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Phase transitions in ferroelectric superlattices

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The construction of artificial ferroelectric superlattices with fine periodicity presents exciting possibilities for the development of new materials with extraordinary properties and furthermore a probe for understanding the fundamental physics of ferroelectric materials. Our superlattices of $PbTiO_3/SrTiO_3$ are prepared on conducting 0.5% Nb doped (001) $SrTiO_3$ substrates using off-axis RF magnetron sputtering. Cross-sectional TEM investigations were performed on several samples and reveal the coherent growth and artificial layering of the samples. Further structural characterization using standard θ-2θ x-ray diffraction was performed on a series of 20 bilayer superlattices in which the $PbTiO_3$ thickness was varied from 54 to 1 unit cells while the $SrTiO_3$layer thickness was maintained at 3 unit cells. Intuitively one expects, as the thickness of the $PbTiO_3$ layers relative to the $SrTiO_3$ layers is reduced, a decrease of the ferroelectric polarization which should result in a concomitant decrease of the average lattice parameter. This is indeed the behaviour we observe for superlattices $PbTiO_3/SrTiO_3$ n/3 where n is greater than 3. However, surprisingly, the 2/3 and 1/3 superlattices display larger average lattice parameters which indicate a recovery of ferroelectricity at very small $PbTiO_3$ layer thicknesses, a finding we confirmed using atomic force microscopy. The experimental finding thus stands in stark contrast to the intuitive expectation of a ferroelectric-paraelectric phase transition in this system as the ferroelectric component is reduced and we find further that the temperature of the ferroelectric-paraelectric phase transition is also greatly modified. Due to the excellent quality of the samples we are able to present the results of a number of detailed structural and electrical characterizations, along with the development of first principles based theoretical models, which cast further light on the fascinating phase transition behaviour of this system. Through this we can gain an insight into how we can understand and control the behaviour of ferroelectricity as the physical dimensions are reduced and the relevant boundary conditions are modified.