

## Evidence for a Thickness-Dependent Crossover from Charge- to Strain-Mediated Magnetoelectric Coupling in $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ / $\text{PbZr}_{0.2}\text{Ti}_{0.8}\text{O}_3$ Thin Film Oxide Heterostructures

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Layered magnetoelectric thin film materials, which exhibit coupling between ferroelectric polarization and magnetization, have received considerable attention for use in a new generation of “spintronic” memories [1]. Coupling at the interface between the layers can occur by various mechanisms, including charge and strain transfer, which are sensitive to local microstructure, chemistry, and defects [2]. The performance of devices for applications such as spin valves and tunnel junctions hinges on quantifying and controlling the local features that mediate coupling.

Here we present a comprehensive structural, chemical, and magnetic analysis of LSMO / PZT / LSMO and LSMO / PZT /  $\text{SrRuO}_3$  (SRO) thin film heterostructures in significantly different charge and strain states. Scanning transmission electron microscopy (STEM) and geometric phase analysis (GPA) are used to directly map local strain, while electron energy loss spectroscopy (EELS) is used to explore valence changes at the LSMO / PZT interface. These results are complemented by depth-resolved polarized neutron reflectometry (PNR) measurements that provide information about the spatial dependence of magnetization [3]. These experimental observations are combined with phenomenological models and first-principles density functional calculations, which reveal the role of local elastic strains on the electronic distribution, to understand the suppression in the LSMO Curie temperature ( $T_C$ ).

Figure 1 shows local strain gradients measured by GPA. It is clear that there is a significant gradient in strain that depends on the thickness of the PZT underlayer. This gradient reaches a maximum near the vacuum surface, where neutron results show the magnetization is most suppressed. Likewise, STEM high-angle annular dark field (HAADF) images and electron energy loss spectroscopy (EELS) shown in figure 2 reveal changes in PZT polarization and an induced change in Mn valence near the interface. This is associated with an interfacial magnetization measured in PNR. From the magnitude of the induced magnetization changes we find local strain dominates coupling beyond the ultrathin (~4 nm) limit of LSMO. This suggests that layer thickness is crucial parameter in determining device performance and we offer ways to tune the geometry of these structures to favor a particular coupling mode.

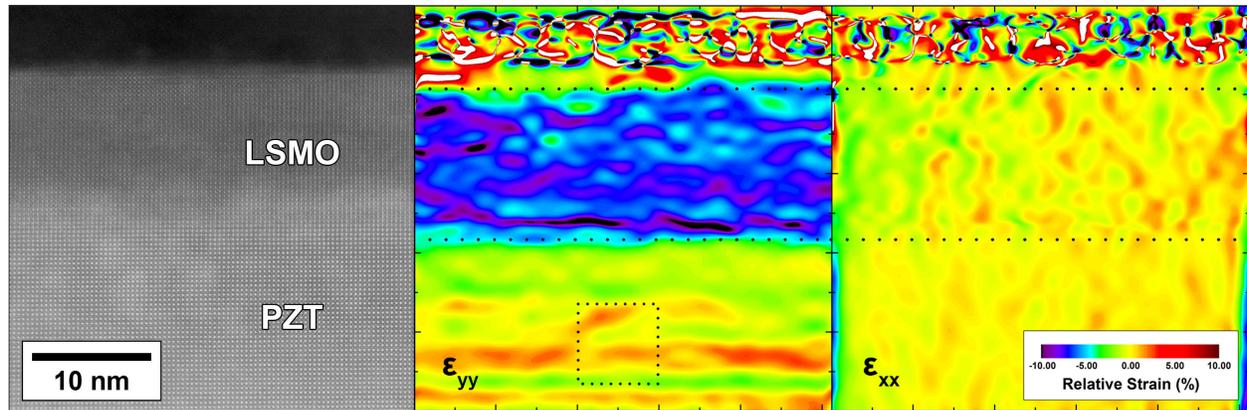
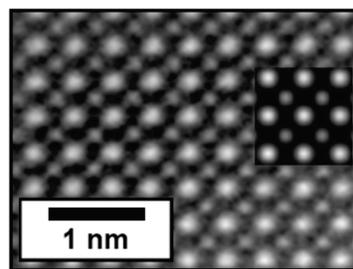


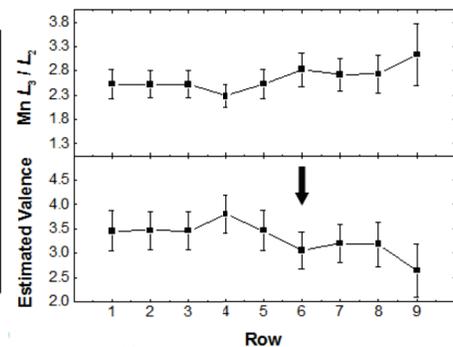
Figure 1: Geometric phase analysis of the LSMO / PZT interface showing out-of-plane strain ( $\epsilon_{yy}$ ) gradients that depends on PZT underlayer thickness. This is responsible for local  $T_C$  suppression.

Figure 2

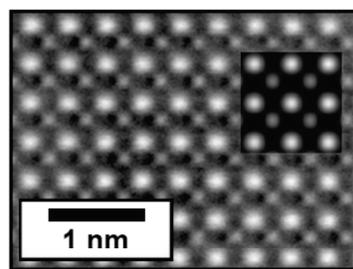
Left: High-angle annular dark field (HAADF) images of the PZT layer showing local polarization near the LSMO interface. The insets show multislice simulations of this same structure.



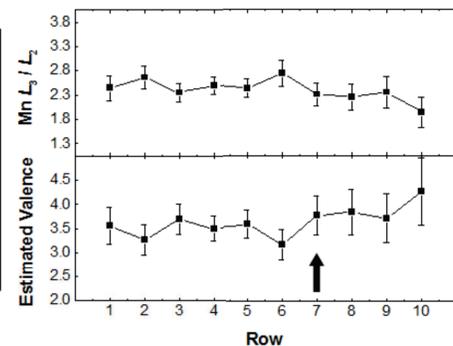
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Right: Mn  $L_{23}$  ratios measured near the LSMO / PZT interface (indicated by arrows), showing changes in Mn valence across the interface induced by charge screening.



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## References and Acknowledgments

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- [2] Vaz, C. A. F. (2012). Electric field control of magnetism in multiferroic heterostructures. *J. of Phys. Cond. Mat.*, 24(33), 333201.
- [3] This research was supported in part by Oak Ridge National Laboratory's Shared Research Equipment (ShaRE) User Program, which is sponsored by the Office of Basic Energy Sciences, U.S. Department of Energy. Polarized neutron reflectometry experiments were performed at the Spallation Neutron Source at Oak Ridge National Laboratory, managed by UT-Batelle, LLC for the U.S. Dept. of Energy. Part of this work was conducted in Drexel University's Centralized Research Facilities. We gratefully acknowledge the support of National Science Foundation Grants #DGE-0654313, #CMMI-1031403, #DMR-0908779, and #DMR-0821406, as well as Office of Naval Research Grant #N00014-1101-0296 / 12PR02130-02.