

Controlling the Anti-Phase Boundary Concentration in $\text{Sr}_2\text{AlTaO}_6$

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Beamline(s): X7A

Introduction: The properties of perovskite dielectric materials show sensitivity to the presence and concentration of anti-phase boundaries (APBs). The ideal double perovskite structure ($\text{A}_2\text{BB}'\text{X}_6$; space group $Fm3m$), has regular alternating BX_6 and $\text{B}'\text{X}_6$ corner-sharing octahedra (i.e., an ordered domain). Ordered domains in strontium aluminum tantalate ($\text{Sr}_2\text{AlTaO}_6$; known as SAT) are known to be separated by APBs where the alternating pattern of corner-sharing AlO_6 and TaO_6 is inverted. The APB concentration in SAT, which can be quantitatively evaluated through analysis of the peak broadening in the X-ray powder diffraction patterns, is known to be sensitive to changes in the synthesis conditions and composition. In this study, daughter compounds of SAT were synthesized containing excess tantalum ($\text{Sr}_{2-x}\text{Al}_{1-x}\text{Ta}_{1+x}\text{O}_6$ where $0.01 \leq x \leq 0.04$) or aluminum ($\text{Sr}_2\text{Al}_{1+x}\text{Ta}_{1-x}\text{O}_{6-2x}$ where $0.01 \leq x \leq 0.07$) in a molten salt flux, and samples were subsequently annealed at 1300°C in an attempt to control the APB concentration.

Methods and Materials: Synchrotron x-ray powder diffraction (XRPD) data was collected at the X7A beamline on all compounds synthesized during this study and XRD grade CeO_2 standard for instrumental line broadening corrections. The samples were sealed into 0.3-mm diameter glass capillaries that were rotated at a rate of 1-2 Hz to reduce any possible preferred orientation of the crystallites. A single crystal Ge (111) monochromator was used to obtain 0.79911-Å radiation. Data was collected using a linear position-sensitive detector (PSD). The PSD was stepped in 0.50° intervals between 8° and 58° two-theta. Counting times for the first 25° was done at 30 s step^{-1} ; for the second 25° , collection was done at 60 s step^{-1} . Initial data refinements were done using the KOALARIET software package that included the refinement program "X-fit". The program "Breadth" was used for diffraction line broadening analysis, as it calculates both volume-weighted and surface-weighted crystallite sizes, ordered domain sizes, and strains using the alternative integral-breadth method.

Results: Initial results indicate samples synthesized using the straight flux and flux/anneal methods by altering the $\text{Al}^{3+}:\text{Ta}^{5+}$ ratio to be aluminum-rich generally show no change in ordered domain size, even though the ordered domain size is greater for the flux/anneal compounds than the straight flux compounds. Aluminum-rich samples synthesized by using the flux method and with subsequent annealing to 1300°C by deviating the $\text{Al}^{3+}:\text{Ta}^{5+}$ ratio show a decrease in ordered domain size as the mole % of aluminum increases. SAT with a 50:50 Al^{3+} to Ta^{5+} ratio made via the flux only appears to be anomalous. Recent additional analyses have shown that the effect seen in figure two has proven to be difficult to reproduce.

Conclusions: It is possible that deviating the ratio of Al^{3+} to Ta^{5+} so that the compounds are aluminum-rich allows the control the concentration of APBs in the double perovskite $\text{Sr}_2\text{AlTaO}_6$. Additionally, allowing the Al^{3+} to Ta^{5+} ratio to deviate so the compounds are tantalum-rich shows no change in the ordered domain size, thus no change in the concentration of APBs. Subsequent experiments show difficulty in preparing the tantalum-rich compounds without tantalum oxide containing a minor amount of a tantalum oxyfluoride impurity. Additional heating (flux/anneal synthetic method) of these compounds shows similar trends, with larger ordered domain sizes and exsolved impurity phases. Further studies are needed to investigate the reproducibility of the results so the ideas developed in this study can be applied to materials with significant dielectric properties; if so, this pathway could be used to custom-make better dielectric materials.