Statement of Research

K.M. Taddei

Abstract: The mastery of existing and the discovery of new quantum materials (materials which exhibit macroscopic manifestations of quantum phenomena) are vital to the development of new and improved technologies. This research proposal lays out a progressive tiered approach to uncover guiding principles behind the discovery and optimization of quantum materials. Starting with a newly discovered family of quasi-one-dimensional spin-triplet superconductors, the various compositional forms will be used to study superconductivity against the nuclear and electronic structure. The recently identified structural and magnetic instabilities will be tuned allowing for a new test of candidate microscopic theories (density-wave, pair-density wave, quantum critical point) in a material with radically different structure than the often-studied compounds. This work will be greatly aided by the material's quasi-one-dimensionality which allows for easier more complete theoretical treatment and computational modeling. As the next step, the results from these studies will be used with recent theories to predict candidates for spin-triplet superconductivity – a form of superconductivity with substantial potential in spintronics and qubit design. Finally, the vast resources compiled in computational materials repositories will be used to identify new quantum materials using a parallel approach – looking for materials with band structures predicted to engender quantum phenomena.

Science Case

Quantum materials have been identified by the US DOE in numerous round tables and basic needs workshops as key to future technologies (1; 2; 3). This is evident as materials exhibiting topological states, SC and quantum spin liquid entanglement offer not incremental improvements in existing capabilities but entirely new possibilities – such as spin-transport electronics, new scalable qubits and quantum information processing. However, of significant impediment to the advance of such technologies is a lack of direction in their discovery. Current research is driven by flurries of activity around a few compounds which show promising properties. While this leads to a deep literature for a given system it does not provide important checks of proposed mechanisms nor ways forward to optimize the desired properties in a generalized material (4).

This proposal addresses this lack of direction by leveraging opportunities where theory and experiment have potential for significant overlap. To start, work will focus on a new family of Q1D TSC ($A_xTM_3As_3$ with A = alkali metal, x = 1, 2 and TM = transition metal) whose Q1D structure (shown in Fig. 1(a)) contrasts with the often studied Q2D cuprates and iron-based SC (Fig. 1(b)) (5; 6). The reduced dimensionality provides advantages by enhancing quantum fluctuations through confinement and allowing more direct theory treatments. My recent work has shown these Q1D systems exhibit physics thought relevant to SC (such as magnetic and structural instabilities) yet it remains ill-understood how or if these instabilities correlate with the SC properties (e.g. transition temperature, critical fields etc.) in the Q1D case as they do in the Q2D systems (5; 7). Since the choice of A and TM site ions can tune the SC properties, this question can be addressed through suppressing SC via stoichiometry and studying the corresponding changes in the instabilities (8). $A_xTM_3As_3$ therefore provides a unique system to test proposed microscopic theories of SC which will allow for wider generalizations to help in the discovery of new materials.

Another opportunity in these materials is their realization of TSC (9; 10). In TSC electrons pair into spin-1 states leading to a bevy of possible applications, such as spin-polarizability of the SC current and new qubit design and quantum computing architectures (11; 12). However, TSC is rare in practice and it is

therefore, of great interest to better understand both it and the properties which give rise to it. $A_xTM_3As_3$ has already proven useful in this regard – with recent theory work suggesting its structural elements as inherently giving rise to TSC, namely so-called 'odd-gon' sublattices (e.g. the Cr triangles in Fig. 1(a)) (13). Using these predictions and the results of the previously described work, it is possible to identify new materials which may exhibit TSC and test their viability.

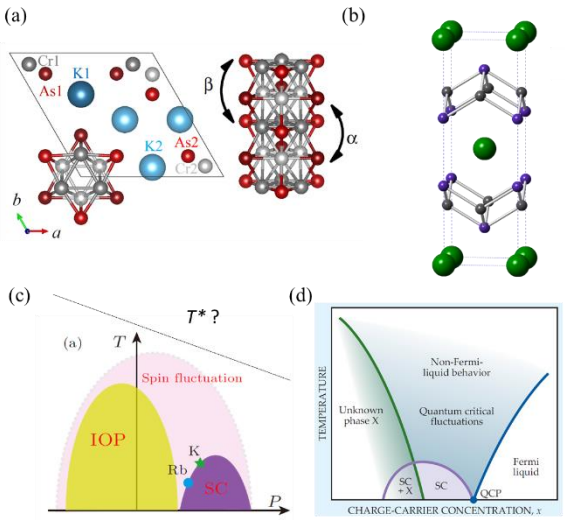

Figure 1: Crystal structure of (a) quasi-one-dimensional $K_2Cr_3As_3$ and (b) quasi-two-dimensional $BaFe_2As_3$. Predicted phase diagram for $A_2Cr_3As_3$ superconductors showing nearby magnetism. (d) Generic phase diagram for unconventional superconductors based of off numerous quasi-two-dimensional systems. See Thrust 1. Panels (c) and (d) are reproduced from references (32) *and* (33)

Looking forward, methodologies to discover quantum materials with possible new phenomena are desirable to advance beyond current device design. Recently, the growth of computing power and the efforts of funding agencies have given rise to a search philosophy which could usher in a new era of strongly-correlated materials (14). Programs such as the Computational Materials Repository, Open Quantum Materials Database and Materials Genome Initiative have begun compiling the results of Density Functional Theory calculations performed on thousands of possible compounds. With such comprehensive and openly accessible collections of predicted electronic structures, it is possible to identify interesting electronic configurations and to search for suitable materials within these databases. One such possibility is the Lifshitz transition (LST) in which changes to the energies of conducting bands can lead to discrete changes in the topology of the phase space available to electrons (i.e. Fermi surface, Fig. 3) (15). At the point of the LST, divergences can arise in derivative properties leading to exotic behaviors. Recently, the LST has been identified as a candidate for unconventional SC, heavy-fermion liquid physics in Kondo-Heisenberg systems and for many novel topological behaviors such as topological insulators, SC and the associated Weyl, Dirac and Majorana fermions (16; 17; 18; 19). Therefore, the discovery of new systems which exhibit this transition is of great interest to the unearthing of new exotic states.

Research Plan

Thrust 1: First, the known compositions of $A_2Cr_3As_3$ (with A = Na, K, Rb, Cs) will be synthesized. In this series of compositions, the SC transition T_c decreases with increasing alkali size and it is ill-understood what leads to this effect. This author's recent work has identified both short-range structural and dynamic magnetic instabilities in the $K_2Cr_3As_3$ compound, the latter of which couples to both SC and the structure. This gives a clear path for studying how these two instabilities interact with SC by determining their relative strengths as functions of A – and therefore T_c – using neutron and x-ray scattering techniques. As of yet, no experimentally determined phase diagram for these '233' materials exist: this first work will provide this much needed map of the relevant phase space. As a next step the newly discovered Mo analogues will be synthesized and studied. These materials have higher T_c but display an opposite trend as a function of T_c with A. By studying both materials and the relative strength of the short-range correlations it will be possible to determine if there is a correlation to T_c and whether it is possible to form a fully ordered phase in any of these materials. This will provide vital insight on how the instabilities interact with SC relative to the iron-based and cuprate SC. Furthermore, these two

instabilities are not strongly coupled to each other – an important advantage over the two-dimensional systems where their coupling is a confounding effect for understanding SC. Additionally, current work in collaboration with theory groups is identifying expected signatures of the TSC state in the dynamic spin-susceptibility (as probed by neutron scattering). When concluded this will afford a straight forward path to confirm the nature of the SC state in these materials and set the stage for the second part of this plan.

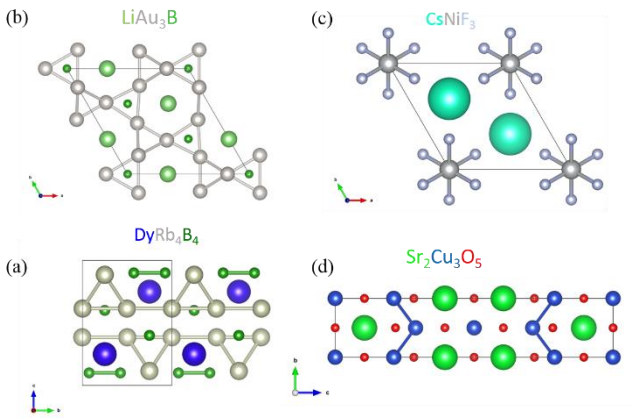

Figure 2: Crystal structures of identified quasi-one-dimensional odd-gon materials (a) $DyRh_4B_4$, (b) $LiAu_3B$, (c) $CsNiF_3$ and (d) $Sr_2Cu_3O_5$. Each material has a one-dimensional transition metal sublattice and exhibits either ferromagnetism ($CsNiF_3$) or predicted, observed or incipient yet ill-understood superconductivity ($DyRh_4B_4$, $LiAu_3B$ and $Sr_2Cu_3O_5$). See Thrust 2.

Thrust 2: Recent theory has suggested the 'odd-gon' sublattice of the Cr atoms (sublattice units with an odd number of sites) naturally gives rise to a spin-polarized state. Using this as a starting point, I have identified other materials with 'odd-gon' and Q1D *TM* sublattices to start the search for new TSC, namely $DyRb_4B_4$, $LiAu_3B$, $Sr_2Cu_3O_5$ and $CsNiF_3$ (Fig. 2) (20; 21; 22; 23). These materials show promising properties with the former three being, predicted or proximate to SC respectively and the latter exhibiting ferromagnetic order. Using the first three, the SC state will be characterized, and it will be determined if it is spin-triplet using transport and susceptibility measurements looking for signatures of line nodes in the SC state and critical fields in excess of the Pauli pair breaking limit. For the latter, doping and pressure will be used to attempt destabilization of the ferromagnetic order with the intention of tracing out a phase diagram like those shown in Fig. 1. The information gained from the first thrust will be employed here to better direct possible routes to establish TSC as well as a feedback to test the conclusions thus drawn.

Thrust 3: In preliminary work, I have identified several candidate transition-metal dichalcogenide materials (TMD) (formula MX_2 where M and X represent a transition metal and a chalcogenide respectively) from the CMR which show promise as a starting point for my program (14). To filter through the hundreds of examined compounds three criteria were used to identify interesting compounds: exhibition of the appropriate band structure, analogous 'sister' compounds which have reduced electronic topology (i.e. Fermi surface) and established synthesis procedures with relative accessibility. $CrTe_2$, $NiSe_2$, $TiTe_2$, and $HfTe_2$ all meet these criteria – providing a starting point with some extant veracity (24) (25) (26) (27). Of the families considered in the CMRs, the TMDs are particularly suited for studying LST as they straddle the line between metals and semi-conductors or semi-metals (28). This provides an obvious path to tune electronic behavior via shallow valence/conduction bands. Specifically, many compounds in this family are present in 'sister' pairs where formulas of both MSe/Te_2 and MS_2 exist with the former being metallic and the latter a semi-metal (fig. 3). Using these sister materials as endmembers one can easily tune the electronic band structure ostensibly crossing through LST as well as a metal to semiconductor transition and possible magnetic transitions. Indeed, many compounds in this family have led to exciting discoveries recently – including superconductivity, quantum confinement and topological states – indicating them as good starting points (27) (29) (30).

Each of these compounds and its sister material will be synthesized and characterized with transport measurements and laboratory-based diffraction sources. Once the endmembers' stability, synthesis procedure and probability for a LST are determined (the latter of which is addressable through comparison of the endmembers transport properties), series will be synthesized doping between the endmembers (e.g. $M\mathrm{Te}_{2-x}\mathrm{S}_2$) (31). These series will then be studied using the powerful and somewhat comprehensive techniques of neutron and x-ray scattering to fully elucidate structural/magnetic behavior and crucially to understand the underlying dynamics and possible critical phenomena expected around the LST.

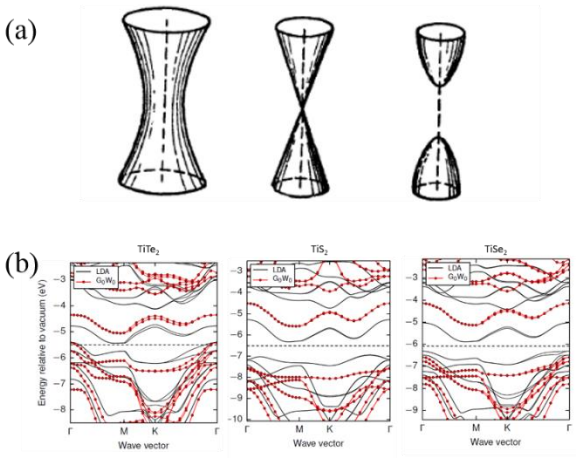

Figure 3: (a) Diagram of a Lifshitz transition showing a pinching off of a cylindrical Fermi surface (from (15)). (b) Calculated band structures for three Ti containing transition metal dichalcogenides showing multiple pathways to dope through a Lifshitz transition (from (14)). See Thrust 3.

Background: As a researcher, I have developed the crucial suite of experience necessary to implement this program. As a graduate student with a joint position at Northern Illinois University and Argonne National Laboratory (ANL), I developed extensive experience with difficult synthesis procedures and transport characterization working with the Emergent Materials group to synthesize myriad doping series of the iron-based superconductors. Furthermore, with my appointment in ANL's Neutron and X-Ray Scattering group, I became proficient in scattering techniques and their implementation in the study of quantum phenomena in correlated materials.

As a Post-doctoral associate at Oak Ridge National Laboratory, I have continued my work on unconventional SC as well as built my expertise as a neutron scatterer. Here I have performed much of the original work identifying structural and magnetic instabilities in the $K_2Cr_3As_3$ material described above. This work has involved learning the varied implementations of neutron scattering including spectroscopy and total scattering to uncover both temporal and spatial short-range correlations. Unlike my previous position, I have not had access to synthesis laboratories which has caused me to develop my abilities to find and nurture collaborations. This included work with synthesis groups within Oak Ridge as well as around the world. More recently I have started collaborations with two theory groups to understand SC in these materials working closely to use my experimental results to develop the theoretical descriptions and then using their predictions to direct my research.

For this later position rather than joining an existing project, I conceived, designed and still currently lead my own research program. This has developed my independence and ability to lead multi-group collaborations. Furthermore, as a post-doc I took a leadership position within the ORNL post-doc community running and being elected to the ORNL post-doc association's executive committee and serving as the vice-chair of its research committee. In these roles I planned research seminars, tours of user facilities, developed a new event teaching post-docs how to engage the public in their research, acquired increased funding for the association from ORNL management as well as organized and ran the annual post-doc research symposium. As such I have the independence and leadership skills to design and run and independent research program.

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