Defect- and Interface-Driven Behavior in Oxide Thin Films

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Complex and functional oxides possess a wide range of properties, providing platforms for tunable and emergent behavior via interfaces, defects, and reduced dimensionality. In this talk, I will discuss the effect of defects on magnetic properties and the emergence and role of unique phonons in thin oxide films. These examples cover a range of experimental and theoretical collaborations spanning multiple projects and career stages.

Given the sensitivity of oxide magnetism to local crystal structure, growth conditions may lead to unexpected or undesirable outcomes. In molecular-beam epitaxy (MBE)-grown Co-substituted $BaTiO_3$ films [1], despite confirmed uniform distribution, the films remain non-magnetic due to oxygen-vacancy-mediated spin coupling. In heterostructures, thin nominally conductive magnetic layers may be especially defect sensitive. In pulsed-laser deposition (PLD)-grown $(SrTiO_3)^5$-$(SrRuO_3)^n$-$(SrTiO_3)^5$ films [2], transport measurements indicate that the $n = 2$ structure is conductive and ferromagnetic, but the $n = 1$ structure is insulating and non-magnetic. Scanning transmission electron microscopy (STEM) reveals Ru-Ti intermixing in the $n = 1$ structure, which density functional theory (DFT) shows leads to AFM-like insulating states. For $(CaRuO_3)^n$-$(SrRuO_3)^1$-$(CaRuO_3)^n$ films [3], as n decreases, local structural perturbations near the SRO layer cause an insulating and magnetic state, suggesting an oxide engineering pathway. As another route, DFT predicts that carrier doping can couple magnetic and polar behavior, enabling switchable spin textures in h-$LuFeO_3$/MnSTe [4].

Like the deviations from bulk behavior seen above at thin-film interfaces, emergent phonon behavior can also occur as the superlattice layer thickness decreases. In PLD-grown $(SrTiO_3)^n/(CaTiO_3)^n$ superlattices [5], STEM imaging and spectroscopy of the structure and phonon modes along with optical spectroscopy measurements show the emergence of interface modes with decreasing n, which eventually become the dominant vibrational response. These observations were corroborated by extensive DFT calculations to understand structural changes and potential defect effects on the localization of the phonon modes. Similar techniques were used to probe related systems like grain boundaries [6]. Lastly, the role of phonons in the non-resonant excitation and temperature dependence of emission in optically active Er_2O_3 films and how modifying the phonons may affect emission will be discussed [7].

As illustrated, defects, interfaces, and reduced dimensionality are not just perturbations to ideal behavior, but ingredients that either prevent or enable unique properties depending upon the system. Understanding how these perturbations manifest and how to control them provides a route from explaining unexpected behavior to engineering functionality.

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