SAND2011-8169C

Role of Interfaces and Defects on the Thermal Properties of Complex Oxide Thin Films

Jon F. Ihlefeld,[1] Patrick E. Hopkins,[1,2] Harlan Brown-Shaklee,[1] John Leonard,[3] Brian M. Foley,[2] James LeBeau,[3] Bryan Huey,[4] Carolina Adamo,[4] Darrell G. Schlom,[5] and Jon-Paul Maria[3]

[1] Sandia National Laboratories, [2] University of Virginia, [3] North Carolina State University, [4] University of Connecticut [5] Cornell University

Motivation

Thermal and thermoelectric properties of oxides have recently garnered increased interest as an environmentally friendly alternatives to conventional *p*-block-based thermoelectrics. Many oxides have competitive power factors, but high thermal conductivities limit *ZT* values and utility in energy harvesting applications. In this work we have studied the effect of interfaces and defects on the thermal conductivity of $SrTiO_3$, $BiFeO_3$, and $ZnGaMnO_4$ thin films.

Samples Studied

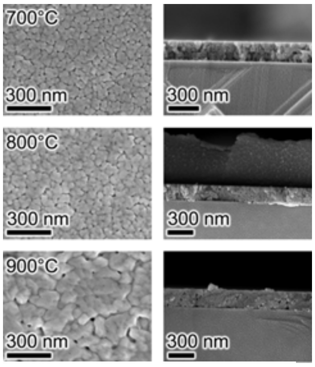
- Polycrystalline $SrTiO_3$ on sapphire (CSD)
  - Porous grain size series
  - Dense grain size series
- Epitaxial $BiFeO_3$ on $SrTiO_3$ (001) (MBE)
  - Engineered domain structures
  - Stoichiometry series
- $Zn(Ga,Mn)_2O_4$ (PLD)
  - Phase separating nanostructures

Sample Details/Measurement

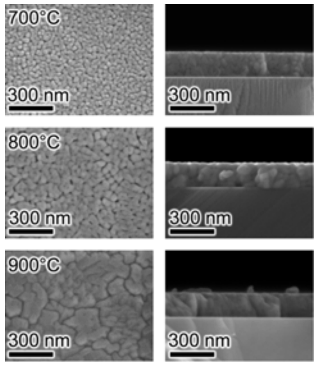
- $SrTiO_3$
  - 0.15M chelate chemistry with methanol solvent
  - Spin cast and fire every 3 layers (porous) fire every layer (dense) at 700°C on 0001- oriented sapphire substrates for 1 hour in air to coarsen grain size
- $BiFeO_3$
  - Reactive MBE with metallic Bi and Fe sources and O_2/O_3 reactive gas atmosphere at $1x10^{-6}$ Torr background pressure
  - Variable substrate temperature with fixed bismuth flux (modify Bi:Fe ratio) on $SrTiO_3$ (111) substrates. Samples are single domain (0001)-oriented.
  - Non-vicinal $SrTiO_3$ (001) and vicinal 4° miscut toward <011> for 2 domain variants and toward <110> for a single domain variant. Virtually all domain walls are 71°.
- $ZnGaMnO_4$
  - Pulsed laser ablation on MgO (001) substrates
  - End-member $ZnGa_2O_4$ and $ZnMn_2O_4$ films prepared in same manner
- Time domain thermoreflectance used to characterize thermal conductivities

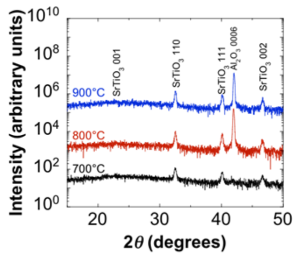
$SrTiO_3$ Thin Films

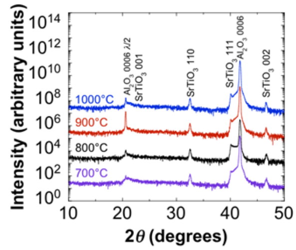
Porous $SrTiO_3$

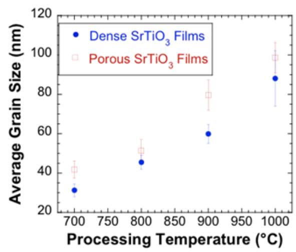


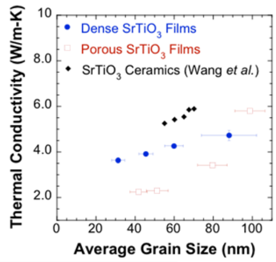
Dense $SrTiO_3$





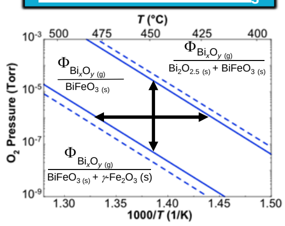


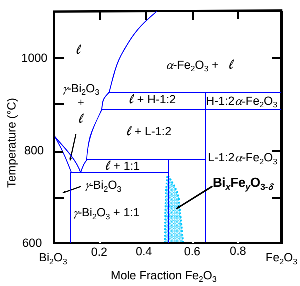


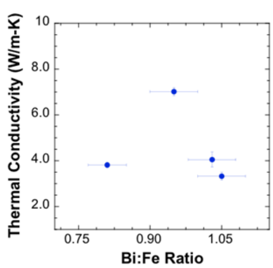


$BiFeO_3$ Thin Films

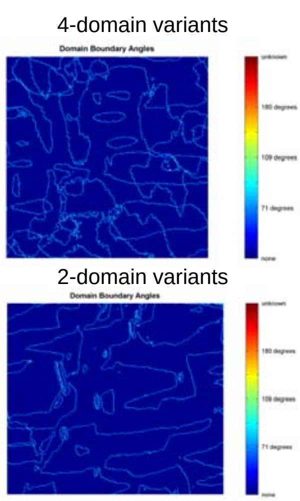
0001-Oriented $BiFeO_3$

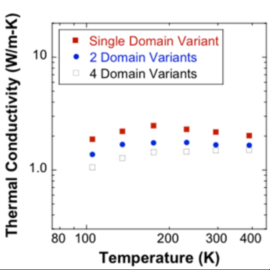




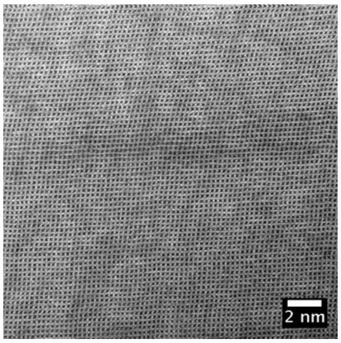


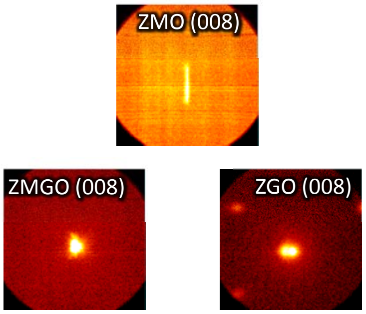
001_p-Oriented $BiFeO_3$



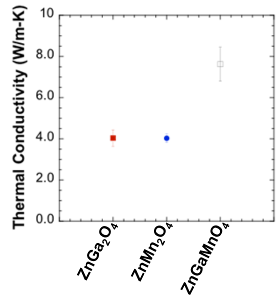


$ZnGaMnO_4$ Thin Films





Cross-Plane Thermal Conductivity



Conclusions

- Porosity and grain size greatly affect the thermal conductivity of $SrTiO_3$ thin films
- Tuning grain size may be an attractive approach for thermoelectric preparation
- Stoichiometry greatly affects thermal conductivity in $BiFeO_3$ single crystal films
- Ferroelastic domain boundaries act as phonon scattering sites in $BiFeO_3$ single crystal films
- $ZnGaMnO_4$ films display higher cross-plane thermal conductivities than the end member compositions. This is likely due to improved crystalline quality in the phase separating material.

This work was supported by the Laboratory Directed Research and Development program at Sandia National Laboratories. Sandia National Laboratories is a multi-program laboratory operated by Sandia Corporation, a wholly owned subsidiary of Lockheed Martin Corporation, for the United States Department of Energy's National Nuclear Security Administration under Contract DE-AC04-94AL85000