Introduction

Quantum magnetic fluctuations stand at the heart of modern condensed matter physics. Their ubiquity belies a complexity and variety that couples together concepts from a vast array of topics, from the coherence of superfluidity and superconductivity (*1, 2, 3*) to the massively entangled disorder of quantum spin liquids (*4*), and sometimes leaving the bounds of condensed matter physics entirely (*5*). Because of the importance of quantum magnetism, there is a particular emphasis on finding and understanding magnetic ground states where quantum fluctuations are so strong that they overcome magnetic order, and on understanding the phase transitions from such states to more conventional ones (*6*). **The research conducted in my laboratory will go directly into the growth of materials and experiments to better understand the types of states that are possible when such fluctuations are present in a metallic host. My emphasis will be on exploring these states and understanding the phase transitions that lead to them in rare-earth based R_2T_2X (R = rare earth element, T = transition metal element, X = main group element) quantum magnets, where quantum magnetic fluctuations are brought on through geometric frustration, electronic correlations, and dimensionality.**

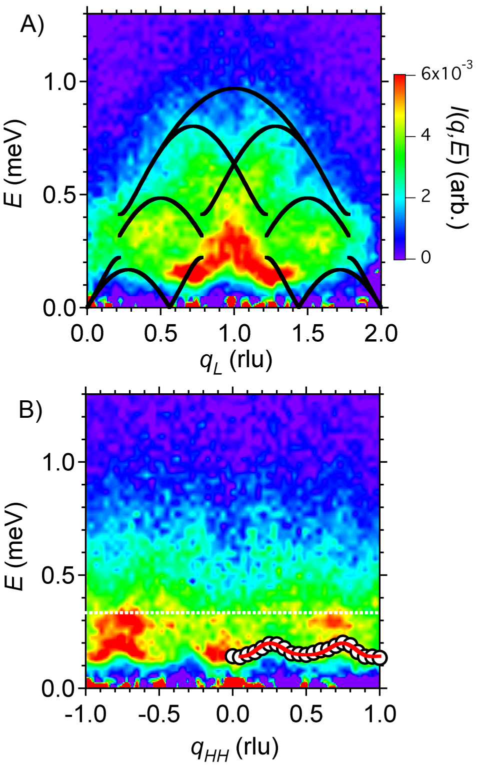

Fig. 1: (A) Neutron scattering spectrum of Yb_2Pt_2Pb for momentum along the 1-dimensional c-axis at $B = 1.5$ T. Any $B > 0.5$ T creates a spinon fermi surface, modifying the allowed 2 spinon excitations (black lines). A confined 2 spinon bound state is observed at the zone center. (B) Spectrum at $B = 1.5$ T with momentum perpendicular to the c-axis. Along with the quantum continuum, we observe a longitudinally polarized dispersing excitation associated with the bound state (points and fit). Both measured at $T = 0.1$ K. From (*7*).

We will use established techniques to grow these crystals in house, making crystal growth a central thrust my research program. With these samples, I will use the neutron scattering techniques that I have learned as a student and postdoc to explore the energy and momentum dependencies of these fluctuations. Neutron scattering is a unique spectroscopic tool that directly probes modulated magnetization on energy scales ranging from 0.1 to 1000 meV (*8*), making it ideal for studying dynamic magnetic phenomena in bulk materials such as those that will be grown in my lab. These experiments will be done at neutron scattering facilities in the US and around the world where I have cultivated expertise and research collaborations throughout my scientific career.

The major thrust of research in my laboratory will focus on the novel quantum magnetic ground states in the the R_2T_2X materials. This family of materials is extremely diverse (*9, 10, 11, 12, 13*), with the most famous example being Yb_2Pt_2Pb (*14*), a large moment Ising antiferromagnet where the fundamental low energy magnetic excitations are astonishingly, spinons on one dimensional chains (*15, 7*). There are a vast number of avenues that can be explored in this field by tuning the quantum magnetic properties through crystal growth techniques and the use of external parameters such as temperature, magnetic field, and pressure.

Prior Research

As a postdoctoral researcher at Brookhaven National Laboratory (via Stony Brook University), Texas A&M University, and now at the Stewart Blusson Quantum Matter Institute at the University of British Columbia, I have focused on crystal growth and neutron scattering with two main materials, Yb_2Pt_2Pb and the quantum critical magnet YFe_2Al_{10}. Yb_2Pt_2Pb is an antiferromagnetic metal with large, orbitally dominated Yb magnetic moments at low temperatures (*16, 17*). We have unambiguously demonstrated through neutron scattering that Yb_2Pt_2Pb – despite the seemingly classical magnetic moments of the Yb^{3+} ions – is a one-dimensional quantum spin chain material (*15*). I have made measurements of the low energy magnetic excitation continuum in Yb_2Pt_2Pb on high resolution neutron spectrometers to study how an applied magnetic field creates spinon bound states and a longitudinal interchain mode, condensed matter analogues to quantum chromodynamics and the Higgs Boson (*5, 7*). In the structurally related but more two dimensional compound Yb_2Si_2Al, we have demonstrated the presence of a quantum phase transition at $T = 0$ and explicitly shown the mixed Yb-valence character of this material, hinting at the connections between dimensionality, magnetic fluctuations, and electronic correlations in determining novel ground states (*9*).

A member of a different material family entirely, YFe_2Al_{10}, is a two-dimensional correlated electron magnet that naturally sits very near a quantum critical point (*19*). I have measured the quantum critical (QC) fluctuations directly using neutron spectroscopy, a very challenging project. We have demonstrated the separability of the energy and momentum of these fluctuations, painting a remarkable picture of quantum criticality in YFe_2Al_{10} where the fluctuations are completely local with no spatial correlations (*18*). The quantum critical dynamics are entirely temporal and very likely a manifestation of a topological phase transition associated with the formation of magnetic moments and potentially a realization of the two dimensional XY model (*20*).

As a graduate student, my work focused on crystal growth and experiments on the magnetically mediated superconductor UPt_3 (*21*). Using the vertical floating zone technique, I grew some of the highest quality UPt_3 crystals ever produced. Using small angle neutron scattering (SANS) from the vortex lattice (VL) in the superconducting mixed state, I probed the nodal structure of the superconducting gap (*22*) and also uncovered an unknown VL metastability in UPt_3, which has interesting consequences for the Fermi surface and weak antiferromagnetic order, the subject of ongoing work in my former group. My polarized neutron diffraction experiments measured the electronic spin susceptibility in UPt_3 across the superconducting transition temperature (*23, 24*). These experiments confirm that the spin susceptibility of UPt_3 does not change across T_c, indicating that the superconducting pair wave function is an equal spin paired triplet state. My triple axis spectroscopy measurements demonstrated that the antiferromagnetic order in UPt_3 is not nearly as well understood as was once thought and that quasi elastic magnetic fluctuations may be intrinsic in the clean limit rather than static order. This study is also ongoing and has important theoretical consequences for symmetry breaking. My high quality crystals were used by a wide variety of collaborators, most notably for Polar Kerr effect measurements which demonstrate definitively that the low temperature superconducting phase breaks time reversal symmetry (*25*) and for Josephson tunneling, which show that the same phase is in an odd parity chiral state (*26*). Finally, I will add that in my group as a grad student, I participated in the experiments and scientific discussions surrounding many ultra low temperature measurements on superfluid ^3He, the paradigm unconventionally paired system where magnetic fluctuations provide the pairing glue. The most notable of these experiments used silica aerogel to provide correlated defect scattering, stabilizing different superfluid phases (*27, 28*).

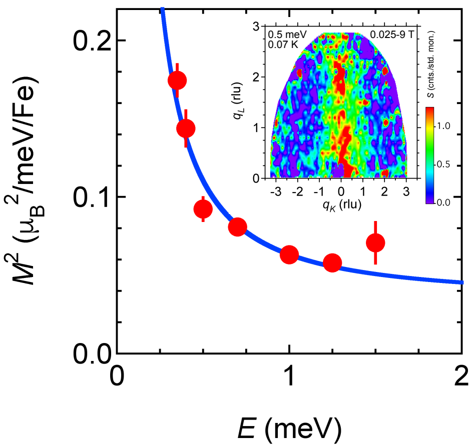

Fig. 2: (A) Power law divergence of the energy dependence of the fluctuating magnetization in YFe_2Al_{10} measured by neutron scattering at $T = 0.1$ K and $B = 0.025$ T, a hallmark of quantum criticality. (Inset) Color plot showing the difference of the momentum dependence of the scattering measured at $B = 0$ and 9 T and E=0.35 meV, indicating local magnetic fluctuations. Both from (*18*).

Emergent Quantum Fluctuations in 221 materials

My current research interests focus on novel quantum magnetic fluctuations. This could perhaps refer to well understood fractionalized quantum excitations in a system that might be naively projected to host quasi-classical fluctuations; it might also refer to particularly exotic quantum magnetic excitations that emerge from a host with more straightforward quantum magnetic properties; or it could refer to fluctuations that come from a host that has very little magnetic character in the first place. Yb_2Pt_2Pb provides a perfect example of the former two. As a compound with large Yb^{3+} moments, one would naively expect quasi-classical excitations rather than the spinons that actually dominate the low energy magnetic properties. And from those relatively straightforward spinons, we clearly see longitudinal excitations of the antiferromagnetic order parameter, akin to the "amplitude-ons" seen in superconducting condensates, other two dimensional quantum magnets (*29*), and even the Higgs boson (*5*). The final scenario above is perfectly described by the quantum critical fluctuations seen in YFe_2Al_{10}, where the novel magnetic dynamics are brought on entirely through electronic correlations. **In my laboratory, we will study all such quantum magnetism, the unusual non Fermi liquid phases where these sorts of dynamics are typically found, and the transitions that link these phases – which often map onto the most important theoretical constructs in physics today, such as topological phase transitions (*30*), one dimensional Luttinger liquid physics (*31*), the two dimensional XY model (*20*), *etc.* – to more trivial and better understood ones**.

There is a considerable, largely unexplored phase space in the R_2T_2X materials. Restricting this discussion to just the simplest cases – compounds where the rare earth element is Yb or Ce – there are more than 29 known materials with closely related crystal structures. These structures are topologically equivalent to the Shastry-Sutherland lattice (SSL) (*32*), with the rare earth elements forming orthogonal pairs in two dimensional layers (Fig. 3A). This lattice is geometrically frustrated for magnetic interactions and has been studied extensively both experimentally and theoretically (*33, 34*). For most of these R_2T_2X materials, very little is known, often only crystal structure and incomplete magnetic measurements. From what is known, these metallic materials span a wide range of compositions and therefore electronic properties, giving an extremely varied physical terrain to explore. Many families of materials are studied due to the wide range of physics observed on non-trivial crystal structures; the rare-earth 227 pyrochlore oxides for instance, feature members that

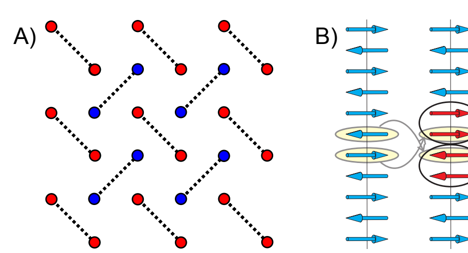

Fig. 3: (A) Illustration of the Shastry-Sutherland lattice, with orthogonal pairs of planar atoms. (B) Illustration of how electron exchange on an orbital AFM chain leads to the creation of 2 spinons. From (*15*).

host phases with strong long range order to spin ice phases and spin liquid candidates (*35*). That the crystal growth of the R_2T_2X compounds is rather much more simple is a great advantage in comparison, as is the fact that the R_2T_2X crystals are good, robust metals, lending them well to measurements under pressure and introducing free charge and therefore electronic correlations as an important degree of freedom for the entire series. I will add that the SSL problem is well established theoretically, making this an excellent topic for theory collaboration, particularly in the context of quantum spin liquids, an active area of current research in many materials (*36*).

My research group will use established techniques to grow R_2T_2X crystals. Typically, they grow readily from flux, which I have relied on heavily on as a postdoc. Other standard techniques, such as arc melting, solid state reactions, and perhaps even floating zone refining will be used when appropriate. Several of the stoichiometric R_2T_2X materials naturally host the quantum excitations that I am interested in. Exploiting the vast array of compounds in this family by chemical doping – either through inducing chemical pressure effects or altering the electronic correlations by changing the free charge – is a straightforward and obvious direction to pursue. Both of these routes will fundamentally alter the magnetic ground states themselves and in some cases couple these states to the conduction electrons. There are many directions that can be simultaneously pursued in this varied landscape and I envision several distinct but complimentary PhD thesis projects ongoing at all times. **Although crystal growth from flux can be a labor intensive process, it is luckily a fantastic area for undergraduates to participate in research.** The skills needed are well within a motivated undergraduate's capabilities and I will make full use of undergraduate help in our efforts. As with any crystal growth lab, sample characterization is essential. **We will leverage university user facilities for X-ray diffraction, scanning electron microscopy, and any other available tools for structural and compositional information.** We will utilize standard techniques in our own laboratory such as heat capacity, DC and AC resistivity, and AC and DC magnetic susceptibility, and magnetization to fully characterize our materials.

When interesting materials are uncovered, in addition to our in house characterization, we will use a two pronged approach to understand the quantum magnetic properties. **We will use neutron scattering to study the momentum and energy dependence of the magnetic fluctuations directly.** Inelastic neutron scattering is a technique that has been used to measure the dispersion of quantum magnetic fluctuations in an array of materials too numerous to list here. Neutron diffraction is also a powerful tool to understand underlying magnetic order and my research group will exploit this technique when appropriate. These experiments will be done at neutron scattering facilities in the United States: mainly the Spallation Neutron Source at Oak Ridge National Laboratory in Tennessee, the Center For Neutron Research at the National Institute for Standards and Technology in Maryland, and the High Flux Isotope Reactor, also at Oak Ridge. Additionally, I have extensive experience in neutron scattering facilities all over the world and we will use international neutron sources with instruments that have exceptional capabilities when appropriate, such as the cold neutron spectrometers at the ISIS neutron source at Rutherford Appleton Laboratory in England.

The second prong will be to **tune these fluctuations through *in situ* application of stress and strain to study their phase transitions.** Small piezo electric actuators allow one to continuously apply several GPa of pressure, both positively and negatively, on millimeter sized samples at low temperatures (*37*) with devices that are now commercially available. This induces a considerable amount of strain in typical metals such as the R_2T_2X compounds, which will substantially modify their electronic structure and therefore changing the quantum magnetic behavior, giving insight into the fluctuations and the correlated electronic states hosting them. The technique lends itself to a variety of experimental probes – resistivity, AC susceptibility, and even NMR (*38*) are known to work well – and the experimental geometry can be easily integrated with a hall sensor magnetometer (Fig. 4A). It is a long term, but very challenging goal to integrate continuous tuning of pressure into neutron scattering as well.

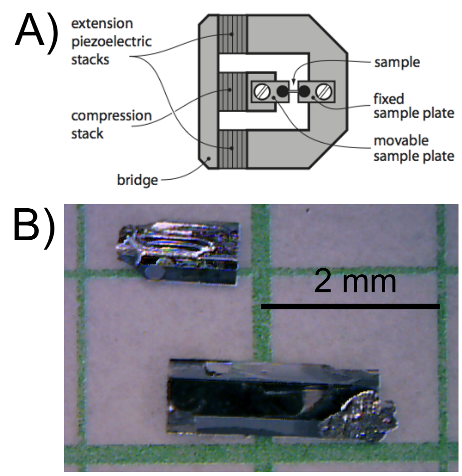

Fig. 4: (A) Sketch of working concept for commercial devices for in situ tuning large applied stresses and strains to a single crystal sample. From (*37*). (B) Small Ce_2Ge_2Mg single crystals grown from flux.

There are several materials that my group can start measuring immediately with these techniques. Yb_2Pt_2Pb is the most obvious candidate for strain experiments, to strengthen or weaken the interactions leading to the one dimensional fluctuations with positive or negative force. Ce_2Ge_2Mg (Fig. 4B) is almost certainly also a one dimensional spin chain material with slightly different Hamiltonian than Yb_2Pt_2Pb, although neutron scattering measurements do not yet exist to confirm this. Ce_2Ge_2Mg has the advantage that the antiferromagnetic ordering temperature is more accessible (10 K, rather than 2 K in the case of Yb_2Pt_2Pb) making it a simpler job to study the influence of antiferromagnetic order on the fluctuations and look for emergent excitations brought on my quasiparticle confinement. There are many mixed valence materials in this family that could be induced to have larger, fluctuating moments, such as Yb_2Si_2Al which is driven to a relatively large local moment with Mg doping (*39*), and in which I have recently uncovered a magnetic resonance with neutron scattering. And I am eager to begin with Ce_2Cu_2In, Ce_2Au_2In (*13*) and the $Yb_2Pd_2In_{1-x}Sn_x$ series (*11*), in the context of both neutron scattering and strain to name just a few places to start. And these projects do not include exploring members of this series with other rare-earth elements, $U_2Pd_2In_{1-x}Sn_x$ among other examples (*10*), which introduce new and nearly completely unexplored magnetic and electronic possibilities.

Further Experiments

It's always a good idea to have a variety of research alternatives. In addition to the experiments proposed above, I have several experimental ideas that can be started quickly in the interest of proceeding with experiments immediately at the beginning of my appointment.

Antiferromagnetic Order in UPt_3

Antiferromagnetic order in UPt_3 at $T_N = 5$ K has remained mysterious for more than 25 years due mainly to the small size of the ordered moment ($\sim 0.02\ \mu_B$/U atom) and the fact that it is only reliably observable in scattering experiments (*21*). From my work as a graduate student, there is evidence to suggest that what is thought to be bulk static order is in reality, a completely dynamic phenomena in the clean limit. It is certain, however, that doping the Pt site with small amounts of Pd greatly increases the size of the ordered moment, making it observable without neutron scattering (*21*). I will use the strain technique discussed above to mimic the chemical pressure effects of Pd doping, allowing for detailed, continuous measurements on the emergence of the ordered moment from the fluctuations, with the aim being to understand the phase transition between these ordered phases.

Low dimensional fluctuations in other compounds

Although I will begin research on the R_2T_2X materials immediately, there is no need to restrict studies on low dimensional fluctuations to these materials. There are a wide variety of materials that could be adapted for tunable strain experiments. The rare earth perovskite material $YbAlO_3$ is in many ways the insulating twin to Yb_2Pt_2Pb (*40*). As $YbAlO_3$ is part of a larger "1-1-3" series of materials with unexpected quantum dynamics in several members (*41*), experiments under uniaxial pressure would be illuminating. An even more fundamental study would be to examine the limits of quantum magnetism as these insulators are doped into metals. Looking a bit to the past, the rare earth tetraboride compounds are largely 2-dimensional, with the rare earth elements arranged in the SSL in 2-D planes (see (*42*) for summary). There are many magnetic phases in these materials due to strong frustration, but the dynamics for much of the series are thought to be described by an effective $S = 1/2$ theory and an Ising model, similar in many ways to Yb_2Pt_2Pb. With more "classical" magnetic frustration dominating much of the series, an interesting problem would be to explore the limits of quantum magnetism through external tuning. Since many in this family grow readily from flux and would seem to be ripe for study in my laboratory.

Supply of samples for collaborators

Throughout my career I have been firmly involved in supplying samples for scientific collaborations and developing and participating in collaborations through our crystal growth operation will play a key part in my research program. Broadening the experimental expertise on a problem through collaboration is critical and I have personal connections with experts in NMR, scanning squid techniques, scanning tunneling microscopy/spectroscopy, various Josephson junction techniques, thermal conductivity, and a host of X-ray scattering techniques just to name a few. Among the notable samples I have prepared are crystals of UPt_3 that were used to map the nodal structure of the superconducting gap by directional Josephson tunneling (*26*), and crystals of UPt_3 used in measurements of the polar Kerr effect that show time-reversal symmetry breaking in the low temperature superconducting B-phase, while the A-phase which marks the onset of superconductivity $T = T_c$ is time reversal invariant (*25*). With just Yb_2Pt_2Pb, I have recently sent crystals to collaborators in China for measurements of thermal expansion and the de Hass van Alphen effect and to Germany for optical spectroscopy and Mössbauer spectroscopy, to name only a few examples, all of which are on going collaborations. Collaborations help to uncover new and unexpected directions for our own studies and I will happily work to cultivate experimental collaboration both inside and outside of my institution.

More challenging crystal growth

As a graduate student, I produced some of the highest quality UPt_3 crystals in the world using the floating zone technique. There are many interesting physical problems that could be more readily studied with similar large, high quality crystals. I have a longer term goal to make such crystals of many materials – other magnetically mediated superconductors in particular – to study the coupling of quantum magnetic fluctuations to conduction electrons via neutron scattering. To make definitive conclusions, such experiments require very large and very high quality samples, which are extremely challenging to make. It would require an investment in both time and money for developing refining capabilities, even with the use of external growth facilities with extraordinary capabilities such as PARADIGM at Johns Hopkins University (https://occamy.chemistry.jhu.edu/tour/paradim/index.php), with no guarantee of success. As a professor, I will leverage my extensive contacts with other crystal growers as well as the knowledge base built in my own lab, with an eye towards undertaking these challenging types of growth and refinement in the future.

Materials and experimental directions that are as yet unknown

Inevitably in any crystal growth lab, interesting and unforeseen directions for study will be uncovered in the course of growing crystals. My lab will not be limited to narrow confines but will rather be open to and excited about new directions as they appear in the course of developing our research program. The flexibility of our growth techniques, in house measurement capabilities, and spectroscopic techniques leave no doubt that through crystal growth, we will find new, unexpected, intellectually stimulating, and productive avenues for study. Further, I am excited about the prospect of developing expertise in new experimental techniques that complement my existing experimental tools – resonant inelastic x-ray scattering, for example – as my group builds an institutional knowledge base and gains new insight into our materials.

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