In-situ transmission electron microscopy investigation of stress-induced reversible charged domain walls in ferroelectrics

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Ferroelectrics have been extensively utilized in electronic devices like actuators, capacitors and memories due to their unique electric hysteresis performance. Charged domain walls (CDWs) in ferroelectrics barely exist because of their high electrostatic energy. Head-to-head and tail-to-tail domain structures lead to bound charges at the domain walls that restrains the formation of CDWs at the equilibrium state. Theoretical simulations and experimental measurements demonstrated that dense CDWs can effectively enhance the piezoelectric response of ferroelectrics [1]. The conductivity of CDWs is tunable, which is critical for some applications. Recent investigations showed that CDWs formed only when an electric field was applied [2]. Here we demonstrate that mechanical stress can also be used to introduce CDWs, which avoids a potential issue of current leakage caused by an applied electric field.

In our study, we applied an in-situ straining transmission electron microscopy (TEM) technique to explore the possibility of introducing CDWs in a barium titanate ($BaTiO_3$) single crystal via mechanical stressing. Pillars for TEM observation were prepared using the focused ion beam technique. Fig.1 (a) shows a TEM image of domains in $BaTiO_3$ before stressing. They are determined to be 90° domains based on the crystallographic orientation of the domain walls. Compressive stressing led to the formation of a high density of nanoscale domains separated by domain walls that were aligned with the [100] direction (Fig.1 (b)). These new domain walls are all CDWs because of their head-to-head or tail-to-tail domain structures. The original domain structure returned after the full retraction of external stress (Fig.1 (c)).

Fig.2 presents a schematic diagram demonstrating the evolution of 90° domains under mechanical stress. Compressive stressing forced the formation of fine purple domains within the originally yellow domain areas through 90° ferroelastic domain switching, which produced a high density of positive and negative bound charges near the domain walls. Removing the external stress led to the disappearance of the CDWs and the original domain structure returned. This is the first experimental evidence demonstrating that mechanical stress can be used to introduce reversible CDWs. Phase field modeling is used to simulate the dynamic process of the formation of CDWs. Our results are undoubtedly of great importance for the investigation of electromechanical properties of ferroelectrics.

References:
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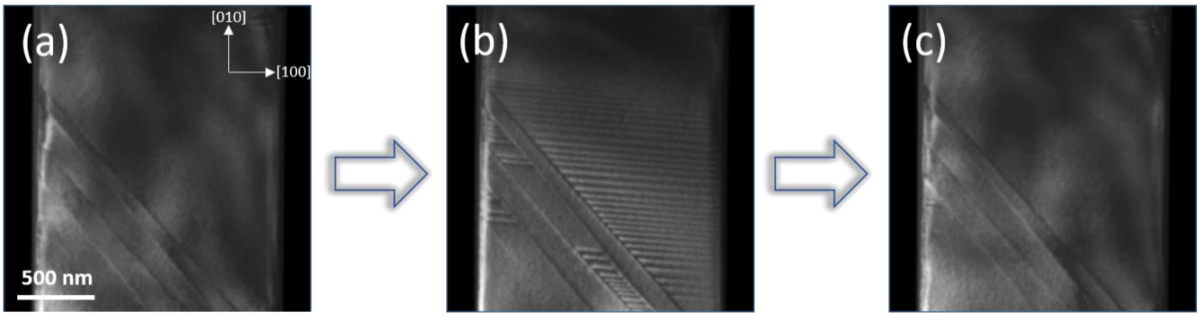

Fig. 1: TEM images of domain structures in $BaTiO_3$ before (a), during (b) and after (c) mechanical stressing.

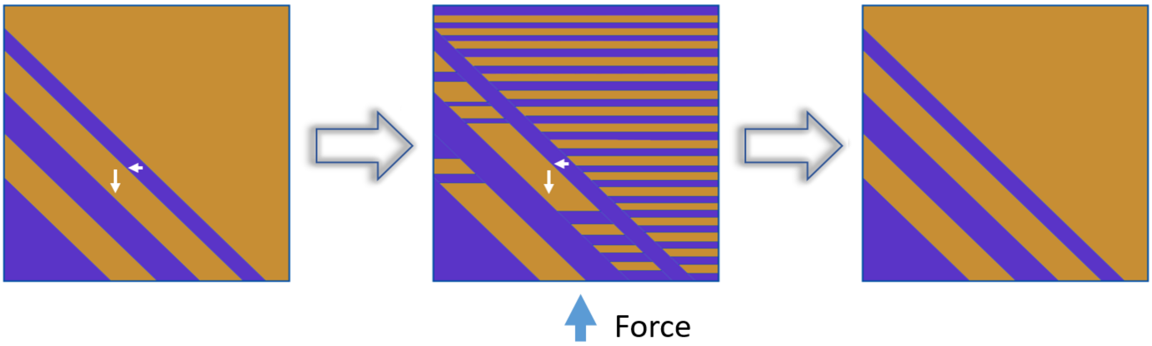

Fig. 2: A schematic diagram of the 90° domain structures in $BaTiO_3$ corresponding to Figure 1. The white arrows indicate the polarization of the domains. Two types of domain areas exist: the yellow and purple domains that have their polarization pointing downward and leftward, respectively.