Journal of Optoelectronics and Advanced Materials Vol. 6, No. 3, September 2004, p. 1059 - 1063

FIRST ORDER REVERSAL CURVES AND HYSTERESIS LOOPS OF FERROELECTRIC FILMS DESCRIBED BY PHENOMENOLOGICAL MODELS

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The increasing parts of the non-symmetric P(E) hysteresis loops of PZT ferroelectric films (First Order Reversal Curves - FORC), were used in order to calculate the FORC distribution in which a separation of the reversible and irreversible contributions to the polarization was obtained. The role of the frequency of the applied field on the switching properties of the ferroelectric films were analyzed by this method. A phenomenological model was used to describe the P(E) hysteresis loops. Using the model parameters determined from the fit of P(E) major loops, minor loops and the FORCs were simulated, at various frequencies and amplitudes of the applied field. The calculated FORC diagram was compared with the experimental one and the limits of the model were discussed.

(Received April 19, 2004; accepted June 22, 2004)

Keywords: Ferroelectric switching, First order reversal curves, Phenomenological model

1. Introduction

The method of investigation of hysteretic systems by means of First Order Reversal Curves (FORC) diagrams initially proposed for geological ferromagnetic systems and lately for recording media, spin glasses and ferrofluids [1-3] was recently applied for ferroelectric films [4-5]. The method involves the measurement of minor hysteresis loops obtained by cycling the sample between saturation and a variable reversal field. Being sensitive to various parameters, including characteristics of the ferroelectric system but also characteristics of the field history and characteristics, the experimental FORC diagrams contain a rich information. In order to disclose the information contained in these diagrams, modelling tools are used and the role of the model parameters on the FORC diagrams are separately investigated [6-8]. Various techniques have been employed for describing the hysteretic behaviour of ferroelectrics [9-11]. Among them, the semi-macroscopic theories combining the phenomenological approach with energy relations give reasonable results and have a small number of effective model parameters. The phenomenological approach initially proposed for soft magnetic materials by Jiles and Atherton [12] was successfully applied for describing the hysteresis loops of PMN ferroelectric relaxors [13]. In this paper, the experimental FORC diagrams obtained for $Pb(Zr,Ti)O_3$ (PZT) thin films are compared with the ones simulated on the basis of the phenomenological model of Jiles-Atherton, with the approximation of Deane [14]. An interpretation of the mechanisms involved in the observed patterns is proposed.

2. Experiment and definition of the FORC diagrams

$Pb(Zr,Ti)O_3$ (PZT) films obtained by Rf - sputtering technique having Pt electrodes were used for experiments [15]. Single pulse hysteresis measurements in 500Hz triangular signal with

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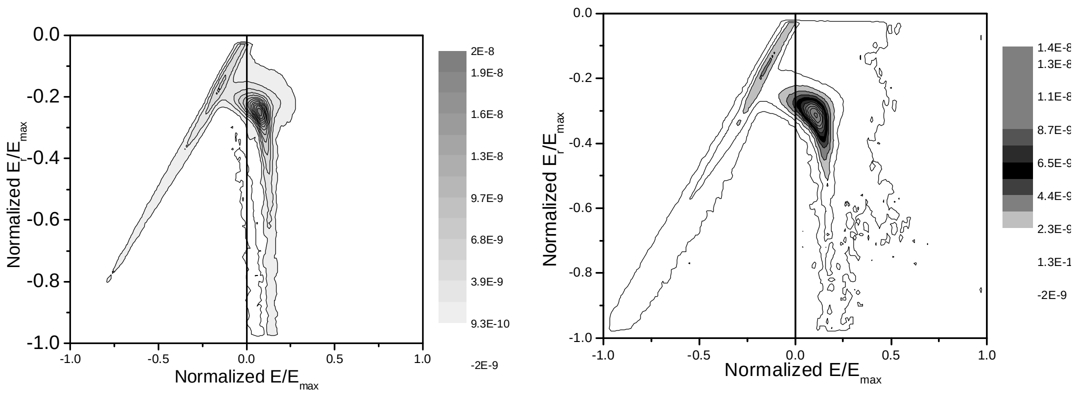
10V maximum amplitude have been performed using a FCE ferroelectric tester (Toyotechnica) according to the sequence: (i) saturation under a positive field $E \geq E_{sat}$ =400kV/cm; (ii) ramping the field down to a reversal value E_r, when the polarization follows the descending branch of the Major Hysteresis Loop (MHL); (iii) increasing the field back to the positive saturation, when the polarization is a function of both the actual field E and of the reversal field E_r [4-5]. The FORC family starting on the descending MHL branch is denoted as $p^{-}_{FORC}(E_r, E)$, similarly, the FORCs $p^{+}_{FORC}(E_r, E)$ can be obtained using the ascending branch of the MHL [4]. The FORC diagram represents a contour plot of the FORC distribution, defined as the mixed second derivative of polarization with respect to E_r and E [2]:

$$\rho^{-}(E_r, E) = \frac{1}{2}\frac{\partial^2 p^{-}_{FORC}(E_r, E)}{\partial E_r \partial E} = \frac{1}{2}\frac{\partial}{\partial E_r}\left[\chi^{-}_{FORC}(E_r, E)\right], \tag{1}$$

in which $\chi^{-}_{FORC}(E_r, E)$ are the differential susceptibilities measured along the FORCs. The distribution $\rho(E_r, E)$ describing the variation of polarization with respect to changes in both the reversal field E_r and the actual electric field E and it was determined using the numerical method proposed in [2]. By changing the coordinates of the FORC distribution from *(E_r,E)* to *$\{E_c=(E-E_r)/2, E_i=(E+E_r)/2\}$*, where E_c and E_i play the role of local coercive and interaction field respectively, $\rho(E_c, E_i)$ becomes identical with the Preisach distribution [16] for systems satisfying the classical Preisach model conditions [17]. The experimental FORC diagrams obtained for PZT film at the frequencies f = 5 Hz and f = 500 Hz are presented in Fig. 1 (a-b). They clearly show reversible and irreversible components with a maximum at E_{rM}=-0.3E_{max}, E_M=0.1E_{max} which has the meaning of the fields for the highest number of switchable units causing the main contribution to the polarization. An asymmetry of the of the irreversible FORC along E_M=0.2E_{max} was explained as due to self-polarizing effect in the film [4-5]. A higher spreading and diffusion of the irreversible part of the FORC distribution is found for higher frequency together with an increasing of the reversible component. The position of the maximum of the irreversible part is slightly shifted towards higher fields for f = 500 Hz. In the present paper, the best fit parameters of the phenomenological model Jiles-Atherton [12], with the approximation of Deane were used for simulating the FORC polarization and FORC diagrams at the two frequencies.



a b

Fig. 1. Experimental FORC diagrams of PZT films at frequencies: (a) 5 Hz and (b) 500 Hz.

3. Theory

In the Jiles-Atherton approach [12] the *P(E)* hysteresis loops was modeled in two steps [12]. Firstly, the ideal anhysteretic polarization is calculated by considering a classical Boltzmann statistics for the probability of dipoles to occupy specific energy states. The hysteretic effect is later added by considering the deviations from the anhysteretic state due to the restriction of domain wall movement by pinning sites in the ferroelectric. In the presence of a scaling polarization P_1, the assumption that dipoles can orient in any direction yields the Langevin formula:

$$P_{an} = P_1 + P_S \left[\coth\left(\frac{E_e}{a} \right) - \frac{a}{E_e} \right] \tag{2}$$

where P_S is the saturation polarization, a is a constant and E_e is the effective field considered as:

$$E_e = (E - E_0) + \alpha(P - P_0) \tag{3}$$

in which αP quantifies the field contributions due to the interdomain coupling and stress effects and (E_0, P_0) are the bias field and respective polarization considered to account for the asymmetries of the hysteresis loops. The hysteresis curve $P(E)$ is obtained by including the reversible P_{rev} and irreversible polarization P_{irrev} resulting from the translation and bending of domain walls pinned at the inclusions of material:

$$\frac{dP_{irrev}}{dE} = \tilde{\delta} \frac{P_{an} - P_{irrev}}{k\delta - \alpha(P_{an} - P_{irrev})} \tag{4}$$

where, within the approximation proposed by Deane [14]:

$$\tilde{\delta} = \begin{cases} 1, \{dE > 0 \; and \; P > P_{an}\} or \{dE < 0 \; and \; P < P_{an}\} \\ 0, otherwise. \end{cases} \tag{5}$$

The parameter k quantifies the average energy required to reorient the domains and it is assimptotically approximated by the coercive field in the soft materials. In our simulations, k parameter was chosen as a Gaussian function of the values of the electric field. The total polarization is thus:

$$P = P_{rev} + P_{irrev}, \text{ in which: } P_{rev} = c(P_{an} - P_{irrev}) \tag{6}$$

The final equation of Jiles-Atherton model used by us in our simulation was:

$$\frac{dP}{dE} = \tilde{\delta} \frac{(1-c)(P_{an} - P)}{\delta k(1-c) - \alpha(P_{an} - P)} + c\frac{dP_{an}}{dE} \tag{7}$$

where $\delta = 1$ on the ascendent and $\delta = -1$ on the descendant branch of the hysteresis loop.

4. Results and discussion

The experimental MHL loops of PZT films at the two frequencies have been firstly fitted with the present model. A good fit was obtained for f=500Hz (Fig. 2 b) and a reasonable one for f=5Hz (Fig. 2 a), due to the space charge effects altering the switching polarization and manifesting in the measured P(E) loops mainly at low-frequency.

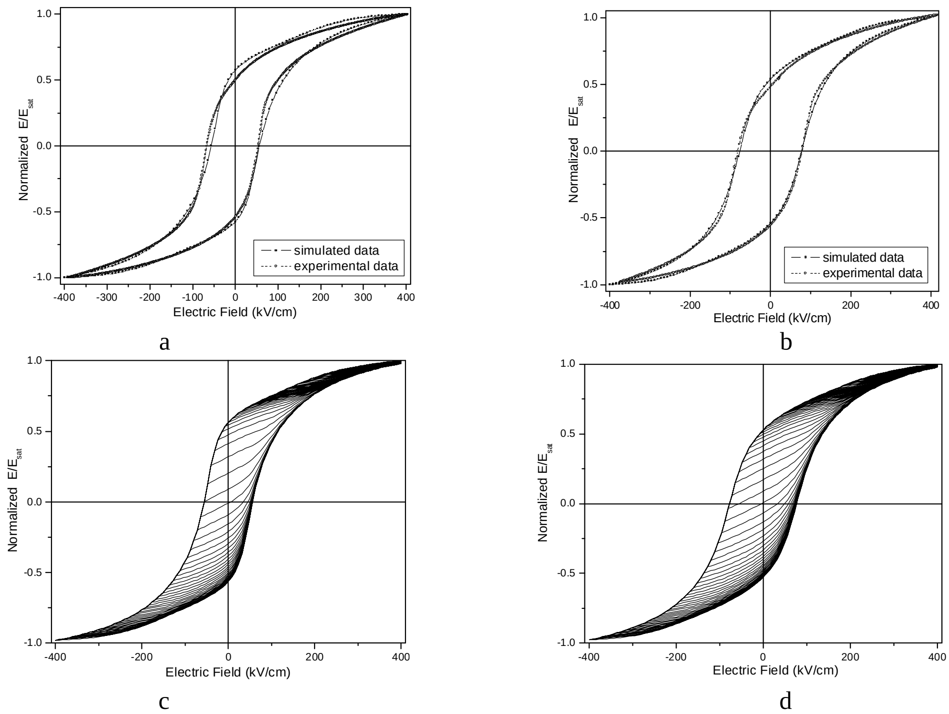


Fig. 2. Experimental, best-fit MHL at (a) 5 Hz and (b) 500 Hz frequency of electric field and computed FORCs on the basis of phenomenological model at: (c) 5 Hz and (d) 500 Hz.

Using the best fit values determined from the MHL, the FORCs were simulated (Fig. 2 c, d). The computed FORC diagrams (Fig. 3 a, b). show similar behaviour with the experimental ones: a reversible component with a maximum at E=0 and an irreversible part located at the same field values as the experimental ones. The model is also able to describe the asymmetry of the FORC diagrams along the E_r axis which previously was considered as coming from the preferential polarization of the switching units located at the bottom electrode – film interface which can easily switch in one direction, but they need higher and distributed fields for switching in the opposite one [4]. This effect, recently found in bulk materials and described within Landau models seems to be more related to intrinsic characteristics of the switchable system than to an interface effect [18]. The model is also able to describe the effect of increasing frequency on the FORCs causing a shift of the maximum towards higher fields and a spreading of the distribution. Due to the fact that the switchable dipoles are not able to follow the fast variation of the applied fields at higher frequency, an apparent higher coercivity is found in the simulated FORCs, like in the experimental diagrams (Fig. 1 b).

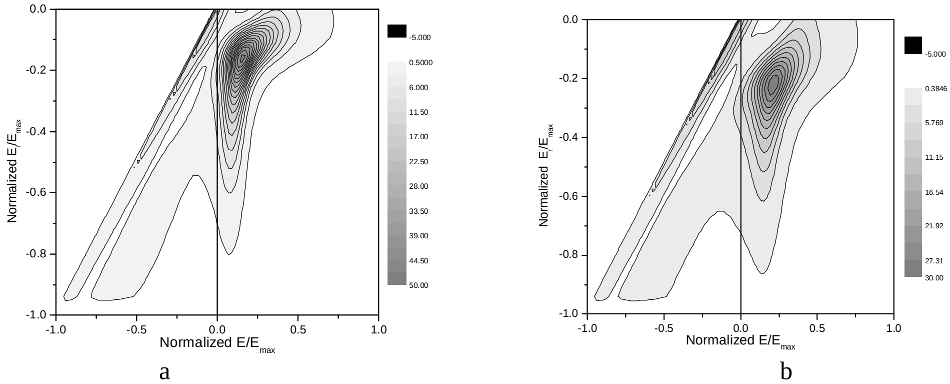


Fig. 3. FORC diagrams computed on the basis of the phenomenological model at (a) 5 Hz and (b) 500 Hz.

5. Conclusions

The experimental FORC distribution obtained for PZT films at different frequencies were compared with ones computed on the basis of a phenomenological model. The Jiles-Atherton theory in the version proposed by Dean was used for the fitting of the MHL. Using the best values fits, the FORCs were simulated. The computed FORC diagrams show similar behavior with the experimental ones. The model is able to describe correctly the reversible and irreversible components on the $P(E)$ loops, the asymmetry of the FORC diagrams along the E_r axis and the effect of increasing frequency on the FORC diagrams.

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