

Probing Mobile-point-defect-mediated Nanodomain Evolutions in Ferroelastic-ferroelectrics Under High Stress with *In-situ* TEM

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Ferroelastic-ferroelectrics are multi-functional materials with attractive applications such as actuator, memory devices and flexible/wearable electronic devices [1-7]. This class of intrinsically brittle materials exhibits unique unconventional deformation mechanisms that could be potentially utilized to engineer novel electric-mechanical components. Notably, the close-correlated domain evolutions and phase transformations in ferroelastic-ferroelectrics is reported to generate a complex hierarchical structure that is responsible to the superelastic deformation behaviors of the materials at nanoscale [7]. By applying high stress to the material, hierarchical nanodomain evolutions can be introduced into ferroelastic-ferroelectrics, effectively tuning their properties. However, the complex nanodomain evolutions are challenging to understand: the domain mobility, the distributions of local strains and mobile point defects at domain walls, and the growth of the bundle domain structures have been discussed for a long time with controversy [1,2]. Small scale mechanical *in-situ* TEM observations provide unique real-time capability of capturing the nanodomain evolution while the stress field is applied. Here, by applying *in situ* TEM mechanical tests couple with 4D-STEM techniques that are capable of generating nm-resolved strain mapping in an aberration-corrected transmission electron microscope [1,4,5,6], we studied free-standing single-crystal BaTiO₃ and PMN-PT sub-micrometer pillars, to show the mechanism of the mobile-point-defect-mediated nanodomain evolutions (Figure 1) in ferroelastic-ferroelectrics under high stress. This reversible domain-mediated deformation mechanism allows for superelastic deformation of nominally rigid single crystal oxide nanowires. The use of 4D-STEM to dynamically map the ferroelectric domain structure and local strain state over large fields of view is transformative to our ability to understand this complex deformation behavior in a quantitative manner.

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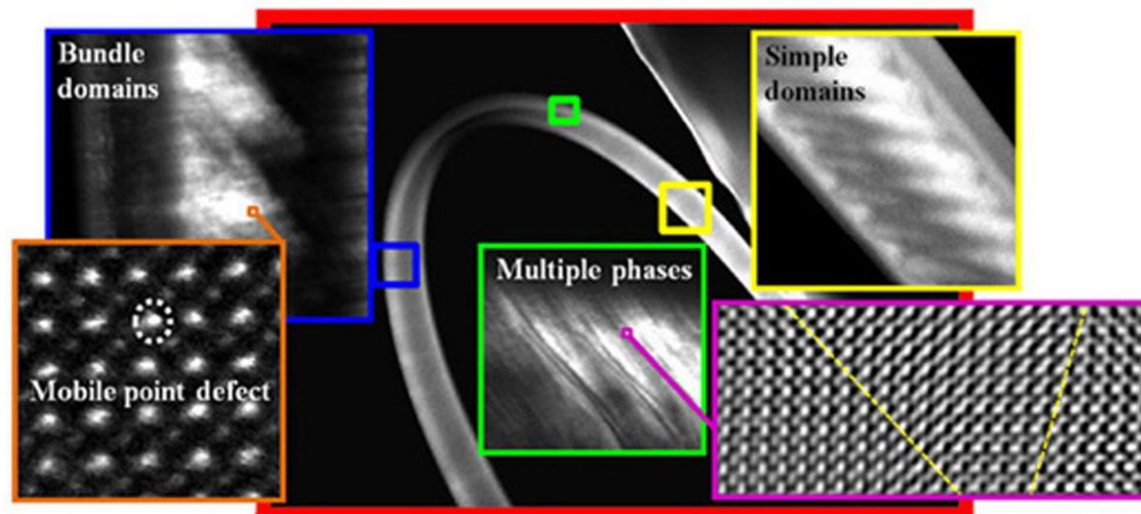


Figure 1. The mobile-point-defect-mediated nanodomain evolutions in a free-standing single-crystal BaTiO₃ sub-micrometer pillar under heavily bending.

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