Tunable, Multi-State Switching in Ferroelectric Thin Films

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With CMOS scaling approaching its fundamental size limitations, it is imperative to explore novel information processing techniques with increased functional diversities and new modalities of operation. Recent work in oxide electronics has suggested that adaptive computational systems, such as neuromorphic devices, which mimic the human brain with dynamic self-adjustment operations, could dramatically increase computational density and power efficiency beyond Boolean computations[1,2]. In particular, the realization of adaptive neuromorphic devices requires multi-value states in devices that could be tuned in a non-volatile manner. Thus a number of different materials such as ferroelectrics, ferromagnetics, and phase-change materials that exhibit multiple states have been considered as possible candidates for such applications. Prior work on ferroelectrics has primarily relied on extrinsic approaches to stabilize stochastic and partial switching to accomplish multi-state function[3,4]. Here, we explore the potential for tunable, multi-state polarization via intrinsic control of switching kinetics. In this work, 80 nm $La_{0.7}Sr_{0.3}MnO_3$ / 100 nm $Pb_{0.2}Zr_{0.8}TiO_3$ / 20 nm $La_{0.7}Sr_{0.3}MnO_3$ / $SrTiO_3$ (001) and (111) heterostructures were prepared using pulsed-laser deposition. Upon the application of Positive-Up-Negative-Down (PUND) pulse series, it is noted that (111)-oriented heterostructures exhibit a three-state switching process wherein the intermediate state appears with an ~50% polarization change from the initial state, while (001)-oriented films show conventional bipolar switching. Furthermore, we observe that the duration of the intermediate state in (111)-oriented films can be tuned by pulse width where smaller pulse widths induce a more stable intermediate state within a larger voltage range. Our results also indicate that by tuning the delay time between two pulses, namely, adjusting the rate of driving voltage, the intermediate states can be tuned across more than 10 distinctive polarization states between the fully-poled initial and final states. In addition, these tunable intermediate states are deterministically reproducible and highly stable, showing less than 5% polarization change over 3000 sec. in retention measurements. In order to understand the structural origin of the switching mechanism, we performed piezoresponse force microscopy to explore the evolution of domain structures for these different intermediate states. Two distinctive twinning structures are observed in these intermediate states: Type-I twinning structures which are fully-poled and Type-II twinning structures which are half-poled. The evolution in the volume fraction of these two types of twinning structures leads to a continuous polarization state change that gives rise to multiple intermediate states. Furthermore molecular dynamics (MD) simulations are performed to reveal the mechanism of volume fraction change of twinning structures with the rate of driving force. MD studies suggest that two kinetically distinctive switching processes are present in (111)-oriented films including a fast ferroelectric switching process that occurs directly between fully down-poled to fully up-poled Type-I twinning structures, and a slow ferroelastic switching that occurs via the pathway from fully down-poled Type-I twinning structures to half up-poled Type-II twinning structures and back to fully up-poled Type-I twinning structures. Due to the kinetic nature of these two switching processes, tuning the rate of driving voltage can effectively control the volume fraction of domains that undergo each type of switching process and therefore give rise to tunable intermediate states that possess different fractions of twinning structures.

References:

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