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Research Statement

The increasing energy demand and the negative environmental impact of fossil fuels present a formidable challenge to our global society. Such challenge must be addressed by developing sustainable energy technologies based on advanced functional materials, including solar cell, batteries, thermoelectrics, thermal energy storage/management and so on. My research contributes to this endeavor by establishing a better understanding of the structure-properties relationship in several material systems and investigating novel materials for energy and electronic applications.

My Ph.D. research has focused on eco-friendly silicides for thermoelectric (TE) power generation and cooling. Higher manganese silicides (HMS) with complex Chimney Ladder crystal structure are promising TE materials due to their unusually low thermal conductivity (κ). By inelastic neutron scattering measurements and density functional theory calculations, I have first discovered the low-energy optical phonon modes in HMS, which can enhance phonon scattering and reduce κ. In addition, I have improved the TE performance of HMS by ~60% through combining heavy element alloying and nanostructuring. These high-performance silicides were further used to fabricate the first silicide TE devices in U.S. These studies offer new insights into developing novel materials/devices for TE waste heat recovery.

After receiving my Ph.D. in December 2014, I am currently working as a postdoctoral fellow at UT Austin. I have worked on three independent research projects since then. The first one is focused on low-dimensional magnetic materials for spin caloritronics. I have grown several high-quality antiferromagnetic oxide crystals, and quantified the large electron spin mediated κ. Moreover, a better understanding of the interaction of spin excitations with phonons, grain boundaries and impurities has been established. The second project is to investigate novel materials for thermal management. Using the steady state and Raman lock-in methods, I have discovered an ultra-high bulk κ approaching 1000 W/mK in cubic boron arsenide, which is found to be the first known semiconductor with such high κ. In addition, I have developed several new ceramic oxide electrolytes with high lithium ion conductivity and ultra-small interfacial resistance for all-solid-state lithium ion batteries. These research experiences have expanded my knowledge and prepared me to be an effective researcher in interdisciplinary sciences.

My research has resulted in eleven first-authored (including first-coauthored) papers published in *Science*, *Nature Communications*, *PNAS*, *Advanced Energy Materials* and so on. During my eleven years of academic training, I have mastered skills of bulk and nanoscale physical property measurements, single-crystal growth, spark plasma sintering, and neutron scattering.

My future work will primarily focus on developing materials for energy and spintronic applications. To achieve this, I will synthesize novel functional materials by both crystal structure and nanostructure engineering. I will also probe quasi-particles transport mechanisms in these materials by combining experiment and theory. These efforts, involving materials science, chemistry, mechanical engineering and solid-state physics, will be achieved in the following proposed projects.

1. **Spin and phonon transport in 2D heterostructures derived from misfit layer compounds**

Motivation: Heterostructures play a crucial role in high-speed nano-electronic and spintronic devices and are essential elements in semiconductor industries. However, only limited combinations of materials, including graphite, h-BN, and transition-metal dichalcogenides, have been investigated so far. Moreover, spin and phonon transport in heterostructures remains to be better understood.

The proposed research**:** The purpose of this work is to synthesize novel 2D heterostructures derived from misfit layer compounds and investigate their spin and phonon transport at nanoscale. Misfit layer compounds are naturally comprised of a 2D arrangement of atoms that consist of several layers of atoms, leading to an incommensurate crystal structure bound by weak van der Waal interactions between interleaved layers. One misfit compound I have grown previously is the spin ladder compound $Sr_{14}Cu_{24}O_{41}$ (***PRB*** **2016,** ***PRB*** **2017**). This proposal is focused on several complex systems, including antiferromagnetic $Sr_{14}Cu_{24}O_{41}$, thermoelectric $(LaS)_{1.2}CrS_2$, and metastable compound $(BiSe)_m(TiSe_2)_m$, which will be grown by chemical vapor transport method or mechanical exfoliation. I will investigate the layer thickness and layer configuration-dependence of spin and phonon transport in these heterostructures by suspended micro-bridge method and Raman scattering. According to my previous neutron scattering measurements, the phonons in two sublattices are weakly coupled to each other in $Sr_{14}Cu_{24}O_{41}$ (***PRB*** **2016**). I will verify whether the in-plane quasiparticle transport is relatively independent in each sublattice. In addition, I will study the effect of interaction between sublattices on spin and phonon transport by applying external pressure or chemical doping.

The broader impacts: The successful design of 2D heterostructures derived from misfit materials could be a harbinger for the discovery of a new class of 2D materials with exotic physical properties. The work can create opportunities for studying spin and phonon transport by harnessing the unusual features of atomically thin materials, which show potential applications in new-generation thermoelectircs, sensors and spintronics.

***Possible sponsors*:** NSF (Division of Materials Research; Emerging Frontiers and Multidisciplinary Activities); DOE (Office of Science-Basic Energy Sciences); Army Research Office (Multidisciplinary University Research Initiative)

2. Investigating Majorana fermions in quantum spin liquids for quantum computing

***Motivation*:** Exotic particles called Majorana fermions, which are their own antiparticles, have potential applications in quantum computing. However, their existence has yet to be definitively confirmed because of obstacles in experiments and the lack of detailed theoretical information.

The proposed research*:** The overall research objective is to investigate the nature of Majorana particles in quantum spin liquids, which show exotic excitations associated with quasiparticle fractionalization and topological order. To detect the charge-neutral Majorana modes, low-temperature thermal transport measurements is a powerful tool. Possible existence of Majorana modes has been recently reported in $\alpha-RuCl_3$ via thermal Hall measurement. I will hunt Majorana fermions in other quantum spin liquids, such as 3D hyper-honeycomb β-Li_2IrO_3 and post-perovskite $CaIrO_3$, which can be grown by chemical vapor transport method and flux method, respectively. According to a recent theoretical calculation, additional peak in κ due to itinerant Majoranas can be observed. I will search for Majorana particles in these spin liquids by longitudinal thermal conductivity measurements. I have performed such measurements on several 1D spin liquid compounds $Sr_{14}Cu_{24}O_{41}$, $SrCuO_2$, and Ca_2CuO_3 (PRB* 2017, *PRB Rapid* 2017**) as well as cubic boron arsenide (***Science* 2018**). Inelastic neutron scattering will further be used to investigate the magnetic excitations in single crystals (***PRB* 2016**), revealing the nature of Majorana modes. Moreover, the interaction of Majorana fermions with other quasipartilces remains elusive, which can be revealed by detailed temperature, magnetic field and pressure dependent thermal transport measurements.

The broader impacts: Understanding the nature of Majorana fermions is important for revealing novel aspect of strongly correlated topological quantum matters. The study can open up new applications, such as the emerging field of quantum computing.

***Possible* sponsors:** NSF (Division of Materials Research); DOE (Office of Science-Basic Energy Sciences); Office of Naval Research (Multidisciplinary University Research Initiative).

3. Enhancing thermoelectric performance by embedding magnetic nanostructures

***Motivation*:** Thermoelectric materials, which can convert temperature gradients directly into electricity and vice versa, have received renewed interest for waste heat recovery and refrigeration applications. Current studies on improving TE performance have limited on manipulating transport of charge carriers and phonons by chemical doping and nanostructuring. The figure-of-merit *ZT* values for most materials are less than 2, generally around 1 due to inter-related TE properties.

The proposed research: This research aims to achieve high *ZT* over 2 by introducing secondary magnetic nanostructures, which can alleviate the inter-dependence between electron and phonon properties in classical thermoelectric materials. The phonon scattering can be enhanced as a result of magnetic fluctuations and increased grain boundaries. In the meantime, the charge transfer between the nanostructure and matrix can be used to modify the electronic properties of nanocomposites. The multiple scattering of electron by magnetic fluctuations can also lead to the enhancement of Seebeck coefficient. In this work, I will demonstrate this by synthesizing silicides nanocomposites with embedded superparamagnetic Co or Fe_3O_4 nanoparticles. In my previous study, silicides have been investigated for high-temperature TE applications (**Adv. Energy Mater. 2014, Nat. Commun. 2015, *J Alloy Compd.* 2015**). In order to achieve a coherent or semi coherent interface between the nanopartilces and matrix, a melt-quenching-annealing method will be employed, as has been used for fabricating MnSi/HMS nanocomposites (***J. Mater. Chem. C* 2015**). The effect of the size and amount of magnetic nanoparticles on TE properties will be investigated to optimize TE performance.

The broader impacts: Introducing the spin degree of freedom can effectively manipulate charge carrier and phonon transport in TE materials, which can realize *ZT* over 2 in some magnetic nanocomposites. The established novel mechanisms for *ZT* enhancement will also facilitate the development of next-generation of TE materials/devices.

***Possible sponsors*:** DOE (Office of Science); NSF (Chemical, Bioengineering, Environmental and Transport Systems); NASA Science Mission Directorate

4. Quantifying effect of grain boundaries on Li-ion conduction in solid electrolytes for all-solid-state batteries

***Motivation*:** All-solid-state lithium ion batteries using nonflammable solid electrolytes are considered to be the next-generation device for electrochemical storage. The grain boundaries have a significant impact on solid electrolytes and are key hurdles that must be overcome for their commercial applications. However, such grain boundary effect on ionic transport is not fully understood.

The proposed research*:** This study focuses on quantifying the grain boundary effect on ionic transport in solid electrolytes. The good candidate materials may have anisotropic lithium diffusion channels along different crystallographic directions, such NASICON-type material $LiZr_2(PO_4)_3$. I have previously prepared this compound and garnet solid electrolytes by solid-state reaction followed by spark plasma sintering and achieved a high ionic conductivity and a low interfacial resistance (PNAS* 2016, *JACS* 2018**). I will further investigate the effect of grain size and grain boundary chemistry on ionic transport. The effect of grain orientation on ionic transport will also be investigated. The textured samples can be fabricated by multiple hot deformation (***PRB* 2017**). Moreover, I will grow large-size crystals of solid electrolytes using the traveling-solvent floating zone method. The grain boundary resistance will be quantified by comparing polycrystals directly with single crystals. In addition, all-solid-state lithium ion batteries will be assembled using the obtained single crystals or grain-boundary-engineered polycrystals with high ionic conductivity.

The broader impacts: The obtained results can provide valuable fundamental understanding of the role of grain boundaries on ionic thermal transport and how tailoring the microstructure can lead to the high ionic conductivity in solid electrolytes.

***Possible sponsors*:** NSF (Chemical, Bioengineering Environmental and Transport Systems); DOE (Materials Science and Engineering Division)

In summary, by studying the transport of quasiparticles, including electrons, phonons, spins, and ions in materials at the macro-, micro- and nanoscale, a better understanding of the structure-properties relationship will be established in bulk and nanostructured materials. The obtained knowledge will offer new insight into the development of high-efficiency devices for energy and spintronic applications.