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Probing nanoscale ferroelectricity by ultraviolet Raman spectroscopy[1]

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Conventional vibrational spectroscopies operating in visible and infrared range fail to measure the phonon spectra of nanoscale ferroelectric structures because of extremely weak signals and the overwhelming substrate contribution. In this talk, application of ultraviolet (UV) Raman spectroscopy for studies of lattice dynamics and ferroelectric phase transitions in nanoscale ferroelectrics will be presented. We demonstrate that UV Raman spectroscopy is an effective technique allowing the observation of phonons and determination of the ferroelectric phase transition temperature (T_c) in nanoscale ferroelectrics, specifically, $BaTiO_3/SrTiO_3$ superlattices having the ferroelectric $BaTiO_3$ layers as thin as 1 unit cell, and single $BaTiO_3$ layers as thin as 4 nm. $BaTiO_3/SrTiO_3$ superlattices and ultrathin $BaTiO_3$ films studied were grown by molecular beam epitaxy on $SrTiO_3$ as well as $GdScO_3$ and $DyScO_3$ substrates. Excellent epitaxial quality and atomically abrupt interfaces are evidenced by X-ray diffraction and high resolution transmission electron microscopy. UV Raman results show that one-unit-cell thick $BaTiO_3$ layers in $BaTiO_3/SrTiO_3$ superlattices are ferroelectric with the T_c as high as 250 K, and induce the polarization in much thicker $SrTiO_3$ layers adjacent to them. The T_c in superlattices was tuned by hundreds of degrees from $\sim$170 to 650 K by varying the thicknesses of $BaTiO_3$ and $SrTiO_3$ layers. Using scandate substrates enables growth of superlattices with systematically changed coherent strain, thus allowing studying the stress effect on the ferroelectric phase transitions. UV Raman data are supported by the thermodynamic calculations of polarization in superlattices as a function of temperature. The work was done in collaboration with A. Soukiassian, W. Tian, D.G. Schlom, Y.L. Li, L.-Q. Chen, X.X. Xi (Pennsylvania State University), A. Bruchhausen, A. Fainstein (Centro Atomico Bariloche & Instituto Balseiro, Argentina), R. S. Katiyar (University of Puerto Rico), A. Cantarero (University of Valencia, Spain), K.J. Choi, D.M. Kim, C.-B. Eom (University of Wisconsin), H.P. Sun, X.Q. Pan (University of Michigan), S.M. Nakhmanson; K.M. Rabe (Rutgers University), Q.X. Jia (Los Alamos National Laboratory)

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