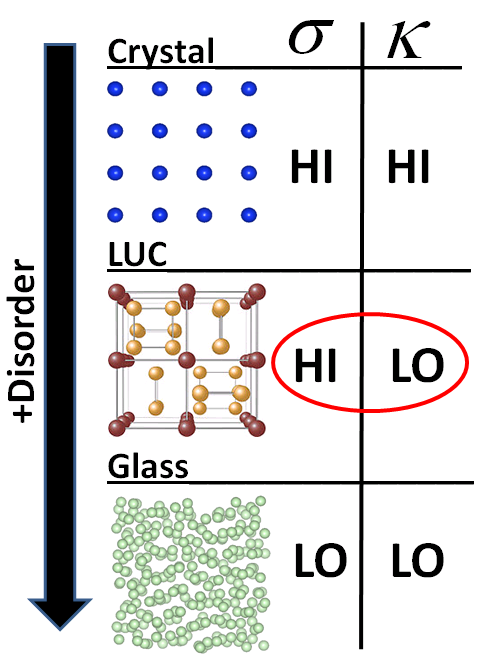
**Project Title:** Atomistic Modeling of Thermal Transport in Large Unit Cell Crystals for Thermoelectric Application

**Research Team:**

Graduate Student: Jason Larkin, Mechanical Engineering  
Faculty Advisor: Associate Professor Alan McGaughey, Mechanical Engineering  
Affiliated Faculty: Professor Kenneth D. Jordan, Department of Chemistry & Visiting Professor Wissam Al-Saidi, Department of Chemical and Petroleum Engineering (University of Pittsburgh)

**Project Description**

*large unit cellforing efficiency ofenergy generation*

Thermoelectric energy generation - the transformation of waste heat into useful electricity - is a promising source of sustainable energy [cite]. Thermoelectric materials directly convert a temperature difference into an electric voltage as a result of their intrinsic (atomic-level) electronic and thermal properties. The performance of a thermoelectric device can be quantified through the the *thermoelectric figure of merit*, ***ZT* =** , where *T* is the average device temperature, *S* is the Seebeck coefficient (a ratio of the induced thermoelectric voltage for an applied temperature difference), is the electrical conductivity, and is the thermal conductivity. For thermoelectric devices to be competitive with other power generation requires *ZT* > 3 [cite]. Achieving this performance is challenging because the electrical and thermal properties in *ZT* are coupled in the majority of materials. Using nanostructuring to reduce while maintaining good electrical properties has been identified as one possible strategy. Producing such nanostructured materials (e.g., superlattices), however, is costly. An emerging area of study is the use of large unit cell crystals.

Figure 1: LUC crystals can maximize ZT. ZT.

Large Unit Cell crystals have an ordered (crystalline) structure, but the basic building block (unit cell) of the crystal has a large number of distinct atoms (Figure 1) [cite]. They are thus effectively disordered over length scales relevant to thermal transport, and as such, their thermal conductivities can be as low as a glass [cite]. The unique feature of large unit cell materials is that they are still ordered for electron transport, which results in a large and *ZT*. Available large unit cell crystals have ZT<3 [cite] and more research is critically required to improve their thermoelectric efficiency.

Improving the ZT of large unit cell crystals through thermal conductivity reduction requires a detailed atomistic analysis. The computational tools and resources available in my research group, the Nanoscale Transport Phenomena Laboratory (NTPL) provide the required framework. First, classical simulations using Molecular and Lattice Dynamics (MD and LD) will be performed [cite]. Classical indicates that quantum effects are not included [cite]. The classical simulation results will then be used to design *ab initio* (from first principles) simulations. *Ab initio* simulations include quantum effects explicitly and have very high accuracy [cite]. The *ab initio* simulation results will be directly compared to experiment to more efficiently search the LUC crystal design space.

Naïve/important questions that should be answered on the first page: Why not just do quantum to start? Why are simulations necessary -> Why not just do experiments? What materials will you consider?

**Research Plan**

***Objective 1****: Use Molecular and Lattice Dynamics to explore LUC/disordered crystal design space.*

As the name suggests, large unit cell materials require a large number of atoms to model accurately. Size effects will be initially tested to ensure converged results, as in [Galli]. These test results will be valuable when designing time-efficient *ab initio* simulations.While these simulations can make experimentally accurate predictions, they are very computationally expensive [cite]. Simulations are limited by the computational density of the resources available, but typically simulations can allow for 100s-1000s of atoms [cite]. This makes it important to test size effects using computationally inexpensive methods developed by the NTPL group. This will allow us to design efficient ab-initio simulations to run on the ERDC supercomputing resources (keep?).

***Objective 2:*** *Design ab-initio simulations to predict the thermal properties of realistic LUC crystals.*

Ab-initio simulations will be run on the extensive resources NTPL has at the ERDC (keep?). In particular, Density Functional Theory (DFT) and Density Functional Perturbation Thoery (DFPT) will be used to analyze the vibrational properties of LUC crystals []. From these vibrational properties the thermal properties of LUC crystals can be predicted, including the thermal conductivity [cite]. Previous measurements suggest that system crystal sizes of roughly 1000 atoms can give accurate results [Galli]. This is well within the capabilities of the ERDC resources, whose number of processors is roughly 15,000 (eh, keep?).

***Objective 3:*** *Report ab-initio simulation results to publishing journal(s) and experiments to improve ZT over existing LUC/disordered materials.*

These above objectives can be used iteratively, with data from real experiments, to efficiently search the LUC design space. ***Ab-initio simulations can predict LUC materials with even lower thermal conductivities.*** For example, modifying LUC crystals with heavy atoms and/or bonding disorder has been shown to be promising mechanisms for reducing thermal conductivity [cite]. Colloboration with NTPL’s adjacent lab group Thermal Energy Engineering Laboratory led by Jon Malen will allow the comparison of our ab-initio simulation with real experimental results, each method assisting the other.

**Relationship to CIT/ICES Strategic Focus**

Atomistic simulation of large unit cell materials fits directly into CIT’s nanotechnology initiative. This project is aligned with ICES’s Center for Multiscale Modeling for Engineering Materials (CM2EM) and Center for Nano-enabled Device and Energy Technologies (CNXT), both of which McGaughey is affiliated with. The work provides a compelling example of multi-scale modeling by demonstrating how classical-level (Molecular and Lattice Dynamics) simulations can inform quantum-level (ab-initio) simulations, maximizing the impact of both.

**Present and Future Funding**

Preliminary funding for this project was through a CIT Dean’s fellowship and the PITA grant “Multi-scale modeling of phase change materials,” with Jason Larkin as the supported graduate student. He does not have support in place for the 2010-2011 academic year.

The Dowd fellowship will enable the team to complete the development of interatomic potentials for a wide range of Ge-Sb-Te materials. With these interatomic potentials in hand, the team will be competitive for funding from NSF’s Thermal Transport Processes (ENG-CBET) and Electronics, Photonics & Device Technologies (ENG-ECCS) programs, SRC’s Device Sciences – Modeling and Simulation program (a call will open in Spring 2011), and CMU’s DSSC. The team will collaborate with CMU’s Jim Bain (ECE, DSSC), who does experimental work on PCM materials and devices and Jon Malen (ME), who thermally characterizes micro- and nanoscale materials and devices. Together, they will apply for funding from NSF’s Interdisciplinary Research (IDR) program.

**References**

Just listing out the references I have so far that are applicable.

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