

Intrinsic Defects Pinned at the Domain Wall in Lithium Niobate

G. Stone¹, H. Steigerwald², K. Buse², W. Sohler³, V. Gopalan⁴, S. Phillpot⁵, V. Dierolf¹

1. Physics Department, Lehigh University, 16 Memorial Drive East, Bethlehem, PA 18015, USA

2. Institute of Physics, University of Bonn, D-53115 Bonn, Germany

3. Applied Physics, University of Paderborn, D-33098 Paderborn, Germany

4. Department of Materials Science and Engineering, Pennsylvania State University, University Park, PA 16802, USA

5. Department of Materials Science and Engineering, University of Florida, Gainesville, FL 32611, USA

Ferroelectric domain engineering is a powerful technique that can be used to create nonlinear optical devices based on quasi-phase matching. However, creating domains smaller than $1\mu\text{m}$ poses significant challenges. This is partially due to an incomplete understanding of the properties of the ferroelectric domains; particularly in regards to the interaction of the domain wall and intrinsic defects, which determine the domain shapes and growth dynamics. Such defects are present in large numbers in congruent LiNbO_3 crystals due to lithium deficiency. Due to the ionic nature of these intrinsic defects, they constitute defect dipoles that can be oriented in different ways in respect to the ferroelectric axis. In particular, after a domain inversion at room temperature these dipoles may not reorient along with the ferroelectric polarization and find themselves in an energetically unfavorable, “frustrated,” alignment that creates strain in the material. It has been found that such strain is released over time and by thermal treatments as shown by the observation of characteristic shifts in the Raman spectra [1].

The focus of our work was the investigation of the interaction of the intrinsic defects with domain walls using scanning confocal Raman microscopy. For these studies, it is critical to use well-annealed samples that are free of the (above mentioned) strain-induced Raman effects in the bulk crystal and charge induced effects [2]. In such samples, changes of the Raman spectra are limited to the close vicinity ($\approx \pm 250\text{nm}$) of the domain wall (see Fig. 1). The observed changes are spectral shifts of the Raman peaks from their positions in the bulk crystal. The directions of these shifts closely resemble those seen in freshly created domains [1]. However, at the domain wall these shifts are permanent and withstand thermal treatments. The degree of shift at the domain wall is dependent on the stoichiometry of the sample and hence the number of intrinsic defects due to lithium deficiency. These results lead to the conclusion that at the domain wall, the defects and their alignment are permanently fixed. This finding is consistent with the modified energetic of defects at the domain wall found in simulations [3].

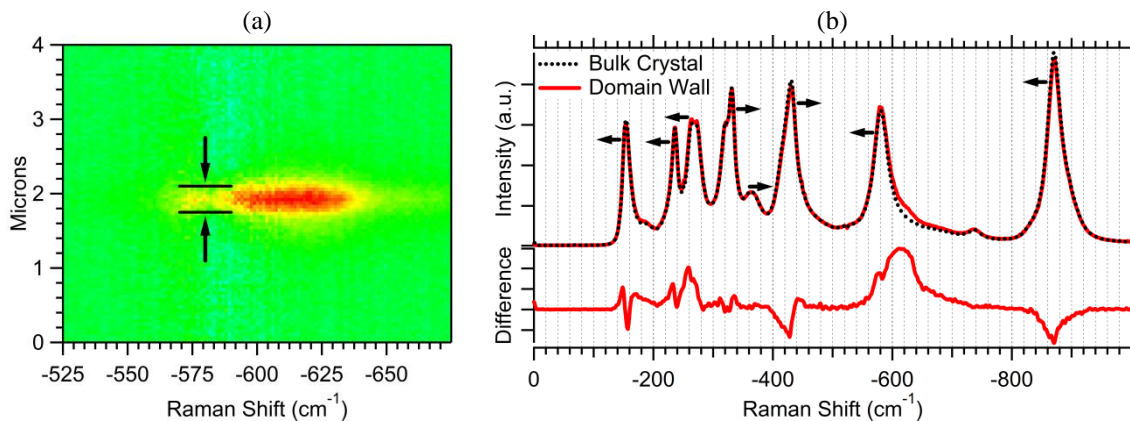


Fig. 1 (a) The changes in the Raman spectra near the $\text{E}(\text{TO})_8$ modes at a domain wall. (b) A comparison of the Raman spectrum taken from the bulk crystal and the domain wall along the z-axis in congruent Lithium Niobate. The derivative type shapes in the difference spectrum clearly indicate the shifts. Arrows indicate the direction of the shifts of the Raman peaks.

References

- [1] J. G. Scott, S. Mailis, C. L. Sones, and R. W. Eason, “A Raman study of single-crystal congruent lithium niobate following electric-field repoling,” *Applied Physics A* **79**, 691-696 (2004)
- [2] P. Capek, G. Stone, V. Dierolf, C. Althouse, and V. Gopalan, “Raman studies of ferroelectric domain walls in lithium tantalate and niobate,” *Physica Status Solidi (C)* **4**, 830-833 (2007).
- [3] H. Xu, D. Lee, S. B. Sinnott, V. Gopalan, V. Dierolf, and S. R. Phillpot, “Interactions of Defects and Domain Walls in LiNbO_3 – Insights from Simulations,” *IOP Conference Series: Materials Science and Engineering* **15**, 012003 (2010)