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D. P. Chen and J.-M. Liu

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Dynamic hysteresis of tetragonal ferroelectrics: The resonance of 90°-domain switching

D. P. Chen and J.-M. Liu

Laboratory of Solid State Microstructures, Nanjing University, Nanjing 210093, China
Institute of Advanced Materials, School of Physics, South China Normal University, Guangzhou 510006, China

Consequently, the following power law scaling behaviors are realized via the nucleation-and-growth process based on the ferroelectric resonance mode may be called as the dipole switching mode for \( \frac{A}{2} \equiv \frac{x}{2} \) switching: avalanche-like switching of those dipoles. As a straightforward argument, this mode, coined as the cluster switching mode, is schematically shown in Fig. 1(c) left column. The spectrum \( A_{d}(\omega) \) arising from this mode may be single-peaked too and plotted by the dashed dot line. The spectrum \( A(\omega) \) should be a sum of \( A_{d}(\omega) \) and \( A_{d}(\omega) \).

In this letter we address this issue. We start from the Ginzburg-Landau phenomenological theory for a tetragonal FE lattice. In our calculation, we take lattice constant \( a_{0} \) (~1.0 nm), Landau coefficient \( a_{0} \) in the free energy, and polarization \( P_{0} \) of the object FE material at \( T = T_{f} \) far below \( T_{c} \) as reference, where \( a_{0} = a_{10}(T_{f} - T_{c}) \) and \( a_{10} > 0 \). All free energy terms are normalized by term \( [a_{0}]^{2} P_{0}^{2} \) which has an energy unit. Thus, one has normalized \( T = T/T_{c} \) with \( T_{0} = T_{c}/(T_{f} - T_{c}) \). We perform the Monte Carlo simulation on a two-dimensional (2D) \( L \times L \) square lattice with periodic boundary conditions (\( L \) and all spatial coordinates are normalized by \( a_{0} \), using the Metropolis algorithm. On each site, an electric dipole \( P(r) = (P_{x}, P_{y}) \), normalized by \( P_{0} \), and an elastic displacement vector \( u(r) = (u_{x}, u_{y}) \), normalized by \( a_{0} \),
where dimensionless factor $t_{dp} = (8\pi\kappa/\varepsilon_0)^{-1}$ with $\varepsilon$ the dielectric permittivity and we take $t_{dp} = 1$; $r$ and $r'$ are the spatial coordinates. This term favors an anti-parallel alignment of dipoles, and it is of long-range. A realistic calculation is done by either Fourier transform or finite truncation treatment,22 while for 2D lattice this treatment is precise as long as the truncating distance $R$ is big ($R = 8$ in our simulation).

The spatial strain field is facilitated with elastic energy $f_{el}$ yielding

$$f_{el} = \frac{1}{2} C_{11}^* (u_{xx}^2 + u_{yy}^2) + C_{12}^* u_{xx} u_{yy} + \frac{1}{2} C_{44}^* u_{xy}^2$$

with $C_{11}^*$, $C_{12}^*$, and $C_{44}^*$ are the normalized values of the elastic coefficients $C_{11}$, $C_{12}$, and $C_{44}$. Given the linear quadratic coupling of local elastic strain with the local dipole, one has

$$f_{ep} = -(t_{ep} u_{xx} (q_{11}^* P_x^2 + q_{12}^* P_y^2) + u_{xy} (q_{11}^* P_x^2 + q_{12}^* P_y^2) + q_{44}^* u_{xy} P_x P_y),$$

where $t_{ep}$ is a dimensionless pre-factor, $q_{11}^*$, $q_{12}^*$, and $q_{44}^*$ are the normalized values of the electrostrictive coefficients $q_{11}$, $q_{12}$, and $q_{44}$. This term is the core ingredient favoring the 90°-domain structure. Finally, electrostatic energy $f_{ep}$ is given as

$$f_{ep} = -(E_x P_x + E_y P_y),$$

where electric field $E = (E_x, E_y)$ is normalized by term $|\alpha_0| P_0$, taking $E_x = E(t) = E_0 \sin(2\pi \omega t)$ and $E_y = 0$.

We consider two cases: one case with $t_{ep} = 1$, and the lattice is dominated with the 90°-domain structure; the other by setting $t_{ep} = 0$, so the single domain or 180°-domain structure is preferred. The parameters for the simulation are listed in Table I and were used extensively in literature.22,23 The Monte Carlo simulation procedure was described previously,22,24,25 and no details are given here. The finite lattice size effect is negligible as long as lattice size $L$ is much bigger than the 90°-domain width. The unit for frequency $\omega$ is inversely measured by the Monte Carlo step (mcs) which counts $L^2$ dipole flip attempts. One mcs = 1 corresponds to the characteristic rate $\omega_0$ for the dipole flip attempts, which is supposed to follow the thermal-activation law $\omega_0 = \nu \exp(-F_0/k_B T)$, with $F_0$ the potential barrier and $\nu$ the frequency for the basic dipole excitation mode.

We first look at the case of $f_{ep} = 0$. Extensive simulation on the $P$-$E$ loops at various $\omega$ and $E_0$ are performed, and the hysteresis evolves from thin and well saturated loop into square and fat loop until unsaturated round one, with increasing $\omega$, as shown in Fig. 2(a) and discussed in literature.5,7,9 In Fig. 3(a) are plotted the three $A(\omega)$ curves for this case. All these curves show the single-peaked pattern, with the peak height and location shifting upward and rightward with increasing $E_0$. From the log-log plot (inset), one observes the power law scaling in both the $\omega \to 0$ and $\omega \to \infty$ limits. In correspondence, the domain structures at several states

![Diagram](https://example.com/diagram.png)
associated with the $P$-$E$ loop are presented in the left column of Fig. 3. While the initial state is occupied with $180^\circ$-domains (state O), it is seen that state C has the nominal coercive field shows slower shrinking for the single-domain structure than that for the single-domain $\theta \sim 0$ and state F has $\theta \sim \pi$ over the whole lattice, in spite of the local fluctuations. These results show that the $P$-switching is dominated by the dipole switching mode and the resonance occurs at time $\tau_{do}$.

For $f_s > 0$, the lattice is dominated with the $90^\circ$-domains. At $t_{es} = 1$, the simulated loops at several $\omega$ are shown in Fig. 2(b), and the area spectra at three different $E_0$ are presented in Fig. 3(b). No substantial difference in the loop pattern between the two cases $t_{es} = 0$ and $t_{es} = 1$ is shown. The minor differences are (1) the $90^\circ$-domain structure shows a rounder loop pattern, while the single-domain lattice generates a more square-like loop; (2) with decreasing $\omega$, the nominal coercive field shows slower shrinking for the $90^\circ$-domain structure than that for the single-domain structure.

The substantial difference between the two cases is illustrated by $A(\omega)$. For $t_{es} = 1$, it is surprising that $A(\omega)$ at each $E_0$ demonstrates two well defined peaks instead of one peak in the case of $t_{es} = 0$, similar to the solid line in Fig. 1(b). For details, first, the peak at the high-$\omega$ side is located at $\tau_{do}$, similar to the case of $t_{es} = 0$, while the low-$\omega$ peak is located roughly at $\tau_{do} \sim 10^5$ mcs. Time $\tau_{do}$ would be longer if $t_{es}$ is larger. Second, the peaks at $\tau_{do}$ and $\tau_{di}$ increase in height and rightward with increasing $E_0$, while this response is much more significant for the peak at $\tau_{do}$ than that at $\tau_{di}$, indicating that the dynamic response associated with the peak at $\tau_{do}$ is non-robust. Third, $A(\omega)$ at both the low-$\omega$ and high-$\omega$ limits still follows the power-law scaling, as shown in the inset of Fig. 3(b). The scaling exponents are roughly $E_0$-independent but strongly $T$-dependent.

To understand the origin of peak at $\tau_{do}$, one snapshots the domain structures at several states. Referring to state C and state F, it is observed that even passing forward to state B and

![FIG. 2. (Color online) Simulated $P$-$E$ loops for the FE lattice with $t_{es} = 0$ (a) and $t_{es} = 1$ (b). The value of $\omega$ for each loop is labeled numerically with unit $mcs^{-1}$, $T_s = 0.05$.](image_url)

![FIG. 3. (Color online) Evaluated $A(\omega)$ for (a) $t_{es} = 0$ and (b) $t_{es} = 1$ respectively, at three different $E_0$, $T_s = 0.05$. The snapshots of the FE domains for $t_{es} = 0$ (left column) and $t_{es} = 1$ (right column) at states O, C, and F (see Fig. 1(a)). The domain orientations are marked by colors, and angle $\theta$ is defined with respect to the x-axis. The blue and red arrows indicate the domain orientations.](image_url)
state C from state O, the 90°-domain pattern can be somehow maintained, blueprinted by the green stripes in state C. Inside these stripes are accommodated with a number of dipole clusters. At state F, the reversed domain structure still shows the yellow stripe-like pattern which is opposite in polarization to those clusters in state C. This demonstrates that the dipoles in these clusters reverse in an avalanche/collective manner, i.e., the cluster switching mode in these clusters. The reverse process of the 90°-domains as an additional sequence makes substantial contribution to the dynamic hysteresis. In fact, quite earlier experiment already revealed the so-called trans-domain cooperative switching in PbZrxTi1-xO3 ceramics with the 90°-domain structure.26

To separate the contributions from the two modes, one consults to Fig. 4(a) where the evaluated \(A(\omega)\) for \(t_{es} = 1\) and \(t_{es} = 0\) at \(E_0 = 4.0\) are plotted. Although the two spectra are different in the overall sense, their behaviors at the high-\(\omega\) limit remain identical. Keeping in mind that \(A(\omega) = A_d(\omega) + A_{ds}(\omega)\), one has \(A(\omega) = A_d(\omega)\) at \(\omega \to \infty\). It is reasonable to assume that \(A_d(\omega)\) at \(f_{es} > 0\) has the same distribution as \(A(\omega)\) at \(f_{es} = 0\), although their magnitudes are different but proportional to each other. Therefore, \(A_d(\omega)\) and \(A_{ds}(\omega)\) at \(t_{es} = 1\) can be simply obtained, as shown in Fig. 4(b).

As expected, both \(A_d(\omega)\) and \(A_{ds}(\omega)\) show the single-peaked pattern with the peaks located, respectively, at \(\tau_{ds}\) and \(\tau_{ds}\). The ratio \(\tau_{ds}/\tau_{ds}\) is \(\sim 630\), indicating that the cluster switching occurs at much lower \(\omega\) than the dipole switching. In addition, both \(A_d(\omega)\) and \(A_{ds}(\omega)\) exhibit the power-law frequency scaling at \(\omega \to 0\) and \(\omega \to \infty\), respectively, with similar exponents \(z\) and \(b\), thus allowing the same scaling behavior and exponents for \(A(\omega)\). More simulations on cases where \(E(t)\) is applied along directions other than the \(x\)-axis reveal similar behaviors although the scaling exponents may be different. These results suggest that the cluster switching mode can not be ignored for FE materials with 90°-domain structure. However, as \(E_0\) is extremely big or \(T\) is high, this mode will be much less pronounced, as identified in some experiments.8–11 In evidencing the above argument, one looks at the effect of \(T\). We present the \(A(\omega)\) data with \(E_0 = 4.0|z|P_0\) at several \(T^*\) in Fig. 5(a). It is seen that with increasing \(T^*\), the two-peaked pattern transits gradually into single-peaked one. The peak or anomaly at \(\tau_{ds}\) nearly disappears at \(T^* = 0.30\).

We evaluate the power-law scaling exponents (\(x\), \(b\)) at different \(T^*\) and present them in Fig. 5(b). While \(b = 1.0\) is roughly \(T\)-independent, \(x\) increases with increasing \(T^*\), from \(x \sim 0.23\) at \(T^* = 0.01\) to \(x \sim 0.52\) at \(T^* = 0.50\), and then tends to be saturated. In fact, for the low-\(\omega\) behavior, the high quality PbZrxTi1-xO3 epitaxial thin films showed \(E_r(\omega) \sim \omega^\gamma\) with \(\gamma\) increasing with increasing \(T\), while the measured \(P_r(\omega)\) remains well saturated,11 consistent with the present simulation. Similar results were obtained in PbZrxTi1-xO3 ceramics.13,27 Theoretically, it was argued that \(x\) is \(T\)-dependent in the similar way as revealed experimentally.14 On the other hand, exponent \(\beta\) is insensitive to \(T^*\) and \(\beta = 1.0\) seems to be universal. In the present case, no substantial contribution from the cluster switching mode in the high-\(\omega\) limit is observed, thus \(\beta = 1.0\) applies. It is also understood that thermal fluctuations would have more remarkable impact on the cluster switching mode than on the dipole switching mode, resulting in disappearance of the former mode in the high-\(T\) range. Regarding the amplitude exponents \(m\) and \(n\), it is obtained that exponent \(n \sim 2.0\) applies to all the cases, which is nearly \(T\)-independent either. However, given a fixed \(T\), the evaluated \(m\) at \(f_{es} = 0\) is smaller than that at \(f_{es} = 0.05\). It increases slightly with increasing \(T\).

**FIG. 4.** (Color online) (a) Simulated \(A(\omega)\) for \(t_{es} = 0\) and \(t_{es} = 1\). (b) Simulated \(A(\omega)\) for \(t_{es} = 1\), and its two components \(A_d(\omega)\) and \(A_{ds}(\omega)\). \(T^* = 0.05\) and \(E_0 = 4.0|z|P_0\).

**FIG. 5.** (Color online) (a) Simulated \(A(\omega)\) for \(t_{es} = 1\) at different \(T^*\) as labeled. (b) As-evaluated scaling exponents \(x\) and \(b\) as a function of \(T^*\) (the dashed lines indicate the theoretically predicted exponents). \(E_0 = 4.0|z|P_0\).
So far, no clear evidence supporting the existence of the cluster switching mode and its significance is available. However, it is still possible to unveil some indirect evidences. Consulting to available data on the dynamic hysteresis in several FE materials,8–11,13 one finds that exponent $a$ is smaller when $E_{0}$ is relatively lower. Keeping in mind that it is technically difficult to access very low frequency for measuring $A_{c}(\omega)$, those data used for evaluating exponent $a$ were obtained at $\omega > 0.1$ Hz. This $\omega$-range may overlap with the resonance frequency for the cluster switching mode. Therefore, the evaluated $a$ would be smaller at lower $E_{0}$ since $A_{d}(\omega)$ becomes more remarkable. Surely, in realistic FE materials, the domain structure is more complicated than pure 90°-domains, making an experimentally identifying of $A_{d}(\omega)$ tougher.

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