechanical coupling coefficient k_p, ferroelectric materials have gained widespread application in engineering. In particular, the perovskite-type ferroelectric thin films are very promising for application in microelectronics as high dielectric capacitors [1, 2], piezoelectric sensors [3, 4], electro-acoustic transducers [5], high frequency SAW devices [6, 7], ultrasonic sensors [8] and many others [e.g. 9–13]. It is common knowledge that the basic physical properties of those materials depend on the chemical constitution, atomic structure, domain structure and microstructure that, in turn, depend on technological conditions of the preparation process. However, their practical application depends on the degradation properties such as a low voltage breakdown, fatigue and ageing of ferroelectric that have been pointed out as major problems of ferroelectric films affecting their lifetime.

Fatigue is the decrease in the switchable polarization with the increasing number of polarization reversals. Several mechanisms for fatigue have been proposed for both the bulk and thin film ferroelectrics including dendrite formation of oxygen deficient filaments [14], domain pinning by defects [15, 16], space – charge accumulation at the film – electrode interface or domain boundaries under the repeated polarization switching, grain-boundaries compensating for the externally applied voltage, polarization screening by defects [17–20] and locking domains by electronic charge trapping centres (electron domain pinning) [18, 19]. There are far more approaches to the explanation of the fatigue phenomenon e.g. the formation of a-domain wedges and micro-cracks due to the piezoelectric deformation [21], the pinning of domains at grain boundaries triggered by the migration of pores [22]. A simple model for ferroelectric fatigue based on the Landau theory has been also developed [23–25]. The model suggests the formation of mesocopic domain/defect structures consisting of opposing domains stabilised by planes of charged defects.

Although the fatigue properties have been studied for a long time and they can be significantly diminished in PZT thin films by using appropriate electrode materials [16, 26–31], a fundamental understanding of the phenomenon is still lacking and the information is still insufficient for both a quality improvement of ferroelectric devices and their lifetime predictions. Therefore, degradation mechanisms should be studied for the lifetime prediction, as well as for the material development.

The influence of illumination on the polarization switching has been observed for a number of ferroelectrics with significant photosensitivity; it is known as the polarization switching or simply photoswitching [32]. An analysis of the published data [17–19, 21, 32, 33] shows that the nature of photoswitching in different ferroelectrics is basically the same: an increase in the density of free carriers by photogeneration of nonequilibrium carriers reduces the coercive field E_c and the switching time τ_s. The total number of the 180° domains participating in the polarization switching process increases and so does the mobility of the 180° domain walls.

It should be also noted that all of the known studies of the effect of fatigue of ferroelectric thin films were carried out on sandwich-type structures (conducting or covered with a conducting layer substrate – ferroelectric film – conducting upper electrode). In this case, the emphasis is laid, in our opinion, on the inhomogeneity of the object under study in the direction normal to its surface that makes it difficult to interpret the results.

In the present work the symmetrical planar electrodes were used to study the effect of the photoactive light on fatigue.

2. Samples and method of investigation

Polycrystalline $Pb(Zr_{0.53}Ti_{0.45}W_{0.01}Cd_{0.01})O_3$ films of about 2×10^{-6} m in thickness were obtained on the substrates made of stainless steel and dielectric radioceramics (Al_2O_3 with additives) [34, 35]. The measurements were performed using Al electrodes of a thickness of 0.3×10^{-6} m deposited by evaporation in vacuum (Fig. 1).

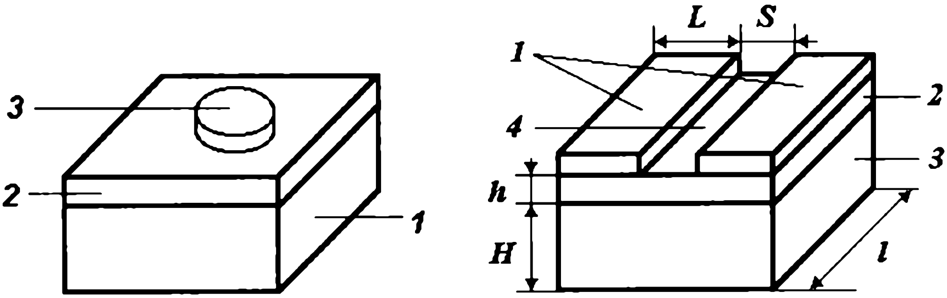

Fig. 1. a) The sandwich-type capacitor scheme: *1* – the stainless steel substrate of 100×10^{-6} m in thickness; *2* – the ferroelctric thin film; *3* – the disk-shaped Al electrode of 1.76×10^{-6} m^2 in area. b) The planar capacitor scheme: *1* – Al electrodes of 1×10^{-3} m in length (l) and 2×10^{-3} m in width (L); *2* – the thin ferroelectric film of 2×10^{-6} m in thickness (h); *3* – the Al_2O_3 polycrystalline substrate of 1×10^{-3} m in thickness (H); *4* – the dielectric gap of 8×10^{-6} m in thickness (S).

The dielectric hysteresis loops were recorded according to the Sawyer–Tower scheme "modified" by the application of an amplifier of small signals due to the small capacitance of planar capacitors $(1.5 - 5.0) \times 10^{-12}$ F.

To switch the polarization repeatedly the sinusoidal field of frequency ranging from 20 to 20×10^4 Hz was used. The relative error of the calculation $\Delta P_0/P_0$ was at 5%.

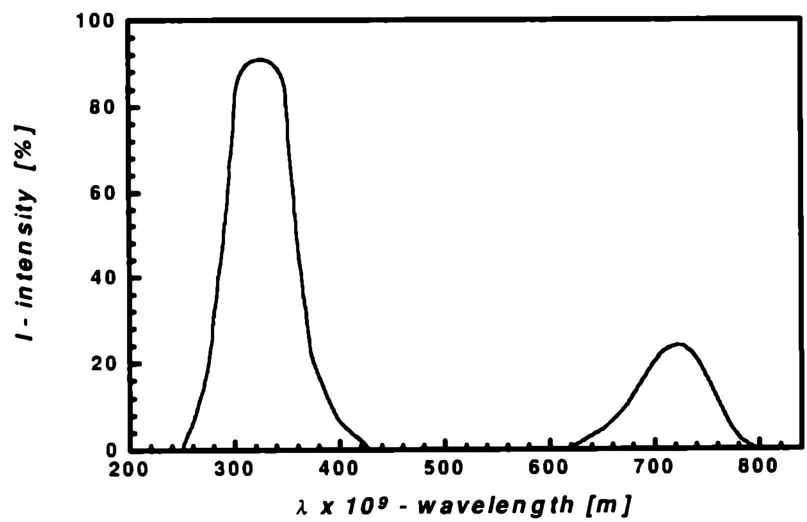

Fig. 2. The spectral characteristics of the filter used.

Since the band gap of the PZT materials is 3.6 eV [17], the quartz lamp and the filter that did not transmit the visible or almost all the infrared radiation ($\lambda = 250 - 420 \times 10^{-9}$ m; $E = 4.95 - 2.94$ eV) were used (Fig. 2) to induce non-equilibrium photocarriers to the conduction band in PZT films through the band-band transition [17]. According to the estimations of the absorption coefficient of the PZT films carried out for the wavelength of 325×10^{-9} m, the optical penetration depth is about 130×10^{-9} m [17]. Therefore the non-equilibrium photocarriers affect the PZT films near the Al-PZT interface.

The possibility to determine the remanent polarization and coercive field values in the measurements with planar electrodes was established previously [36].

3. Results and discussion

The values of the remanent polarization P_r of the thin films on the metal and dielectric substrates were, practically, the same ($20.0 - 24.5 \times 10^{-2}$ C/m^2 and $19.2 - 23.4 \times 10^{-2}$ C/m^2, respectively). However, the coercive field value E_c of the thin films deposited on the metal substrates was higher by c.a. an order of magnitude ($15 - 25 \times 10^6$ V/m and $3 - 5 \times 10^6$ V/m, respectively) than those of the thin films obtained on the dielectric substrates. Such a great difference in the E_c values seems to be due to the formation of a transition layer with pyrochlore structure on the boundary between the substrate and the thin film with the perovskite structure [37, 38].

Figure 3 shows the fatigue characteristics of the films on metal substrates (sandwich-type capacitors) at the frequencies of 10^2 Hz (down-triangles) and 10^4 Hz (circles). As can be seen, with increase of the frequency, the fatigue decreases as in the case of switching by bipolar rectangular pulses [15, 39–41] and triangular voltage pulses [31].

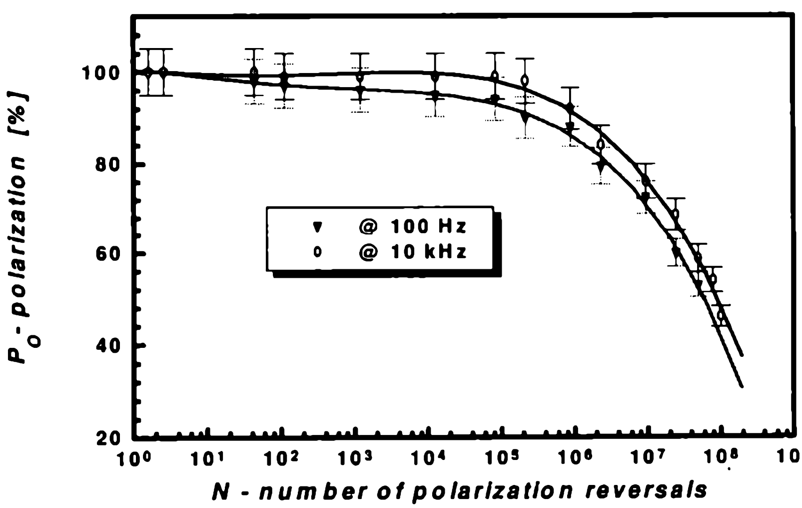

Fig. 3. The fatigue characteristics of the thin films deposited on the metal substrates at the frequencies of 10^2 Hz (triangles) and 10^4 Hz (circles). The field amplitude $E_a = 20 \times 10^6$ V/m.

Figure 4 shows the fatigue characteristics of the thin films grown on the dielectric substrates (planar capacitors) in the dark state (squares) and on continuous illumination with photoactive light (circles). Figure 5 represents the same curves for the higher frequency of the switching field. One can see that UV-illumination of the thin films accelerates the fatigue process.

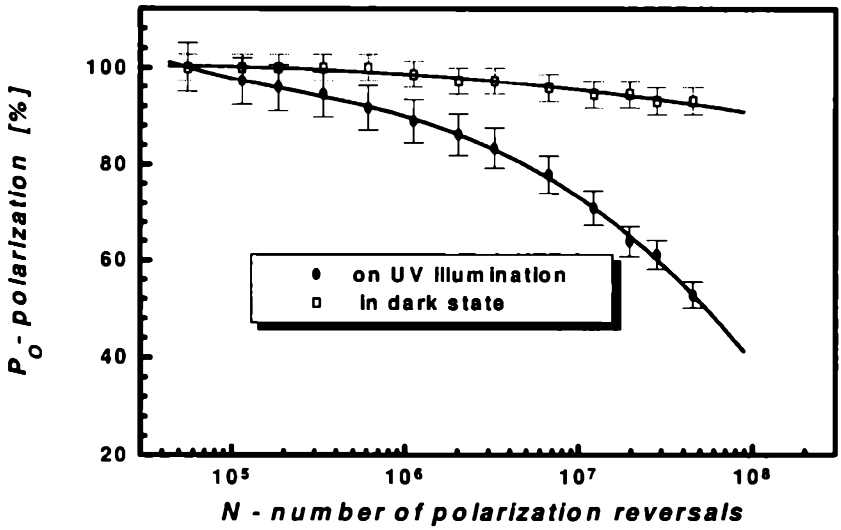

Fig. 4. The fatigue characteristics of the thin films deposited on dielectric substrates obtained at the frequency of 10^3 Hz and the electric field amplitude of 10×10^6 V/m with no UV-illumination (squares) and under illumination with the photoactive light (circles).

It has been argued by DIMOS *et al.* [19] that switchable polarization can be suppressed by generating and subsequent trapping of electronic charge carriers. That electronic charge trapping at the domain walls can lock those walls and lead to the suppression of the switchable polarization in the PZT films subjected to electrical fatigue [18, 19] what is consistent with our results. Similar results on suppressing the process of the 180° polarization switching by UV-illumination were obtained for $PbTiO_3$ single crystals [31] and other bulk ferroelectric materials [42].

It should be noted, however, that the "additional" fatigue induced by the photoactive light has proved to be, in practice, completely reversible. In other words, if in the course of a run similar to that the results of which are represented in Fig. 5 (the curve with stars), the illumination was stopped and the switching was continued after several cycles of switching n_o, then, after $D_n = 2.4 \times 10^6$ cycles of switching, the sample went into the state corresponding to $n_o + D_n$ cycles of switching for the curve with squares in Fig. 5.

Of special importance is the fact that the films on metal substrates (sandwich-type capacitor) were fatigued more rapidly than those on dielectric substrates (planar capacitor), though they were prepared under the same conditions. This evidences the major contribution of transition layers to the development of the fatigue process that agrees well with the model of fatigue proposed in [15, 16].

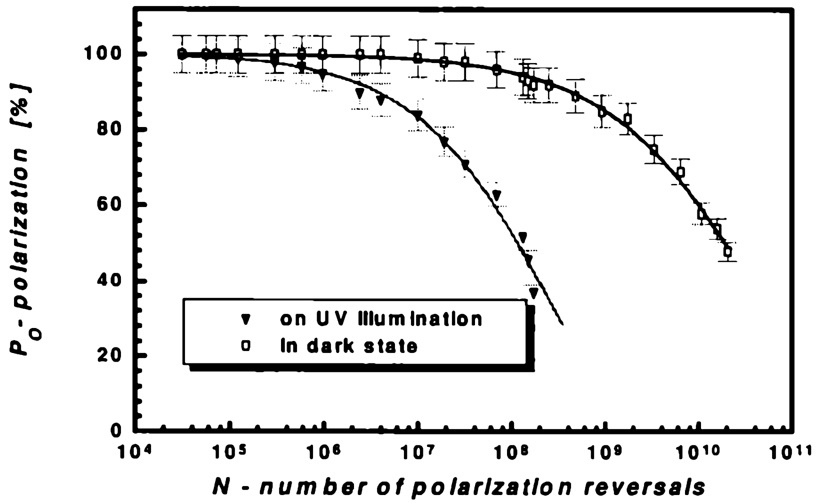

Fig. 5. The fatigue characteristics of the thin films deposited on dielectric substrates obtained at the frequency of 10^3 Hz and the electric field amplitude of 10×10^6 V/m with no UV-illumination (squares) and under illumination with the photoactive light (triangles).

4. Conclusions

In the present paper, the results of particular fatigue measurements performed on RF-sputtered PZT films of about 2×10^{-6} m in thickness are presented. Polarization switching characteristics obtained as a result of the application of sinusoidal a.c. electric fields were studied for both the sandwich-type thin film capacitors and the planar-type ones. The planar-type structure enables the study of the influence of the free charge carries induced by UV-illumination on the planar capacitor gap. It has been ascertained that (i) the fatigue of the thin PZT films became noticeable after 10^6 switching cycles for a sandwich-type structure and after 10^9 switching cycles for the thin film capacitors with planar electrodes, (ii) the film illumination during polarization switching accelerates significantly the fatigue process and (iii) the additional fatigue induced by the photoactive light was completely reversible.

Nevertheless, the authors realise that the results of the experiments presented in this paper are of tentative character: there is no experimental evidence available about the effect of the light intensity, temperature, field strength amplitude, etc. Therefore we can hardly make more general conclusions. However, even at this step of the investigations it is safe to say that the effect of fatigue is largely determined by the concentration of the free charge carriers as well as by the presence of transition layers.

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