Scaling on hysteresis dispersion in ferroelectric systems

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The hysteresis area as a function of frequency of the time-varying external electric field—hysteresis dispersion—for ferroelectric $Pb(Zr_{0.52}Ti_{0.48})O_3$ is measured, and the Monte-Carlo simulation on the hysteresis dispersion for a model ferroelectric lattice is performed too. We demonstrate the scaling behavior of the single-peaked hysteresis dispersion for the two ferroelectric systems, predicting a unique effective characteristic time for the domain reversal. This characteristic time shows an inversely linear dependence on the field amplitude as long as the amplitude is high enough that the reversible domain rotation response is negligible. © *2001 American Institute of Physics.*
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Although the problem of domain reversal in ferroelectrics (FE) has been one of the classic topics in the framework of first-order phase transitions,[1] it is attracting special attentions, mainly because of the development of high-speed FE-random access memories and other advanced FE-based electronic devices.[2] Consequently, the domain reversal has been performed and observed *in situ* by means of an atomic force probe or other high resolution probes.[3–5] The domain reversal in these devices is driven under time-varying electric field (ac-type or pulse type), but earlier studies on domain reversal kinetics paid less attention to the frequency dependence. In fact, the domain reversal can be explained by the nucleation-and-growth mechanism.[1] We present a brief remark on this issue under the ac field. Given an external electric field $E(t)$ at time t, says $E(t)=E_0\sin(2\pi ft)$ where E_0 is amplitude and f is frequency, the nucleation rate for new domains can be predicted with a characteristic time τ_n. Also the velocity for a new domain boundary can be defined, with a characteristic time τ_g. Nevertheless, it is well accepted that nucleation and growth occur concurrently and the two sequences overlap each other.

It is physically reasonable to define a third time τ_e, in order to characterize the kinetics of domain reversal. As an assumption, $\tau_e=\sqrt{\tau_n\tau_g}$, which may be called the effective characteristic time. On the other hand, the hysteresis is always observable in accompany with the domain reversal. Similar to the dielectric spectrum, we define the frequency dependence of hysteresis area $A(f)$ at any given E_0 as the hysteresis dispersion. In this letter, we study the scaling behavior of the hysteresis dispersions for $Pb(Zr_{0.52}Ti_{0.48})O_3$ (PZT), from which the kinetics of the domain reversal is understood in a phenomenological sense. Our main conclusion is that the characteristic time τ_e is physically definable and demonstrated by the scaling behavior. This conclusion is supported with our Monte-Carlo (MC) simulation on model ferroelectric lattice.

The experiment was performed by applying the Sawyer–Tower technique to measure the hysteresis loops of (001)-textured PZT samples under various $E(t)$ with f covering 10^{-2}–10^5 Hz and E_0 from 0 to 45 kV/cm. The sample fabrication and the procedure of Sawyer–Tower measurement were described previously.[6] Our MC simulation starts from a two-dimensional squared $L\times L$ lattice with periodic boundary conditions. Each site of the lattice is imposed on with a displacement vector $\mathbf{u}_i$ as the electrical polar vector whose direction is randomly defined on the lattice plane. $\mathbf{u}_i$ at each site is thus proportional to the local spontaneous polarization and subjected to a double-well potential with the nearest-neighboring interaction taken into account. The lattice Hamiltonian can be written as[7]

$$\tilde{H}=\sum_i\left(\frac{p_i^2}{2m}-\frac{a}{2}\mathbf{u}_i^2+\frac{b}{4}\mathbf{u}_i^4\right)-U\sum_{\langle i,j\rangle}\mathbf{u}_i\cdot\mathbf{u}_j-\sum_i\mathbf{E}(t)\cdot\mathbf{u}_i, \tag{1}$$

where $\langle i,j\rangle$ represents that over the nearest neighbors is summed once, p_i is the momentum at site i, a, and b are the double-well potential parameters, U is the ferroelectric ordering factor, m is the mass. $\mathbf{E}(t)$ defines the base direction to which the direction of $\mathbf{u}_i$ or $\mathbf{u}_j$ refers. The Metropolis algorithm employed in our MC simulation was reported previously.[8] The lattice parameters are $L=512$, $kT=1.0$, $a=20$, $b=200$, $U=1.5$ and $\max(\mathbf{u}_i)=0.5$, with 24 orientations being taken by $\mathbf{u}_i$. The lattice polarization $P=(1/L^2)\Sigma\mathbf{u}_i\cdot(\mathbf{E}/|\mathbf{E}|)$ is obtained by averaging over four rounds of data from different seeds for random number generation. The MC is scaled in a unit of mcs and $PE(t)$ is normalized by temperature unit of kT.

As an example, we present in Figs. 1(a) and 1(b) the measured $P-E$ loops for the same PZT sample and the simulated loops for the model lattice, respectively, at various val-

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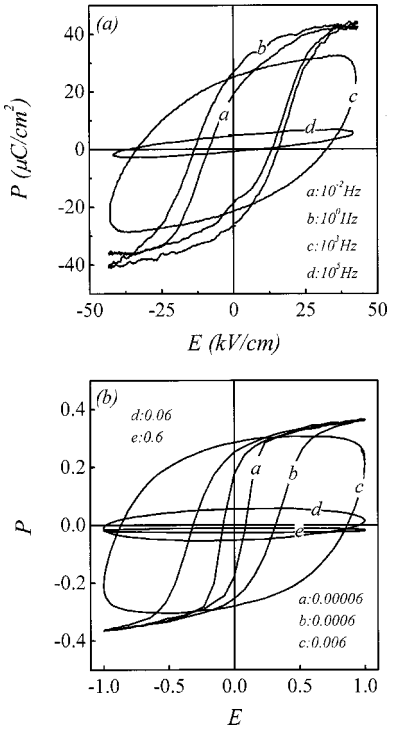

FIG. 1. Hysteresis loops at different f measured for PZT (a) and simulated for the model lattice (b) under fixed field amplitude E_0. The inserted numbers show the values of f.

ues of f but fixed E_0. As expected, the dependence of the hysteresis shape and area on f is remarkable. An excellent similarity in loop shape and its evolution between the two systems is also revealed. At very low f, the loop area is smaller, with lower coercive field and well-saturated tip. With increasing f, the loop area becomes larger (loops a,b) and the loop finally evolves into a tilted ellipse with rounded corner (loop c). Further increasing f results in significant shrinking of the loop along the polarization axis (loop d) before finally collapsing into a tilted line.

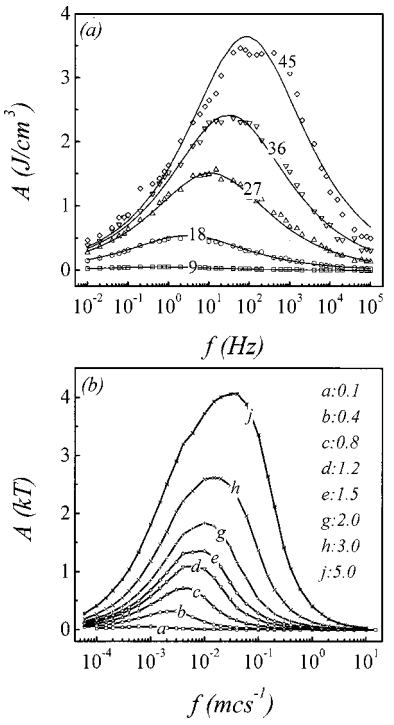

FIG. 2. Hysteresis dispersion $A(f)$ measured for PZT (a) and simulated for the model lattice (b) under different field amplitude E_0. The inserted numbers show the values of E_0.

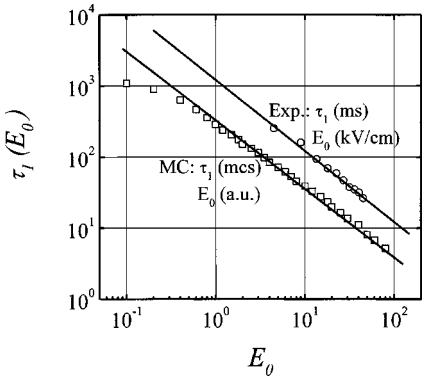

FIG. 3. Unique effective characteristic time for PZT (label "Exp.") and the model lattice (label "MC"). The units for τ_1 and E_0 are indicated in the figure.

It may not be convincing that the PZT loop shows an elliptic pattern at a low frequency ($f \sim 10^3$ Hz), since the well-saturated loop even if f is $\sim 10^7$ Hz or higher was reported.[1] However, it should be emphasized here that the E_0 dependence of the loop shape and area (to be shown later) is significant too, or even more than the f dependence. In our experiments, the maximum E_0 is only 45 kV/cm, far lower than 10^3–10^5 kV/cm applied in the reported measurement.

The measured and simulated hysteresis dispersion curves $A(f)$ at different E_0 are plotted in Figs. 2(a) and 2(b), respectively. For both systems, the dispersion shows a single-peaked pattern with the peak value raising and peak-position increasing when E_0 increases. This single-peaked pattern indicates that a unique characteristic time for domain reversal is probably definable. Suppose the two quantities, τ_n and τ_g, are very different, the dispersion would have two peaks, one appearing at $f \sim \tau_n^{-1}$ and the other at $f \sim \tau_g^{-1}$. Otherwise, the dispersion $A(f)$ at different E_0 should be scalable by the unique characteristic time. To check this scalability, the simplest scheme is to evaluate an arbitrary n-th momentum $S_n = \int_0^\infty f^n A(f)\,df$. However, the integration can not be converged unless $A(f)$ decays faster than $f^{-(n+1)}$ as $f \to \infty$. By redefining the dispersion using $\log(f)$ as a variable, we have the following scaling parameters:[9]

$$
\begin{aligned}
&\gamma = \log(f),\\
&S_n(E_0) = \int_{-\infty}^{\infty} \gamma^n A(\gamma, E_0)\,d\gamma, \quad n = 1,2,...,\\
&\gamma_n(E_0) = S_n(E_0)/S_0(E_0),\\
&n_2(E_0) = S_2(E_0)/S_1^2(E_0),\\
&\tau_1^{-1} = 10^{\gamma_1},
\end{aligned}
\tag{2}
$$

where γ is the modified frequency, γ_n is the nth characteristic frequency [unit: $\log(s^{-1})$], n_2 is the scaling factor, and τ_1 is the effective characteristic time. The evaluated scaling factor $n_2(E_0)$ becomes E_0 independent for both system as long as E_0 is not very small. This independence predicts the scalability of dispersion $A(\gamma)$ by a one-parameter scaling func-

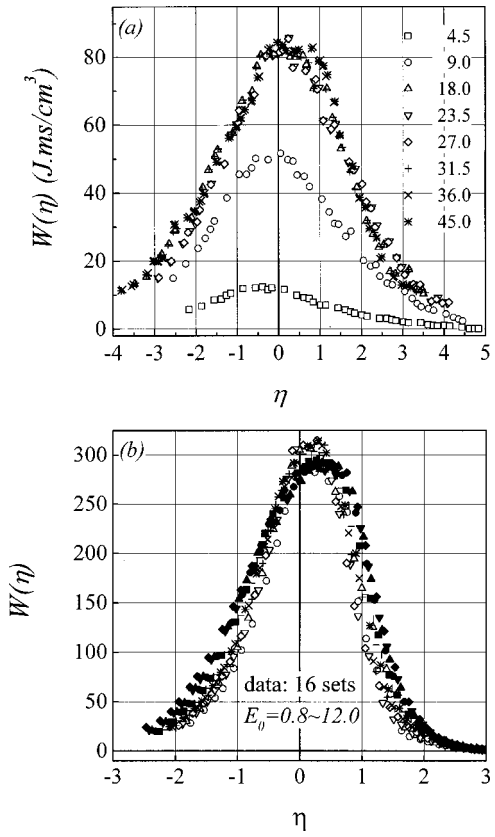

FIG. 4. Scaling-plotting of hysteresis dispersions for PZT (a) and the model lattice (b). The inserted numbers are the values of E_0.

tion. The dependence of τ_1 on E_0 is given in Fig. 3 where the two straight lines represent the inverse linear relationship. The as-defined τ_1^{-1} in Eq. (2) seems to be linearly correlated with E_0 as long as the latter is not very small.

To construct the one-parameter scaling function, we consider the fact that the only variable for $A(f)$ is frequency f, which is the reciprocal of time. Its dimensionality can not be other than one. Therefore, the scaling function of the following generalized form yields

$$W(\eta)=\tau_1 A(\gamma,E_0),$$
$$W(\eta)=f_1^{-1}A(\gamma,E_0), \tag{3}$$

with the scaling variable $\eta=\log(f\tau_l)$ and the effective characteristic frequency $f_1=\tau_1^{-1}$.

Replotting all the measured and simulated dispersion curves $A(\gamma)$ according to Eq. (3) produces Figs. 4(a) and 4(b), respectively. Except for the cases of very small E_0, all the curves fall onto the same curve within the statistical errors, demonstrating the scaling property of the hysteresis dispersion for both systems. This also indicates that for the domain reversal in PZT and the model lattice investigated here, respectively, there indeed exists a unique characteristic time which is τ_l or a quantity proportional to τ_1, by which the hysteresis dispersion can be uniquely described if the general scaling function $W(\eta)$ is available.

The failure to scale the dispersion at very small E_0 is ascribed to the fact that time τ_1 is much shorter than that given by relationship $\tau_1 \propto E_0^{-1}$. Generally speaking, the domain reversal for spin system at very low field, say, at a field close to the stationary coercivity, may be dominated by reversible domain rotation instead of irreversible domain boundary migration. Because the former has a much faster response speed than the latter, the as-derived τ_1 is, of course, shorter. As for the experimental relevance for the relationship $\tau_1 \propto E_0^{-1}$, previous studies on domain nucleation and growth in $BaTiO_3$ under dc field[10] revealed that the nucleation rate can be expressed as $p(1/ms)\propto E_0^{2/3}$, so that $\tau_n \propto E_0^{-2/3}$ can be predicted. The domain boundary migration velocity takes the form $\nu \propto E_0^{4/3}$, from which $\tau_v \propto E_0^{-4/3}$ is derived. Therefore, we have the linear relationship $\tau_1 \propto \tau' = \sqrt{\tau_n \tau_v} \propto E_0^{-1}$, consistent with our findings for PZT and the model lattice. The collected data by Scott *et al.* on $KNbO_3$ also supported this relationship with the typical switching time of $\sim 10^{-1}$ ms for domain reversal under a field of 100 kV/cm.[2,11] Note here that τ_1 is just the effective characteristic time, which is proportional to the realistic time, τ_n or τ'.

In summary, we have demonstrated the scaling property of the hysteresis dispersion for ferroelectric systems from the measured hysteresis for PZT and the Monte-Carlo simulated domain reversal sequence for a ferroelectric model lattice. An effective unique characteristic time is predicted, with which the domain reversal can be fully described. It has been revealed that the characteristic time is inversely proportional to the amplitude of the applied field as long as the latter is not very small.

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[1] J. D. Gunton, M. San Miguel, and P. S. Sahni, in *Phase Transitions and Critical Phenomena*, edited by C. Domb and J. L. Lebowitz (Academic, London, 1983), Vol. 8, p. 1.
[2] J. F. Scott, Ferroelectr. Rev. **1**, 1 (1998).
[3] A. Gruverman, O. Auciello, and H. Tokumoto, Annu. Rev. Mater. Sci. **28**, 101 (1998).
[4] G. D. Hu, J. B. Xu, and I. H. Wilson, Appl. Phys. Lett. **75**, 1610 (1999).
[5] M. Alexe, C. Harnagea, W. Erfurth, D. Hesse, and U. Gösele, Appl. Phys. A: Mater. Sci. Process. **A70**, 247 (2000).
[6] J.-M. Liu, H. P. Li, C. K. Ong, and L. C. Lim, J. Appl. Phys. **86**, 5198 (1999).
[7] T. Janssen and J. A. Tjion, Phys. Rev. B **24**, 2245 (1981).
[8] J.-M. Liu, Q. C. Li, W. M. Wang, X. Y. Chen, G. H. Cao, X. H. Liu, and Z. G. Liu, J. Phys.: Condens. Matter **13**, L153 (2001).
[9] A. Craevich and J. M. Sanchez, Phys. Rev. Lett. **18**, 1308 (1981).
[10] H. L. Stadler and P. L. Zachmanids, J. Appl. Phys. **34**, 3255 (1963); **35**, 2895 (1964).
[11] J. F. Scott, R. B. Godfrey, C. A. Araujo, L. McMillan, H. B. Meadows, and M. Golabi, in *Proceedings of the International Symposium on Application of Ferroelectrics* (IEEE, Piscataway, 1986), p. 569.