Investigation of Ferroelectric Properties of Oxide Superlattices at Low Temperatures

How do strain, dimensionality, and polarization compete in the low-dimensional structure, $(SrTiO_3)_n(BaTiO_3)_mSrO$?

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Abstract

The oxide superlattice $(SrTiO_3)_nSrO$ has been identified as a promising tunable dielectric for modulating frequency signals, especially at 5G gigahertz frequencies, due to its epitaxial strain to enhance tunability and defect mitigating structure that reduces charged point defects. The next generation of this material, $(SrTiO_3)_n(BaTiO_3)_1SrO$ thin films, combines two ferroelectric materials, $SrTiO_3$ and $BaTiO_3$, in a superlattice broken up with non-ferroelectric SrO layers. This study explored the in-plane and out-of-plane ferroelectric and dielectric properties of the $(SrTiO_3)_n(BaTiO_3)_1SrO$ thin films at temperatures 80 K – 340 K for n=1, 3, and 5. Understanding the competing $SrTiO_3$'s and $BaTiO_3$'s polarizations' interactions can be useful for optimizing the material for planar and vertical devices. Measurements involved the use of Cr/Au interdigitated electrodes for in-plane characterization and a $SrRuO_3$ bottom electrode with Cr/Au circular top electrodes for out-of-plane characterization. The data was collected by combining ferroelectric and dielectric measurement software with a low temperature probe setup. By investigating dielectric constants with temperature and measuring ferroelectric hysteresis loops at 80 K, we found that in-plane shows a clear ferroelectric transition around 170-180 K and clearly shows ferroelectric properties at 80 K. Out-of-plane measurements show high leakage and did not clearly indicate ferroelectric properties.

Introduction/Background

Telecommunications companies have been exploring gigahertz frequencies for 5G cell phone technologies as fertile territory to address our ever-increasing need for more data.

Prof. Darrell Schlom's group at Cornell University identified the Ruddlesden-Popper phase, $(SrTiO_3)_nSrO$ (see Figure 1 for structure), as a candidate for its low loss in bulk and reduced point defects. The resulting paper found $(SrTiO_3)_nSrO$ phases exhibited high performance levels and was highly tunable for n>= 3 at frequencies up to 125 GHz. The superlattice periodicity, n, can additionally be exploited to achieve high performance at room temperatures. [1]

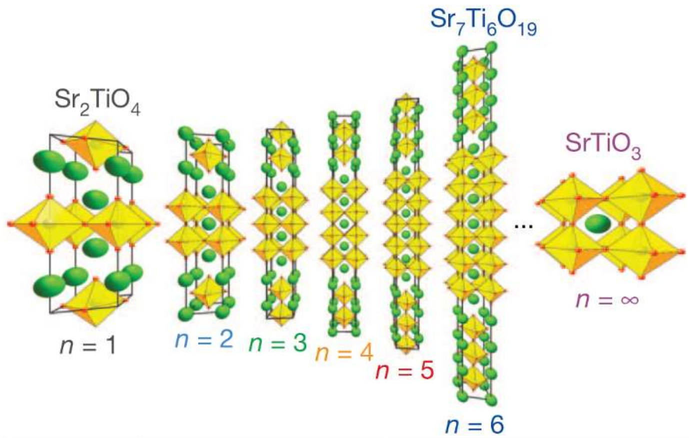

Figure 1 Structure of $(SrTiO_3)_nSrO$[1]

Objectives

In this project I measured the ferroelectric properties at low temperatures, where the polarization is strongest, of $(SrTiO_3)_n(BaTiO_3)_mSrO$ (structure shown in Figure 2) thin films, a candidate material for high-frequency applications.

This Ruddlesden-Popper superlattice combines two ferroelectric materials, $SrTiO_3$ and $BaTiO_3$, in a superlattice broken up with non-ferroelectric SrO layers. The addition of $BaTiO_3$ allows for further complexity and optimization of lattice constant and polarizability. Grown on the substrate (110) $DyScO_3$, the $(SrTiO_3)_n(BaTiO_3)_mSrO$ thin films have competing ferroelectric polarizations, $SrTiO_3$ being in plane, and $BaTiO_3$ out of plane. This could be useful for optimizing the material for planar or vertical devices if we can understand how these two materials interact with each other (See Figure 2). I had been measuring the in-plane and out-of-plane ferroelectric and dielectric properties in the low-dimensional structure of $(SrTiO_3)_n(BaTiO_3)_1SrO$ thin films for n = 1, 3, and 5.

Through this project we will better understand the electronic properties of the ferroelectric Ruddlesden-Popper materials.

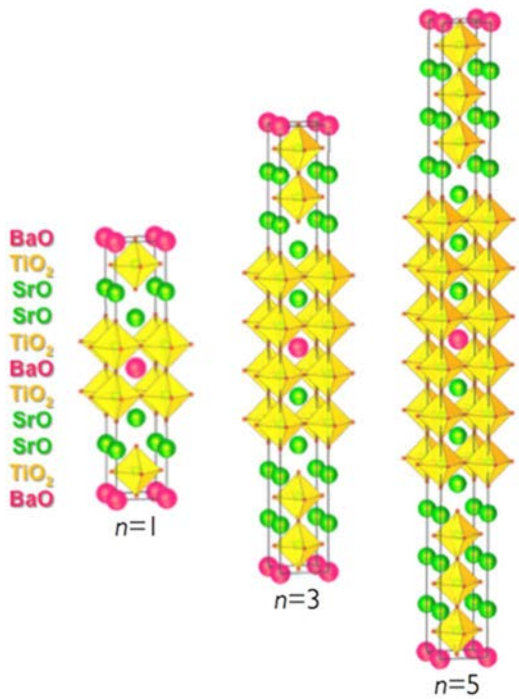

Figure 2 Structure of $(SrTiO_3)_n(BaTiO_3)_mSrO$[2]

Results

By investigating dielectric constants between the temperatures 80 K and 340 K and measuring ferroelectric hysteresis loops at 80 K, we found that in-plane samples show a clear ferroelectric transition around 170K at about 1000 (see Figure 3) for n=3 and around 180 K at about 1100 for n=5 (note: n as in $(SrTiO_3)_n(BaTiO_3)_1SrO$). In-plane samples for n=3, 5 are clearly ferroelectric at 80 K (see Figure 4). Additionally, out-of-plane measurements have high leakage (see Figure 5) and did not clearly indicate ferroelectric properties for any value of n (see Figure 6, for further explanations and graphs see Appendix). To lower dielectric leakage for the out of plane devices, recommendations are to use more oxide compatible material for both electrodes and to use smaller pad sizes if possible (the size used was 150 μm in diameter).

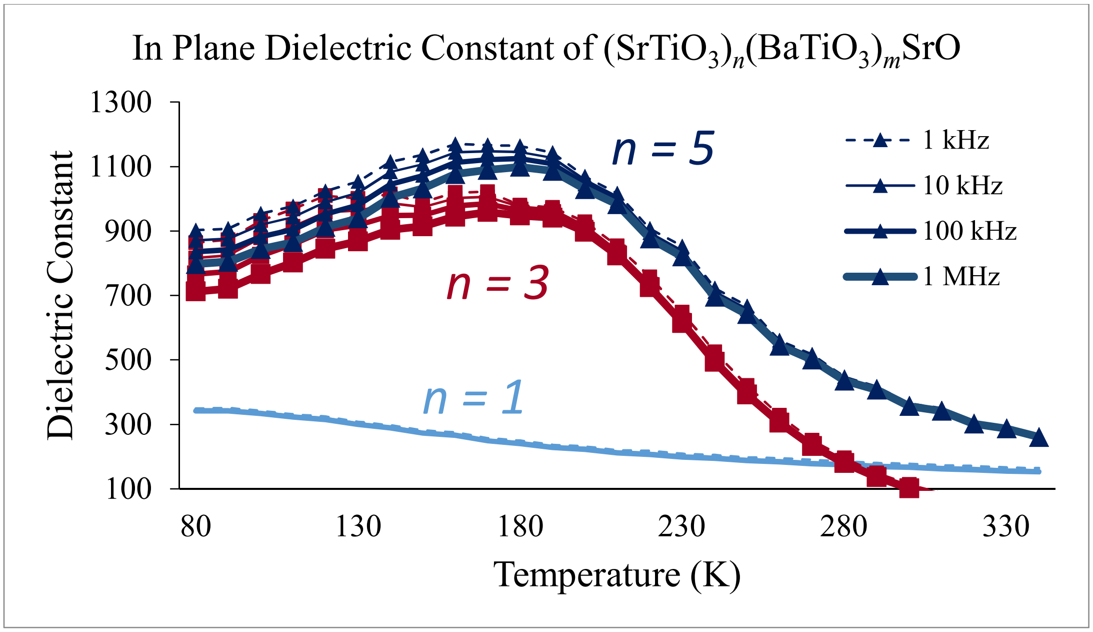

Figure 3: For n=1, there is no peak that would be indicative of a ferroelectric transition. In contrast, for n=3 and for n=5, there are peaks indicating ferroelectric properties.

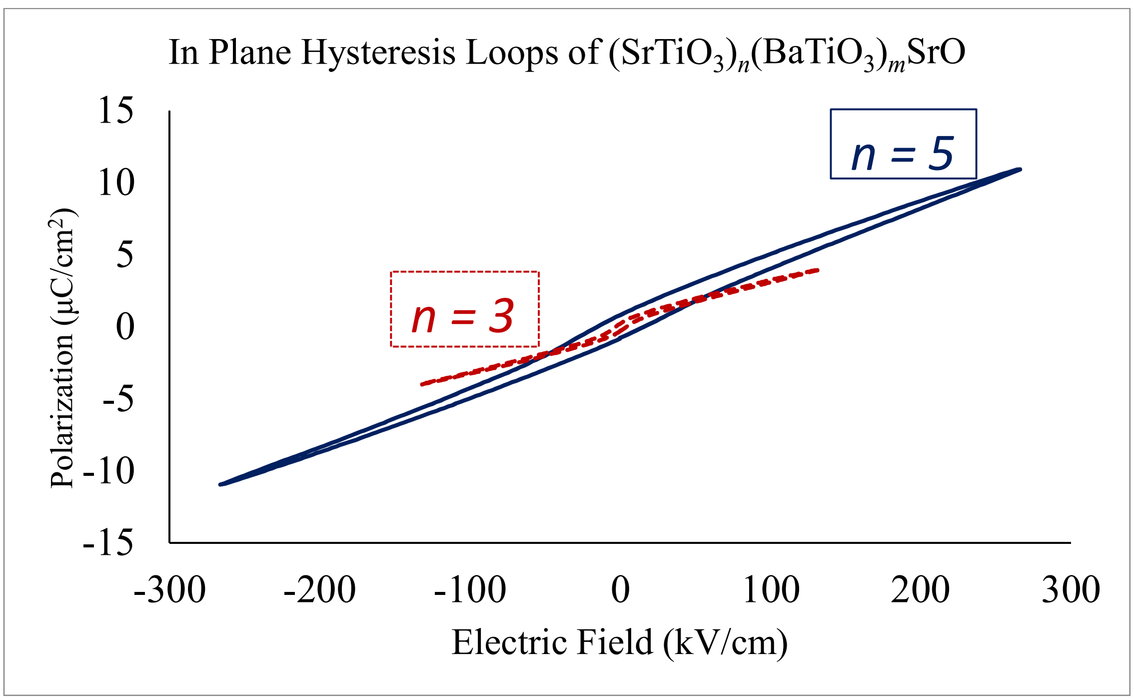

Figure 4: The shapes of the hysteresis loops indicate presence of ferroelectric properties (though probably not that strong).

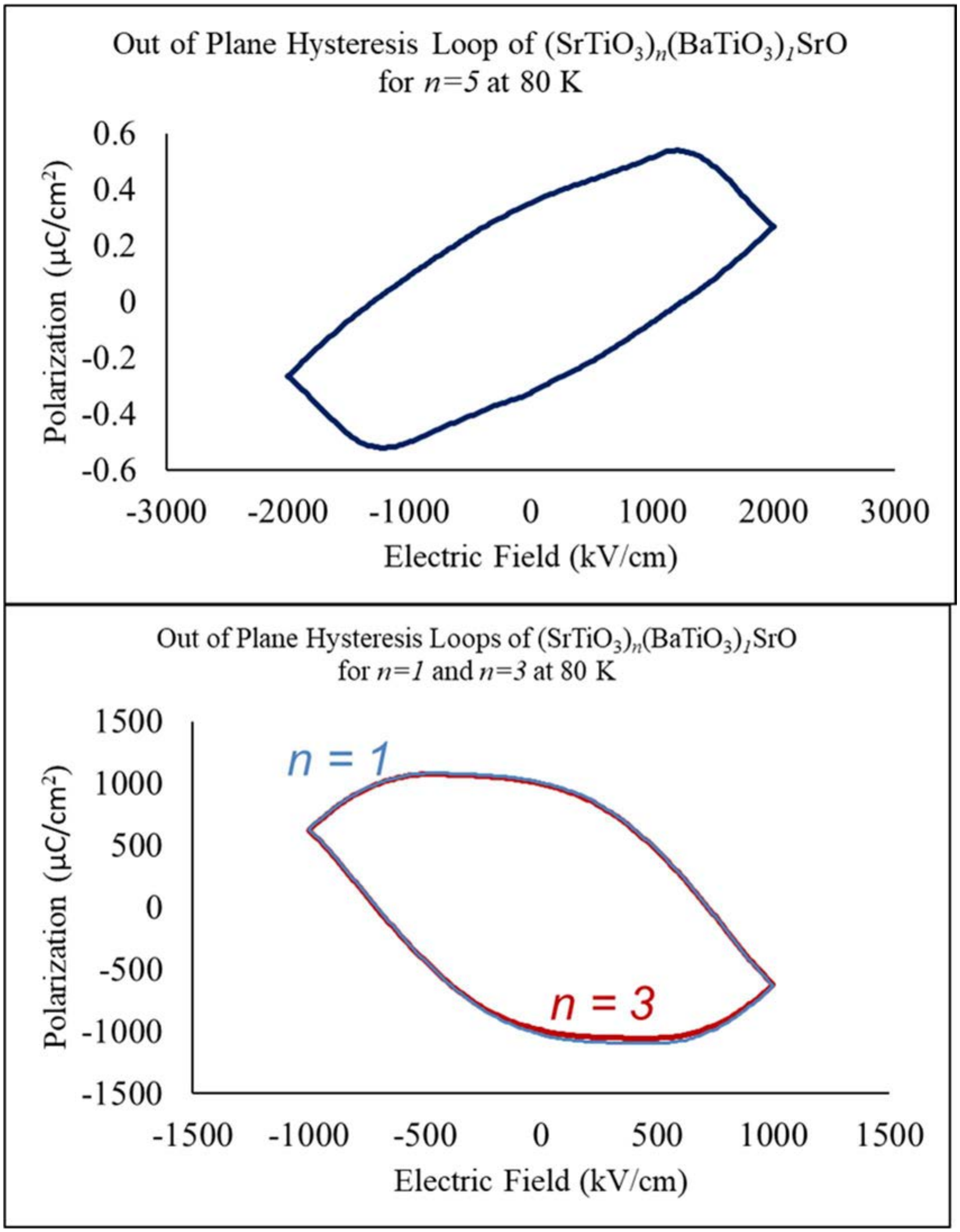

Figure 5:The top graph for the sample n=5, while indicating reasonable looking polarization values, indicates leakage from the shape. This leakage is made much more clear in the n=1 and n=3 samples in the bottom graph, which shows unreasonable polarization measurements.

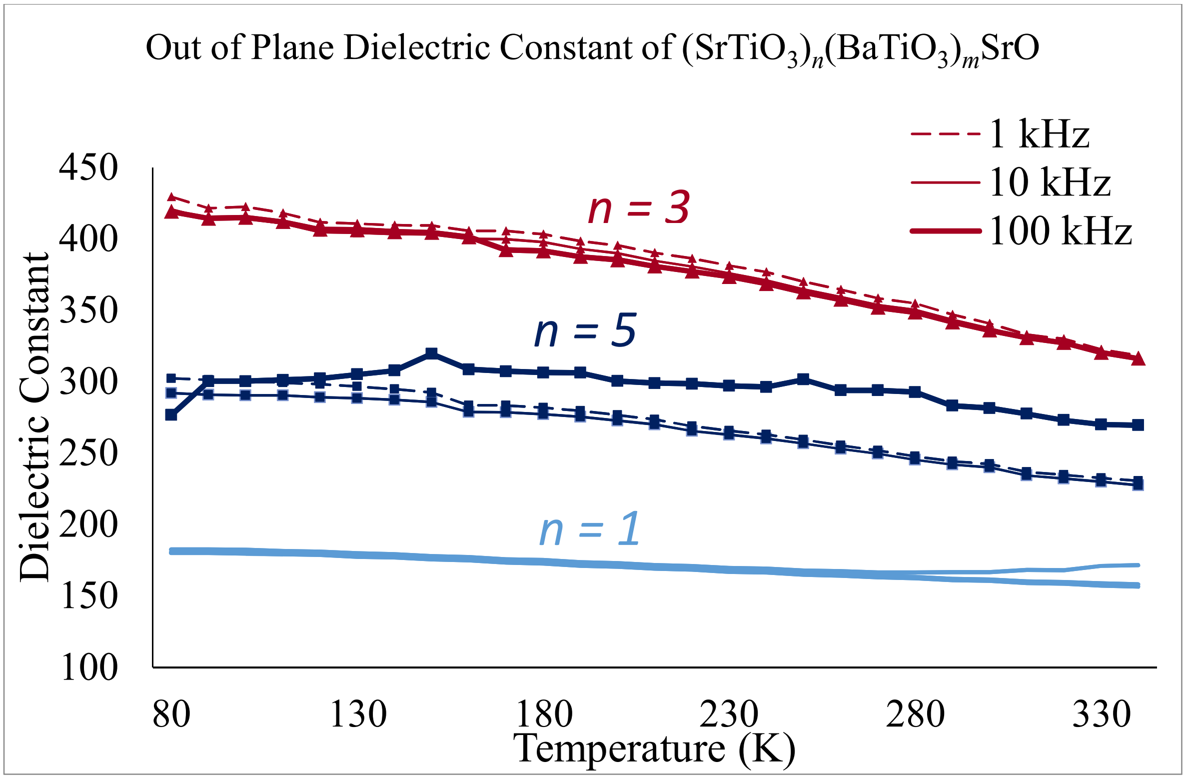

Figure 6: For all of the samples measured, there is no clear peak, and therefore, ferroelectric transition indicated. Compared to in-plane, out-of-plane dielectric constants are much lower.

Methods

Natalie Dawley grew the $(SrTiO_3)_n(BaTiO_3)_1SrO$ on (110) $DyScO_3$ and Kaynan Goldberg of North Carolina State University patterned the resulting samples with Cr/Au electrodes. Figure 7 shows the ferroelectric and dielectric testing devices on the blue cart. The cryoprobe contacted the surfaces for measurements.

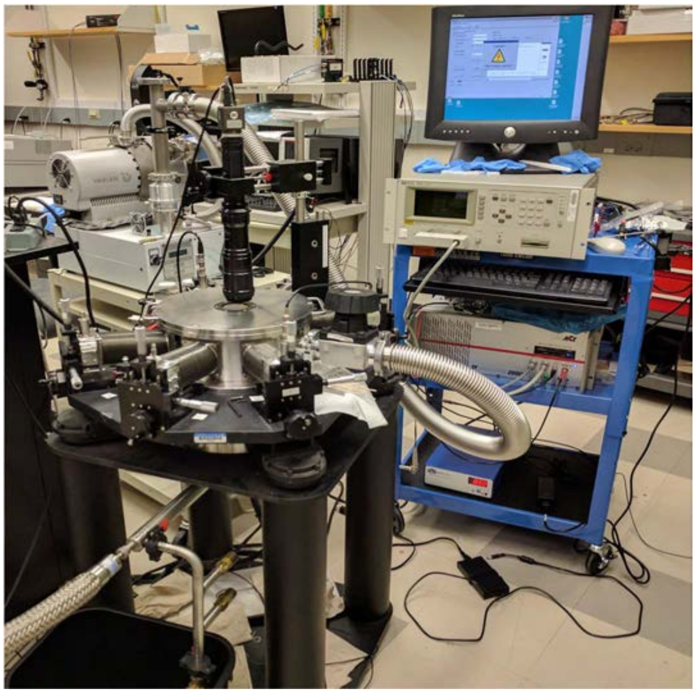

Figure 7: The cryoprobe, on the left, was used to contact the electrodes and allowed for measuring at low temperatures. The cart, to the right, has the dielectric tester at the top, and the ferroelectric tester on the shelf below.

I combined a ferroelectric tester device (TF-Analyzer 2000) and a dielectric tester (4284A Hewlett Packard LCR meter) with a cryoprobe. For the in-plane measurements I used to probe tips to contact Cr/Au (chromium/gold) interdigitated electrodes for each temperature (see Figure 8). For out-of-plane measurements I used probe tips to contact a Cr/Au circular pad and a corner pad used to contact the bottom $SrRuO_3$ (strontium ruthenate) layer (see Figure 9).

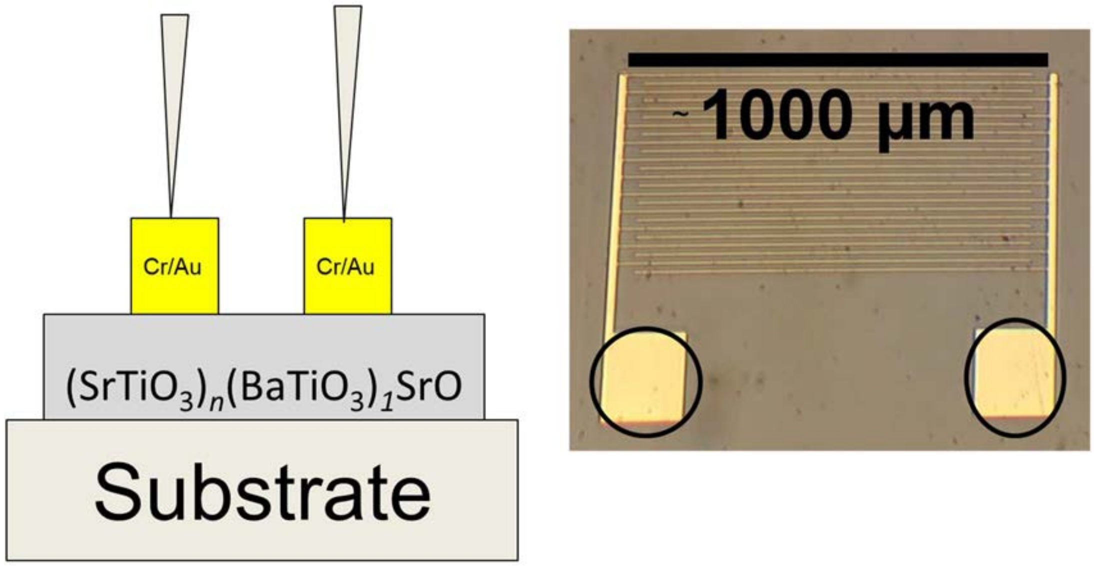

Figure 8: On the left, the structure of the in-plane sample can be seen. The $(SrTiO_3)_n(BaTiO_3)_1SrO$ thin film (with thickness of 100nm) was deposited on the $DyScO_3$ substrate. The Cr/Au electrodes were pattened as interdigitated electrodes, and the probe tips contacted these electrodes. On the right, the interdigitated electrodes used is shown, with contact points circled.

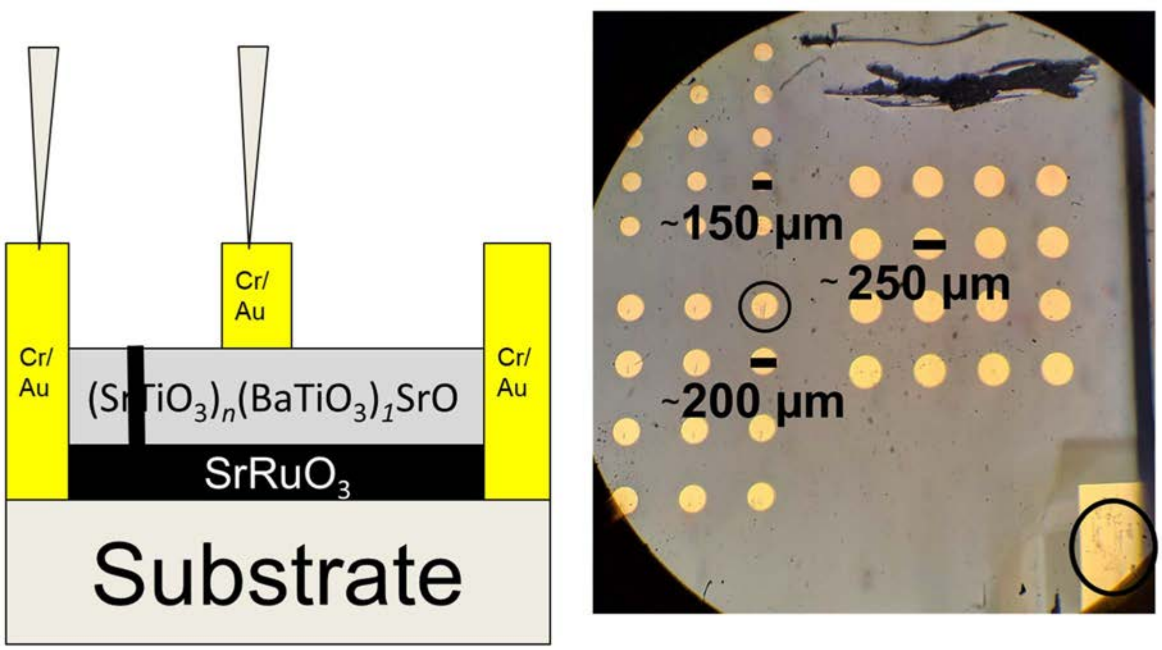

Figure 9: On the left, the structure of the out-of-plane is shown, which is similar to in-plane except for the deposited 15 nm layer of $SrRuO_3$. To contact the $SrRuO_3$ layer, the bottom electrode, a probe tip contacted the corner Cr/Au. The other tip was placed on a circular pad as the top electrode. The right shows the circular pads, their sizes, and the corner pad. Contact points are circled.

Measurements were all collected, graphed, and analyzed in Excel files. For the in-plane dielectric measurements, the analysis methods I used for these measurements came from a paper by Kidner et al.[2], where equations 10, 11, and 12 were used to determine the dielectric constant for each of the interdigitated electrodes:

For the out-of-plane dielectric constants, since the pads we used were similar to a parallel plate capacitor, we used the equation $C = \kappa \varepsilon_o A / d$.

References

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2. Kidner, N. J., Homrighaus, Z. J., Mason, T. O. & Garboczi, E. J. Modeling interdigital electrode structures for the dielectric characterization of electroceramic thin films. *Thin Solid Films* **496,** 539–545 (2006).

Acknowledgements

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Appendix

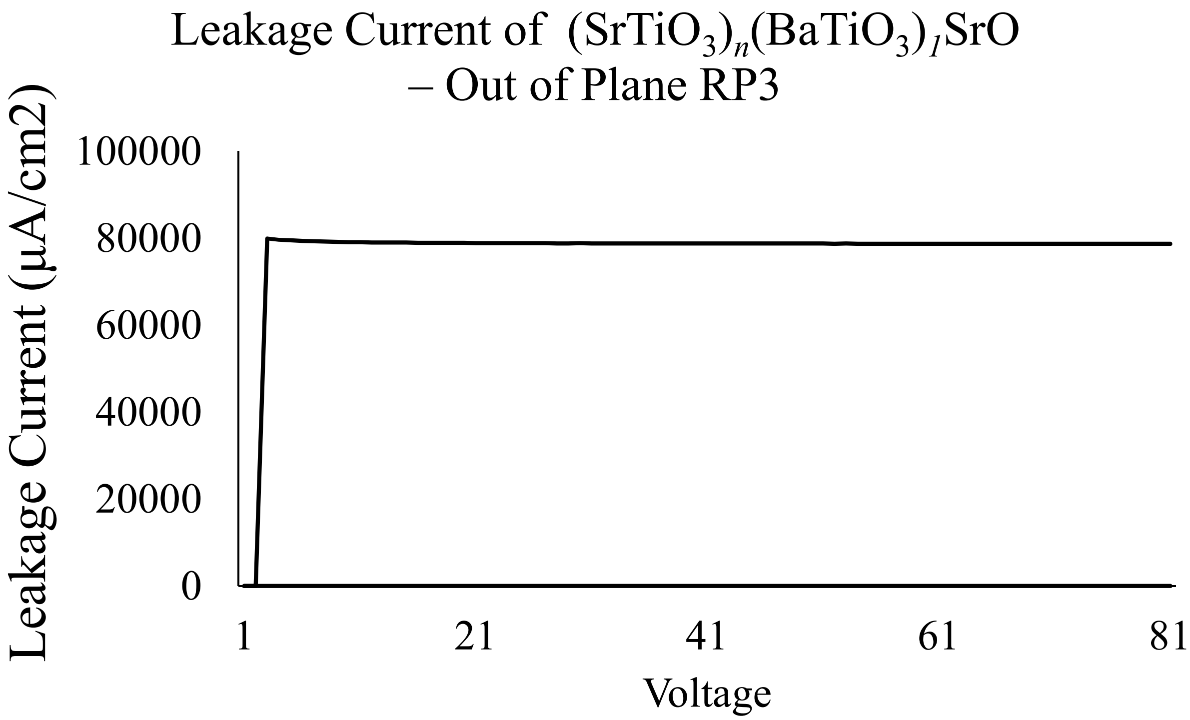
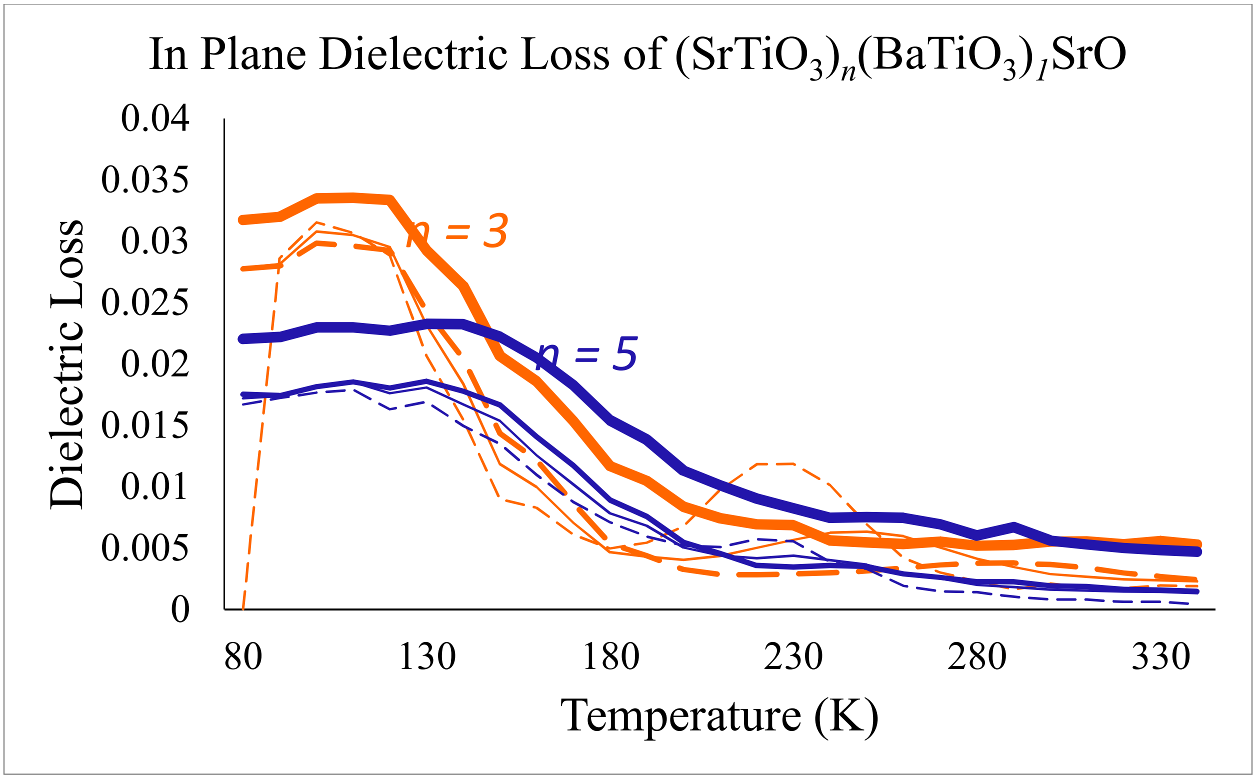


Figure 10: This measurement shows that for even voltages as low as perhaps 5V, the leakage current measured is extremely high.



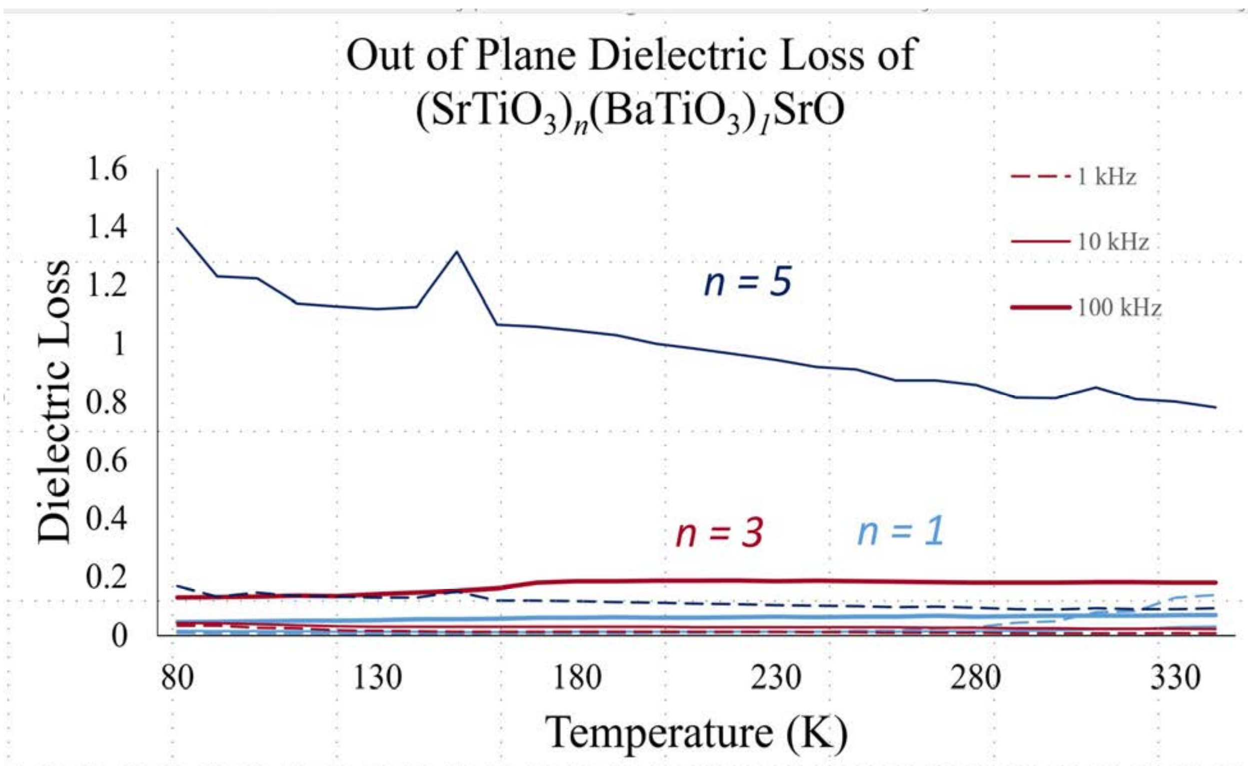


Figure 11: Comparing the top graph and the bottom graph, it is very clear that out-of-plane overall has greater dielectric loss, especially for n=5.