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LETTER TO THE EDITOR

Scaling of dynamic hysteresis in ferroelectric spin systems

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Abstract
We present a systematic study of dynamic hysteresis in ferroelectric spin systems by Monte Carlo simulation based on the ferroelectric DIFFOUR model, Sawyer–Tower hysteresis measurement of $YBa_2Cu_3O_7$(YBCO)$PbTi_{0.48}Zr_{0.52}O_3$ (PZT)/YBCO thin film capacitors and the phenomenological approach to the $(\Phi^2)^2$-model system. The hysteresis area A as a function of frequency f of the external field E is single-peaked, due to the full spin relaxation at low f and suppressing effect at high f for the domain switching. It is revealed from the simulation that the hysteresis area A can be scaled with a dynamic scaling function $A(f)/A_m = F(x)$, which is symmetric to $x = 0$, where $x = \log(f/f_m)$ with $f = f_m$ the position of the peak $A_m = A(f_m)$. The scaling gets support from the experiment and is consistent with the prediction of the $(\Phi^2)^2$-model.

The problem of nonequilibrium first-order phase transitions has been the subject of extensive investigations over the past decades. In addition to the main stem of the study, i.e. kinetics of nucleation and growth as well as late stage coarsening of the domains, the system order parameter as a function of a varying external field yields a hysteresis that represents the energy dissipation during one cycle of domain reversal [1]. However, the latter problem has been studied less [2,3]. When an external field E with frequency f and amplitude E_0 is applied, the system, far below the Curie point, has an order parameter response which is very dependent of both f and E_0, resulting in a very different hysteresis in shape and area. The dynamic hysteresis deals with the external field dependence of the hysteresis and associated parameters, such as hysteresis area, remnant order parameter and coercivity. This problem is now also of interest from the point of view of applications, mainly due to the fact that the high-speed memory devices in which ferroelectric and ferromagnetic thin films are involved are being developed extensively [4].

The earlier studies focused on kinetic Monte Carlo (KMC) simulation of the dynamic hysteresis effect based on the Ising model and the phenomenological description in the scheme of the Landau free-energy approach [5–9]. Our understanding of the dynamic hysteresis is in the initial stage. It was proposed [6,7] that the hysteresis area A responded to varying f and E_0 in a well-defined way as f is quite low and can be expressed as $A \propto f^m \cdot E_0^n$, while the dynamic behaviour of the hysteresis away from the low-f range has rarely been studied, except for the preliminary theoretical results proposed by Rao *et al* [6]. $A(f)$ increases first as f is low and then decreases at high f, after passing across the maximal A_m at $f = f_m$. The low-f effect is ascribed to the dynamic relaxation of domains and the high-f behaviour can be explained by the serious suppression of the domain reversals.

Systematic experiments on the dynamic hysteresis performed recently on ferroelectric $Pb(Zr_{0.52}Ti_{0.48})O_3$ (PZT) revealed distinguishable differences between the experiment and the theoretical prediction [10]. In this letter, we perform a systematic study of this problem. We start from the Landau-type approach of discretized spin systems and perform the KMC simulation on the dynamic hysteresis phenomenon. We compare the simulated results with our experimental data on PZT thin film capacitors and numerical modelling based on the two-dimensional $(\Phi^2)^2$ continuum model [6]. A dynamic scaling relation which normalizes the response of A against f and E_0 in a simple way is proposed.

Our KMC simulation starts from a two-dimensional squared $L \times L$ lattice with a periodic boundary condition applied. Each site of the lattice is imposed with a displacement vector u_i for an electrical polar. u_i is thus proportional to the local spontaneous polarization. According to the so-called DIFFOUR model, u_i at each site is subjected to a double-well potential with an additional nearest-neighbouring interaction taken into account. The lattice Hamiltonian can be written as [11]:

$$\tilde{H} = \sum_i \left(\frac{p_i^2}{2m} - \frac{a}{2}u_i^2 + \frac{b}{4}u_i^4 \right) - U \sum_{\langle i,j \rangle} u_i \cdot u_j - \sum_i E \cdot u_i \qquad (1)$$

where $\langle i, j \rangle$ represents the nearest neighbours summed once, p_i the momentum at site i, a and b the double-well potential parameters, U the ferroelectric ordering factor, m the mass and $E = E_0 \sin(2\pi f \cdot t)$ the electric field pointing to the base direction by which the direction of u_i or u_j is determined. The three terms in the equation represent the energy of site i under the double-well potential, the nearest-neighbouring electrical interaction and static energy, respectively.

The Metropolis algorithm is employed in our KMC simulation in which domain reversal is achieved via re-imposing the magnitude and direction for u_i. At each site u_i is only permitted a 180°-flip, but its magnitude is chosen at random. The initial lattice consists of a number of domains of average diameter R. Inside each domain all sites have the same u_i, both in direction and magnitude, but the direction of u_i differs from one domain to another. The lattice polarization

$$P = \frac{1}{L^2} \sum_i u_i \cdot \frac{E}{|E|}$$

is obtained by averaging over four sets of data from different seeds of random number generation. The KMC is scaled in a unit of mcs.

To check the simulated results, the dynamic hysteresis of a PZT thin film capacitor—a typical ferroelectric spin system—is measured using the Sawyer–Tower method and a detailed description of the thin-film deposition and hysteresis measurement was given previously in [10].

Figures 1(a) and 1(b) present a series of simulated and measured hysteresis obtained at various f but fixed E_0. The measured hysteresis is reproduced in a satisfactory way by the

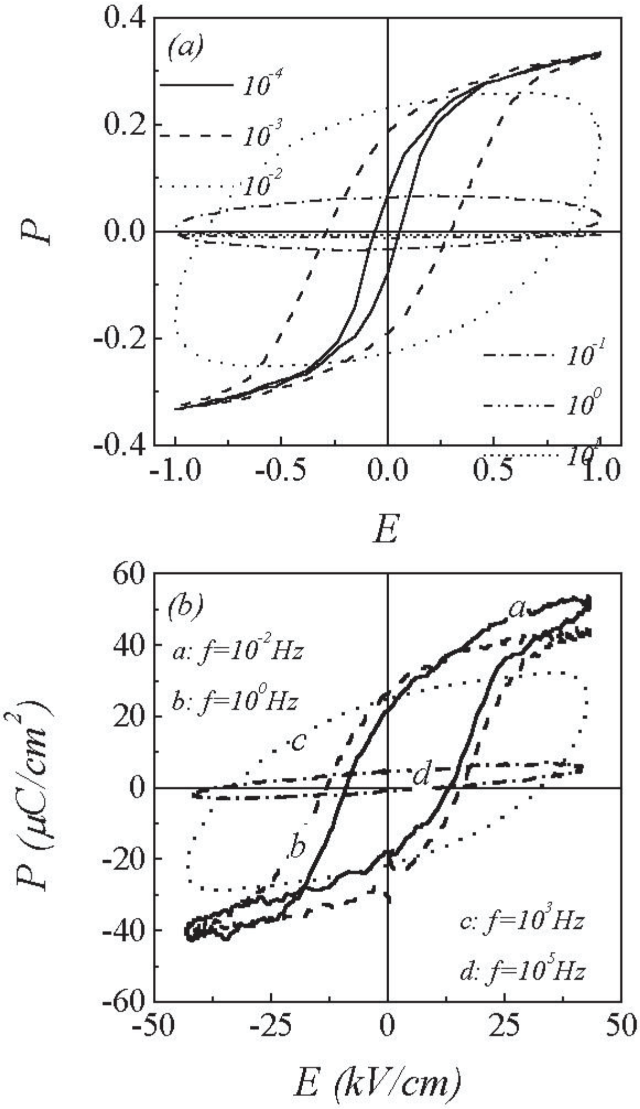

Figure 1. Hysteresis loops for the DIFFOUR model (a) and PZT thin film capacitor (b) at various frequencies of the external field E.

simulation, looking at the hysteresis shape and its evolution with f. At very low f, the system takes a very thin rhomb-like loop which evolves with increasing f into broad-rhomb and tip-round fat loop. The elliptical pattern is observed for the loop at high f before it finally collapses into a horizontal line when f is extremely high. Both the simulated and measured data show that the remnance P_r and hysteresis area A grow first with f as f is very low and then decrease with f after reaching their maximal values, respectively. $P(E = E_0)$ falls down with increasing f whiles coercivity E_c is increased linearly. We do not need to mention that the single-peaked pattern of $P_r(f)$ and $A(f)$ shifts upward and towards the right when E_0 increases. In figures 2(a) and 2(b) the simulated and measured $A(f)$ at various E_0 are plotted,

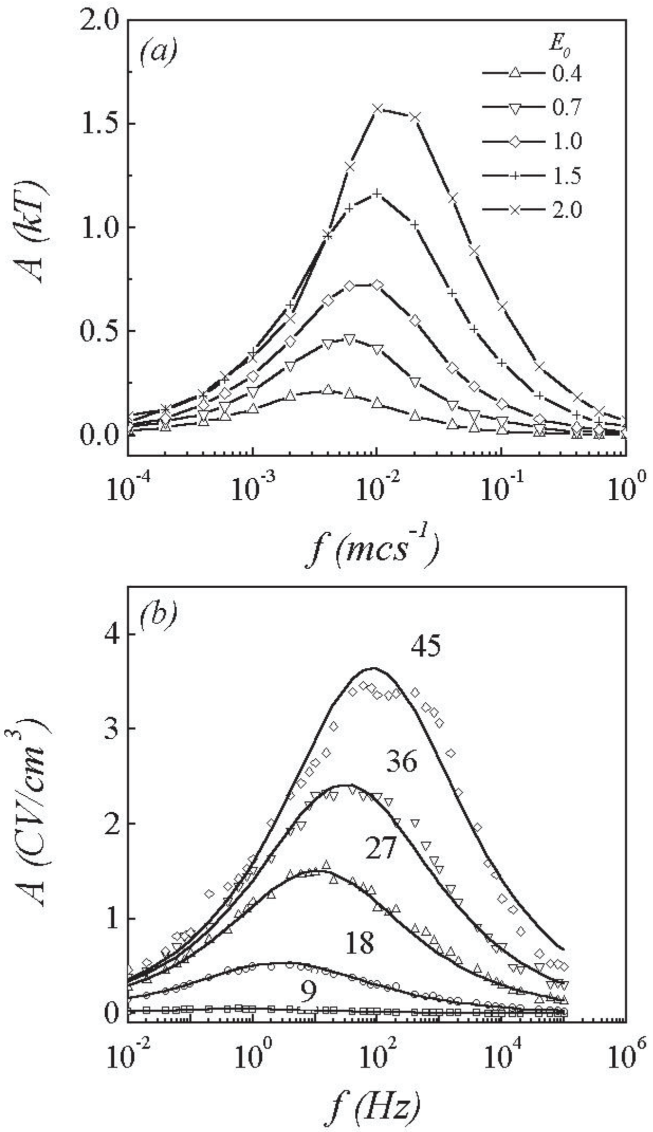

Figure 2. Hysteresis area A as a function of frequency f at various values of E_0 from (a) the simulation and (b) the experiment on PZT capacitor (the inserted are values of E_0).

where the f-axis is in log-scale. An excellent similarity between the simulated and measured $A(f)$ curve and its evolution with increasing E_0 is shown again, while noting that the time scale used for the simulation and real measurement is different. For each case, $A(f)$ shows a single-peaked pattern with the maximal area A_m locating $f = f_m$. If looking at the individual curves, one finds the almost perfect symmetry of each curve $A(\log f)$ around axis $f = f_m$.

When both simulated and measured $A(f)$ data at very low f can be well fitted with $A \propto f^m \cdot E_0^n$ with $m = 1/2$ and $n = 2/3$, no frequency-dependence of $A(f)$ in other cases has been proposed. Considering the perfect symmetry of each $A(\log f)$ curve with respect to

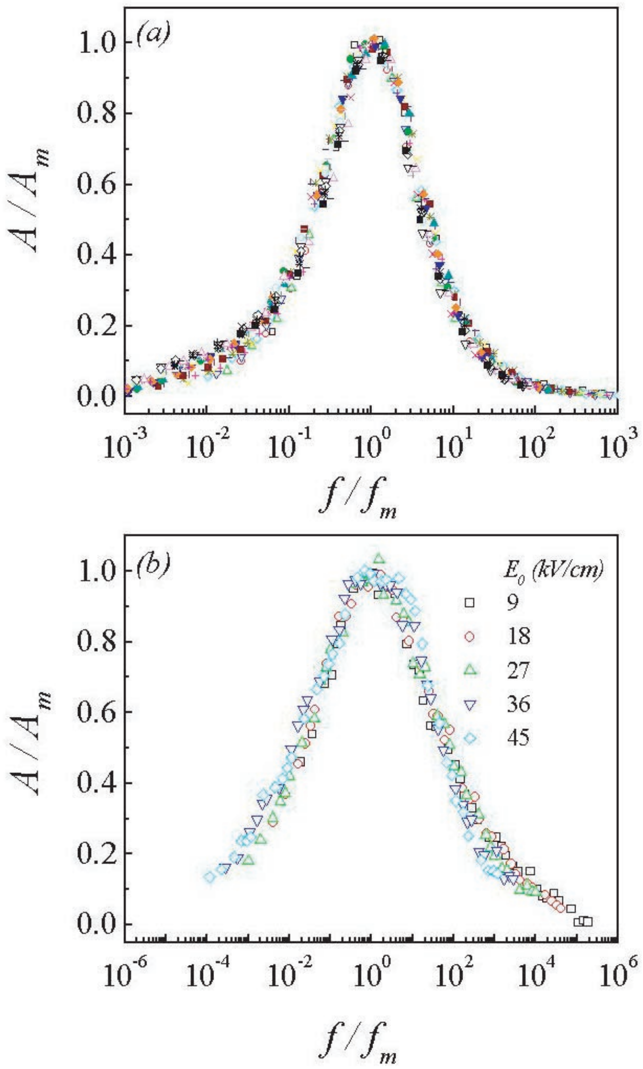

Figure 3. Normalized hysteresis area A/A_m as a function of normalized frequency f/f_m for the DIFFOUR model (a) and PZT thin film capacitor (b) at various values of E_0. 32 sets of data at different E_0 (from 0.1 to 80.0) are plotted together in (a).

axis $f = f_m$, it is natural to perform a dynamic scaling of the overall data. Let us apply a scaling transform:

$$\tilde{A} = A/A_m \qquad \tilde{f} = f/f_m \tag{2}$$

onto all simulated and measured data, yielding $\tilde{A}(\tilde{f})$ as presented in figure 3(a) for 32 sets of simulated data of different E_0 values (from 0.1 to 80.0), and in figure 3(b) for the five sets of measured data obtained at different E_0 values (from 9 kV cm^{-1} to 45 kV cm^{-1}). Note here that the hysteresis is already over-saturated as $E_0 = 80$, even at very high frequency. It is interesting to find that all simulated data sets fall onto the same curve with high reliability, as shown in figure 3(a). This predicts the existence of a scaling function symmetric to $k = 0$:

$$\tilde{A} = F(k) \qquad k = \log(f/f_m) \tag{3}$$

applicable for dynamic hysteresis of ferroelectric spin systems. Our simulation on various systems defined with different parameters a, b, U and R reproduces equation (3), demonstrating the universality of the scaling behaviour of the dynamic hysteresis.

A direct support of equation (3) first comes from the experiment on PZT thin film capacitors. Performing the scaling to the measured data presented in figure 2(b) produces similar universal scaling behaviour as shown in figure 3(b). The different shape in the scaling function between the simulation and experiment are attributed to the difference in time-scale. In fact, the f-axis in figure 2(b) covers eight orders of magnitude, while just four orders of magnitude for f-axis are covered in figure 2(a), keeping in mind the excellent consistency in shape between the simulated and measured $A(f)$. It is therefore demonstrated that the dynamic hysteresis in ferroelectric spin systems can be scaled with a simple function $F(k)$ which is symmetric to $k = 0$.

From our simulation the two characteristic parameters for the scaling, A_m and f_m, as a function of E_0 can be well defined, as shown in figures 4(a) and (b). The monotonous growth of A_m and f_m with increasing E_0 over a wide E_0-range is established in the simulation (sim : A_m and sim : f_m) and roughly supported from the experimental data on PZT capacitors (exp : A_m and exp : f_m) although for the latter the data are not enough for full support. If definable, the following power-like dependencies can be established:

$$\begin{aligned} A_m &\propto E_0^m \\ f_m &\propto E_0^n \end{aligned} \tag{4}$$

with positive exponents m and n which seem to depend respectively on the system parameters. It should be noted here that the $f_m \sim E_0$ dependence is seriously slowed down as E_0 is very high, as predicted from the simulation, while such a saturating tendency is not yet achieved in our experiment on PZT due to technique difficulty. There is not enough experimental data on the $f_m \sim E_0$ relation to give a strong support of the simulated behaviour.

In order to explain this interesting scaling behaviour, one may investigate the response of an N-component $(\Phi^2)^2$ model with $O(N)$ symmetry to an oscillating field E. The system order parameter set Φ is non-conserved and its evolution with time t obeys the Langevin equation:

$$\frac{\partial \Phi_\alpha}{\partial t} = -\Gamma \frac{\delta F}{\delta \Phi_\alpha} + \eta_\alpha \tag{5}$$

with the Gaussian white noise η_α as:

$$\begin{aligned} &\langle \eta_\alpha(x,t) \rangle = 0 \\ &\left\langle \eta_\alpha(x,t) \cdot \eta_\beta(x',t') \right\rangle = 2\Gamma \delta_{\alpha\beta} \delta(x - x') \delta(t - t') \end{aligned} \tag{6}$$

where $\alpha\beta = 1, 2, \ldots, N$, respectively; x is the spatial coordinate and Γ is the mobility for the spin-lattice relaxation which is of $\sim 10^7$ Hz on the order of magnitude for real solids, F is the free-energy function ($(\Phi^2)^2$ type) [6]:

$$F = \int \mathrm{d}^3 x \left[\frac{1}{2} J (\nabla \Phi_\alpha)(\nabla \Phi_\alpha) + \frac{r}{2} (\Phi_\alpha \Phi_\alpha) + \frac{u}{4N} (\Phi_\alpha \Phi_\alpha)^2 - \sqrt{N} E_\alpha \Phi_\alpha \right] \tag{7}$$

where Φ is an N-component vector and J is the interaction between two components; $r = T - T_c^{TF}$ where T_c^{TF} is the mean-field critical temperature; u is the pre-factor and counts the contribution of the second order nonlinear interaction and $u = -2\pi^2 (T_c - T_c^{TF})$. Since $\Phi_\alpha \Phi_\alpha$ scales as N, each term in the bracket scales as N, therefore so does the free energy. We assume that the external field $E_\alpha = E \cdot \delta_{\alpha,1} = E_0 \sin(2\pi f \cdot t) \cdot \delta_{\alpha,1}$, pointing to

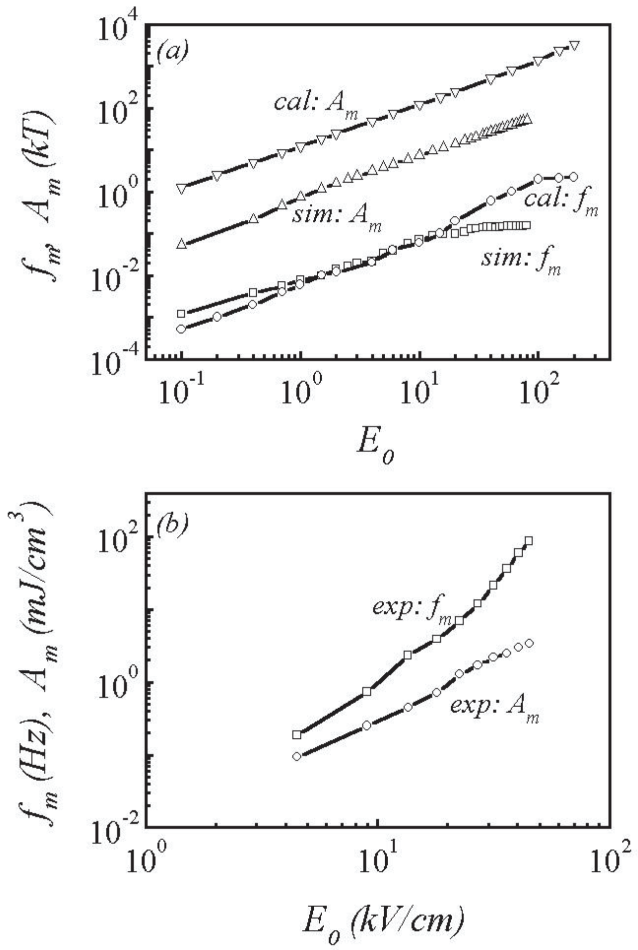

Figure 4. (a) Dependence of parameters A_m and f_m as a function of E_0 as evaluated from the simulation and the N-component $(\Phi^2)^2$ model with $O(N)$ symmetry, (b) the measured E_0-dependence of A_m and f_m for PZT thin film capacitor.

axis $\alpha = 1$. Equation (5) is equivalent to an infinite hierarchy of differential equations for the cumulants of Φ_α. In the $N \Rightarrow \infty$ limit, this infinite hierarchy of differential equations is truncated and the following coupled integrodifferential equations are obtained [6]:

$$\begin{aligned}
\frac{\mathrm{d}P(t)}{\mathrm{d}t} &= \frac{1}{2}\left[P(t)A(t) + E_0 \sin(2\pi f \cdot t)\right] \\
A(t) &= -\left[r + u \cdot P^2(t) + u \cdot S(t)\right] \\
S(t) &= \frac{1}{2\pi^2}\int_0^1 q^2 C_T(q,t)\mathrm{d}q \\
\frac{\mathrm{d}C_T(q,t)}{\mathrm{d}t} &= -\left[q^2 - A(t)\right]C_T(q,t) + 1
\end{aligned} \tag{8}$$

where $P(t)$ is the component of the order parameter P along $\alpha = 1$, i.e. magnetization or polarization and $C(q,t)$ the correlation function which has the transverse component $C_T(q,t)(\alpha \neq 1)$ and longitudinal component $C_L(q,t)(\alpha = 1)$.

An analytical solution to equation (8) is unavailable, thus a numerical analysis is needed. For the initial conditions for equation (8), the spin system is assumed to remain at the equilibrium state (as $T < T_c$ and $E \sim 0+$) at $t = 0$. So we have:

$$\begin{aligned} P(0) &= \left[\frac{r - r_c}{u}\right]^{1/2} \\ C_T(q,0) &= q^{-2} \\ C_L(q,0) &= q^{-1}\tan^{-1}\left(\frac{q}{2}\sqrt{\frac{2P}{E_\alpha}}\right) \\ C_{\alpha\beta}(q) &= \langle \Phi_\alpha(q)\Phi_\beta(q)\rangle = 0 \qquad \alpha \neq \beta. \end{aligned} \tag{9}$$

The numerical calculation is performed in order to evaluate $P(t)$ so that the dynamic hysteresis over wide ranges of f and E_0 is obtained. The calculated hysteresis shows the same shape evolution with f and E_0, consistent with the simulated and measured ones. Figure 5 presents the evaluated $\tilde{A}(\tilde{f})$ at a series of E_0 and the scaling performance according to equation (3). The evaluated f_m and A_m as a function of E_0 are presented in figure 4(a) as a comparison (cal : A_m and cal : f_m). At first glance, the calculated results exhibit an excellent consistency with the simulated and measured ones, and thus successfully explain the scaling behaviour of dynamic hysteresis in ferroelectric spin systems. In particular, the quantitative consistency between the simulated scaling and calculated one can be established.

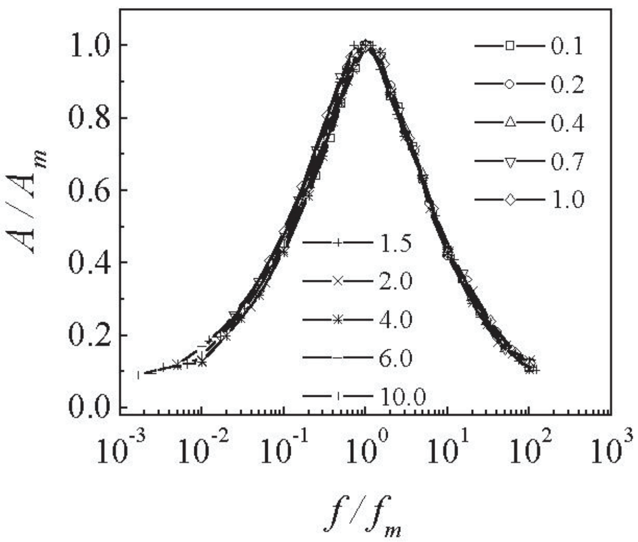

Figure 5. Normalized hysteresis area A/A_m as a function of normalized frequency f/f_m for the N-component $(\Phi^2)^2$ model with $O(N)$ symmetry (10 sets of data plotted together).

The above proposed scaling can be, in principle, extended to a three-dimensional case and the output remains the same. The scaling phenomenon reflects an intrinsic kinetic competition between the spin relaxation through the ground-state fluctuations and the delayed response of the spin reversal to the varying external field. The physical picture is quite clear although the symmetric scaling has been revealed here for the first time. The N-component $(\Phi^2)^2$ model gives a phenomenological but physically essential approach to this competition, as shown in equations (5) and (7). Moreover, PZT, as one of the standard soft-mode activated ferroelectric spin systems, has indeed been observed to exhibit such a scaling behaviour.

In conclusion, we have presented a detailed investigation on dynamic hysteresis in ferroelectric spin systems. The MC simulation based on the DIFFOUR model has revealed a significant dependence of the hysteresis on both frequency and amplitude of the external field and a scaling behaviour of the dynamic hysteresis has been proposed. This scaling property has been confirmed with the experimental measurement on PZT thin film capacitors over a wide range of external field frequency using the Sawyer–Tower method. The N-component $(\Phi^2)^2$ model with $O(N)$ symmetry has been applied to successfully explain the proposed scaling behaviour.

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