Antiferroelectric polarization switching and dynamic scaling of energy storage: A Monte Carlo simulation

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The polarization-electric field hysteresis loops and the dynamics of polarization switching in a two-dimensional antiferroelectric (AFE) lattice submitted to a time-oscillating electric field $E(t)$ of frequency $f$ and amplitude $E_0$, is investigated using Monte Carlo simulation based on the Landau–Devonshire phenomenological theory on antiferroelectrics. It is revealed that the AFE double-loop hysteresis area $A$, i.e., the energy loss in one cycle of polarization switching, exhibits the single-peak frequency dispersion $A(f)$, suggesting the unique characteristic time for polarization switching, which is independent of $E_0$ as long as $E_0$ is larger than the quasi-static coercive field for the antiferroelectric–ferroelectric transitions. However, the dependence of recoverable stored energy $W$ on amplitude $E_0$ seems to be complicated depending on temperature $T$ and frequency $f$. A dynamic scaling behavior of the energy loss dispersion $A(f)$ over a wide range of $E_0$ is obtained, confirming the unique characteristic time for polarization switching of an AFE lattice. The present simulation may shed light on the dynamics of energy storage and release in AFE thin films.

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I. INTRODUCTION

It was not until 1951 when Kittel investigated and put forward related phenomenological theory on antiferroelectric (AFE) materials as a subclass of ferroelectric materials.1 In an AFE lattice, unlike a ferroelectric (FE) one, adjacent electric dipoles are oriented in opposite directions. Thus, its total spontaneous polarization $P$ is zero under zero electric field, since the adjacent dipoles cancel out each other. Accompanying with the double-loop hysteresis of electric polarization $P$ against electric field $E$, an AFE system shows quite different characteristics from an FE system. The phase transitions, domain structure, and polarization switching of an AFE lattice, exhibiting some analogies with those of an FE lattice, also depend strongly on temperature $T$, electric field $E$, microstructural details, and so on.2,3 It is noted that these issues have been much less concerned due to the limited practical applications of AFE materials in comparison with FE materials.

Nevertheless, one exception is the application potentials of AFE materials in high-energy-storage capacitors, which was suggested as early as 1961 by Jaffe and has been continuously investigated mainly from experimental aspects.4–8 When an electric field $E$ is applied to an AFE material in a capacitor structure, the induced AFE-FE phase transitions take place, which thus stores the input energy. After the field is removed, the reversal FE-AFE transitions occur rapidly,9 leaving the input energy to be stored inside the capacitor at the open-circuit state and allowing its rapid release upon the connection to external loads. The advantage of AFE energy storage and release lies in the fact that an AFE material usually has relatively high dielectric constant but also high dielectric breakdown field on one hand, and on the other hand the field-induced double-loop hysteresis has a very small remnant polarization, giving rise to high energy release efficiency. In other words, except for a small heat loss due to the polarization switching, most of the input energy can be released in this cycle. Additionally, AFE materials possess good fatigue endurance.10,11

So far more than 100 AFE materials have been synthesized, among which PbZrO3 (PZO) represents one of the most popular ones.3,12–14 In spite of the attractive high-energy-storage application potentials of these materials, substantial dependences of the energy storage performance on the AFE-FE-AFE transition cycle and microstructural details have been repeatedly investigated,6,11,15–17 raising quite a few of issues on the complexity in optimizing the energy storage performance. The three most concerned issues are: (1) the stored energy density $W_0$; (2) the energy release efficiency $\eta$ which is defined as the ratio of releasable energy density $W$ over $W_0$ ($\eta = W/W_0$) in one hysteresis cycle; and (3) the thermal and frequency stability and the energy storage/release cycling lifetime. Physically, the three issues are relevant with the so-called dynamic hysteresis in an AFE system. This dynamic hysteresis deals with the polarization switching behaviors in response to temperature $T$ and time-varying electric field $E(t)$ by generating a hysteresis from which the two parameters $W$ and $W_0$ can be evaluated, where $A = W_0 - W$ is the hysteresis loop area $A = \int PdE$ in one cycle and efficiency $\eta = 1 - A/W_0$. In particular and as a conventional scenario, the dynamic hysteresis treats the frequency dispersion $A(f)$ and amplitude dependence $A(E_0)$, or...
simply $A(f, E_0)$, at a given $T$, where parameters $f$ and $E_0$ are defined by time-varying electric field $E(t) = E_0 \sin(\omega t)$ with $\omega = 2 \pi f$ and $t$ the time. This physics can be schematically illustrated in Fig. 1 where a model double-loop hysteresis for a typical AFE material is sketched.

Surely, a thorough understanding of the dynamic hysteresis for an AFE lattice and associated polarization switching becomes imperative for efficient energy-storage device design. Our target is to attain the largest stored energy density $W$ and highest energy release efficiency $\eta$ simultaneously. At the same time, the thermal stability (i.e., $T$-dependence of $A(f; E_0)$) is also important, which will be discussed elsewhere. However, very differently, the dynamic hysteresis for conventional FE materials has been extensively investigated since 1990s,18–20 These studies revealed the intrinsic relationship between the dynamic hysteresis and FE domain switching. For an FE lattice, the domain switching can be kinetically described by a mono-dispersive domain size distribution and domain wall motion speed, resulting in the well-known single-peak frequency dispersion $A(f)$, while the domain wall motion speed is dependent of field amplitude. In details, for a time-varying $E(t)$, the frequency dispersion $A(f)$ of an FE lattice can be scaled by

$$A(f) = A_0 + E_0^p |f|^g \left(\frac{f}{E_0}\right)^c,$$

where $A_0$ is the area in the $f \rightarrow 0$ limit counting the effect from non-dynamic origins, $a$, $b$, and $c$ are the scaling exponents, and $g$ is a non-monotonic function which meets $g(x) \Rightarrow 0$ as $x \Rightarrow 0$ or $\infty$.19,21 This scaling reflects the nature of a unique characteristic time $\tau$ for domain switching at a given $E_0$. In the simplest cases, this scaling behavior can be approximately described in the two extreme ends of frequency $f$:18,22

$$\begin{cases} A(f, E_0) \propto E_0^{2/3} f^{1/3} & \text{as } f \rightarrow 0 \\
A(f, E_0) \propto E_0^c f^{-1} & \text{as } f \rightarrow \infty, \end{cases}$$

noting that the scaling exponents to $E_0$ and $f$ may be different from the stated values here, as discussed for various theoretical models19,21,23 and experiments on different materials.24–26

It is clear that the scaling behavior grasps the core nature of domain switching for an FE system.

The dynamic hysteresis and scaling behavior discussed earlier may not be applicable directly to an AFE lattice. For two stubbornly coupled antiparallel FE sublattices, no clear domain concept can be defined in the AFE state unless one sublattice is at least partially switched by electric field. The AFE-FE switching sequence may be kinetically scaled by one characteristic time ($\tau_{c,1}$) too. This switching sequence completes the AFE-FE transitions. Subsequently, the FE domains in the whole lattice may experience growth and coarsening, which is scaled by another characteristic time ($\tau_{c,2}$). Therefore, it seems that the dynamic hysteresis for an AFE lattice may not be scalable by one characteristic time but two times if any.

In fact, some experiments on the scaling behavior of AFE systems were reported. Kim and Kim proposed two scaling relations of the AFE hysteresis area $A(f, E_0)$ in a mixed AFE betaine phosphate-arsenate (BP0.99Na0.01) crystal at a frequency below 200 Hz.27 Chen et al. established the scaling behavior of energy storage density $W$ for the saturated double-loop hysteresis of Pb0.99Nb0.02[(Zr0.60Sn0.40)0.95Ti0.05]O3.28 However, these experiments covered relatively narrow $f$-range, which is insufficient for evaluating the dynamic hysteresis. More experimental data covering broader $f$ and $E_0$ ranges are needed, which will be helpful for not only checking theoretical model if any but also revealing the underlying physical scenario for the dynamic hysteresis. On the other hand, there have been only a few simulation reports on the polarization switching of AFE materials.29,30 In addition, no simulation on the scaling studies of AFE materials has been available to the best of our knowledge. We are only aware of the work of Misirlioglu et al. which deals with a two-dimensional Ising model to simulate the FE-AFE hysteresis.31

In this work, we address the polarization switching and its scaling behavior in a model AFE lattice. For ferroelectrics, one may adopt phase-field model to obtain the domain structures and to track its evolution.32 However, it is rather difficult to simulate the situation of AFE.33 As discussed earlier, we can simply divide an AFE lattice into two coupled FE sublattices and consider a coupling energy between them to favor anti-parallel dipole alignment. We start from the Landau–Devonshire phenomenological theory on a tetragonal AFE lattice and present domain structures at different states associated with a double-loop $P$-$E$ hysteresis. Subsequently, we pay attention to the energy loss, i.e., hysteresis area $A(f, E_0) = \int |P|dE$, and the recoverable energy $W = |E|dP$ (upon an electric discharging), which are both sketched in Fig. 1. It will be shown that the frequency dispersion can still be scaled by one characteristic time instead of two times if field amplitude $E_0$ is sufficiently large over the quasi-static field for AFE-FE transition, $E_{AFE1}$, as shown in Fig. 1. Based on the simulation results, one may derive some conclusion on the energy storage/release as a function of $f$ and $E_0$.

Prior to the model description and results presentation, the difficulty in experimental checking of our simulation results should be mentioned. As subsequently shown, in spite of some qualitative consistencies between our proposed scaling behaviors and existing experimental data, to be discussed

![FIG. 1. A sketch of energy storage for an AFE system as shown in a double-loop P-E hysteresis, where the coercive field $E_{AFE1}$ for the AFE-FE transition and field $E_{AFE2}$ for the FE-AFE transition are marked.](image-url)
later, a quantitative comparison becomes impossible since so far no any physical parameters in the framework of the Landau–Devonshire phenomenological theory on any AFE system are available. The available experimental data are insufficient for a convincing comparison. Therefore, our simulation conclusion would appeal for further experimental verification, especially with respect to AFE domain structure and dynamic hysteresis.

II. MODEL AND SIMULATIONS

A. Model description

We utilize the Monte Carlo simulation to track the dynamic hysteresis driven by field \( E(t) \) for a two-dimensional \( L \times L \) AFE lattice.\(^{34-36} \) The lattice consisting of two interpenetrated FE sublattices labeled \( \alpha \) and \( \beta \), respectively, is applied with the periodic boundary conditions. Each site in sublattice \( \alpha \) is surrounded with four nearest neighbor sites in sublattice \( \beta \) and four next nearest neighbor sites in \( \alpha \) and vice versa. On each site are imposed two order parameters: electric dipole vector \( P(x,y) = (P_x, P_y) \), normalized by \( P_0 \), and displacement vector \( u(x,y) = (u_x, u_y) \), normalized by lattice constant \( a_0 \). This lattice is employed as an approach to an AFE thin film lattice where all electric dipoles are aligned in the \( x \)-\( y \) (\( a \)-\( b \)) plane. This approach seems to be over-simplified but our main motivation is to make a qualitative insight into the energy storage/release associated with an AFE thin film capacitor.

Based on the Landau phenomenological description of a tetragonal FE lattice,\(^{37} \) our simulation starts from the total free energy on this AFE lattice

\[
F_{\text{total}} = F_{ab} + \sum_{\alpha, \beta} F_{ld} + F_{gr} + F_{ela} + F_{ele},
\]

where \( F_{ab} \) is an exclusive term associated with the exchange interaction between sublattices \( \alpha \) and \( \beta \), while \( F_{ld} \), \( F_{gr} \), \( F_{ela} \), and \( F_{ele} \) are the Landau potential, gradient energy, elastic energy, and electric energy of sublattice \( \alpha \) or \( \beta \), respectively.\(^{37} \) For simplicity, \( F_{gr}, F_{ela}, \) and \( F_{ele} \) consider only the interaction between electric dipoles from the identical sublattice.

For accounting the AFE interaction, we follow the standard treatment of term \( F_{ab} \) which favors the anti-parallel alignment of the nearest dipole pairs\(^{1} \)

\[
F_{ab} = J \sum_{\langle i,j \rangle} P_i \cdot P_j,
\]

where \( \langle i,j \rangle \) means a summation over all the nearest-neighboring dipole pairs and \( J > 0 \) is the antiferroelectric coupling constant normalized by Landau coefficient \( |z_0| \). Thus, all free energy terms are normalized by the energy unit \( |z_0| P_0^2 \).

Similar to previous simulations on FE lattice,\(^{37} \) for each FE sublattice, the Landau free energy polynomial function is expanded up to the sixth-order

\[
F_{ld} = A_1(P_x^2 + P_y^2) + A_{11}(P_x^2 + P_y^2) + A_{111}(P_x^2 + P_y^2) + A_{12}(P_x P_y + P_y P_x),
\]

where the Landau coefficients are represented in coefficients \( A \) with various subscripts and \( A_1 = A_{10}(T - T_0) \), where \( T_0 \) is the critical temperature for the paraelectric to FE transition of the corresponding FE sublattice.

The lowest-order expression of the gradient energy is

\[
F_{gr} = \frac{1}{2} \left[ G_{11}(P_x^2 + P_y^2) + G_{12}(P_x P_y) + G_{44}(P_x P_y)^2 \right],
\]

where \( P_{ij} = \partial P_i / \partial \xi_j \) and coefficients \( G_{ij} > 0 \). Subsequently, term \( F_{ela} \) is defined as

\[
F_{ela} = \frac{1}{2} C_{ij} u_i u_j - \frac{1}{2} C_{44} u_x u_y - \frac{1}{2} C_{xy} u_y u_x,
\]

where \( u_j = \partial u_j / \partial \xi_j \), \( u_j = \partial u_j / \partial \xi_j \), and strain \( \eta_{xy} = Q_1 P_x^2 + Q_2 P_y^2 \), \( \eta_{yz} = Q_3 P_x^2 + Q_2 P_y^2 \), \( \eta_{xxy} = Q_4 P_x^2 P_y^2 \), and coefficients \( C_{ij} \) are the elastic coefficients and \( Q_{ij} \) are the electrostrictive coefficients. It should be mentioned that the total elastic energy \( F_{ela} \) given in Eq. (7) looks quite different from that reported in literature.\(^{38,39} \) In fact, the total elastic energy \( F_{ela} \) is usually written as \( F_{ela} = 1/2 C_{ijkl}(\epsilon_{ij} - \epsilon_{0ij}) \) (\( \epsilon_{ij} - \epsilon_{0ij} \)) where \( C_{ijkl} \) is the elastic stiffness tensor, and \( \epsilon_{ij} \) and \( \epsilon_{0ij} \) are the total strains and stress-free strain, respectively, with the tensor subscripts \( (i,j,k,l) = 1, 2, 3 \).\(^{39} \) This energy term can be re-formulated into three parts. The first part is related only with polarization \( P \) which can be merged into the Landau free energy term. The second part counts the conventional elastic energy and the third one counts the electrostriction energy, i.e., the second part of Eq. (7).\(^{39} \) While the details of this re-formulation are omitted here, Eq. (7) is actually equivalent with the original formulation of \( F_{ela} \) for the latter two parts, while the first part is included into the Landau term Eq. (5). In fact, Eq. (7) has also been often employed.\(^{40} \)

Finally, term \( F_{ele} \) consisting of dipole–dipole interaction and electrostatic energy can be written as

\[
F_{ele} = \frac{1}{4 \pi \varepsilon_0} \sum_{\langle i, j \rangle} \left[ \frac{P_i \cdot P_j}{|r_i - r_j|^3} - \frac{3P_i \cdot (r_i - r_j) [P_i \cdot (r_i - r_j)]}{|r_i - r_j|^5} \right] - \left( E_i P_x + E_j P_y \right),
\]

where the pre-factor \( (1/4 \pi \varepsilon_0) \) is taken for normalization purpose and field \( E(t) \) is applied along the \( x \)-axis. Instead of using the Ewald summation scheme for this long-range term, we adopt a more tractable calculation done by finite truncation treatment. It is noted that for a two-dimensional lattice, this treatment is accurate as long as the truncating distance \( R \) is big (\( R = 8 \) in our simulation).\(^{41,42} \)

As mentioned earlier, one key problem with the present simulation is that so far neither phenomenological theory other than the above scheme on AFE lattice is available nor physical parameters for those AFE materials have been reported. As a qualitative approximation, all the physical coefficients except the AFE coupling constant \( J \) in Eq. (4) are chosen following the reference system BaTiO\(_3\), which

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are listed in Table I.\textsuperscript{37} Unfortunately, the choice of all the coefficients regarding BaTiO$_3$ instead of any real AFE system makes a quantitative comparison with experiments impossible. Parameter $J$ is treated as a variable in the simulation so that a representative double-loop hysteresis can be generated in a quasi-static condition at low $T$. In our case, it is shown that $J=1.00$ is appropriate in terms of qualitative agreement with the major features of an AFE system. In fact, the most concerned AFE material Pb(Zr$_{1-x}$Ti$_x$)O$_3$ with $x \ll 0.5$ has similar perovskite structure as BaTiO$_3$.

### B. Procedure of simulations

The MC simulation is performed in two steps. First, we simulate an AFE lattice using the gradual annealing method. A lattice with randomly distributed $P$ and $u$ at a high $T \gg T_0$ is submitted to the thermal evolution toward the equilibrium, step by step down to a sufficiently low $T$. In order to relax the electric dipoles and elastic strains in the lattices, we update the values of $P$ and $u$ by the standard Metropolis procedure, in which the trial dipole orientation is chosen at random. Considering that the response time of elastic strain is far shorter than that for dipole relaxation, we give ten chances to its strain relaxation every time we relax one electric dipole.\textsuperscript{41,42} As a result, the initial dipole configuration of sublattice $x$ is a ferroelectrically ordered state with all the dipoles aligned along the $x$-axis, while those from sublattice $b$ are oriented in the opposite directions. The annealing process continues for $10^6$ mcs with 1 mcs standing for $L^2$ dipole flip attempts.

Second, the AFE lattice is then submitted to field $E(t)$ below $T_p$. The instant polarization $P(t)$ averaged over the whole lattice configuration is plotted against $E(t)$ to produce the $P$-$E$ hysteresis. For the high-$f$ cases, the $P$-$E$ hysteresis may be time-dependent in the initial periods of cycling and the presented hysteresis is different from the quasi-stable one in the very late stage. In general, the first 10 loops were discarded and the subsequent 20 loops were taken for the data averaging of $P(t)$. Finally, the loop area $A(f, E_0)$ data package are evaluated. In our simulations, we pay our attention to the cases of $E_0 > E_{AF1}$, while the other cases will be addressed in the future due to the different physics.

### III. RESULTS AND DISCUSSION

#### A. Domain structures and evolution

We have performed extensive simulations on the evolution of dipole configuration and dynamic hysteresis over broad ranges of variables $f$, $E_0$, and $T$. For each sublattice, one can define the so-called FE domains, and such a domain refers to a region in which most dipoles align along one direction although minor dipoles may align differently.

We first look at the domain structure in various states (A, B, C, and D) of a typical double-loop hysteresis, as shown in Fig. 2(a) where $f = 4.88 \times 10^{-4}$ mcs$^{-1}$ and $E_0 = 8.0$ at $T = 0.2$. The domain patterns of the two sublattices $x$ and $b$ are plotted in Fig. 2(b). The domain orientations are marked by colors and red arrows, and angle $\theta$ is defined with respect to the $x$-axis. Two major features deserve mention here. First, at state A, adjacent dipoles from different sublattices ($x$ and $b$) are oppositely aligned, constituting an AFE state. However, each sublattice shows its own FE domain structure. For example, at state A, sublattice $x$ does not exhibit a mono-domain but multi-domain structure, similarly does sublattice $b$. Second, at state B, all dipoles antiparallel with external field are completely switched, and an FE state over the whole lattice is reached. State C allows some local dipole fluctuations in the two sublattices, i.e., small cluster-like FE domains of opposite polarization embedded in the FE matrix. This evolution becomes more remarkable and those cluster FE domains grow and coarsen in size at state D in comparison with them at state C.

The above highlighted evolution of the AFE configuration in association with the double-loop hysteresis seems to hint two characteristic sequences. One is the dipole reversal of the two sublattices along the path from state A to state B. The other is the appearance of cluster-like domains in the oppositely aligned matrix along the path from state B to states C and D. Indeed, the FE domain patterns and sizes of the two sublattices following the two paths are quite different. However, the two sequences are actually miscible, suggesting that the dipole switching sequences in the two sublattices are coherent and synchronous. It is thus implied that the polarization switching over the whole lattice may be possibly described by one characteristic time. This conjecture will be confirmed by the hysteresis dispersion and scaling analysis.

#### B. Dynamic hysteresis and energy storage

Now we investigate the dynamic hysteresis along two lines. One is the frequency dispersion $A(f)$ at given $E_0$ and the other is the $E_0$-dependence at given $f$. The representative examples at $T = 0.2$ are plotted in Figs. 3(a)–3(d). For the cases of $E_0 (~5.0)$ only slightly larger than $E_{AF1}$, as shown in Fig. 3(a), the double-loop feature can be observed only in the low-$f$ range and the hysteresis evolves into the inclined fat single loop shape with increasing $f$. The loop area $A(f)$

<table>
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<th>Parameter (unit)</th>
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<th>Parameter (unit)</th>
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<td>$A_{12}$ ($g_{12}$)</td>
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<td>$g_{12}$</td>
<td>$G_{12}$ ($g_{12}$)</td>
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<td>$g_{144}$</td>
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<tr>
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<tr>
<td>$L_{a0}$</td>
<td>64</td>
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<td>$g_{11}$</td>
<td>$L_{a0}$</td>
<td>1.00</td>
</tr>
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first increases and then decreases. For the high-$E_0$ cases ($E_0 = 8.0 > E_{AF}$) shown in Fig. 3(b), qualitatively similar features to the low-$E_0$ cases can be identified while the double-loop feature may sustain until a much higher $f_0$. On the other hand, the $E_0$-dependence exhibits different characters, as shown in Figs. 3(c) and 3(d), respectively, for the low-$f$ and high-$f$ cases ($f = 2.4 \times 10^{-7} \text{ mcs}^{-1}$ and $2.4 \times 10^{-4} \text{ mcs}^{-1}$). It is seen that the double-loop feature remains nearly unchanged with increasing $E_0$ in the low-$f$ cases. However, the hysteresis can become fat upon increasing $E_0$ in the high-$f$ cases, where the double-loop shape disappears at large $E_0$, suggesting more complicated evolution of the domain structure in the high-$f$ range.

The dynamic hysteresis discussed earlier suggests the significant dependences of the energy storage/release on $f$, $E_0$, and $T$. To evaluate these dependences, we present the releasable energy $W$ as a function of $E_0$ at different frequencies in Fig. 4(a) for $T = 0.2$ and Fig. 4(b) for $T = 0.01$. At a relatively high $T$ ($T = 0.2$), $W$ is enhanced with increasing $E_0$ in the low-$f$ range and then tends to be saturated in the large $E_0$ range. However, a monotonous decreasing of $W$ with increasing $E_0$ is identified in the high-$f$ range due to the gradual disappearance of the double-loop feature. The hysteresis becomes expanded and fat with increasing $E_0$, leading to reduced $W$. These results are qualitatively consistent with the experimental observations.

Physically, it is reasonable to argue that a larger $E_0$ always brings about larger $dE/dt$ and shorter time for domain

FIG. 2. (a) A simulated $P$-$E$ hysteresis at $f = 4.88 \times 10^{-4} \text{ mcs}^{-1}$ and $T = 0.2$, given $E_0 = 8$. Various states in the hysteresis are labeled with A, B, C, and D, respectively. (b) The snapshots of the domain structures for sublattice $\alpha$ (left column) and sublattice $\beta$ (right column) at states A, B, C, and D. The domain orientations are marked by colors and red arrows, and angle $\theta$ is defined with respect to the $x$-axis. Dark yellow represents the polarization along the $x$-axis, and blue stands for the polarization along the $-x$-axis. The light blue areas between the dark yellow and blue domains mark the domain walls. The magnitude of electric dipole vectors in white areas is zero.

FIG. 3. Simulated $P$-$E$ loops for the AFE lattice at $T = 0.2$: (a) $E_0 = 5$ at various $f$, (b) $E_0 = 8$ at various $f$, (c) $f = 2.4 \times 10^{-7} \text{ mcs}^{-1}$ with different $E_0$, and (d) $f = 2.4 \times 10^{-4} \text{ mcs}^{-1}$ with different $E_0$.

FIG. 4. The recoverable energy density $W(E_0)$ calculated from the $P$-$E$ loops simulated for the low-$f$ and high-$f$ cases with (a) $T = 0.2$ and (b) $T = 0.01$. The value of $f$ for each curve is labeled numerically with the unit $10^{-6} \text{ mcs}^{-1}$. 
switching during field-driven transitions at any fixed $f$, resulting in more remarkable hysteresis effect. Given a relatively high $T$ and in the low-$f$ cases where characteristic relaxation time $\tau \ll f^{-1}$, the enhanced $W$ with $E_0$ in the low-$E_0$ range is mainly ascribed to the field-driven AFE-FE transitions, which lead to much larger polarization $P$, but the double-loop hysteresis remains roughly unchanged in shape. In the high-$f$ cases where $\tau \sim f^{-1}$, the decayed $W$ with $E_0$ comes from the increased remnant polarization due to the intensified hysteresis effect with gradual disappearance of the double-loop feature, as shown in Fig. 3(d). If temperature $T$ is reduced down so that $\tau \sim f^{-1}$ becomes available even at extremely low $f$ ($\sim 10^{-7}$ mcs$^{-1}$), it is inevitable that energy $W$ as a function of $E_0$ always falls down in the $f$-range covered here above $\sim 10^{-7}$ mcs$^{-1}$. This is the reason for the observations shown in Fig. 4(b).

C. Hysteresis dispersion scaling

To evaluate the characteristic relaxation time as a function of $E_0$ at different $T$, we plot the evaluated dispersion $A(f)$ for various $E_0$ given $T=0.1$ and 0.4 in Figs. 5(a) and 5(b), respectively, where frequency $f$ covering from $10^{-7}$ mcs$^{-1}$ to $10^4$ mcs$^{-1}$ is presented in the logarithmic scale. It is noted that the smallest $E_0$ is still larger than $E_{AF1}$. Referring to earlier works on the frequency dispersion on FE lattice, several points deserve mention here. First, similar to the FE lattice, all the dispersion curves at the two temperatures exhibit the single-peak pattern, with the peak coordinate designated as $(f_m, A_m)$, suggesting a resonant state of the polarization switching at $f \sim f_m$. In other words, the characteristic time $\tau$ for polarization switching is $\tau \sim f_m^{-1}$. Second, it is found that peaked area $A_m$ increases with $E_0$, but the peaked frequency $f_m$ does not show remarkable shifting toward the high-$f$ side with $E_0$, very different from the FE lattice. The identifiable $E_0$-dependence of $f_m$ is very weak indeed. Third, the $T$-dependences of both $f_m$ and $A_m$ are also very weak although a slight shifting of $f_m$ toward the high-$f$ side with increasing $T$ can be detected.

Given the single-peak dispersion behavior, one can rescale these dispersions at various $E_0$ following the simple exponential scaling relationships:

$$A'(f') = A(f)/E_0^a$$

$$f' = f/E_0^c$$

where $a$ and $c$ are the scaling exponents that can be obtained from the best fitting of the simulated data so that the whole set of dispersion curves at a given $T$ can fall onto a master curve.

The best fitted results give the scaling exponents $a = 1.00 \pm 0.05$ and $c = 0.05 \pm 0.05$ for $T < 0.3$, and $a = 1.05 \pm 0.05$ and $c = 0.05 \pm 0.05$ for $T > 0.3$ in the present simulations. The rescaled dispersion curves for $T = 0.1$ and 0.4 are plotted in Figs. 5(c) and 5(d), respectively, demonstrating that these curves do fall onto one master curve within the simulation uncertainties. The scaling of the data implies at least three facts: (1) the frequency dispersion $A(f)$ can be indeed scaled by simple power-law transforms based on a single parameter $E_0$ over a broad range of $E_0$; (2) time $\tau$ shows very weak dependence of $E_0$; and (3) time $\tau$ shows very weak dependence of $T$ as long as $T \ll T_0$.

To understand the reason for such weak $E_0$-dependence of characteristic time $\tau$, we come back to the hysteresis itself and correlate it with the kinetics of polarization switching in the characteristic time language. As discussed earlier, one refers to Fig. 2(a) and starts from state A where the AFE order is the ground state. Given fixed $f$, $E_0$, and $T$, field $E(t)$ drives the evolution of the lattice from state A to state B via the AFE-FE transition. The corresponding characteristic time for such a transition is denoted as $\tau_{AFE-FE}$. Subsequently, the dynamic hysteresis proceeds from state B to states C and D in sequence via the FE-AFE transition. The corresponding characteristic time is denoted as $\tau_{FE-AFE}$. Clearly, one expects that a well-expanded and fat hysteresis will be developed if $\tau_{FE-AFE} > \tau_{AFE-FE}$, otherwise, a slim and thin hysteresis is inevitable if $\tau_{FE-AFE} \ll \tau_{AFE-FE}$.

For comparing the time scales $\tau_{FE-AFE}$ and $\tau_{AFE-FE}$, one discusses the single dipole switching sequence as a zero-order approximation of the polarization switching, considering the domain growth is much faster than the domain nucleation for typical FE systems. The dipole switching for an FE lattice and an AFE lattice is shown in Fig. 6(a). We consider the first quadrant of the hysteresis. For the FE lattice where all dipoles spontaneously align rightward and the AFE lattice where all dipoles are driven to align rightward, a decreasing electric field $E$ (its positive direction aligns rightward) may trigger one rightward dipole (cycled) to flip leftward (red arrow). The energy change associated with such a flip is

$$\Delta H = \Delta H_{FE} - 2p \cdot E_0$$

for FE lattice

$$\Delta H = \Delta H_{AFE} - 2p \cdot E_0$$

for AFE lattice,

where $\Delta H_{FE} > 0$ and $\Delta H_{AFE} < 0$ are, respectively, the changes of interactions between this dipole and its neighbors upon
such a flip for the FE lattice and AFE lattice. The characteristic dipole switching time $\tau$ can be roughly expressed as

$$\tau = \tau_0(T) \exp (\Delta H/k_B T),$$

where $k_B$ is the Boltzmann constant and $\tau_0$ is the characteristic flip time for a free dipole at a given $T$. For the AFE lattice, it is clear that the relation $\tau_{FE,AFE} < \tau_{AFE,FE}$ is always true, qualitatively illustrating why the peak frequency $f_m$ is only weakly $E_0$-dependent although the peak height $A_m$ is strongly $E_0'$-dependent. The weak $T$-dependence of $f_m$ can be understood in a similar way.

For a better illustration of the above scenario, we present in Fig. 6(b) the $P$-$E$ hysteresis loops corresponding to $f_m \sim 6.25 \times 10^{-3}$ mcs$^{-1}$ at five different values of $E_0 > E_{AF1}$. Here, the electric field is re-scaled by $E_0$ and it is seen that the five loops are roughly overlapped in spite of delicate differences among them. While the delicate differences reflect the weak $E_0'$-dependence, the overlapping reflects the $E_0'$-independence. This property is favored for AFE storage energy applications and one need not take care of the frequency sensitivity.

**D. Energy storage density**

Finally, we discuss the energy storage density $W$ in response to variations of $f$, $E_0$, and $T$ for the present system. The simulated results are summarized in Figs. 7(a) and 7(b) for two different $T$. Based on the dynamic hysteresis analysis, it is obvious that the storage density $W$ always decays monotonously with increasing $f$ over the whole $f$-range.

Nevertheless, such decay shows distinct features in different $f$-subranges. The rapid decay occurs in the intermediate $f$-subrange, while a plateau is observed in the low-$f$ subrange and the storage density becomes nearly zero in the high-$f$ subrange. An unexpected fact is that the frequency threshold beyond which the decay becomes rapid is lower when $E_0$ is larger, suggesting that the frequency stability of the energy storage is worse at larger $E_0$.

The continuous decay of $W$ outside the high-$f$ subrange can be well understood considering the relationship $W(f) = W_0(f) - A(f)$, while the total energy density $W_0(f)$ is relatively insensitive to $f$ as $f < f_m$. As $f > f_m$, the hysteresis shrinks along the polarization axis rapidly, leading to serious suppression of $W_0$ and thus less and less energy can be stored and released. Besides, given the scaling behavior of $A(f, E_0)$, it would be interesting to check the possible scaling over $W(f)$ at different $E_0$. Similarly, we propose

$$W'(f') = W(f)/E_0^m$$

$$f' = f/E_0^c,$$

where exponents $m$ and $n$ correspond to exponents $a$ and $c$ in Eq. (9) on scaling of dispersion $A(f)$. The best fitting of all the $W(f)$ data gives rise to $m = 0.05 \pm 0.05$ and $n = -1.00 \pm 0.05$ for $T = 0.1$, and $m = 0.20 \pm 0.05$ and $n = -0.90 \pm 0.05$ for $T = 0.4$. The re-scaled data are plotted in Figs. 7(c) and 7(d) for the two temperatures and it is seen that the scaling on data at $T = 0.1$ is slightly better than that at $T = 0.4$. These scaling behaviors confirm again the single characteristic time for polarization switching in the present AFE system.

What should be noted is that the scaling behaviors as revealed in the present model rely on the assumption that the exchange interaction is completely attributed to the adjacent dipoles from different sublattices. Surely, for realistic AFE materials, the antiferroelectric coupling interactions mechanism is much more complicated, rendering a simulation tougher. Besides, we only consider the 2D thin film lattice with in-plane AFE domains here, which also deserves.
IV. CONCLUSION

In summary, we have simulated the electric field driven AFE-FE transitions and domain structural evolution in association with the double-loop hysteresis for a two-dimensional model AFE system, based on the Landau phenomenological theory by using Monte Carlo method. The dynamic hysteresis over broad ranges of parameters $f$, $E_0$, and $T$ for the cases of $E_0 > E_{AF}$ and $T \ll T_0$ have been carefully investigated. It is revealed that the energy storage density $W$ is strongly dependent on $f$, $E_0$, and $T$, consistent with some experimental observations. Both the frequency dispersion $A(f)$ and energy storage density $W(f)$ can be scaled by simple power laws based on a single parameter $E_0$, which confirms that one characteristic time $\tau$ is enough for the description of polarization switching over the whole lattice. The weak $E_0$-dependences of characteristic time $\tau$ and monotonous decaying behaviors of energy storage $W$ in different $f$-subranges are discussed and explained qualitatively. The present work raises strong appealing for experimental investigation of the dynamics of field-driven AFE-FE inter-transitions in AFE materials.

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