



Tailoring SrTiO₃ Thermoelectric Properties Through *n*-Type Doping and Processing Science

SAND2013-1701C

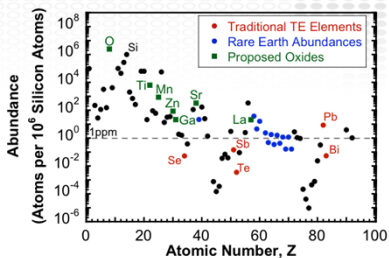
Jon Ihlefeld,¹ Harlan Brown-Shaklee,¹ Peter Sharma,¹ Brian Foley,² Patrick Hopkins,² Paul Clem,¹ and Douglas Medlin¹

¹Sandia National Laboratories

²University of Virginia

Exceptional
service
in the
national
interest

Motivation
Conventional thermoelectric components are comprised of relatively rare and strategically limited elements. This ultimately may limit their utility to non-industrial or niche applications



Oxide ceramics have recently garnered increased interest as thermoelectric materials for high temperature energy harvesting applications. Perovskites, including CaTiO₃, SrTiO₃, and BaTiO₃, are interesting candidates for thermoelectrics because their electrical and thermal behavior can be tailored with A-site and B-site dopants. Further, the perovskites can accommodate multiple dopant atoms that can reduce phonon heat transport while simultaneously generating free carriers. It is necessary to identify methodologies for reducing thermal conductivity without sacrificing thermopower and electrical conductivity of *bulk* oxides.

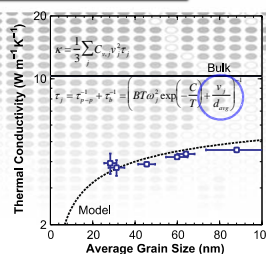
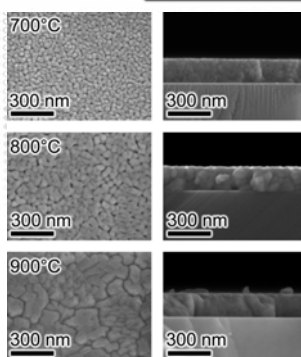
Goals

1. Determine limits of SrTiO₃ thermal conductivity scaling via grain size reduction – thin films
2. Determine whether SrTiO₃ thermal conductivity can be significantly reduced by introducing A-site disorder and
3. Evaluate the impact of Bi and La co-doping on electrical transport

Samples Studied

1. Thin film SrTiO₃ grain size series on sapphire substrates
2. Bismuth and lanthanum doped SrTiO₃ ceramics were prepared by traditional ceramic processing techniques and heat treated in reducing atmospheres to generate *n*-type thermoelectrics

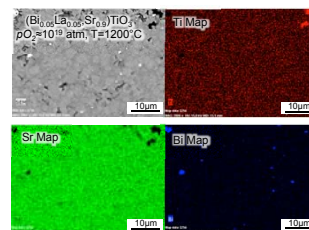
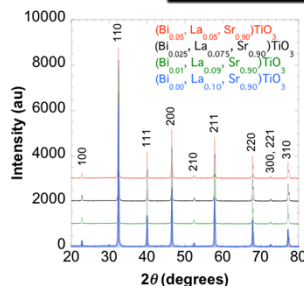
SrTiO₃ Scaling Effects – Thin Films



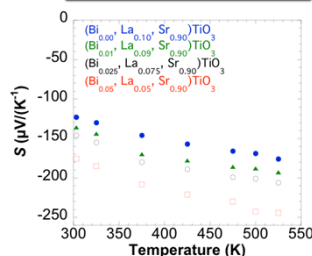
- Grain boundary scattering dominates
- 3X decrease in thermal conductivity by utilizing nano-scale grains

Foley et al. Appl. Phys. Lett. 101, 231908 (2012)

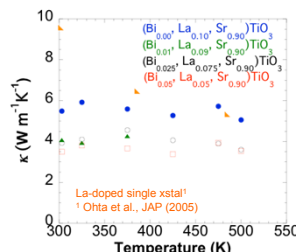
Bulk Ceramic (La,Bi)-SrTiO₃



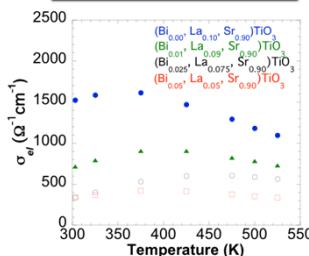
S, T>300K



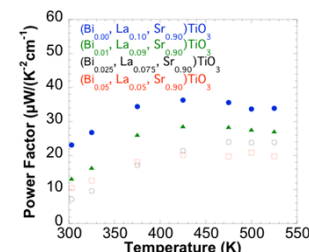
κ, T>300K



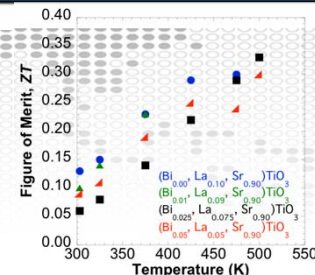
σ_{el}, T>300K



S²-σ_{el}, T>300K



Thermoelectric Performance



Summary of Performance at 500K

Bi (%)	La (%)	S (μV/K)	σ _{el} (Ω ⁻¹ cm ⁻¹)	k _{th} (W/m-K)	PF (μW/cm-K ²)	ZT
0	10	-169	1180	5.0	33.7	0.33
1	9	-188	780	780	27.6	-
2.5	7.5	-201	589	3.6	23.8	0.33
5	5	-243	354	3.5	20.9	0.30

High Temperature Measurement Capability



- All platinum wiring ensures operation across broad atmosphere conditions (pO₂ from 10⁻²¹ to 1 atm)
- Temperature capabilities to 1200°C
- Entirely automated control and measurement

Program Impact

- Developed model to determine scaling effects on thermal conductivity
- Bismuth effectively reduced thermal conductivity when it replaced lanthanum. Favorable power factor and figure of merit values were obtained for (Bi, La)-SrTiO₃ despite a drop in σ_{el} with respect to La-SrTiO₃
- Unique high temperature plus controlled atmosphere Seebeck and σ_{el} probe developed
- 2 papers published
 - Foley, et al., Appl. Phys. Lett. 2012
 - Campion, et al., J. Amer. Cer. Soc. 2013
 - 1 under review, 2 in preparation
- US Provisional Patent Application: 61/765,227 High ZT Bi-Doped Perovskite Thermoelectrics

Future Work

- Measure thermoelectric properties up to 1300K using newly developed high temperature/controlled atmosphere probe