Deterministic Control over Symmetry States in Mixed Phase $BiFeO_3$

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Under a sufficiently large compressive epitaxial strain imposed via substrate, thin films of $BiFeO_3$ transform into functionally active, nanoscale mixtures of tetragonal-like (T) and rhombohedral-like (R) monoclinic phases.[1] The interfaces between these R and T phases are highly elastically strained, remaining almost dislocation free despite substantial differences in strain and polarization.[1,2] The fact that the films are also energetically in a mixed phase space, should result in profound property malleability or plasticity,[3,4] where the relative proportion of phases in the films and therefore their resulting properties can be readily varied in a continuous manner in response to external forces.

We investigate the use of local applied stress as a tool for achieving deterministic reversible control of the mixed phase populations in $BiFeO_3$ thin films and subsequently as a means to exert direct control over the piezoresponse behaviour. We show that the phase population can be reversibly controlled through selective application of stress and electric field to increase the proportion of R and T phase respectively. This observed competition between phases is shown to be consistent with first-principles based effective Hamiltonian simulations employed to evaluate the thermodynamic competition under the influence of epitaxial strains, applied uniaxial stresses and electric fields. We experimentally verify the theoretical predictions by systematic application of uniaxial stress and electric field through an atomic force microscopy probe, illustrating deterministic control of coexisting phases in $BiFeO_3$ films. This enables quantitative predictions of the phase population of films grown to a range of compressive epitaxial strains and how these films will respond to external forces. Further study of the hysteretic behaviour of the films under applied stress demonstrates control over the electric field induced switching and provides a robust understanding of the observed R-T phase transitions. The work demonstrates how the property malleability of mixed phase $BiFeO_3$ can be exploited to achieve deterministic control over phase populations and resulting functionality, presenting an invaluable tool for growth of films with targeted functional properties. Similar approaches to those presented here are envisaged to be broadly applicable to thin functional oxide films in general.

References

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