Research Statement

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1 INTRODUCTION

Computers help to manage the operation of society or enterprise, provide a tool to process large amounts of data, and help individuals to entertain themselves or connect with people around the world. As the data size gets bigger and the process/operation become increasingly complicated, faster computing is desired. Quantum computation which manipulates quantum bits (qubits) using quantum phenomena such as quantum superposition and entanglement provides much faster computing environment. n number of qubits can process 2^n different states simultaneously while the classical computers process n states with n number of bits. The development of quantum computation has been initiated and in its early stage [1-3]. It is important to identify prospective quantum computing materials and understand quantum phenomena within the materials.

I propose to establish a research program to create and investigate novel quantum ground states and the resulting properties of potential materials for quantum computation. Examples of such materials are magnets with spin liquid state or superconducting state, in which quantum entanglement plays a major role. My research takes advantage of my expertise on crystal growth and full characterization of samples. I will mainly use modern synchrotron-based resonant x-ray scattering (REXS) technique, coherent x-ray imaging techniques, and elastic/inelastic neutron scattering techniques to study the structural/magnetic ground states and quantum entangled states. I will also use complementary in-house x-ray diffraction, transport, and magnetization measurement techniques. These will aid a thorough understanding of the quantum phenomena occurring in the sample.

In the following, I will describe some **representative** projects and their timelines that I would lead as an Assistant Professor at Texas Tech University.

2 RESEARCH TOPICS

RESEARCH TOPIC #1. FRACTIONALIZED SPIN EXCITATIONS IN MAGNETS.

A quantum spin liquid (QSL) is a state in which quantum fluctuations prevent the spins from entering a long-range ordered state at zero temperature. The QSL state cannot be described by the broken symmetries associated with conventional ground states, and the spins become fractionalized and entangled [4, 5]. A high degree of entanglement is one of the defining properties of QSLs. *QSLs can exhibit topological states with fractionalized excitations, which may be utilized in advanced quantum computing applications* [6]. Spinons can freely propagate in the large phase space and thus spinon excitations form a continuum. Inelastic neutron scattering (INS) techniques have successfully revealed an excitation continuum in QSL candidate materials, considered to be the most striking direct evidence of the fractionalized excitations [5, 7-11]. For example, in *Spin-Liquid-Like State in the Triangular Lattice Antiferromagnet $TbInO_3$ (in preparation for publication)*, I observed broad magnetic excitations centered around the triangular Brillouin zone (BZ) boundary which appears gapless and dispersion-less in energy spaces in a newly discovered quantum spin liquid candidates $TbInO_3$ (Fig. 1). This observation is consistent with the excitation continuum, the signature of the fractionalized excitations.

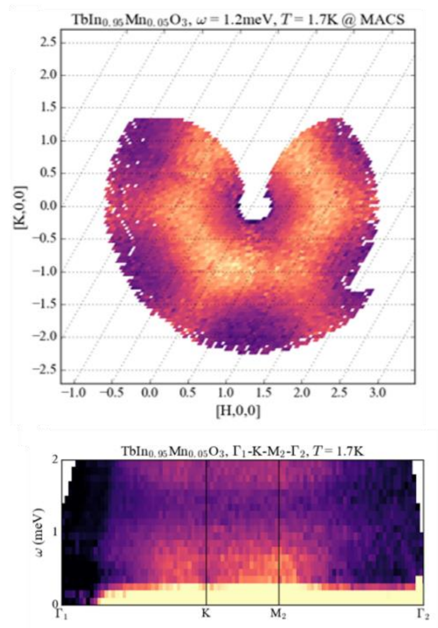

Figure 1. (Top) Broad spectrum in momentum space. Signals are broadly centered around the triangular BZ boundary. (Bottom) Excitation continuum in energy space along the high symmetry directions.

The fractionalized excitations in the QSL states are realized in frustrated antiferromagnets, such as magnets with Kagome lattices, honeycomb lattices, and triangular lattices, but only a handful of QSL candidates have been reported. Much theoretical effort has been made to model the QSL states and resulted in various possible QSL states possessing different properties, for instance, some models showing gapped excitations and others with gapless excitations [12-17]. Theories can also vary noticeably depending on the lattices, and many studies have focused on Kagome and honeycomb lattices with spin-1/2 moments where the quantum fluctuation is strong. Recently, spin-1 antiferromagnet [18, 19] was claimed to show a possible QSL behavior and because of significantly reduced quantum fluctuations with S = 1 in this material, it attracts many scientists in the field and is under special scrutiny. This magnet is based on a triangular lattice which is also a rare example of QSL although the first QSL state was found in the triangular organic antiferromagnet [20]. Therefore, it is important to investigate the true ground state of such materials not only to establish a groundwork for QSL in S > 1/2 magnetic systems but also to reconcile the discrepancies in theories and facilitate a thorough understanding of the QSL state.

I propose to study the spin excitations in $Ba_3MSb_2O_9$ (M = Ni, Cu). These materials crystallize in $P6_3mc$ perovskite structures, and M layers form a triangular lattice. These compounds exhibit strong antiferromagnetic interactions with large negative Curie-Weiss temperatures [18, 21] but Ni^{2+} and Cu^{2+} spins (S > 1/2) do not order magnetically down to very low temperatures (T < 1K) [18, 19, 21, 22]. Several studies attempted to investigate the spin-liquid-like state in these materials with polycrystalline samples. There has been yet no direct probe of the quantum spin liquid state – the fractionalized excitations.

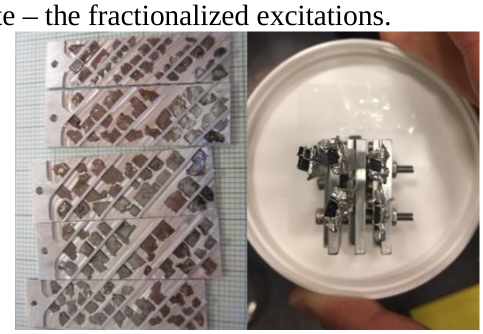

Figure 2. Photos of coaligned single crystals of (left) $CaFe_2As_2$ and (right) $TbInO_3$. I was involved in aligning and assembling of crystals.

We will grow single crystals of $Ba_3MSb_2O_9$ (M = Ni, Cu) compounds. Transport and magnetic properties will be studied on single crystals. Expected single crystals are small for neutron experiments. We will co-align several samples to make a set of well-oriented single crystals, totaling approximately 500 mg, which is an adequate amount for inelastic neutron scattering (INS) [see Fig.2 for examples of co-aligned crystals]. Then we will perform the INS measurements on single crystals with or without applied magnetic fields, or with polarized neutrons. Our project should elucidate the nature of the QSL state in these compounds and could potentially unravel a deep understanding of the QSL state for a general spin, *S*.

RESEARCH TOPIC #2. QUANTUM SPIN DYNAMICS AT A QUANTUM CRITICAL POINT.

Superconducting quantum qubits are one of the early realizations of quantum computing. For the advancement of superconducting quantum computation, the superconductivity should be understood primarily. In antiferromagnets, as the magnetic transition is suppressed to zero temperature ($T_N \rightarrow 0$), a zero-temperature instability may exist between the antiferromagnetic (AFM) and paramagnetic states. This is called an antiferromagnetic quantum critical point (AFM QCP) and accompanied by a state of quantum criticality: a quantum superposition of order and disorder. This quantum fluctuation can lead an exotic ground state such as the superconductivity (SC) in the famous cuprate, heavy-fermions, and Fe-based superconductors. Because the quantum criticality not only exists at $T = 0$ limit but also persists at finite temperatures as shown in Fig. 3, it has been argued that the quantum fluctuations at the AFM QCP are the key for understanding the superconductivity [23–30]. However, studying quantum critical behavior in antiferromagnetic superconductors is challenging because of the emergence of SC: states become inaccessible via various experimental probes. Inelastic neutron scattering can be a good tool to investigate the quantum criticality near the AFM QCP.

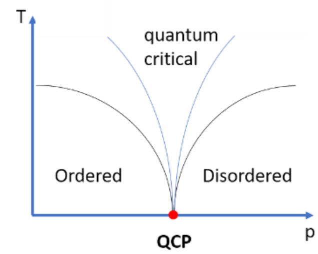

Figure 3. Schematic T-p phase diagram showing QCP and a region of quantum criticality. T is temperature and p indicates external parameters, such as pressure or doping.

In *Spin dynamics near a putative antiferromagnetic quantum critical point in Cu-substituted BaFe₂As₂ and its relation to high-temperature superconductivity (Phy. Rev. B 92, 214404, 2015)*, I identified a putative AFM QCP by performing systematic neutron diffraction measurements on $Ba(Fe_{1-x}Cu_x)_2As_2$ and inelastic neutron scattering measurements on non-superconducting $Ba(Fe_{1-x}Cu_x)_2As_2$ with $x = 0.043$. We found that the dynamic spin-spin correlation length increases rapidly, $\xi \sim T^{-0.32(5)}$ as $T \rightarrow 0$, and we observed the ω/T scaling behavior, the hallmark of quantum criticality at the AFM QCP [23–30] (see Fig. 4). The theory for magnetic quantum phase transitions [31–34] for spin-density wave transitions in a 3D system predicts the scaling as $\xi \sim T^{-3/4}$ and no ω/T scaling of spin fluctuation spectra at/near an AFM QCP. In contrast, in a 2D system, the theory predicts temperature- and energy-independent dynamic correlation length and ω/T scaling at/near an AFM QCP. However, none of the models has been resolved by experimental observation in the Fe-based superconductors.

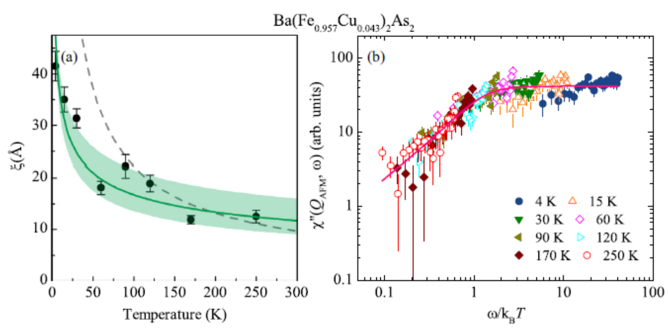

Figure 4. (a) The dynamic spin-spin correlation length. The solid line demonstrates the best fit and the shaded area indicates the error range of the fit. The dashed line shows the theoretical prediction. (b) The universal ω/T scaling plot. The solid line describes the best fit.

I propose to tackle this problem with a smart material design. I plan to study the spin dynamics at/near the AFM QCP in the Fe-based superconductors, especially in Co and Cu co-doped $Ba(Fe_{1-x-y}Co_xCu_y)_2As_2$ compounds. The under-doped $Ba(Fe_{1-x}Co_x)_2As_2$ compounds show 3D spin dynamics and optimal/over-doped $Ba(Fe_{1-x}Co_x)_2As_2$ exhibit quasi-2D spin fluctuations. As Cu is additionally introduced in already-superconducting (under-doped or optimal-doped) $Ba(Fe_{1-x}Co_x)_2As_2$ compounds, we can tune the system to either be 2D or 3D and drive the system close to the AFM QCP. We will perform a systematic inelastic neutron scattering on these $Ba(Fe_{1-x-y}Co_xCu_y)_2As_2$ compounds. I believe that the proposed study will reveal a deep connection between the quantum criticality and the superconductivity, which will improve our understanding of superconductors and superconducting qubits.

RESEARCH TOPIC #3. ANTIFERROMAGNETIC DOMAIN FORMATION IN EXOTIC STATES.

The first step in the application of antiferromagnetic QSL or SC materials is a fabrication of qubits. Since those are magnetic materials, knowledge of antiferromagnetic domain formation and its dynamics is required. It is widely accepted that antiferromagnetism can compete with such exotic ground state (QSL or SC) of the matter. For instance, in *Unconventional pairing in the iron arsenide superconductors (Phys. Rev. B 81, 140501(R), 2010)*, we showed that the antiferromagnetism and superconductivity compete to each other, and antiferromagnetic ordering survives in the SC state in the same volume of the sample. In *Imaging antiferromagnetic antiphase domain boundaries using magnetic Bragg diffraction phase contrast (Nat. comm. accepted, 2018)*, we demonstrated a new x-ray imaging technique developed by Valery Kiryukhin at Rutgers University, showing the antiferromagnetic antiphase domain boundaries in $Fe_2Mo_3O_8$ compound (See Fig.5). Using this new technique, we can directly map AFM domains and domain walls of many types, such as collinear or cycloidal AFM, without any numerical reconstructions.

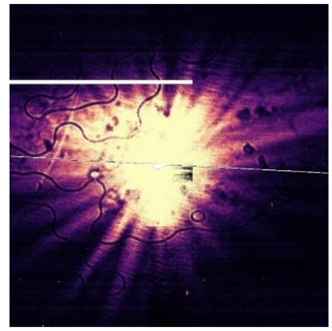

Figure 5. Single-exposure image at the magnetic (0,0,1). Black wavy lines are images of the antiphase domain boundaries.

I propose to study the antiferromagnetic domains and domain walls in the antiferromagnets with exotic ground states using the resonant x-ray magnetic diffraction imaging techniques. I will start this project with well-known antiferromagnetic superconductors $Ba(Fe_{1-x}Co_x)_2As_2$ compounds. They undergo a structural transition from a high-temperature tetragonal to a low-temperature orthorhombic structure, which introduces 4 different orthorhombic twin domains. The size of the twin domains can be as small as the current limit of the imaging technique (an order of microns). This problem can be overcome by applying tensile or compressive stress. It is known that the domain population can be greatly modified so that one can get more than 80% of one domain over other domains with external stress [35, 36]. The schematic of the experiment is shown in Fig. 6. We will apply compressive stress and take snapshots of AFM domain boundaries across the superconducting transition temperature, which will reveal the interconnection between AFM domains and superconductivity. After successful attempts, we will expand this project to other magnetic materials with exotic ground states.

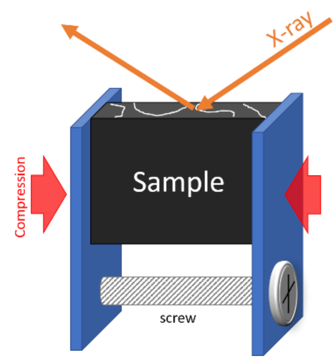

Figure 6. Schematic of resonant x-ray diffraction imaging experiment under compressive stress.

3 TIMELINE OF THE RESEARCH PROJECT

For the early years of my research program, we will get samples from many collaborators, including, but not limited to, Sang-Wook Cheong (Rutgers University), Paul Canfield (Iowa State University), and Robert Birgeneau (UC Berkeley). In addition, I have single crystal samples of various Fe-based superconductors that I grew during my postdoc with Robert Birgeneau, which are readily accessible.

Neutron scattering experiments will be performed at various large-scale facilities such as High-Flux Isotope Reactor (Oak Ridge, TN), Spallation Neutron Source (Oak Ridge, TN), NIST center for Neutron Science (Gaithersburg, MD), and the University of Missouri Research Reactor (Columbia, MO).

X-ray scattering/imaging experiments will be done at facilities like Advanced Photon Source (Argonne, IL) and National Synchrotron Light Source 2 (Brookhaven, NY).

My long-term goal is to set up laboratories for sample growths and characterizations, adding a new invaluable research powerhouse to the current efforts at Texas Tech University. Sample growth will include synthesizing oxide materials as well as intermetallic materials, using the floating zone growth technique and the high-temperature solution growth technique. We will have in-house characterization tools to measure physical properties (PPMS), SQUID magnetometers (MPMS), and crystal structures (high-power XRD machine).

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