Domain Engineering in Ferroelectric Tricolor Superlattices Probed by X-ray Diffraction

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The potential of ferroelectric nanodomains has stimulated intense research in order to contribute to the fundamental knowledge of ferroelectric materials at the nanoscale. In order to understand the future challenge for ferroelectric nanodomains, it is necessary to develop the structures to produce these nanodomains. Although nanoscale ferroelectric thin films can exhibit nanodomain structures, their manipulation is delicate due to the polarization-screening charges at the film surface that can perturb the domain structure. The superlattice geometry can overcome these difficulties and provides an ideal system to investigate domains at the nanoscale. By alternating individual ultra-thin ferroelectric layers with other ferroelectric, paraelectric layers, it is possible to produce ferroelectric domains which are isolated from surface or film-substrate interfaces, and to change the electrostatic domain boundaries as well as the mechanical strain. We have investigated tricolor superlattices based on two ferroelectrics ($PbTiO_3$, $PbZr_{0.2}Ti_{0.8}O_3$) and paraelectric $SrTiO_3$ [N. Lemée et al., ACS Appl. Mater. Interfaces, 7, 19906 (2015)]. In this original geometry of 4 layer periodicity, an ultrathin $SrTiO_3$ layer (1 nm thick) modifies the electrostatic boundary conditions between the ferroelectric layers and produces a depolarization field. Combined with the electrostatic effect, there is competition between different polarization orientations via different strain states in the ferroelectric layers. Indeed we have determined that the $PbZr_{0.2}Ti_{0.8}O_3$ layers are compressively strained whereas the $PbTiO_3$ and $SrTiO_3$ layers are under tensile strain. We have studied the ferroelectric domains in these superlattices, using the combination of standard reflection geometry and grazing incidence X-ray diffraction which provides a powerful technique to investigate the polar nanodomain structure. We have evidenced the presence of 180° ferroelectric nanodomains of alternate "up" and "down" polarization, produced to minimize the electrostatic energy arising from the $SrTiO_3$ induced depolarizing field. Moreover, we demonstrate that a polarization rotation takes place in the tricolor superlattice due to the polarization competition in the ferroelectric layers, and we show that this polarization rotation is associated to a monoclinic Mc phase as revealed by the splitting of the (HHL) and (H0L) reciprocal lattice points. Finally, we have also investigated the temperature dependent behavior, from room temperature to above the ferroelectric transition temperature, of the polar nanodomains. Diffuse X-ray scattering combined with numerical simulations [A. Boulle, I. C. Infante and N. Lemée, J. Appl. Cryst., 49, 845 (2016)] provides information about the thermal evolution of the period disorder as well as the roughness and thickness of the domain walls. The research discussed in this talk shows that these tricolor superlattices provide a powerful tool to study the role of electrostatic and strain effects in the control of ferroelectric nanodomain structures.