Unleashing Strain Induced Ferroelectricity in Complex Oxide Thin Films via Precise Stoichiometry Control

Ryan C. Haislmaier, Everett D. Grimley, Michael D. Biegalski, James M. LeBeau, Susan Trolier-McKinstry, Venkatraman Gopalan and Roman Engel-Herbert

Strain tuning has emerged as a powerful means to enhance properties and to induce otherwise unattainable phenomena in complex oxide films. However, by employing strain alone, the predicted properties sometimes fail to emerge. In this work, the critical role of precise stoichiometry control for realizing strain-induced ferroelectricity in $CaTiO_3$ films is demonstrated. An adsorption controlled growth window is discovered for $CaTiO_3$ films grown by hybrid molecular beam epitaxy, which ensures an excellent control over the Ti:Ca atomic percent ratio of <0.8% in the films. Superior ferroelectric and dielectric properties are found for films grown inside the stoichiometric growth window, yielding maximum polarization, dielectric constant, and paraelectric-to-ferroelectric transition temperatures. Outside this growth window, properties are severely deteriorated and ultimately suppressed by defects in the films. This study exemplifies the important role of precise compositional control for achieving strain-induced properties. Untangling the effects of strain and stoichiometry on functional properties will accelerate both fundamental discoveries yet to be made in the vast materials design space of strained complex oxide films, as well as utilization of strain-stabilized phenomena in future devices.