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Noise and finite size effects in multiferroics with strong elastic interactions

E. K. H. Salje,[1] X. Ding,[2] and Z. Zhao[2]
[1]*Department of Earth Sciences, University of Cambridge, Cambridge CB2 3EQ, United Kingdom*
[2]*State Key Laboratory for Mechanical Behavior of Materials, Xi'an Jiaotong University, Xi'an 710049, China*

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The size dependence of yield point assisted ferroelastic switching is dominated by the appearance of a minimum size where the domain switching by external strain is swamped by noise which is determined by internal jamming processes. The lower ferroelastic lateral cut-off size was found in computer simulations to be 200 × 202 unit cells for hard materials and 40 × 42 unit cells for soft materials. The corresponding length scales are 16 nm and 4 nm, respectively. These lengths are greater than the minimum length to sustain a twinned sample (∼1 nm). Elastic interactions modify the switching behavior of multiferroics at larger lengths but do not prevent elastic switching above these noise cut-offs. © *2013 AIP Publishing LLC* [http://dx.doi.org/10.1063/1.4802787]

Increasing miniaturization leads to two limitations for multiferroic memory devices: first, they will not work if their size is too small to sustain domain rearrangements and hence disallow hysteretic switching. This length is usually around 1 nm. The second limitation stems from the increase of random noise switching with decreasing size. This noise swamps the functional switching below a critical length scale. In magnetic and electric materials, the size limitations mean that small magnets have no sliding magnetic domain boundaries and become magnetically hard.[1–3] In ferroelectric thin film, the minimum film thickness is a few nm below which the dead layer, surface relaxations, and other modifications destroy ferroelectricity.[4–7] Here, we will focus on elastic systems because elasticity is involved in virtually all multiferroics. A natural lower elastic cut-off length is expected to be equivalent to the thickness of ferroelastic domain walls, namely, some 1 nm.[8,9] Here, we will argue that noise destroys ferroelasticity at slightly longer length scales.

Elastic interactions are non-local and shape dependences appear at all lengths. The reason is that elastic dipolar correlations decay with distance as $1/r^3$ which means that the elastic energy in 3-dimensional scales as $\int 1/r^3\, r^2\, dr \sim \ln(R/d)$, where R and d are the largest and smallest length scale considered.[10,11] This logarithmic size dependence modifies the domain patterns[12] but does not destroy ferroelastic switching. The reason is that moving ferroelastic twin boundaries have thicknesses of some ca. <1 nm and can be accommodated in nanocrystals (while their structural characteristics may change for small system sizes[13]). Thin films remain ferroelastic at a thickness of 1 atomic layer.[14] What is not known is: what happens if such thin layers are restricted laterally, e.g., when the thin film is a square with a side length of d. Has d a critical cut-off for ferroelasticity? It is the purpose of this paper to convey a surprising answer: while switching persists to very small values of d, the limiting factor becomes the noise of the switch (near the yield point) when complex and often unwanted domain structures obscure the "useful" hysteresis. The value of d is below 100 atomic distances in hard ferroelastics and below 40 interatomic distances for soft systems.

Our simulations are based on a 2-dimensional layer of interacting particles where ferroelasticity is produced by nonlinear springs in the diagonal of a square lattice[15] (Fig. 1).

This model was first introduced to show that external shear strain will switch domains at temperatures well below the transition point and that such switching needs a coercive strain which depends on boundary conditions, etc.[17,18] We consider strain driven switching (i.e., "hard" boundary conditions) rather than stress driven switching (i.e., "soft" boundary conditions) because nano scale devices are expected to use epitactical shear strain from substrates to generate strain. The fundamental difference for the dynamic behaviour was discussed in Ref. 19. Switching leads to "precursor" and "aftershock" signals where domain structures change before and after the critical strain (∼yield strain) is surpassed.[20] These events define "noise" with a

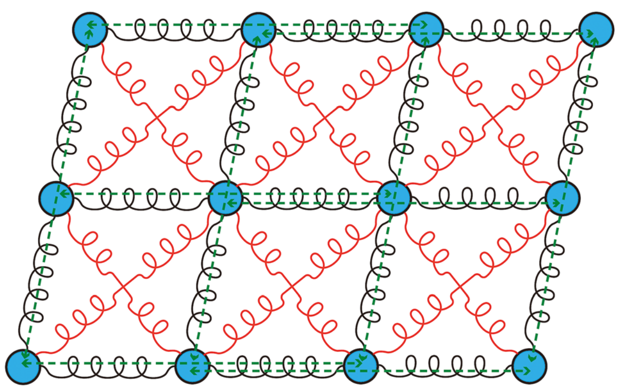

FIG. 1. Interatomic potential for a generic ferroelastic model. The model contains nearest-neighbor (black springs), next-nearest-neighbor (red springs), and third-nearest-neighbor (green lines) interactions ensure a spontaneous shear of the unit cells. The springs between the nearest-neighbors and third-nearest neighbors define the elastic background and define the thickness of interfaces. The red Landau springs (interaction between next-nearest neighbors) define the double well potential of the ferroelastic phase transition (see Ref. 15). They define a second order phase transition inspired by the transition of $SrTiO_3$ (after Ref. 16). The interatomic interaction of this potential are listed as follows: $V_{hard}(r) = 20(r-1)^2$ and $V_{soft}(r) = 10(r-1)^2$ $(0.8 \le r \le 1.2)$, $V(r) = -10(r-\sqrt{2})^2 + 2000(r-\sqrt{2})^4$ $(1.207 \le r \le 1.621)$, $V(r) = -(r-2)^4 (1.8 \le r \le 2.2)$.

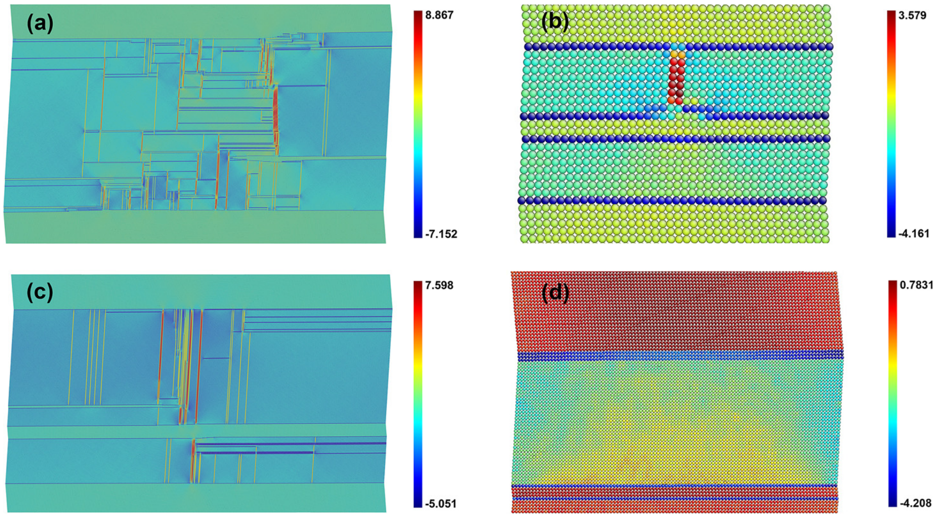

FIG. 2. Soft (a,b) and hard (c,d) ferroelastic thin films under external strain with corner lengths 1000 (a,c) and 40 (b) and 100 (d) atomic distances.

power law statistics of the squared time derivatives of the total energy E, $(dE/dt)^2$. We run this model in the a-thermal regime[15] for a soft and a hard configuration as functions of the lateral dimension d. In Fig. 2, we show a sequence of snapshots of a hard and a soft system.

Visual inspection of the patterns in Fig. 2 shows that the soft pattern are more complex with higher numbers of domain crossings.[17,18] Small soft systems still show complex switching while the switching disappears for small hard systems. We now explore the noise spectra of the switching process. The formal noise statistics are as follows: the probability of a noise event P(J) (J = jerk energy) follows a power law statistics $P(J) \sim J^{-\varepsilon}$ with size dependent cut-off lengths. The exponent is >2 for hard systems and ca. 1.6 for soft systems. Note that experimental values were also found in the range of 1.3–1.6.[12,21–24] The waiting time distribution is exponential for almost all cases, only for small soft systems, we find correlations and a power law distribution with

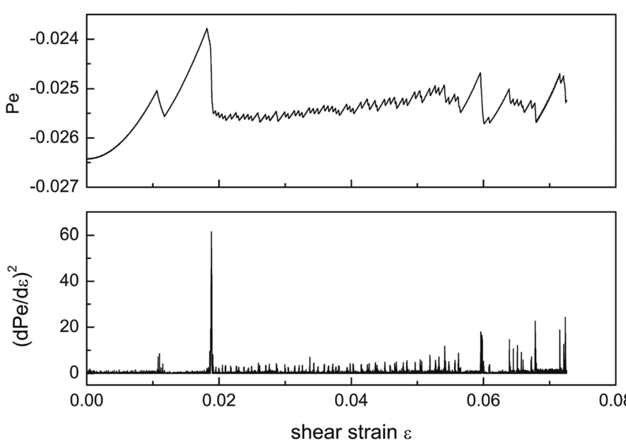

FIG. 3. Mechanical spectrum for a hard system with 200×202 particles. Increasing strain leads to an increase of the elastic energy pe until the yield strain is reached ($\varepsilon \approx 0.019$). At this point, the elastic deformation collapses by a massive change of the domain pattern. Further strain increase will lead to changes of these domain patterns with smaller a noise signals $(dpe/dt)^2$. Controlled switching becomes impossible when this noise level is at the same level as the collapse at the yield strain between the upper yield energy (−0.024 just before the switch) and the lower yield energy (−0.0255 just after the switch). In Fig. 3, the "functionally controlled" switch near $\varepsilon \approx 0.019$ dominates the spectrum, with decreasing system size the noise events increase while the main switch event decreases.

an exponent near 2. The attenuation of the noise pattern is provided by the acoustic phonons, which are included in our simulations. The crucial question is now: when is switching possible in a possible device assembly? To answer this question, we plot the strain dependence of the total energy and the noise derivatives as shown representatively in Fig. 3.

We can now define the switching ability by plotting the size dependence of the upper and lower yield stresses for the hard and soft systems. The plots stress versus strain in Fig. 4 show that the contrast between the upper and lower yield points diminishes rapidly with decreasing system size, a critical size where controlled switching becomes impossible, reached at $d \approx 200$ for hard and $d \approx 50$ for soft crystals. For small sizes below the cut-off, we find that the noise at strains larger than the yield strain in Fig. 3 is in the same order of magnitude as the main jump at the yield point.

If we translate the number of atoms into interatomic distance with radii of 0.08 nm, we find a lower cut-off for ferroelastic switching to be 16 nm for hard and 4 nm for soft crystals. The absolute values will further depend on the strain rate, etc. It is clear, however, that ferroic switching is possible for sizes down to very small scales, even when elastic interactions are an important ingredient for the ferroic interactions. Switching becomes impossible due to unwanted noise of the pattern formation and not due to the expected effect that the sample is simply too small to contain a ferroelastic domain wall. These results mirror some recent results in ferroelectric thin films[25] where $PbTiO_3$ thin films were grown as islands on $SrTiO_3$. These samples showed ferroelectric hysteresis loops at sizes below 50 nm. Simulations in Ref. 26 showed ferroelectric switching down to lateral sizes of 10 nm although with a much greater coercive field. Piezo-force microscopy (PFM) switching of domains with lateral length of some 10 nm has been reported in Ref. 27. Similar observations are expected in ferroelastic materials although we expect that the twin pattern would decay into a multitude of smaller domains under shear, experimental arrangement for the observation of such patterns would be very similar to the PFM experiments of the Chen group.

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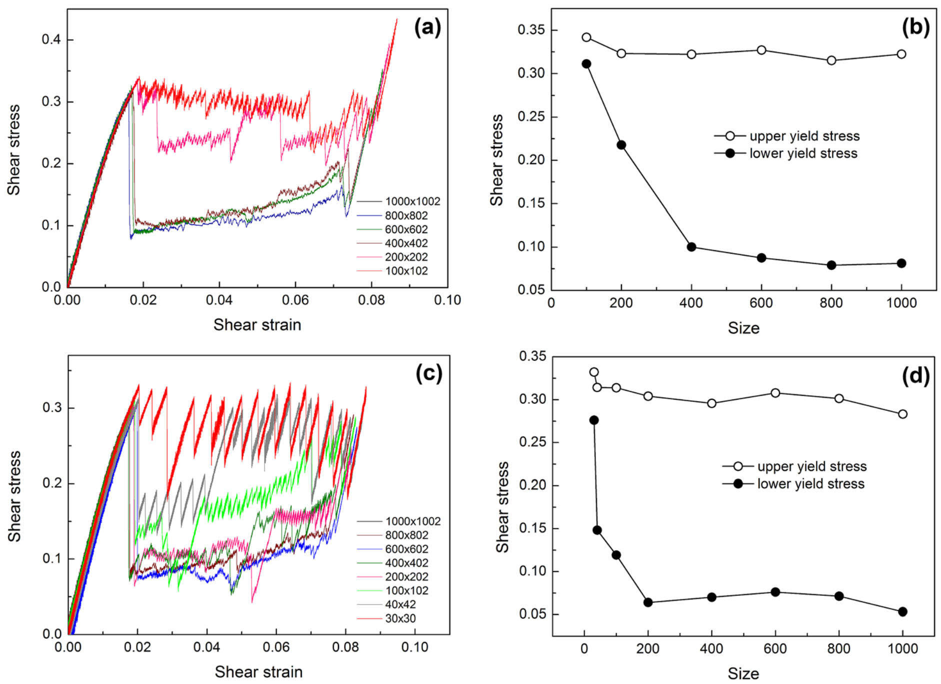

FIG. 4. Mechanical switching spectra for hard (top) and soft (bottom) systems. The difference of the upper and lower yield strain shows the size dependence of the switching process. The critical size is 200 (hard) and 50 (soft) interatomic distances.

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