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Dynamic scaling of hysteresis dispersion in ferroelectrics

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Abstract

The dynamic hysteresis of three representative ferroelectric $Pb(Ti_{0.48}Zr_{0.52})O_3$ (PZT), $Bi_2Sr_2Ta_5O_9$ (SBT) and $Bi_4Ti_3O_{12}$ (BTO) thin films and one 1-3 $Pb_{0.95}La_{0.05}TiO_3$ (PLT)/polymer composites, measured using the Sawyer–Tower (ST) method, is summarized, focusing on the dependence of the hysteresis area as a function of the amplitude E_0 and frequency f of the external electric field E. It is found that the one-parameter scaling of the single-peaked hysteresis dispersion applies to the three types of ferroelectric thin films and a Ginzburg–Landau (GL) model system where the domain reversal can be kinetically described by a unique characteristic time. It is confirmed by all of the three ferroelectric systems that the effective characteristic time for the domain reversal is inversely proportional to the field amplitude, while this scaling does not work for the composite system.

Keywords: Ferroelectric hysteresis; Hysteresis dispersion; Dynamic scaling

1. Introduction

The performance of ferroelectric thin films as employed in many applications is intrinsically related to the dynamics of domain reversal driven under a time-varying periodic electric field E. A comprehensive understanding of the domain reversal dynamics becomes extremely essential taking into account the fact that ferroelectric thin film based memory applications have been attractive in the past decade [1–3]. Currently, two issues deserves attention: first, it is obvious that the domain reversal depends on both amplitude E_0 and frequency f of an external field, say $E = E_0 \sin(2\pi ft)$, where t is time. When f is extremely high or low, the dipole alignment in domains may not be perfect because the relaxation process at low f and dipole-switching prohibition at high f as long as E_0 is not very large [3]. Although quite a few techniques have been developed to characterize the dynamics of domain reversal, we may utilize the area A of ferroelectric hysteresis which records the reversal of domains in a cycle to characterize the dynamics, because the area A represents the energy dissipated during one cycle of domain reversal [4–11]. The details of the hysteresis as labels of the domain reversal are no doubt attractive. The area must be small no matter f is high or low and the maximal area is reached when the domain reversal resonates with the external stimulating field. It is our major argument that such resonance behaviors observed at different E_0 can be normalized by a one-parameter scaling, which has been confirmed in several ferroelectric thin film systems.

To look at the second issue, we may come back to the classical framework of domain reversal: nucleation of new domains and their growth through domain wall motion. Classically, the two sequences are treated separately and each can be described by a characteristic time, t_n and t_g, respectively [12]. A definition of such a characteristic time implies a single-peaked distribution of the domain reversal times. However, for any ferroelectric system, it is hard to imagine a distinct separation of the two sequences in time series, and one must treat them as concurrent processes. Therefore, the second issue is whether a third time t_r is available to characterize uniquely the domain reversal. From the point of view of applications, it is also important to predict the characteristic time t_r given an external field with f and E_0 changing over broad ranges, so that device design can be performed in a reliable manner.

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Along this line, the dynamics of ferroelectric hysteresis for several ferroelectric thin films was studied, including ABO_3-type perovskite ferroelectric $Pb(Ti_{0.48}Zr_{0.52})O_3$ (PZT) [6,7], layered-perovskite ferroelectric $Bi_2Sr_2Ta_5O_9$ (SBT) [9] and Nb-doped $Bi_4Ti_3O_{12}$ (BNT) [11], as well as 1–3 $Pb_{0.95}La_{0.05}TiO_3$ (PLT)/polymer composite system [10]. The microscopic mechanisms of domain reversal for these systems may be a little different from one and another, but it would be interesting to find out the common features of these systems in terms of domain reversal dynamics. We also performed a theoretical approach of the dynamics based on the Ginzburg–Landau (GL) model [8]. While all the pure ferroelectric thin films studied earlier show the scaling behavior of the hysteresis dispersion over a broad range of field amplitude, the composite system does not exhibit the scaling property. This scaling means a simple inversely proportional relationship between the effective time t_r and the field amplitude E_0. In this report, we summarize our investigation on the dynamics of hysteresis, focusing on the scaling and the prediction of the characteristic time for domain reversal in these ferroelectric thin films.

2. Outlines of experiments

The dynamic hysteresis for each thin film was measured using the Sawyer–Tower (ST) method, a detailed description of which was presented earlier [6,7,9–11]. In order to ensure the reliability of the data by the ST method, we compare the hysteresis data by the ST method and the data measured by the standard ferroelectric testing unit RT66A at a hysteresis time of 112 ms (Radiant Technologies Inc., NM, USA). For each sample, it was confirmed that the data from the two methods are very consistent with each other, demonstrating the reliability of the presented ST data.

The thin film samples used for the present experiments were prepared using pulsed laser deposition technique (for PZT) on $La_{0.7}Sr_{0.3}MnO_3$-coated $SrTiO_3$ wafers or onto oxidized Si substrates coated with a bottom electrode consisting of a thin TiO_2 adhesion layer plus a sputter-deposited Pt layer (for BNT). The BST thin films were prepared by sol–gel method on the same wafers as BNT. The 1–3 PLT/epoxy composite samples were prepared by a multi-step sol–gel method [10]. The microstructure and ferroelectric property of these thin films were optimized and detailed description of the preparation procedures was given separately in our earlier reports [6–11]. Pt top electrodes of 200 μm in diameter were sputter-deposited at room temperature with a shadow mask. A 10 min post-annealing at 750 °C was conducted to improve the adhesion of the top electrode with the film surface and relax the stress in the films. This procedure of sample preparation represents the standard one for ferroelectric thin films for NvRAM applications.

For the GL model description of the dynamics of domain reversal and hysteresis, a detailed calculation was performed to search for the possibility of one-parameter scaling. This model presents a powerful support on the scaling hypothesis in the framework of the classical domain reversal dynamics [8].

3. Main results and scaling analysis

3.1. Hysteresis dispersion

As an example, we present in Fig. 1(a–e) the measured area A of hysteresis loops as a function of f (so-called hysteresis dispersion A–f) at several values of E_0 for the four thin film systems, and the calculated dispersion from the GL model, respectively. Without exception, all A–f curves show a single-peaked pattern and the tails at $f \to 0$ and $f \to \infty$ tend to zero. The appearance of the peak pattern is obviously due to the resonance between the domain reversal and external field. However, this single-peaked pattern cannot allow us to claim the unique characteristic time for the domain reversal, or, in other words, the distribution of domain reversal time may not be single-peaked. One needs additional analysis on the dispersion in order to evaluate such a characteristic time and corresponding distribution. We shall come back to this issue later.

From the dispersion curves, one understands that at both low and high-f range, the hysteresis area is small due to the full

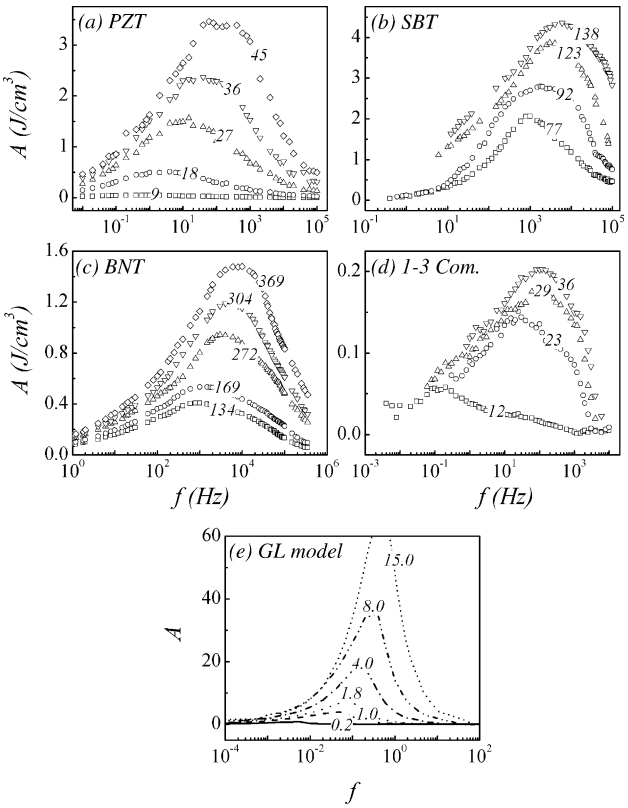

Fig. 1. Measured hysteresis dispersion curves at various field amplitude E_0 for (a) PZT, (b) SBT, (c) BNT, (d) 1–3 composite and (e) the GL model system. The inserted numbers in the plots are the values of E_0.